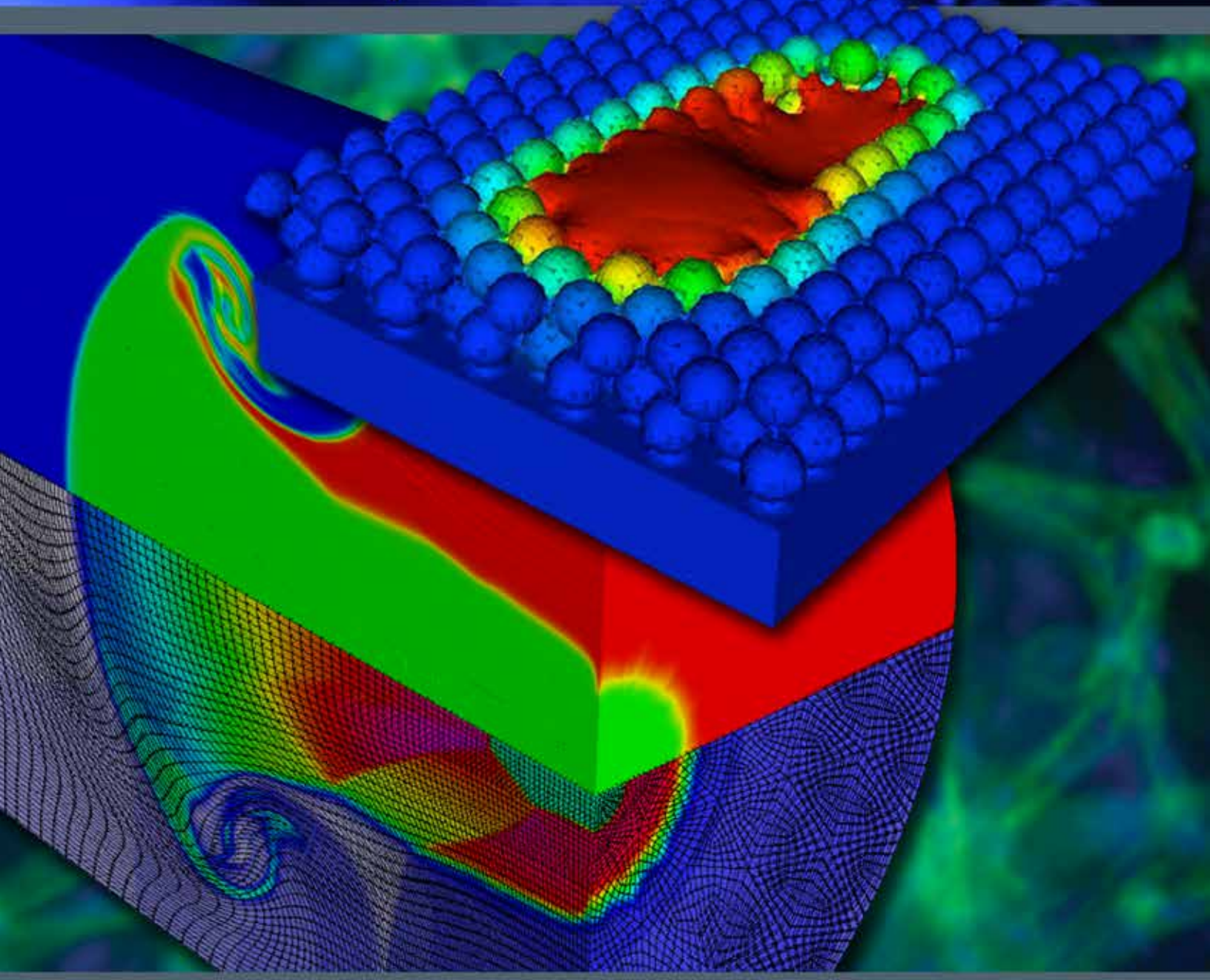
Laboratory Directed Research and Development

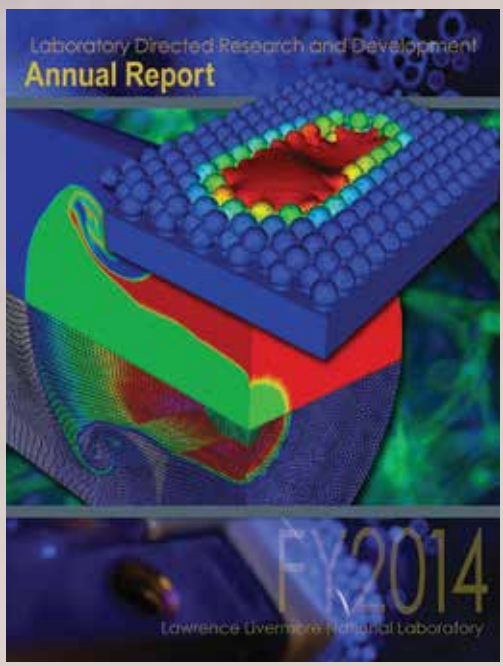
Annual Report



FY2014

Lawrence Livermore National Laboratory

About the Cover



The primary emphasis of the LDRD Program is high-risk, high-reward research that creates innovative technical solutions for some of our nation's most difficult challenges in national and energy security. The cover shows several images from LDRD projects devoted to stockpile stewardship, advanced materials and manufacturing, high-performance computing, energy and climate, and bioscience and biosecurity. The large image in the middle is a simulation of multiple simultaneous physical phenomena of multiple materials depicted with software used to study the behavior of shocked materials and resulting fluid dynamics (LDRD project 14-SI-002). This capability underpins both energy and national security research, as well as computational foundations for a new breed of multiphysics codes being developed for emerging supercomputer architectures. The smaller simulation on top depicts a molten pool from the laser melting of small stainless-steel powder particles barely visible to the naked eye, for a project examining accelerated certification of additively manufactured metals (13-SI-002). The middle background across the front and back covers shows printed human microscopic vessels used to distribute oxygen and nutrients through several layers of tissue, enabling models for measuring the response to unknown chemical and biological agents (14-ERD-005). Finally, along the top and bottom are images of novel fracture-material capsules for use in hydraulic fracturing for natural gas and geothermal production that increase efficiency while minimizing environmental impacts (13-ERD-029).

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The Laboratory Directed Research and Development Program extends its sincere appreciation to the principal investigators of fiscal year 2014 projects for providing the content of the annual report. The program also thanks the following members of the Laboratory Directed Research and Development Office for their many contributions to this publication: Barbara Jackson, administrator; Steve McNamara, computer specialist; Kathy Villela, resource manager; and Kristen Croteau, business manager.

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Science and Technology on a Mission

The Laboratory Directed Research and Development (LDRD) Program was conceived as a bold initiative to ensure that we maintain our scientific and technical vitality. Scientific and technical risk are essential attributes of an LDRD portfolio that expands Lawrence Livermore National Laboratory's capability to serve our national security missions. Our ongoing investments in LDRD continue to deliver long-term rewards for the Laboratory and the nation, supporting the full spectrum of national security interests encompassed by the missions of the Laboratory, the Department of Energy, and the National Nuclear Security Administration. Many of Livermore's programs trace their roots to research thrusts that began under LDRD sponsorship. By keeping the Laboratory at the forefront of research, maintaining and enhancing our core competencies, building new capabilities, and reaching beyond the immediate challenges toward the future, the LDRD Program enables us to pursue cutting-edge science and technology and deliver solutions for the nation's most challenging security issues.

The LDRD Program is the largest single source of internal investment in our future. For fiscal year 2014, the LDRD Program supported 147 projects with an allocation of $78.2M. These projects were selected through an extensive peer-review process to ensure the highest scientific quality and mission relevance. The LDRD projects are consistent with the Laboratory's strategic plan and impact the Laboratory in four distinct ways:

- Attracting and retaining the best and the brightest workforce by conducting world-class science, technology, and engineering
- Maintaining our competency in those core areas where our missions mandate that we must be the best, and evolving these competencies as our missions change—these core competency areas are consistent with the science, technology, and engineering foundations as defined in the Laboratory's strategic plan
- Developing capabilities in focus areas, guided by the strategic plan, where we have chosen to build or expand our expertise to meet our strategic vision
- Looking beyond the immediate programs to future national security challenges

The LDRD Program is a success story. Our projects continue to win national recognition for excellence through prestigious awards, papers published in peer-reviewed journals, and patents granted. With its reputation for sponsoring innovative projects, the LDRD Program is not only a major vehicle for attracting and retaining the best and the brightest technical staff, but for establishing collaborations with universities, industry, and other scientific and research institutions. By keeping the Laboratory at the forefront of science and technology, the LDRD Program enables us to meet our mission challenges, especially those of national security in an evolving global context.

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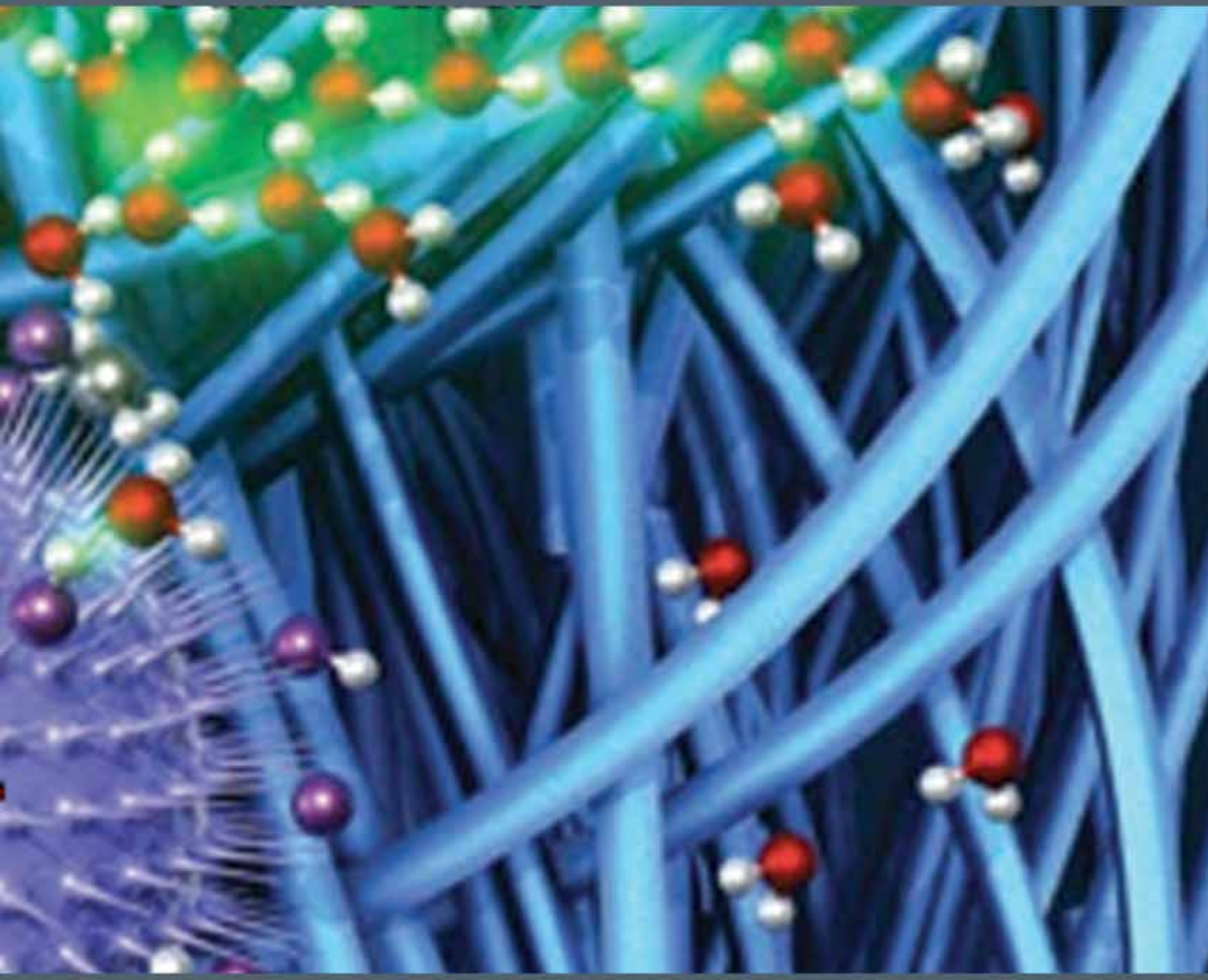


FY2014

Lawrence Livermore National Laboratory

About

Lawrence Livermore National Laboratory

A premier applied-science laboratory, Lawrence Livermore National Laboratory (LLNL) has a mission of strengthening the United States' security by developing world-class science, technology, and engineering.

Lawrence Livermore is renowned for

- Physicists, chemists, biologists, engineers, computer scientists, and other researchers working together in multidisciplinary teams to achieve technical innovations and scientific breakthroughs
- Serving as a science and technology resource to the U.S. government and as a partner with industry and academia
- Pushing the frontiers of knowledge to build the scientific and technological foundation that will be needed to address global security issues of the future

One of three Department of Energy (DOE)/National Nuclear Security Administration (NNSA) laboratories, LLNL is managed by the Lawrence Livermore National Security, LLC. Since its inception in 1952, the Laboratory has fostered an atmosphere of intellectual freedom and innovation that attracts and maintains the world-class workforce needed to meet its challenging science- and technology-based missions.

Laboratory Directed Research and Development

The LDRD Program, established by Congress at all DOE national laboratories in 1991, is LLNL's most important single resource for fostering excellence in science and technology for today's needs and tomorrow's challenges. The LDRD internally directed research and development funding at LLNL enables high-risk, potentially high-payoff projects at the forefront of science and technology.

The LDRD Program at Livermore serves to

- Support the Laboratory's missions, strategic plan, and foundational science
- Maintain the Laboratory's science, technology, and engineering vitality
- Promote recruiting and retention
- Pursue collaborations
- Generate intellectual property
- Strengthen the U.S. economy

Myriad LDRD projects over the years have made important contributions to every facet of the Laboratory's mission and strategic plan, including its commitment to nuclear, global, energy, and environmental security, as well as cutting-edge science and technology and engineering in high-energy-density matter, high-performance computing and simulation, advanced material and manufacturing, data science, lasers and optical systems and energy manipulation.

The *FY 2014 Laboratory Directed Research and Development Annual Report*

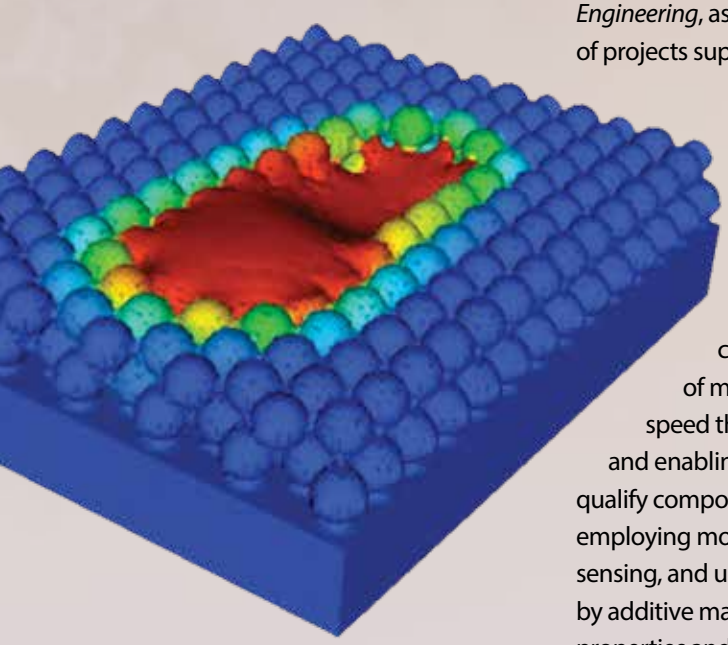
The LDRD annual report for fiscal year 2014 (FY14) provides a summary of LDRD-funded projects for the fiscal year and consists of two parts:

Overview: A broad description of the LDRD Program, highlights of accomplishments and awards for the year, program statistics, and the LDRD portfolio-management process.

Project Summaries: A summary of each project, submitted by the principal investigator. Project summaries include the scope, motivation, goals, relevance to DOE/NNSA and LLNL mission areas, the technical progress achieved in FY14, and a list of selected publications and presentations that resulted from the research. Project summaries for the annual report are organized in sections by research category (in alphabetical order). Within each research category, projects appear for the various groups including Strategic Initiative (SI), Exploratory Research (ER), Laboratory-Wide (LW), and Feasibility Study (FS). Each project is assigned a unique tracking code, an identifier that consists of three elements. The first is the fiscal year the project began, the second represents the project category, and the third identifies the serial number of the proposal for that fiscal year. For example, 14-ERD-100 means the project began in FY14 and falls in the ER project category. The three-digit number (100) represents the serial number for this proposal.

Highlights of Accomplishments for the Fiscal Year

In FY14, the LDRD Program at LLNL continued to be extremely successful in supporting research at the forefront of science, technology, and engineering, providing new concepts for core missions, and creating an exciting research environment that attracts and retains outstanding young talent to the Laboratory. Wide-ranging projects for this fiscal year exemplify LDRD's noteworthy research in support of the Laboratory's long-range strategic science and technology plan, the *Investment Strategy for Science, Technology and Engineering*, as well as for critical national needs. Here, we provide highlight examples of projects supporting various strategic focus areas and core competencies.



Simulation of laser melting of a powder layer showing consolidation by surface tension and gravity and the molten pool in the middle.

Advanced Materials and Manufacturing

In industries such as defense, aerospace, and medicine, the manufacturing processes and materials used to produce critical components must be formally qualified to ensure they perform to specification, as failure could prove disastrous. The extensive empirical testing and evaluation required to develop a material and qualify a component often encompass many thousands of individual tests, at a cost of millions of dollars and 5 to 15 years of effort. Additive manufacturing can speed the development of complex designs, accelerating the development cycle and enabling customization. However, to realize its full potential, the processes to qualify components and certify systems must also be accelerated. An LDRD project is employing modeling, simulation, process optimization, experiment design, in-place sensing, and uncertainty quantification for the accelerated certification of metals produced by additive manufacturing, with the goal of guiding the process to yield optimized properties and performance (13-SI-002). Rather than undertaking exhaustive experimentation, the research team borrows a formula that has proven highly effective for stockpile stewardship work: modeling and simulation paired with targeted experiments and guided by data mining and uncertainty quantification. In turn, the results of this LDRD project may, with additional development effort, be applied to Livermore's nuclear stockpile mission. Nuclear engineers and additive manufacturing experts are presently exploring how their methods could benefit weapons refurbishment endeavors. In the first two years of the project, the team of computational experts has built and begun testing platforms for its multiscale modeling and data-mining efforts. They have

- Demonstrated defect mitigation and computed residual stresses in a part that will be produced
- Validated residual stress predictions with experiments
- Demonstrated an initial modeling capability incorporating surface tension
- Demonstrated powder melt and gas bubble migration and measured powder-bed thermal properties for the powder model

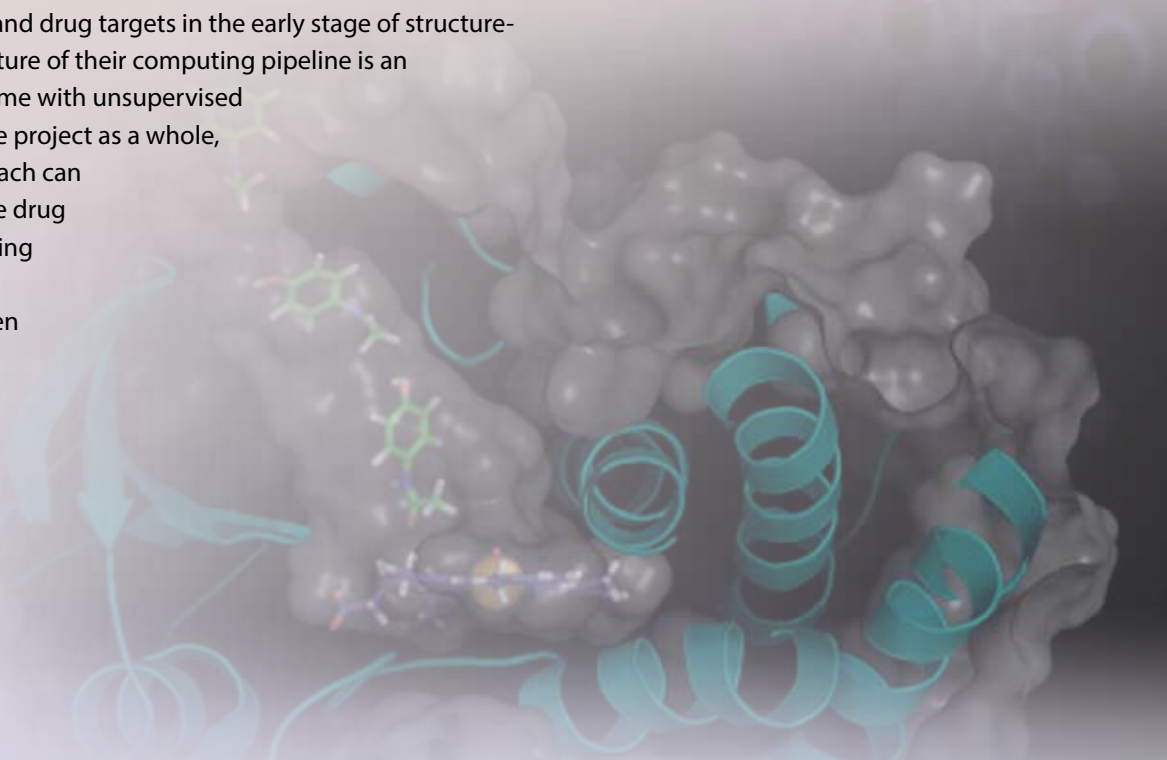
With these platforms, the researchers are progressing toward understanding and optimizing the rapid heating, melting, cooling, and solidification processes at the heart of metal additive manufacturing.

Biosecurity

Utilizing the Laboratory's text-mining capability and world-class expertise in high-performance computing, LDRD researchers are addressing the national need to develop medical countermeasures against emerging bio-threats. This requires accelerating the drug development process. With the project "Computational Advancements in Countermeasures for Emerging Bio-Threats" (12-SI-004), investigators worked to develop capabilities to predict bodily absorption, distribution, metabolism, and excretion of drugs (pharmacokinetics) and adverse side effects in the initial optimization stage to enable successful clinical outcomes for drug candidates. The LDRD project combines systems biology, physiologically based pharmacokinetics modeling, biophysics, computational chemistry, and informatics to create a predictive capability based on a drug candidate's chemical structure. The successful conclusion of this project resulted in

- Establishing a credible capability for an all-computational prediction of drug side effects
- Predicting drug interactions with molecules not targeted
- Linking molecular interactions to pharmacokinetics
- Predicting penetration of the brain's blood barrier
- Providing software as an in-house resource and to external investigators

In a December 2013 *Journal of Chemical Information and Modeling* article, the researchers reported on their high-throughput virtual screening of compound databases using high-performance computing to access the binding affinities between molecular compounds and drug targets in the early stage of structure-based drug design. A notable feature of their computing pipeline is an automated cellular receptor scheme with unsupervised binding-site identification. For the project as a whole, the team determined their approach can outperform, for particular adverse drug reaction classes, the best competing model that uses freely available experimental data. They have been invited to submit a grant to the National Institutes of General Medical Sciences to continue using high-performance computing to predict adverse drug reactions of drugs and drug candidates.



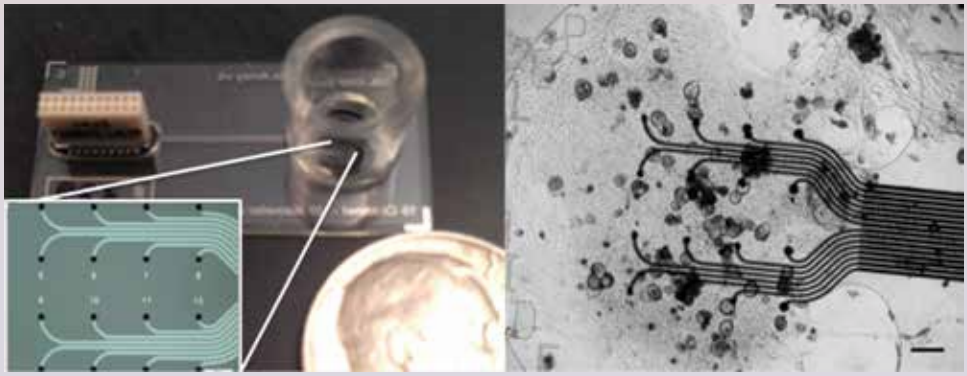
Predicting the binding of drugs to metabolizing enzymes.

Bioscience and Bioengineering

Building upon the success of a research effort to develop a platform for primary human sensory nerve cells that bring information from the body's periphery to the spinal cord (dorsal root ganglion cells), LDRD researchers are developing an "In Vitro Chip-Based Human Investigational Platform" (14-SI-001) to integrate human organ systems into an instrumented, microfluidic platform. Called the iCHIP, it is envisioned to be a highly integrated, multiple-organ, human-relevant in vitro platform (outside the body) to reproduce in vivo (inside the body) physiological response. The research team intends to develop tissue systems that include

- Dorsal root ganglia
- Central nervous system nerve cells
- Blood brain barrier cells that separate circulating blood from the brain extracellular fluid
- Heart tissue

This platform could be used to rapidly assess and predict the toxicity, safety, and efficacy of countermeasures against chemical and biological agents. The team's research will reduce preclinical testing and improve relevance to clinical outcomes with technologies that utilize in vitro platforms with primary human cells organized in a physiologically relevant manner. The platform will also enable investigation of the mechanisms of infection for emerging threats, and it will be used to understand the evolution of threats in human tissue. In FY14, the team has demonstrated the viability of an in vitro collection of human sensory nerve cells for one month, recording reactions to a chemical irritant with an embedded electrode array. Maintenance of nonhuman nerve cells was achieved for more than three months. In addition, they have incorporated an automated fluidic delivery system to the investigational platform, and performed the first correlated simultaneous optical and electrical recording of cells' response to chemical exposure.



Electrode arrays are embedded in an automated fluidic device to record nerve cell response to chemical exposure.

Chemical and Isotopic Signatures

A study of fallout melt glass formation from a near-surface nuclear test published in a July 2014 online edition of the *Journal of Radioanalytical and Nuclear Chemistry* presented major element and actinide composition data from a population of aerodynamically shaped fallout glass samples from a single near-surface nuclear detonation. Work relevant to the Laboratory's core competency in chemical and isotopic signatures was supported by the LDRD project "Improving Resonance Ionization Mass Spectrometry for Next-Generation Nuclear Forensics" (14-ERD-082). The project's aim is to address research issues related to the isotopic analysis of low-abundance materials, such as those found in early solar system materials or nuclear fallout. Today these issues limit the ability to answer fundamental chemistry questions about the genesis of the solar system or to rapidly quantify actinide isotope ratios in fallout. Researchers are using resonance ionization mass spectrometry to rapidly and accurately quantify isotope ratios for materials including plutonium, uranium, magnesium, beryllium, and lithium. The technique is a high-sensitivity, elementally selective, laser-based form of mass spectrometry that offers the potential to determine isotopic composition of materials without sample preparation. The major element compositions of the fallout glass samples they analyzed indicate that

- Composition of local geology is a primary control on the bulk chemistry of the fallout
- Vaporized, residual fuel was incorporated into the melts prior to solidification, likely within seconds, based on uranium isotopic compositions
- Compositions are consistent with two-component mixing between naturally occurring uranium and residual uranium fuel

Although the samples were not the direct result of condensation from the bomb-produced vapor, the samples must have incorporated primary condensates, which dominate the uranium in the glasses examined. This suggests that such glassy fallout materials may be of high value for nuclear forensic investigations. These observations also highlight a need to understand the microscopic-scale features in these materials to unravel the formation processes of condensation, agglomeration, mixing, and diffusion.

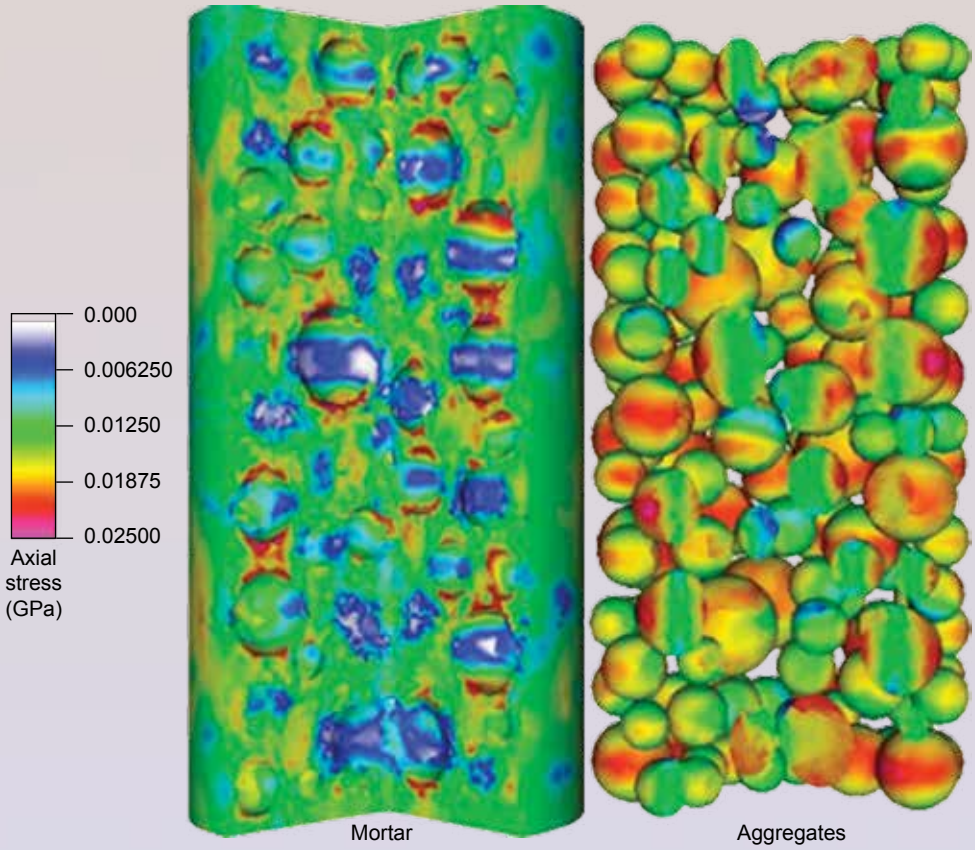
The LION (laser ionization of neutrals) instrument at Lawrence Livermore will accelerate development of resonance ionization mass spectrometry to characterize nuclear materials.

Computational Science and Engineering

In response to the vulnerability of surface facilities, many potential adversaries around the world have constructed spaces deep underground to house particularly important strategic assets. Many of these spaces are presumably intended to hide or protect lethal military equipment and activities, including weapons of mass destruction. The U.S. is confronted with an array of thousands of buried and hardened targets, many of which are beyond the reach of conventional weapons. The objective of a final-year LDRD project was to develop new high-fidelity, three-dimensional modeling capabilities, with computational science and engineering, for predicting conventional penetrator performance against such targets (12-ERD-064). To develop this modeling capability, investigators used a physics-based approach that makes use of small-size simulations to account for material heterogeneities and deformation mechanisms such as fracture, fragmentation, pulverization, and granular mechanics. The goal was to model the response of materials to extreme dynamic-loading environments such as those encountered during the interaction of an earth penetrator with a geologic target or the interaction of a bullet or a shaped charge with ceramic armor. This modeling framework will support the design of advanced penetrating weapons that are smaller, lighter, faster, and more effective against hardened and deeply buried targets. Also, this work will make it possible to design more efficient transparent ceramic armor capable of providing superior protection against a wide range of threats, including shaped charges and improvised explosive devices. The project resulted in

- Development of procedures for simulating discrete fracture and fragmentation for coordinated computations in parallel computer systems
- Simulations of unprecedented details to examine the microscopic structural processes that govern deformation and failure in concrete
- Development of a large-scale model suitable for performing simulations of penetration into concrete and other geologic targets

Researchers are now arranging for a new project for the joint DOE and Department of Defense Munitions Technology Development Program in FY16 that will focus on modeling of concrete for penetration applications at multiple scales, from large to small.



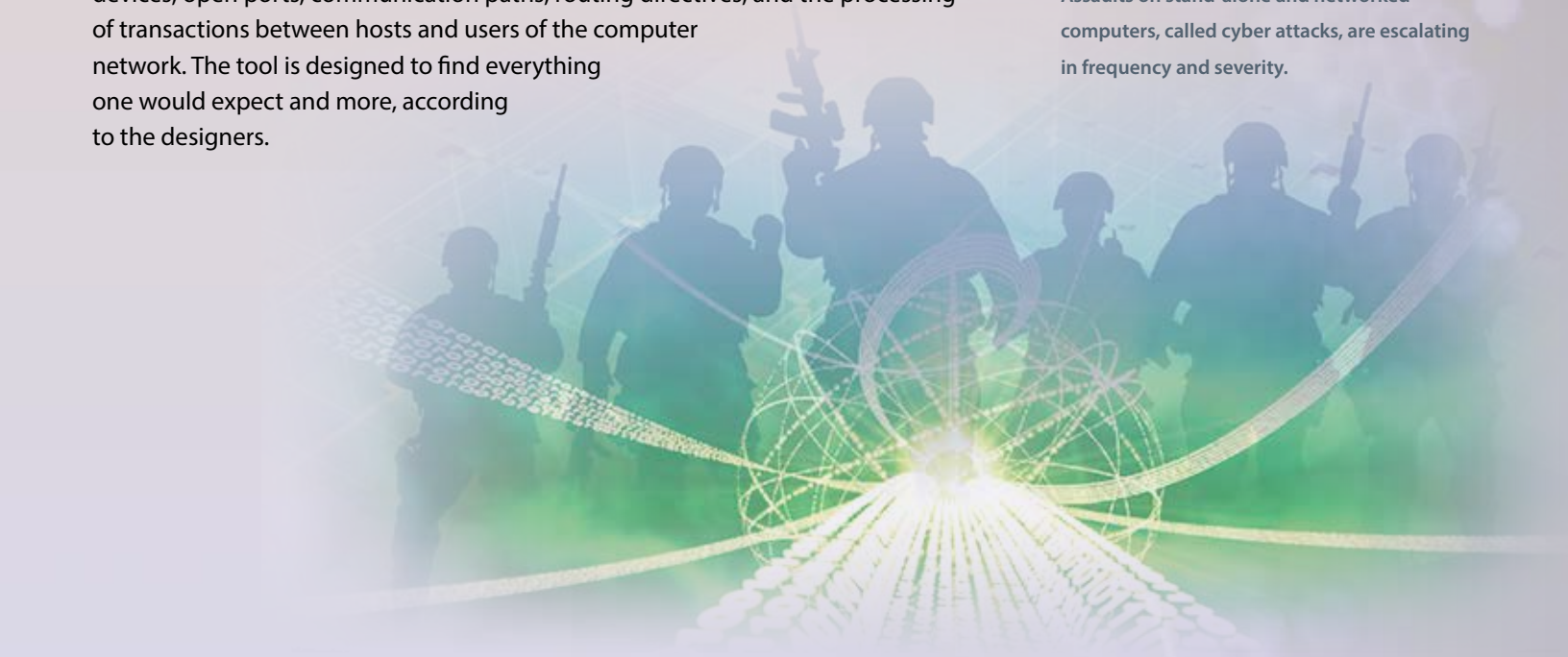
Small-scale simulations of cylindrical concrete samples were used to understand deformation and failure mechanisms to create a model for the response of concrete to a penetrating object.

Cyber Security, Space, and Intelligence

Information warfare is the new art of subverting an enemy in the new battles of the 21st century and beyond. The "Continuous Network Cartography" project (13-SI-004), supporting the Laboratory's cybersecurity strategic focus area, promises to provide network mapping and analytics for the continuous monitoring of computer network components and activities, as well as techniques for mapping and situational awareness to detect noncooperative, complex, or adversarial intrusion, denial, or deception cyber tactics. Researchers propose to build continuous network cartography (mapping) capabilities and analytics that apply machine-learning and statistical methods for understanding network activities. This project also focuses on mapping and inferring hidden or obfuscated network components. These two focus areas directly address the gaps and limitations of today's network-mapping technologies and seek to provide a view of an activity or behavior, enhancing a computer analyst's ability to make timely decisions and effectively change the outcome of a cyber attack. Thus far, the team has

- Created an integrated change-detection framework (dTrend) and evaluated it with real data, as well as presented the dTrend output in an interactive multiple-timescale visualization
- Developed an interactive visualization tool as a step toward incorporating human analysts in the loop
- Created an interactive continuous mapping interface for controlling network mapping setup and execution

Another LLNL cybersecurity tool that received early-stage support from this project was one of eight cutting-edge technologies that was showcased to Silicon Valley venture capitalists under the Department of Homeland Security's Transition to Practice Program. The tool, Net_Mapper, is designed to find anything attached to the network: devices, open ports, communication paths, routing directives, and the processing of transactions between hosts and users of the computer network. The tool is designed to find everything one would expect and more, according to the designers.



Assaults on stand-alone and networked computers, called cyber attacks, are escalating in frequency and severity.

Energy and Climate

Water use in hydraulic fracturing for natural gas production is strongly affected by the need to drive materials, known as proppants, which are typically rounded sand grains, into the created fracture to hold it open during gas production. An LDRD team is developing and demonstrating a new fracture material (13-ERD-029) that will eliminate the environmental problems associated with treating and reusing the water treated with a complex mixture of thickeners and friction reducers that enable the fluid to be sufficiently viscous to move the dense sand particles. The team will develop the materials science and engineering to allow transport and reaction under specific conditions, which could be applied in many other fields. They will also provide experimental support for the necessary engineering and theoretical science, allowing them to demonstrate the applicability of their new fracture material while developing a strong base of new knowledge about fracture flow of particulates and proppants. The goal is to create a neutral-density proppant composed of a reactive material encapsulated in a silicone shell, which reacts within the fracture to become very strong and expansive. Success will improve both the efficiency and environmental impact of natural gas production. In FY14, the team

- Created the first temperature-set proppants, which have a liquid core inside a polymer shell
- Demonstrated that during transport the proppants are malleable and of neutral density, ensuring deep placement
- Demonstrated the proppants set to solids when exposed to temperatures greater than 70°C

The team obtained x-ray images of these proppant capsules inside laboratory samples of Marcellus shale from the Appalachian Basin using an x-ray tomography system, as well as optical images using printed, transparent versions of the same fractures. In the coming year, they will create a mineral filling for the proppant capsules that not only sets, but slowly expands upon curing, and demonstrate scale-up of production to enable large-scale use of these materials. It is expected that the proppants will be initially licensed for use in shale gas operations.

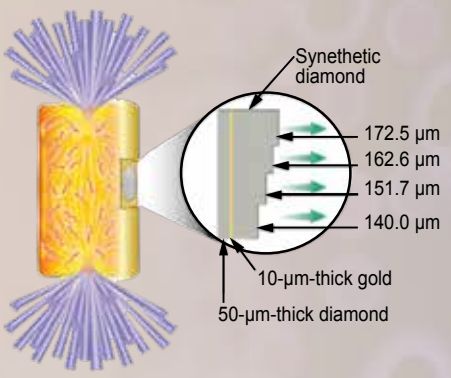
Newly developed fracture capsules for natural gas production being produced with a liquid core inside a polymer shell.

High-Energy Density Science

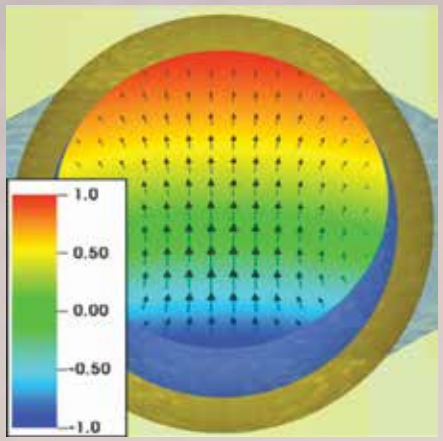
The recent discovery of more than a thousand planets outside our solar system, together with the significant push to achieve inertial-confinement fusion in the laboratory, has prompted a renewed interest in how dense matter behaves at millions to billions of atmospheres of pressure. The LDRD project "Extreme Compression Science" (12-SI-007)

- Developed x-ray diffraction experiments that showed the elemental structure phase transition in magnesium oxide for the first time
- Supported the development of several new diagnostic techniques for high-power laser-driven compression experiments
- Achieved the first-ever shock melting and refreezing diffraction experiment

In a July 2014 *Nature* article, the LDRD researchers describe their ramp-compression measurements for diamond, achieving a peak pressure equivalent to 50 million atmospheres. These equation-of-state data can now be compared to first-principles density functional calculations and theories long used to describe matter present in the interiors of giant planets, in stars, and in inertial-confinement fusion experiments. Their data also provided new constraints on mass–radius relationships for carbon-rich planets. The researchers examined phase transitions or equation of state for iron, tin, molybdenum, iron oxide, silicon dioxide, titanium, bismuth, sodium chloride, aluminum oxide, and aerogels under extreme compressions. The project helped enable target-diffraction diagnostic platforms at Livermore's National Ignition Facility operating at one billion times the pressure at sea level, and resulted in over 20 articles in peer-reviewed scientific journals including *Science*, *Nature*, the *Journal of Applied Physics*, and *Physical Review Letters*. Investigators will take active roles in the development of laser-driven compression experiments at the Linac Coherent Light Source at the SLAC National Accelerator Laboratory at Stanford, the Dynamic Compression Sector at the Advanced Photon Source at Argonne National Laboratory, the European X-Ray Free Electron Laser facility in Germany, and the OMEGA laser in Rochester, New York.



Laser compression experiments employing a layered diamond target that absorbs x-ray energy when laser beams converge inside the target capsule. Measuring the speed at which each sample thickness moves helps researchers better understand material behavior at high pressure.



Computed evaporation-driven velocity field in the hydrogen vapor (shown as arrows) for research into the growth of high-quality hydrogen layers used for laser fusion research.

Inertial-Confinement Fusion Science and Technology

A model of the thermal transport inside of a laser-ignition fusion target shell, as well as a process model to test ideas for improving the rate of producing ignition-quality fuel layers has been developed for an LDRD project examining "Hydrogen Ice Layers for Inertial-Confinement Fusion Targets" (12-ERD-032). Targets for inertial-confinement fusion comprise layers of condensed hydrogen fuel inside spherical capsules. The layers must be easily reproducible and very smooth. Numerous experiments have shown that these requirements can only be met by using a nearly perfect single crystal of solid hydrogen. The formation of these high-quality layers depends on creating and isolating a single crystal of the solid and then slowly cooling the melt to freeze the remaining liquid. The current success rate of this process is subject to the random nature of nucleation and the resulting seed crystal used to grow these layers. This method results in a range of layer qualities, many of which do not meet target specifications. The LDRD researchers worked to develop a deterministic seeding process leading to reproducible high-quality target ice layers. In a *Journal of Applied Physics* article in 2014, the researchers concluded that generation of deuterium–tritium seed crystals in a confined geometry is governed by three effects: self-heating from tritium decay, external thermal environment, and latent heat of phase change at the boundary between hydrogen liquid and vapor. For this LDRD project, the team

- Developed an experimental system that can be used to test the super-cooling of hydrogen on new substrate template materials
- Found that rare-gas solids promote nucleation of solid hydrogen better than other materials and were important in aiding the process of understanding super-cooling effects
- Determined that highly ordered graphite promotes solid nucleation nearly as well as the rare gases, and is more practical to implement

The project researchers will collaborate with the Laboratory for Laser Energetics at the University of Rochester and the Schafer Corporation in Livermore on identifying additional template candidates and testing these with the experimental platform created during this project.

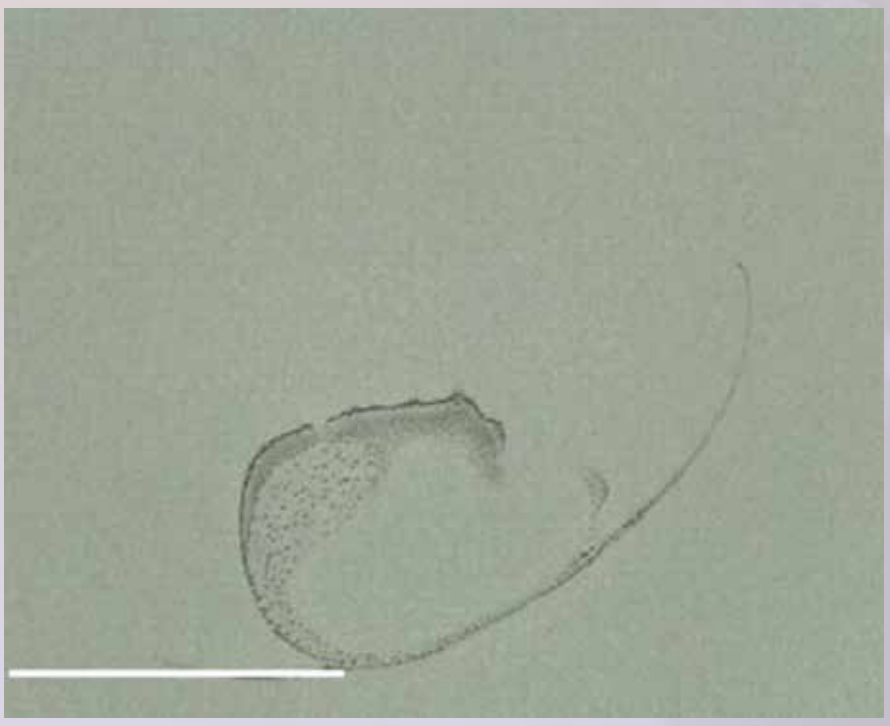
Lasers and Optical Materials Science and Technology

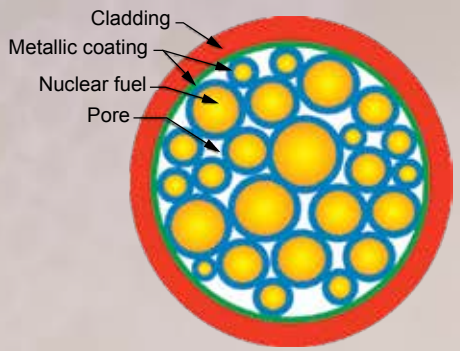
An LDRD project seeking to determine the physical mechanisms of initiation of high-radiant-exposure damage in optical materials for lasers (12-ERD-023), found that increases in the laser damage threshold of fused silica have been driven by the successive elimination of near-surface damage precursors such as polishing residue, fractures, and inorganic salts. In this work, described in a December 2014 *Optics Express* journal article, researchers showed how trace impurities in ultrapure water used to process fused silica optics may be responsible for the formation of carbonaceous deposits. The LDRD researchers used surrogate materials to show that organic compounds precipitated onto fused silica surfaces form discrete damage precursors. The lifetime and performance of optical systems designed to guide high-photon radiation transfer are limited by degradation and damage to key optical components at high-photon radiant exposure, or fluence. Even high-quality optical surfaces without flaws can degrade as a result of extensive multiple-pulse optical stress and can suffer damage from absorption by damage precursors. The mechanisms of this degradation and the nature of these precursors were generally unknown. Researchers employed a suite of integrated tasks that closely link processing, characterization, and modeling to develop a scientific understanding of the mechanisms that govern high-fluence optical damage and degradation, and developed techniques to improve the high-fluence lifetime for optical glasses and other related optical materials. The successful conclusion of this study resulted in

- A new understanding of optical damage and degradation for silica at high pulse fluence and longtime multipulse exposure
- Demonstration of a means to control or mitigate these effects
- Determination that the dominant laser damage precursors at high fluence are microscopic precipitates of trace ionic and organic impurities in processing chemicals
- Determination that defects in these precipitates absorb enough laser energy to reach temperatures that can initiate microscopic-sized damage sites
- Development of processes to reduce the probability of precipitation during wet-chemical processing and drying

Investigators achieved a two-thousandfold reduction in damage density that extends useful operation fluences by almost a factor of two. Laboratory programmatic support will enable them to continue work to fully transfer the optics processes they developed to full-scale optics production and use on the National Ignition Facility at Livermore.

Optical micrograph of residue impurities found in ultrapure process water for laser optics production.





Schematic of an inert metal matrix fuel.

Nuclear Science and Technology

Development of sustainable nuclear energy is critical to the energy security of the U.S. Today, only a small fraction of the enriched uranium that is used to fuel the nation's approximately 104 civilian reactors is actually converted to fission energy—the remaining material is identified as spent nuclear fuel and, rather than being considered for its potential energy, is discarded as waste. An LDRD project on "Ultrahigh-Burn-Up Nuclear Fuels" (12-SI-008) is combining modern computational materials modeling, fabrication, and characterization capabilities and targeted performance-testing experiments to establish the scientific foundation for selecting the optimum fuel type for advanced reactor concepts. Researchers experimentally quantified the stability and kinetics of element phase transformations, inter-diffusion, microstructural evolution, micromechanical properties, and the influence of severe radiation environments on fuel performance. Their work will enable a validated model for advanced nuclear energy materials under extreme conditions of radiation, temperature, and evolving chemistry. The effort extended the Laboratory's capabilities in

- High-energy-density science
- Energy manipulation
- Materials on demand

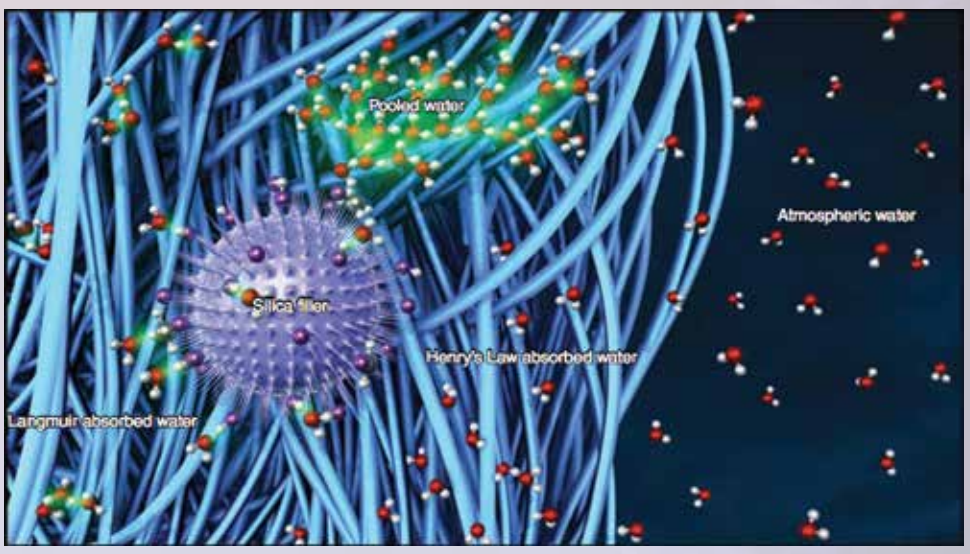
relevant to the core competency in nuclear science and technology. In a February 2014 edition of *JOM* (journal of The Mineral, Metals and Materials Society) the team concluded that electronic-structure calculations and CALPHAD (computer coupling of phase diagrams and thermochemistry) thermodynamic assessments allowed them to study multicomponent alloys and design materials with improved properties, and that their predictions can guide experimental investigations that are usually difficult and costly. This work has contributed to an international database for the Nuclear Energy Agency based in Paris, France. The Korea Atomic Energy Research Institute in South Korea has expressed an interest in their approach to the basic science of ultrahigh-burn-up advanced nuclear fuels. In addition, the project has resulted in over 40 presentations and publications in peer-reviewed scientific publications on alloy behavior and metallic nuclear fuels.

Stockpile Stewardship

Accurate predictions of material compatibilities as a function of age are important in various fields, from designing aerospace components and medical devices to preserving works of art. Some chemical reactions between materials in sealed environments may be benign, but many of them will cause damage and loss of material functionality. Ensuring a safe, reliable, and secure nuclear deterrent also requires scientists to understand weapons performance and the technical issues related to how these systems age. To more closely examine the fundamental chemical transformations that contribute to component aging, LDRD researchers have developed a reactive transport model for assessing the compatibility and chemical kinetics of materials inside nuclear weapons systems with the project "Predicting Weapon Headspace Gas Atmosphere for Modeling Component Compatibility and Aging" (12-ERD-046). The model is based on fundamental physical and chemical properties of the materials and will be versatile enough to apply to different geometries, sizes, and arrangements. Simple diffusion models are too rudimentary for stockpile assessment, so the team developed more advanced mathematical models that incorporate sorption, diffusion, and chemical kinetics to achieve these results. The researchers conducted experiments over a wide range of humidities and temperatures concurrently with model development efforts to verify the model code's accuracy. During the course of the project, the team created

- A new technique for measuring vapor uptake and outgassing
- Multiple-material aging methods
- Moisture-based chemical reaction quantification methods based on quadruple-mass-spectrometry and heat-flow calorimetry
- A dynamic sorption and diffusion model based on absorption, adsorption, and pooling

The team's model will better predict the long-time behavior of weapons materials and allow scientists to develop more robust system components and nondestructive surveillance capabilities for managing the stockpile. The DOE NNSA weapons program will provide support for further development and utilization of this capability, and additional funding opportunities are being explored relevant to munitions technology development, shale-gas production, and countering chemical warfare agents.



Various sorption mechanisms dictate chemical uptake and outgassing in a sealed system, such as Henry's Law, in which a gas is taken in by another material and fills that material's voids. Knowledge of these mechanisms is critical to developing a reactive transport model that can accurately predict component aging and material compatibility of nuclear weapons.

Awards and Recognition

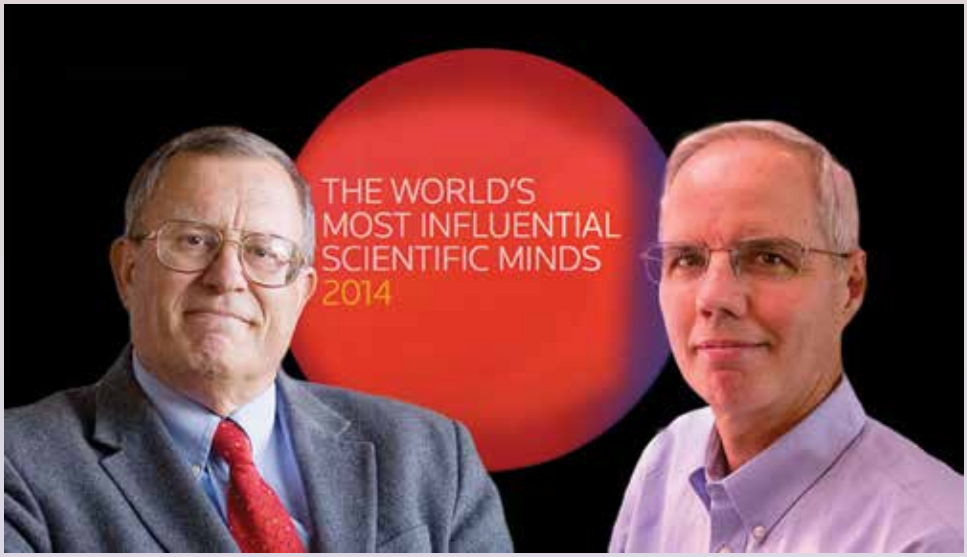
A primary goal of the LDRD Program is to foster excellence in science and technology that will, among other things, attract and maintain the most qualified scientists and engineers and allow scientific and technical staff to enhance their skills and expertise. Laboratory LDRD principal investigators and research teams receive numerous prestigious honors, awards, and recognition for LDRD-funded work. These recent honors attest to the exceptional capabilities, talents, and performances of these researchers, while simultaneously highlighting the success and vitality of the LDRD Program at Livermore.

The World's Most Influential Scientific Minds

Lawrence Livermore scientists Charles Westbrook and William Pitz have been named to the Thomson Reuters list of "The World's Most Influential Scientific Minds."

The list of 3,000 researchers was generated by analyzing citation data over the last 11 years to identify those ranking in the top 1% in citations in their subject area. The two have published numerous research papers on combustion modeling, and their work has been incorporated into codes that simulate combustion in internal combustion engines—codes used by the auto industry and others to optimize engine design, increase efficiency, and reduce emissions.

Westbrook (left) was an LDRD co-investigator for a project on "Local-Scale Atmospheric Reactive-Flow Simulations" (02-ERD-027), among others, and Pitz (right) was a co-investigator for several projects, including "A Hydrogen–Oxygen–Argon Internal Combustion Engine System: The Mechanical Equivalent of a Fuel Cell" (08-ERD-042).



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Optical Society Fellow

Regina Soufli

Researcher Regina Soufli has been elected a fellow of the Optical Society. She was cited for her "significant contributions to the development and characterization of extreme ultraviolet, x-ray, and gamma-ray optics." Soufli has conducted pioneering research in the field of x-ray interactions with matter, publishing methodologies and experimental values for the refractive index of materials in the extreme ultraviolet and x-ray regimes that have been adopted by the scientific community around the world. At LLNL, she has led programs that developed first-of-a-kind extreme ultraviolet and x-ray optics for photo-lithography, solar physics and astrophysics missions for the National Aeronautics and Space Administration, x-ray free-electron lasers, and other high-energy physics applications. She has served as a principal or co-investigator for 10 LDRD projects in x-ray optics, rare-event detection, plasma physics, and space situational awareness. In FY14, Soufli was the principal investigator for an LDRD project investigating multilayer thin-film science for core missions (12-ERD-055). At the Laboratory, Soufli is the 10th current employee and the first woman to be elected an Optical Society fellow, which is limited to less than 10% of the total membership, and the number elected each year is less than 0.4% of current total membership.

Chair of American Nuclear Society Fusion Energy Division

Susana Reyes

Nuclear engineer Susana Reyes has assumed the 2014–15 chairmanship of the American Nuclear Society's Fusion Energy Division, following her election as vice chair in 2012.

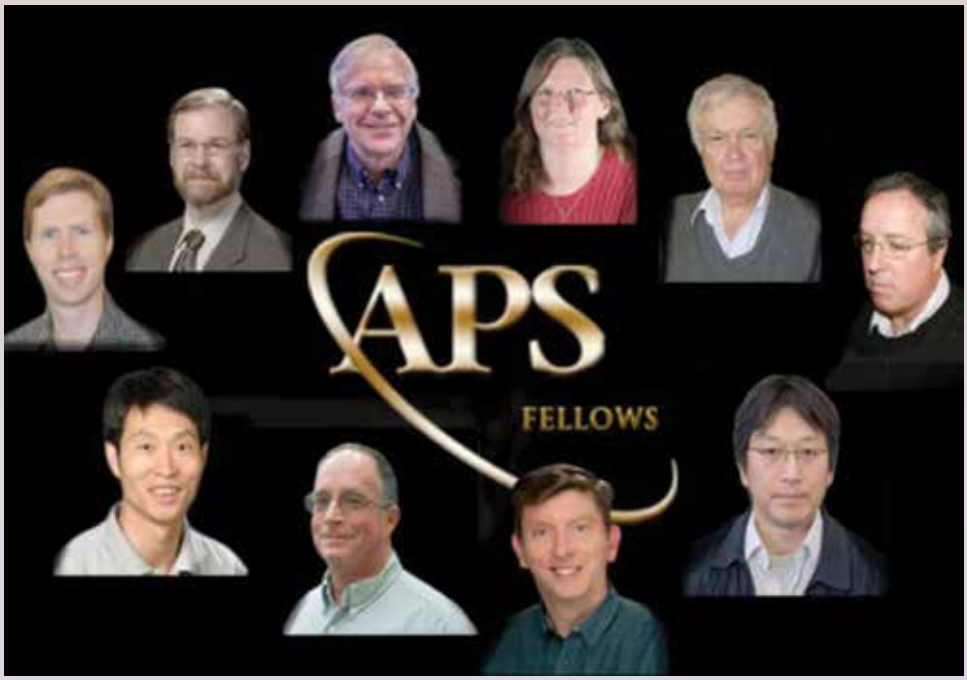
Reyes has more than 12 years of experience in international fusion projects. She joined the Laboratory in 2001 to pursue her interest in fusion science and worked on the safety analysis of inertial fusion energy power plant designs. Since then, she has worked in a variety of fusion research projects, such as the U.S. ITER Test Blanket Module program for the testing of tritium breeding blanket concepts within the ITER magnetic fusion facility, now under construction in Cadarache, France. Reyes is currently the LDRD principal investigator for a next-generation process for tritium recovery from fusion power plant blankets (13-ERD-056).

Reyes earned a master's of science degree in power engineering from the Polytechnic University of Madrid in 1998 and a Ph.D. in nuclear engineering from the UNED University in Madrid in 2001. In 2012, she received the Mary Jane Oestermann Professional Women's Achievement Award from the American Nuclear Society, which recognized her "leadership in developing detailed hazard and safety analyses for both inertial and magnetic fusion facilities, including NIF and ITER, and future power reactors." This award is given annually for outstanding personal dedication and technical achievement by a woman in the fields of nuclear science, engineering, research, or education.

The American Nuclear Society is a scientific and educational organization working to promote the awareness and understanding of nuclear science and technology. Membership comprises 11,000 engineers, scientists, administrators, and educators representing more than 1,600 corporations, educational institutions, and government agencies.

American Physical Society Fellows

Ten LLNL scientists have been selected as 2014 fellows of the American Physical Society. The new fellows represent a wide selection of physics expertise, ranging from laser science to laser hohlraum target capsule design to theoretical solid-state physics. The fellowships are awarded after extensive review and are considered a distinct honor



The American Physical Society named ten Lawrence Livermore researchers as 2014 fellows. Top row from left, Michael Armstrong, Christopher Barty, Raymond Beach, Debbie Callahan, Antonis Gonis, and Frederic Hartmann. Bottom row from left, Yinmin "Morris" Wang, James Tobin, Robert Rudd, and Nobuhiko Izumi.

because the evaluation process, conducted by the fellowship committees of individual divisions, topical groups and forums, relies on nomination and recommendation by candidates' professional peers. Of the ten Laboratory researchers named as 2014 fellows, nine have served as investigators for LDRD projects. Election is limited to no more than one half of one percent of the association's membership for a given year.

- ***Michael Armstrong*** was cited by the Topical Group on Instrument and Measurement Science for outstanding contributions to time-domain experimental methods applied to materials under extreme conditions. He is currently the principal investigator for a high-temperature plasma-chemistry kinetics test bed (14-ERD-077), and has led previous LDRD projects related to high-density hydrogen and laser sensors and diagnostics.

- ***Christopher Barty*** was nominated by the Division of Laser Science for outstanding contributions to time-domain experimental methods applied to materials under extreme conditions, and has been a principal LDRD investigator on projects related to gamma-ray and advanced laser science, including a strategic initiative on precision monoenergetic gamma-ray science (09-SI-004).

- ***Raymond Beach*** was also nominated by the Division of Laser science for seminal contributions to high-average-power diode-end-pumped lasers, including many breakthroughs, widely adopted by the laser community, that have helped push such lasers to higher average powers and efficiencies, and for leadership in developing diode-pumped alkali-vapor lasers, and models for coherent and incoherent photon echoes. Beach has been an LDRD principal investigator for many years, exploring the feasibility of various short-pulse and solid-state lasers and laser applications in manufacturing and national security. His most recent project explored the feasibility of a hybrid rubidium resonance and exciplex pump laser for defense and commercial material-processing applications (10-FS-002).

- ***Antonios Gonis*** was nominated by the Division of Computational Physics for advancing multiple scattering theory electronic structure methods for metals, alloys, and interfaces and for the dissemination of these techniques in condensed matter and materials science. Gonis has been an LDRD principal investigator for many years and most recently lead a team investigating the Coulomb potential in electronic structure calculations (12-ERD-072).

- ***Frederic Hartemann*** was cited for remarkable insights and significant contributions to the physics of coherent radiation interacting with relativistic electrons by the Division of Physics of Beams. He recently concluded an LDRD project on Compton-scattering optimization for ultra-narrowband nuclear photonics (12-ERD-057).

- ***Nobuhiko Izumi*** was cited for outstanding contributions to the development of novel neutron and x-ray diagnostic capabilities for inertial-confinement fusion experiments by the Topical Group on Instrument and Measurement Science. Izumi

has been a co-investigator for several LDRD projects such as laser fast ignition (08-SI-001), high-energy backlighting for high-power laser diagnostics (07-ERD-004), and developing radiography capabilities for fusion-class lasers (05-ERD-006).

- ***Robert Rudd*** was nominated by the Division of Computational Physics for seminal contributions to multiscale modeling of materials physics and science in support of national security. Rudd has been the principal investigator for LDRD projects exploring nanometer-scale mechanics of strength and structure (04-ERD-043) as well as impurity and alloying effects on material strength (08-ERD-035).

- ***James Tobin*** was cited for use of soft x-ray spectroscopy to investigate complex systems, including actinide-based materials, by the Division of Condensed Matter Physics. He has served as the principal investigator for an LDRD project to determine the unoccupied electronic structure of plutonium (04-ERD-105) as well as a project dating back to 1991 that investigated nanometer-scale materials with circular and spin polarization (91-DE-001).

- ***Yinmin Wang*** was cited for his major contributions to the understanding of deformation physics of nanometer-scale crystalline and twinned materials, and for developing effective strategies to enhance the ductility of these superstrong materials for technological applications, including fusion energy targets, by the Division of Material Physics. He has been a member of several LDRD research teams examining transformational materials and is currently the co-investigator for a project developing accelerated certification of additively manufactured metals (13-SI-002).

IEEE Technical Committee on Scalable Computing Young Achievers Award

Abhinav Bhatele

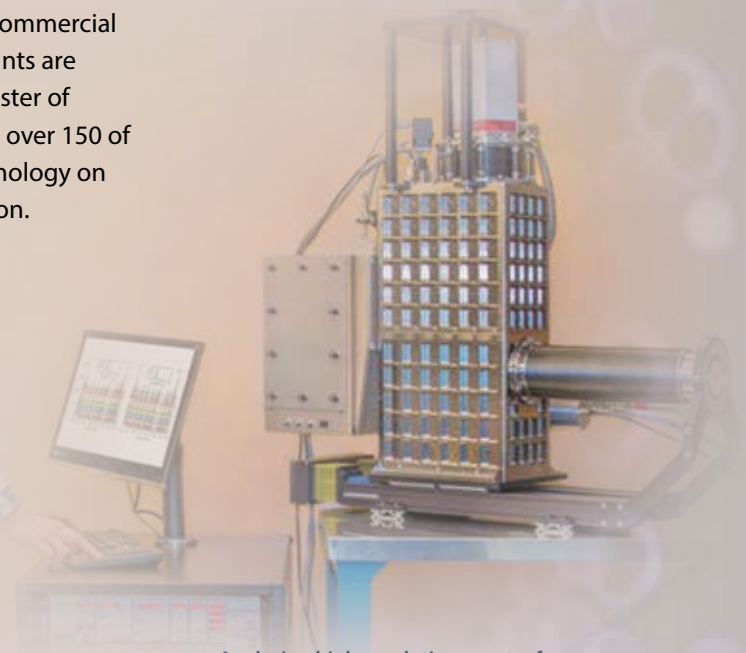
Abhinav Bhatele received the 2014 Young Achievers Award from the IEEE (Institute of Electrical and Electronics Engineers) Technical Committee on Scalable Computing at the November Supercomputing (SC14) Conference in New Orleans. The IEEE award for young achievers in scalable computing recognizes up to three individuals each year who have made outstanding, influential, and potentially long-lasting contributions in the field of scalable computing within five years of receiving their Ph.D. degree. Bhatele is currently the principal investigator for an LDRD project for task mapping on complex computer network topologies for improved performance (13-ERD-055).

R&D 100 Awards

In 2014, LDRD-supported technologies received two of four awards presented to the Laboratory in the R&D 100 Awards competition. The process to select the winners is

demanding and takes almost a year. Awards are often chosen for their commercial potential or enduring value they will bring to the nation. Only 100 entrants are singled out, and often the Laboratory is listed more than once on the roster of winners. Since the Lab began submitting entries in 1978, it has received over 150 of these awards. This continuous recognition epitomizes science and technology on a mission, and shows the level of impact the Laboratory has on the nation.

Superconducting Tunnel Junction X-Ray Spectrometer. The superconducting tunnel junction x-ray spectrometer can measure x-ray energies ten times more precisely than current spectrometers based on silicon or germanium semiconductors. Built in conjunction with STAR Cyroelectronics in Santa Fe, New Mexico, the new spectrometer is a powerful tool for identifying unknown substances, such as traces of evidence from crime-scene samples, impurities in computer-chip materials, and toxic metals in biomedical components. This advanced science and technology was developed over a long period, with early support from LDRD, including a 1992 project on superconducting tunnel-junction x-ray detectors (92-SR-046).



Analyzing high-resolution spectra from the superconducting tunnel junction x-ray spectrometer (shown on the table) to characterize a material sample. The rack contains support electronics for spectrometer operation. (Photo by Roy Kaltschmidt of Lawrence Berkeley National Laboratory.)

Convergent Polishing. Optics for imaging systems, lithography, and fusion research at the National Ignition Facility can now be polished and finished more quickly and economically thanks to the convergent polishing system. The new system can finish flat and spherical glass optics in a single iteration, regardless of the workpiece's initial shape and without operator intervention. This work was supported by the LDRD Program with a project to determine the fundamentals of figure control and fracture-free finishing for high-aspect-ratio laser optics (11-ERD-036).

Optics polisher

Tibbetts Awards

Technology developed by Livermore LDRD teams helped two companies win the U.S. Small Business Administration's Tibbetts Award in 2014, which recognizes economic impact. The awards are given to companies based on whether they have met federal research and development needs, encouraged diverse participation in technological innovation, and increased the commercialization of federal research. STAR Cryoelectronics, LLC of Santa Fe, New Mexico and Inrad Optics of Northvale, New Jersey—both of which have licensed LLNL technologies—are 2 of 25 companies selected for the award. STAR Cryo is using superconducting tunnel junction detectors for high-resolution x-ray spectroscopy and Inrad is using solution-grown organic scintillators for neutron radiation detection.

Lawrence Livermore physicist and LDRD researcher Stephan Friedrich adjusts the operating conditions of the superconducting tunnel-junction x-ray detector.

High-Resolution X-Ray Spectroscopy. The superconducting tunnel junction x-ray detectors operate at very low temperatures, less than 1 K. This enables researchers to measure x-ray energies more precisely to reveal not only the composition of unknown materials, but also the chemical bonding state of elements in the sample. The technology was developed over a long period, with early support from LDRD, including a 1992 project on superconducting tunnel-junction x-ray detectors (92-SR-046). It has many potential applications in both research and industry that range from x-ray astronomy to material analysis. The detector can analyze impurities at the molecular scale, and offer more than a tenfold improvement in energy resolution as compared with conventional silicon or germanium detectors. To operate the detector, which looks like a large computer chip, STAR Cryo also developed an advanced refrigerator technology to precool the detector to 3 K, and then a two-stage adiabatic demagnetization refrigerator to obtain a base temperature below 0.1 K.

Lawrence Livermore physicist and LDRD researcher Natalia Zaitseva examines a single crystal growing in a solution-growth crystallizer developed for production of stilbene crystals for fast neutron detection.

Scintillators for Neutron Radiation Detection. Solution growth applied to stilbene crystals in an LDRD project investigating salicylic acid derivatives as a new class of scintillators for high-energy neutron detection (07-ERD-045) has led to a collaboration with Inrad Optics. The solution-grown crystals are faster and cheaper to produce than stilbene crystals using a melt-growth method. Stilbene is a single crystal used for a type of radiation detection known as pulse-shape discrimination, and can quickly detect neutrons in the presence of a strong gamma-ray background and have good luminescence properties. The melt-growth technique is a high-temperature process in which crystallization is conducted by cooling an initial liquid melt until it becomes a solid. During the solution-grown process developed by LLNL, stilbene is grown using crystallization tanks and seed holders to handle organic solutions to improve the growth process with controlled temperature reduction during the crystal's rotation. These crystals can potentially be used in portable and non-portable neutron radiation detection devices to detect illicit nuclear weapons at ports of entry, security checkpoints, and sensitive city installations, as well as scanning equipment for wide-area sweeping at offshore facilities or onboard ships. They can be used for monitoring nuclear power plants for dangerous or unhealthy levels of radiation from leakage. The solution-grown scintillator technology is important because it demonstrates a more scalable, more economical route for production of a material with superior properties for detection of fast neutrons. The company has begun commercial sales of stilbene

scintillation crystals. In addition to the Tibbetts Award, the LDRD principal investigator on the enabling project (Natalia Zaitseva) and Inrad Optics also were recently honored by the Department of Homeland Security's Domestic Nuclear Detection Office for exceptional contributions to advanced materials development for neutron detection of radiological and nuclear material, as well as receiving a regional award from the Federal Laboratory Consortium's Far West Region Competition. Their work was recognized as an "outstanding commercialization success," and collaborator Inrad Optics also concluded a work-for-others agreement with LLNL to assemble solution-growth equipment.

National Institutes of Health Director's Early Independence Award

Amanda Randles

Computational scientist and LDRD researcher Amanda Randles has received a Director's Early Independence Award from the National Institutes of Health. The award provides funding to encourage exceptional young scientists to pursue "high risk, high reward" independent research in biomedical and behavioral science. Randles will receive about $2.5 million over 5 years. The funding will allow her to pursue research to develop a way of predicting likely sites for cancer to metastasize—a method that combines massively parallel computational models and experimental approaches.

The goal of the project is to develop a method to simulate flow of realistic levels of cells through the circulatory system, thereby gaining insight into mechanisms that underlie disease progression and localization. Building a detailed, realistic model of human blood flow is a formidable mathematical and computational challenge requiring large-scale fluid models as well as explicit models of suspended bodies like red blood cells, which will require high-resolution modeling of cells in the blood stream, and necessitate significant computational advances. Randles will build on the HARVEY computer code, a parallel fluid-dynamics application designed to model hemodynamics in patient-specific geometries, with the goal of further validating computational results through rigorous comparison with in vivo and in vitro measurements.

Through this award, Randles hopes to be able to expand the scope of her projects to address not only vascular diseases, but also the movement of circulating tumor cells in the bloodstream. By studying the impact of cell characteristics on the movement of circulating tumor cells, researchers will gain better understanding of the mechanisms driving cancer metastasis to inform clinical decisions and improve treatment options. Randles, a Lawrence Fellow, works in the LLNL Computation Directorate's Center for Applied Scientific Computing. She is the principal investigator for an LDRD project validating large fluid-dynamics simulations of complex geometries with three-dimensional printing (15-LW-029).

E. O. Lawrence Awards

Two LDRD researchers were 2013 E. O. Lawrence Award laureates for their contributions to the DOE's missions in science, energy, and national security. The award, established to honor the memory of E. O. Lawrence, is intended to recognize mid-career U.S. scientists and engineers for exceptional research and development contributions in support of the DOE.

Stephen Myers

Stephen Myers was recognized for his work on developing seismic monitoring technologies to locate nuclear explosions, and most recently was a co-investigator for an LDRD project on creating optimal fracture networks for energy extraction (11-SI-006). Myers was cited for his leadership in developing the Regional Seismic Travel Time Model and Computing Code that has been used by the Comprehensive Nuclear-Test-Ban Treaty Organization to monitor nuclear events around the world, including North Korea.

Siegfried Glenzer

Siegfried Glazer was recognized for his work in fusion and plasma sciences, and is currently a co-investigator for LDRD project 13-ERD-073 on generation and characterization of matter at extreme gigabar pressures at Livermore's National Ignition Facility. Glenzer and his collaborators were among the first to perform experiments at the facility, beginning with early light in 2004 to full-scale inertial-confinement fusion target capsule experiments from 2008 to 2010. After the successful demonstration of the required target capsule radiation temperature and radiation symmetry, they fielded the first implosions with thermonuclear fuel.

Lawrence Livermore Director William Goldstein, acknowledged the "award continues to validate the impact Lawrence Livermore researchers have made on the science and technology that enhances our national security."

DOE Office of Science Early Career Research Program

Researchers Todd Gamblin and Jennifer Pett-Ridge have been selected by DOE to receive Office of Science Early Career Research Program awards for 2014. The two winners were selected by the Office of Advanced Scientific Computing Research and the Office of Biological and Environmental Research, respectively. These awards provide $500,000 per year for 5 years to support outstanding scientists early in their careers working in disciplines supported by the DOE Office of Science.

Todd Gamblin is a co-investigator for an LDRD project, "Task Mapping on Complex Computer Network Topologies for Improved Performance" (13-ERD-055).

Jennifer Pett-Ridge is a co-investigator for LDRD projects 13-LW-032, "Wonder Bugs and the Carbon Cycle: Characterizing the Carbon Metabolism of Thaumarchaeota" and 14-ERD-038, "Wetlands as a Source of Atmospheric Methane: A Multiscale and Multidisciplinary Approach."

This year, 35 awardees were selected from a pool of about 750 applicants.

Todd Gamblin

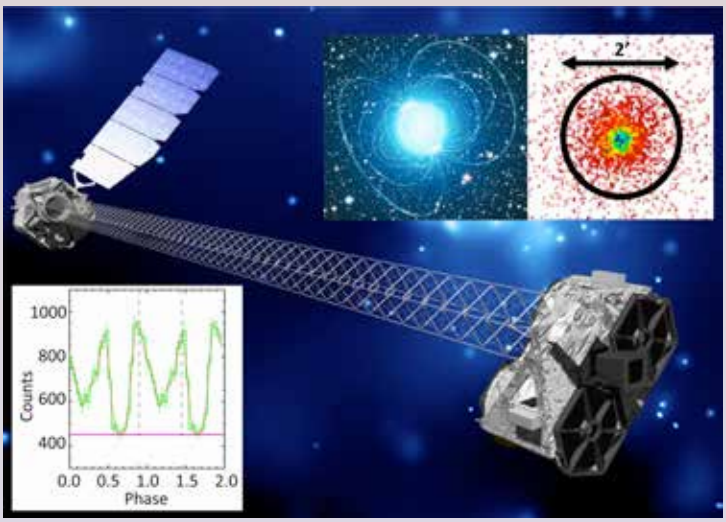
Lawrence Livermore scientist Jennifer Pett-Ridge will receive funding through the U.S. Department of Energy's Office of Science Early Career Research Program for her research in soil microbial communities and carbon cycling in the tropics.

Defense Programs Award of Excellence

Philip Pagoria, an LDRD researcher, was selected in 2014 to receive a Defense Programs Award of Excellence for work performed in 2012 as a member of the TATB Technical Working Group that supports both the DOE and the Department of Defense. The awards are presented by the NNSA Defense Programs for outstanding contributions to the nation's nuclear weapons program. The TATB Technical Working Group was credited with enabling production-scale qualification and establishment of a TATB (triaminotrinitrobenzene explosive) supply that led to the restoration of a full-scale production capability for a material that is critical to the safety of both conventional and nuclear weapon stockpiles. Pagoria concluded an LDRD project in 2014 that examined and developed new energetic materials (12-ERD-066).

European Physical Society High Energy and Particle Physics Prize

Team members Jeffrey Gronberg, Finn Rebassoo, Jonathon Hollar, Mike Albrow, David Lange, and principal investigator Douglas Wright received the 2013 High Energy and Particle Physics Prize from the European Physical Society as part of the ATLAS (A Toroidal LHC Apparatus) and CMS (Compact Muon Solenoid) experimental collaborations for the CERN (European Organization for Nuclear Research) that required the collective efforts of over 3,000 physicists and engineers for each experiment and led to the discovery of a new heavy particle at a mass of around 125 GeV. The observation required the creation of experiments of unprecedented capability and complexity, designed to discern the signatures that correspond to the elusive Higgs boson elementary particle.
The efforts required the use, and in many cases the development, of cutting-edge technologies. In addition, the gigantesque structures were supplemented with appropriate software and computing systems that enabled the analysis of the vast amounts of data that had to be collected. The Livermore researchers were part of an LDRD project exploring the path to discovery at the Large Hadron Collider (12-ERD-051) .



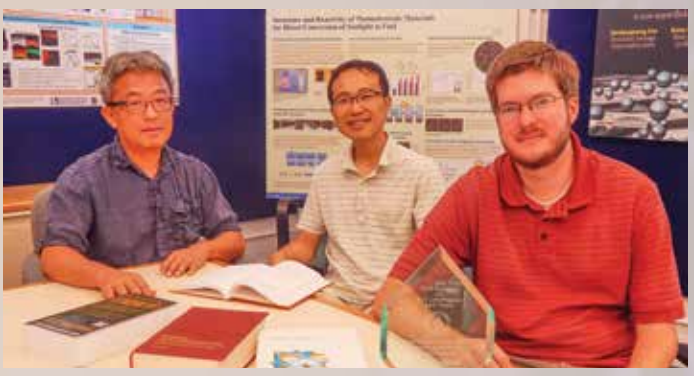
The Nuclear Spectroscopic Telescope Array

NASA Group Achievement Award

In August 2014, LDRD NuSTAR (Nuclear Spectroscopic Telescope Array) science team researchers Julia Vogel, Michael Pivovaroff, and Victoria Kaspi received a NASA Group Achievement Award. Their work was supported by the "Neutron Star Science with the Nuclear Spectroscopic Telescope Array" (13-ERD-029) LDRD project. The NuSTAR mission has already advanced our understanding about how galaxies in the universe form and evolve, and observed some of the hottest, densest, and most energetic objects in the universe, including black holes, their high-speed particle jets, supernova remnants, and the Sun.

DOE Hydrogen and Fuel Cells Program R&D Award

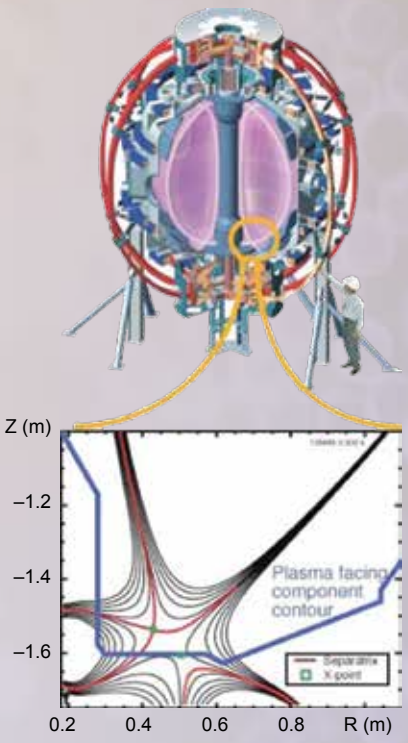
Livermore LDRD researchers Tadashi Ogitsu, Wooni Choi, and Brandon Wood have been recognized by the Hydrogen and Fuel Cells Program of DOE's Office of Energy Efficiency and Renewable Energy in 2014 for their "outstanding dedication and collaboration in photo-electrochemical surface validation." Their ground-breaking work, which integrates state-of-the art tools and methods in materials theory, synthesis, and characterization, has led to the development and validation of novel foundational models of photo-electrochemical solar-hydrogen production and corrosion processes. These models have been crucial to the development of corrosion mitigation strategies for high-efficiency photo-electrochemical devices to meet the DOE's ultimate cost targets in renewable hydrogen production. Livermore LDRD support for the effort was provided by a project that examined the interface between water and semiconductors for photo-electrochemical hydrogen production (11-ERD-073).



Tadashi Ogitsu, Wooni Choi, and Brandon Wood (left to right) were recognized with a DOE 2014 Hydrogen Production and Fuel Cells Program R&D Award from the DOE Office of Energy Efficiency and Renewable Energy.

International Atomic Energy Agency Nuclear Fusion Award Shortlist

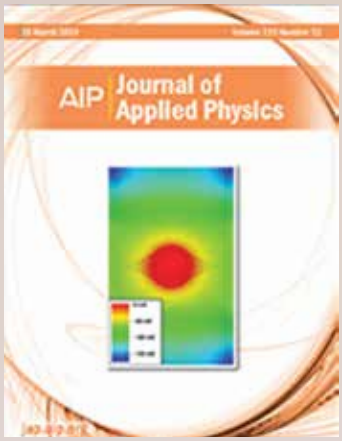
"Taming the Plasma–Material interface with the 'Snowflake' Divertor in NSTX" was selected as one of the top papers for recognition at the International Atomic Energy Agency's Fusion Energy Conference for the 2014 Nuclear Fusion Award. The shortlist consisted of 11 papers judged to be of the highest scientific standard, selected from the journal volume published 2 years previous to the award year. Nominations were based on citation record and recommendation by the board of editors. The snowflake divertor configuration was originally proposed by Dimitri Ryutov in 2007 as a possible solution for the plasma–material interface problem in magnetically confined fusion plasma devices (tokamaks), and research was supported by the LDRD project, "Innovative Divertors for Future Fusion Devices" (08-ERD-019). The project's snowflake research was successfully implemented by LLNL-led research teams in the National Spherical Torus Experiment at Princeton Plasma Physics Laboratory in Princeton, New Jersey and in the DIII-D National Fusion Facility at General Atomics in San Diego, California. The research team led by Dimitri Ryutov also won the R&D 100 Award for "The Snowflake Power Divertor" in 2012.



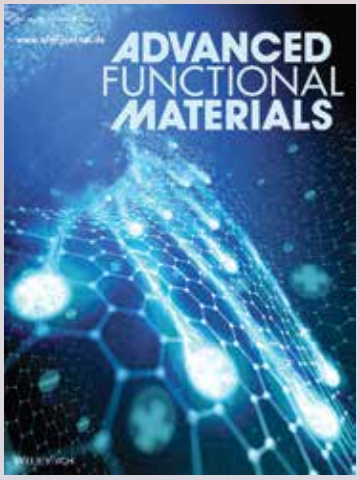
Snowflake divertor

Journal Covers

In addition to the numerous awards LDRD researchers garner each year, innovative science, technology, and engineering is evidenced by both the number of scientific journal articles and front-cover features in high-visibility journals such as *Nature* documenting LDRD Program project results. In 2014, several LDRD projects were featured on the covers of peer-reviewed journals.

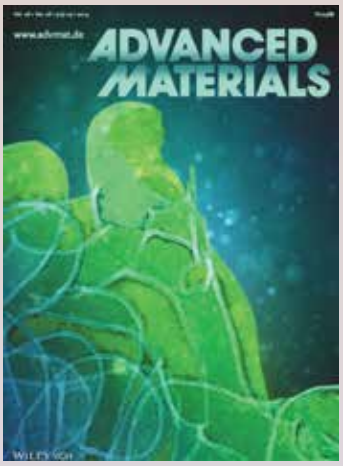


A March 2014 issue of the ***Journal of Applied Physics***, featured research by LDRD researchers Salmaan Baxamusa and colleagues describing the effects of self-heating and phase change on the thermal profile of hydrogen isotopes in confined geometries. The effort was supported by an LDRD project examining "Hydrogen Ice Layers for Inertial-Confinement Fusion Targets" (12-ER-032). The generation of single-seed crystals in place is an important step in the growth of high-quality hydrogen layers in capsules used for inertial-confinement fusion research. It is accomplished by forming a polycrystalline solid layer inside the shell, slowly melting it until a single crystal remains, and subsequently using this crystal as a seed for the growth of a solid hydrogen crystal from its melt. In the paper, the team shows that latent heat effects and by-products of the helium-3 beta decay influence the thermal environment inside the shell, and, thereby dictate the location of the seed crystal.

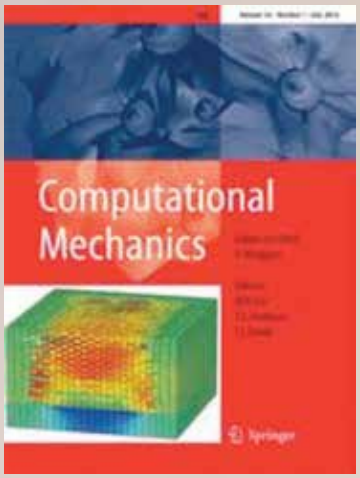


In the cover article of a June 2014 issue of ***Advanced Functional Materials***, LDRD principal investigator Juergen Biener and colleagues showed that the electrical conductance of centimeter-sized three-dimensional nanometer-scale graphene samples (atomic-scale honeycomb lattice made of carbon) can be dynamically controlled by changing the surface charge density. They demonstrated that a fully reversible change in conductance of up to several hundred percent can be achieved by imposing a potential of less than 1 V. The observed conductivity change can be explained by the electrochemically induced accumulation and depletion of charge carriers, plus variation in the carrier mobility because of changes in the density of defects. These results open the door to novel applications of bulk graphene materials such as low-voltage and high-power tunable resistors. Work was supported by LDRD research into dynamic control over electronic transport in three-dimensional bulk nanometer-scale graphene via interfacial charging (12-ERD-035).

The July 2014 cover of ***Nature*** featured an LDRD project in which the researchers describe their ramp-compression measurements for diamond, achieving a peak pressure equivalent to 50 million atmospheres. Diamond, the least compressible material known, provided the first experimental data for constraining condensed-matter theory and planet-evolution models in the terapascal regime (roughly the equivalent of over two million elephants balanced on a single square foot). By realizing the adiabatic conditions of dynamic compression with a loading profile soft enough to avoid shock formation (a nearly fluid-like response of the sample such that strength and dissipation are minimal), the research team was able to document an approach for taking solids to the long-sought high-density conditions of statistical-electron theory. Overall, the research project produced several high-energy-density experimental firsts as well as new diagnostics for "Extreme Compression Science" (12-SI-007).



In a paper featured on the cover of the July 23 online edition of ***Advanced Materials***, LDRD researchers report on the synthesis of ultralow-density, ultrahigh-surface area aluminum oxide and titanium oxide bulk materials that have an interconnected nanometer-scale tubular morphology. The authors have developed an atomic-layer method for making strong, ultralow density materials of novel composition, shown on the cover and based on the LDRD project, "Nanometer-Scale Porous Designer Materials" (13-LW-031). Unlocking the full potential of ultralow-density materials requires the ability to make mechanically robust architectures with deterministic control over cell size, density, and composition. A high strength-to-weight ratio makes interconnected tubular networks an attractive approach for overcoming the decrease in strength and stiffness of low-density materials as their porosity increases.



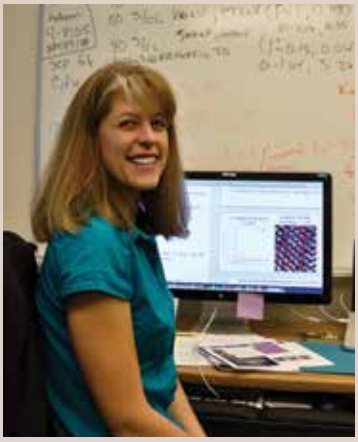
"Implementation of a Thermomechanical Model for the Simulation of Selective Laser Melting," a July 2014 cover article for ***Computational Mechanics***, received support from an LDRD project developing new approaches to accelerated certification of additively manufactured metals (13-SI-002). The paper examined selective laser melting as an additive manufacturing process in which multiple, successive layers of metal powders are heated via laser to build a part. Modeling of the process requires consideration of both heat transfer and solid mechanics. The researchers presented work describing continuum modeling of selective laser melting as envisioned for eventual support of part-scale modeling of the process to determine end-state information such as residual stresses and distortion. Determining evolving temperatures is dependent on the material, the state of the material (powder or solid), the specified heating, and the configuration. Similarly, the current configuration is dependent on the temperatures, the powder-solid state, and the constitutive models. A multiphysics numerical formulation is required to solve such problems. The article described the problem formulation, numerical method, and constitutive parameters necessary to solve such a problem.

Best Geothermal Energy Presentation

Livermore LDRD researcher Wyatt DuFrane was selected for an outstanding technical presentation from the global geothermal community for 2014 by the Geothermal Resources Council. He is a co-investigator for an LDRD project exploring reactive materials for hydraulic fracturing (13-ERD-029). Judging criteria included technical content, quality of visual aids, and presenter's ability to communicate the subject matter at the Geothermal Research Council's 38th annual meeting held Fall 2014 in Portland, Oregon, with over 1,400 attendees from 39 different countries. The presentation was "Smart Tracers for Geothermal Reservoir Assessment," and the principal investigator for the project was Roger Aines. Other cited for the presentation included John Vericella, Eric Duoss, Megan Smith, and Jeff Roberts.

Perspectives from Early-Career Investigators

The LDRD Program is a major vehicle for attracting, enabling, and retaining new technical staff at the Laboratory. Three outstanding LDRD investigators—Heather Whitley, Miguel Morales-Silva, and Gianpaolo Carosi—exemplify LDRD-funded early-career scientists who are already making a difference in their fields.



Heather Whitley

Theoretical chemist Heather Whitley came to the Laboratory in 2007 as a postdoctoral researcher from the University of California, Berkeley. She originally used supercomputer simulations to study the properties of semiconductor nanomaterials that could eventually be applicable to solar cells and other clean-energy technologies. Taking advantage of her supercomputer simulation expertise, Whitley began to use simulations to enhance the fundamental understanding of complex plasmas, and joined the Laboratory in 2011 as a staff member.

In 2012 Whitley received a Presidential Early Carer Award for Science and Engineering, considered the highest honor bestowed by the U.S. government on early-career science and engineering professionals, for applying Monte Carlo techniques to produce very accurate quantum statistical potentials for use in molecular dynamic codes. She currently works to expand the understanding of dense plasma microphysics for both stockpile stewardship and experiments on the National Ignition Facility at Livermore. Whitley has been a co-investigator on two LDRD efforts. In the first, she applied a recently developed massively parallel molecular dynamics code to investigate the validity of specific models for plasma thermal conductivity (12-SI-005). In a second LDRD effort to start in 2015, the molecular dynamics code will be extended to include multiscale models for more accurate simulations of transport across material interfaces (15-ERD-052).

"We're looking at the atomic level to learn about the physics of atoms, ions, and electrons with our simulations," says Whitley." The properties of all matter depend on what happens at this level. We are using simulations to function as a virtual laboratory to achieve high-energy-density regimes difficult to attain experimentally." The research will likely benefit stockpile stewardship by providing a better understanding of the physics underlying aging nuclear weapons. "An ideal way to improve codes is to make connections between theory, simulation, and experiment," adds Whitley." The LDRD process is extremely competitive. Although one must devote a great deal of energy to prepare a proposal, it is very worthwhile to receive funding," observes Whitley." The LDRD Program provides the Laboratory with a way to bring in new talent and ideas."

Miguel Morales-Silva

Physicist Miguel Morales-Silva joined the Laboratory in 2010 after earning his Ph.D. in physics from the University of Illinois at Champaign-Urbana in 2009. During graduate school, Morales-Silva worked on a promising but challenging method (quantum Monte Carlo calculations) to analyze the behavior of hydrogen at high temperatures and pressures, especially when it exhibits metal-like behavior. Livermore's LDRD program provided him the funding to pursue this method at lower temperatures and very high pressures. For his work that enabled the use of advanced computational techniques to study materials at extreme conditions, Morales-Silva received a Presidential Early Career Award for Science and Engineering in 2013.

Hydrogen is the simplest element in the universe but has fascinating properties that have puzzled scientists for decades. "Metallic hydrogen represents the holy grail of high-pressure physics," says Morales-Silva. Despite decades of efforts from experimental groups, details are lacking about this novel state of hydrogen. "It's hard to find answers about metallic hydrogen experimentally. We need supercomputers for this research, but our calculational methods are lagging. We must complement experiments by improving quantum Monte Carlo methods as an alternative to first-principles calculations." He believes the results from an LDRD project for which he is the principal investigator will eventually enhance the accuracy of the Livermore codes and models used in stockpile stewardship science (13-LW-004). In addition, this research may provide scientists with a better understanding of planet formation. Morales-Silva is co-investigator on another LDRD effort that uses quantum Monte Carlo calculations to predict the behavior of elements heavier than hydrogen (13-ERD-067). "We're trying to push the applications of quantum Monte Carlo methods across the periodic table of elements. We want to predict with high confidence the properties of elements solely with simulation."

Morales-Silva says, "The LDRD Program is essential to attracting people doing basic science that will be critical in the future to Livermore programs. I am confident our work will eventually prove useful to the programs. I am doing my best science and writing my best papers." He adds, "Without the LDRD Program, for someone such as myself who is interested in basic science, the Laboratory would be much less attractive. This LDRD has helped me to become an independent researcher and has provided an important first step in my career."

Gianpaolo Carosi

Physicist Gianpaolo Carosi, who received his Ph.D. from the Massachusetts Institute of Technology, came to the Laboratory in 2006. Carosi was co-investigator on a 2009 LDRD project that involved the groundbreaking Axion Dark Matter Experiment, established at Livermore with LDRD support (09-ERD-052). The experiment is currently centered at the University of Washington, and Carosi is a major scientific collaborator. The research team seeks to detect hypothetical dark-matter elementary particles called axions. Dark matter is estimated to make up about 23% of the energy density of the universe (the rest being a mysterious repulsive "dark energy" and ordinary matter). The experiment is designed to find these dark matter axions by measuring their decay into a microwave-frequency photon in the presence of a strong magnetic field.

Because the expected signal from the decay of an axion is so faint, sensitive amplifiers are needed to boost the signal to a detectable level. The LDRD involving Carosi resulted in the design of microstrip superconducting quantum interference devices (SQUIDs), which are tiny rings of superconducting metal that serve as extremely low-noise amplifiers when cooled to near absolute zero. The SQUID amplifiers are designed to magnify the extremely small power signals emitted by axions in a 1-m-tall microwave cavity structure built by Carosi. This promising approach earned him a 2012 DOE Early Career Program award. "Our SQUID amplifier relies on quantum mechanics to make the quietest microwave receiver possible," he says. He compares the SQUID amplifier to an ultrasensitive radio that minimizes static hiss from background noise to allow very weak, distant stations (from the decay of axions) to be heard. Discovery of an axion would further understanding of dark matter, the nature of quantum physics, and the force that binds atomic nuclei. SQUIDS also have potential applications in oil exploration, brain imaging, quantum computing, and secure communications.

Carosi was also part of an LDRD effort to develop rare-event detectors for nuclear science and security (10-SI-015). This project focused on detecting neutral particles such as neutrinos and antineutrinos emitted by nuclear materials and reactors. "We eventually would like to build a worldwide neutrino monitoring program," says Carosi. Remote nuclear reactor monitoring using advanced detectors could revolutionize global nuclear nonproliferation efforts by monitoring reactors hundreds of kilometers away and detecting fissile material from hundreds of meters away. Carosi compares Lawrence Livermore's LDRD process to national competitions for federal research funding. The competition is stiff, and only the very best ideas are awarded funding. He says, "It's one of the most important factors that attracts postdocs, and it allows risky new ideas to percolate."

Program Metrics

Projects sponsored by LDRD contribute significantly to intellectual property, publications, collaborations, and recruitment of postdoctoral researchers at Lawrence Livermore, considering that the program represents a small portion of the Laboratory's total budget. In FY14, LDRD costs at LLNL were $80.1M, which is 5.6% of total Laboratory costs. Here, we present annual performance indicators specified in roles, responsibilities, and guidelines for LDRD at the DOE/NNSA laboratories under DOE Order 413.2B.

Intellectual Property

The number of patents resulting from LDRD-funded research since FY10 and the percentage of total patents that were derived from LDRD research and development is shown in the table below. The fiscal year for which a patent is listed is the year in which the patent was granted—LDRD investment in a technology is typically made several years before the technology is actually patented. Furthermore, although an LDRD-sponsored project makes essential contributions to such technologies, subsequent programmatic sponsorship also contributes to a technology's further development. In FY14, LDRD projects generated 44% of Livermore's total patents, even though the LDRD program was 5.6% of the Laboratory's budget.

Patents resulting from LDRD-funded research as a percentage of all LLNL patents for the last five fiscal years.

Patents	FY10	FY11	FY12	FY13	FY14
All LLNL patents	54	60	78	84	105
LDRD patents	27	32	35	44	46
LDRD patents as percentage of total	50%	53%	45%	52%	44%

Records of invention submitted by LDRD researchers also account for a significant percentage of the total for the Laboratory. Overall, LDRD records of invention for FY10 to FY12 account for 44% of the 728 total. In FY14, there were 86 records submitted at Livermore, with 45 (52%) of those attributable to LDRD-supported projects.

Records of invention resulting from LDRD-funded research as a percentage of all LLNL records for the last five fiscal years.

Record of Invention	FY10	FY11	FY12	FY13	FY14
All LLNL records	160	164	162	156	86
LDRD records	66	59	79	68	45
LDRD records as percentage of total	41%	36%	49%	44%	52%

Finally, LDRD plays a role in producing Laboratory copyrighted material. From FY10 to FY14, LDRD-supported projects accounted for over 27% of the 322 Livermore copyrights. In FY14, there were 73 LLNL copyrights, with 21 (29%) that could be attributed to LDRD research.

Publications in Scientific Journals

The LDRD publications in scientific journals demonstrate that research and development under LDRD furthers the progress of the broad scientific and technical community by contributing new scientific results, innovative technologies, and fundamental breakthroughs. In a typical year, Laboratory scientists and engineers collectively publish around 1,000 papers in a wide range of peer-reviewed journals. In FY14 there were 985 such articles, of which at least 245 (25%) resulted from LDRD projects. Over the last several years, the percentage of LDRD-supported articles has remained relatively consistent, with a 5-year average of over 23% of total Laboratory publications. The following table shows the number of journal articles per fiscal year resulting from LDRD-funded research since FY10, and the percentage of total articles that were derived from LDRD research and development.

Journal Articles	FY10	FY11	FY12	FY13	FY14
All LLNL articles	966	994	1,016	1,155	985
LDRD articles	227	207	230	293	245
LDRD articles as percentage of total	23%	21%	23%	25%	25%

Journal papers resulting from LDRD-funded research as a percentage of all LLNL papers for the last five fiscal years.

Collaborations

External collaborations are essential to the conduct of research and development in LDRD. By collaborating formally and informally with other national laboratories, academia, and industry, LDRD investigators are able to access world-leading facilities and knowledge—both in the U.S. and abroad—and serve as active and prominent members of the broad scientific and technical community. External collaborations are also vital for assembling the best teams for pursuing many research and development opportunities, by complementing LLNL's capabilities and expertise. In addition, LDRD collaborations create strong relationships that are valuable for the Laboratory's pipeline for recruiting scientific and engineering personnel.

The FY14 portfolio included 69 formal LDRD-funded collaborations involving 39 LDRD projects (27% of the total projects funded). Collaborating institutions included the University of California (21% of total collaborators), other academic institutions (69%), and other collaborators such as government agencies and industry (10%). These statistics do not include the numerous informal collaborations that researchers pursue in the course of their LDRD projects.

Postdoctoral Researchers

Because LDRD funds exciting, potentially high-payoff projects at the forefront of science, the program is essential for recruiting top talent in new and emerging fields of science and technology. In FY14, the LDRD Program supported 68% of the Laboratory postdoctoral researchers—there was an average of 139 postdoctoral researchers at LLNL in FY14, of which 95 were supported in some way by LDRD projects. The Laboratory continues significant recruitment efforts to maintain the total number of postdoctoral researchers.

Program

Mission

To fulfill its missions, LLNL must continually invest in the science and technology that form the foundation of its signature capabilities. The LDRD Program, which was established by Congress at all DOE national laboratories in 1991, is LLNL's most important single resource for fostering excellence in science and technology for today's needs and tomorrow's challenges.

According to its Congressional mandate,[1] the purpose of LDRD is to foster excellence in science and technology that (1) supports the DOE/NNSA and LLNL missions and strategic vision, (2) ensures the technical vitality of the Laboratory, (3) attracts and maintains the most qualified scientists and engineers and allows scientific and technical staff to enhance their skills and expertise, (4) helps meet evolving DOE/NNSA and national security needs, and (5) enables scientific collaborations with academia, industry, and other government laboratories.

By enabling LLNL to fund creative fundamental and applied research activities in areas aligned with its missions, the LDRD Program develops and extends the Laboratory's intellectual foundations and maintains its vitality as a premier research institution. The present scientific and technical strengths of LLNL are, in large part, a product of LDRD investment choices in the past.

The value of LDRD to DOE as well as to the country has been clearly articulated. According to a National Academy of Sciences report to DOE in 2012, "A crucial part of the Laboratories' ability to conduct their missions is derived from Laboratory Directed Research and Development (LDRD), the primary source for internally directed R&D funding. Among its other benefits, LDRD provides a major resource for supporting and training staff at each Laboratory."[2] The DOE 2014 report to Congress notes "The LDRD Program provides the laboratories with the opportunity and flexibility to establish and maintain an environment that encourages and supports creativity and innovation, and contributes to their long-term viability. LDRD allows the Department's laboratories to position themselves to advance our national security mission and respond to our Nation's future research needs."[3]

At LLNL in 2014, Laboratory Director William Goldstein and acting Deputy Director for Science and Technology Greg Suski were responsible for the LDRD Program. Execution of the program was delegated to the Senior Advisor to the Director, Rokaya Al-Ayat. The LDRD Program at LLNL is in compliance with DOE Order 413.2B and other relevant DOE orders and guidelines.

[1] *U.S. Department of Energy Order 413.2B, Laboratory Directed Research and Development* (https://www.directives.doe.gov/directives/0413.2-BOrder-b/view) (Retrieved March 18, 2015).

[2] *Managing for High-Quality Science and Engineering at the NNSA National Security Laboratories* (http://www.nap.edu/openbook.php?record_id=13367&page=R1) (retrieved March 18, 2015).

[3] *Fiscal Year 2014 Report on Laboratory Directed Research and Development (LDRD) at the DOE National Laboratories, Report to Congress, December 2014* (http://energy.gov/sites/prod/files/2015/01/f19/2014LDRDReportToCongress.pdf) (retrieved March 18, 2015).

Program Structure

Strategic Initiative

The SI project category, which is open to all Laboratory scientific, engineering, and programmatic staff, focuses on innovative research and development activities that address major specific science and technology challenges of high potential strategic impact for the *Investment Strategy for Science, Technology and Engineering*, and significantly enhance the Laboratory's science and technology base. Projects in this category are usually larger and more technically challenging than those in the other categories. All new and current SIs must be aligned with at least one of the mission focus areas or underlying science, technology, and engineering core competency.

Exploratory Research

The ER project category is designed to help fulfill the strategic research and development needs of a Laboratory directorate (ERD) and must also support and be aligned with the Laboratory's strategic plan. As with all the LDRD project categories, ER proposals must meet the criteria for intellectual merit used across the scientific community, such as importance of the proposed activity to advancing knowledge, capability, and understanding within its own field or across different fields, as well as ensuring the proposed activity suggests and explores creative and original concepts.

Laboratory-Wide Competition

Projects in the LW category emphasize innovative research concepts and ideas and undergo limited management filtering to encourage creativity of individual researchers. The LW competition is open to all LLNL staff in programmatic, scientific, engineering, and technical support areas. Direct alignment with the Laboratory's strategic plan is not required for LW proposals. However, in order to be funded, all LW proposals must be relevant to one or more missions of the DOE and NNSA.

Feasibility Study/Project Definition

This special project category, FS, provides researchers with the flexibility to propose relatively small, short-term projects to determine the feasibility of a particular technical approach for addressing a mission-relevant science and technology challenge. To increase its responsiveness to Laboratory scientists and engineers, the LDRD Program funds FS projects throughout the year, with a one-year funding limit.

Project Competency Areas

Although LDRD projects often address more than one scientific discipline, each project is assigned to 1 of 14 research categories aligned with the Laboratory's science and technology investment strategy. The 14 categories are:

- Advanced Materials and Manufacturing
- Bioscience and Bioengineering
- Biosecurity
- Chemical and Isotopic Signatures
- Computational Science and Engineering
- Cyber Security, Space, and Intelligence
- Energy and Climate
- High-Energy-Density Science
- High-Performance Computing
- Inertial-Confinement Fusion Science and Technology
- Information Systems and Data Science
- Lasers and Optical Materials Science and Technology
- Nuclear Science and Technology
- Stockpile Stewardship

Strategic Context for the FY14 Portfolio

The FY14 LDRD portfolio-management process at LLNL was structured to ensure alignment with the DOE, NNSA, and Laboratory missions. This process involved (1) a top-level strategic planning process to identify strategic science and technology areas for LDRD investment, (2) a call to the Laboratory scientific and technical community for innovative and relevant proposals within the DOE/NNSA mission areas, and (3) a scientific peer-review process to select the highest quality LDRD portfolio from these proposals.

In 2009, the Laboratory director called for the development of a new scientific and technical investment strategy that sets institutional strategic goals and identifies science and technology needs in selected mission focus areas, in fundamental research, and in critical science, technology, and engineering capabilities. The strategy was developed by multidisciplinary teams under the guidance of the deputy director for science and technology. The Laboratory's updated *Investment Strategy for Science, Technology and Engineering* document, which is revised periodically to respond to our evolving mission needs, and set the strategic context for the FY14 LDRD competition. Further strategic context is provided by the *U.S. Department of Energy Strategic Plan, 2014–2018*[4] and by *The National Nuclear Security Administration Strategic Plan, May 2011*.[5] The DOE strategic plan articulates strategic themes for achieving the DOE mission of discovering solutions to power and secure America's future. In FY14, the Laboratory's LDRD Program strongly supported DOE strategic themes:

[4] *U.S. Department of Energy Strategic Plan, 2014–2018.* (http://energy.gov/sites/prod/files/2014/04/f14/2014_dept_energy_strategic_plan.pdf) (retrieved March 18, 2015).

[5] *The National Nuclear Security Administration Strategic Plan, May 2011* (http://nnsa.energy.gov/sites/default/files/nnsa/inlinefiles/2011_NNSA_Strat_Plan.pdf) (retrieved March 18, 2015).

1. **Energy and Environmental Security**—Catalyze the timely, material, and efficient transformation of the nation's energy system and secure U.S. leadership in clean energy technologies
2. **Nuclear Security**—Enhance nuclear security through defense, nonproliferation, and environmental efforts
3. **Scientific Discovery and Innovation**—Maintain a vibrant U.S. effort in science and engineering as a cornerstone of our economic prosperity with clear leadership in strategic areas

The DOE and NNSA oversee the Laboratory's LDRD Program to ensure that it accomplishes its objectives. This oversight includes field and headquarters reviews of both the technical content and management processes.

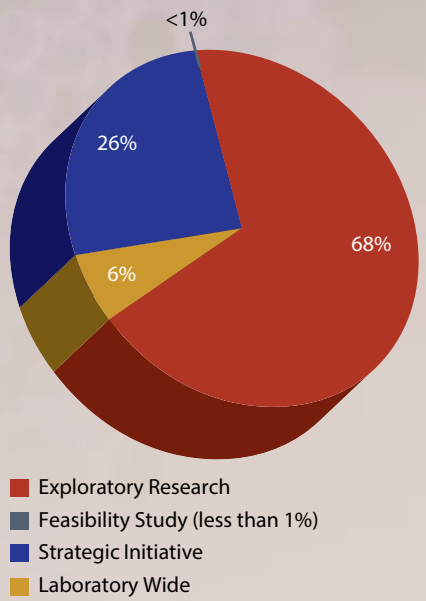
Program Audit by the DOE Office of Inspector General

In February 2014, the DOE Office of Inspector General, Western Audits Division, began an audit at Lawrence Livermore to determine whether NNSA laboratories are effectively managing LDRD projects. The initial investigation at LLNL would determine if fieldwork was also required at Los Alamos and Sandia national laboratories. For the next six months, the auditors examined management reports issued by LLNL and the DOE field office from 2008 to 2014, including reviews of the internal control structure, guidance, regulations, and procedures used to justify LDRD funding for proposals. They also examined the DOE Livermore Field Office and headquarters oversight processes and interviewed a selected set of LLNL investigators.

In addition, the LDRD Program office at LLNL provided a summary data sheet for each research project conducted for the period (from 2008 to 2014), overall LDRD funding, annual reports, and program plans. Final reports and initial proposals were also examined, as needed. Finally, performance measures, peer review processes, as well as Laboratory strategic plans were provided to demonstrate how LDRD projects met LLNL needs and supported missions and benefited the DOE. A particular focus was on the extent to which the DOE and the Laboratory processes provide the necessary internal control to ensure the effective management of LDRD-funded projects.

In July 2014, the DOE Office of Inspector General auditors completed the survey and verification phases of the audit. A final audit report was completed in November 2014, and it was concluded "that LLNL and NNSA had implemented performance/quality and financial monitoring controls." In addition, "Nothing came to our attention to indicate that controls were not in place over initial LDRD project approval and subsequent project management as required by DOE Order 413.2B and LLNL's internal procedures."[6]

[6] *Audit Report —Lawrence Livermore National Laboratory's Laboratory Directed Research and Development Program,* U.S. Department of Energy, Office of Inspector General, Office of Audits and Inspections, OAS-L-15-04, November 2014.



Distribution of funding among the LDRD project categories. Total funding allocated in FY14 was $78.2M.

Structure of the FY14 Portfolio

The FY14 LDRD portfolio was carefully structured to continue the LDRD Program's vigorous support for the strategic vision and long-term goals of DOE, NNSA, and LLNL. The projects described in this annual report underwent a stringent peer-review selection process and received ongoing management oversight.

In FY14 the LDRD Program funded 147 projects for a total allocation of $78.2M. The distribution of funding among the LDRD project categories is shown in the pie chart on the left.

The top bar chart on the next page shows the funding distribution by dollar amount for the 147 FY14 projects—over 65% of the projects were in the $101 to $500K range, with less than 2% falling below $100K. Projects in the $501K to $1M funding range accounted for almost 22% of the total, and over 11% of the projects received more than $1M. The average funding level for the 147 projects was about $532K.

Percentage of LDRD funding and number of projects for each research category for FY14 are shown in the bottom chart on the next page, with the core competency of advanced materials and manufacturing representing the largest project category at 18%, and chemical and isotopic signatures being the smallest at less than 1% of the total number of projects.

Strategic Initiative

In FY14, the LDRD Program funded 13 SI projects. Although the SI category represented just about 9% of the total number of LDRD projects for FY14, it accounted for over 26% of the budget. The SI projects were funded up to $2.7M.

Exploratory Research

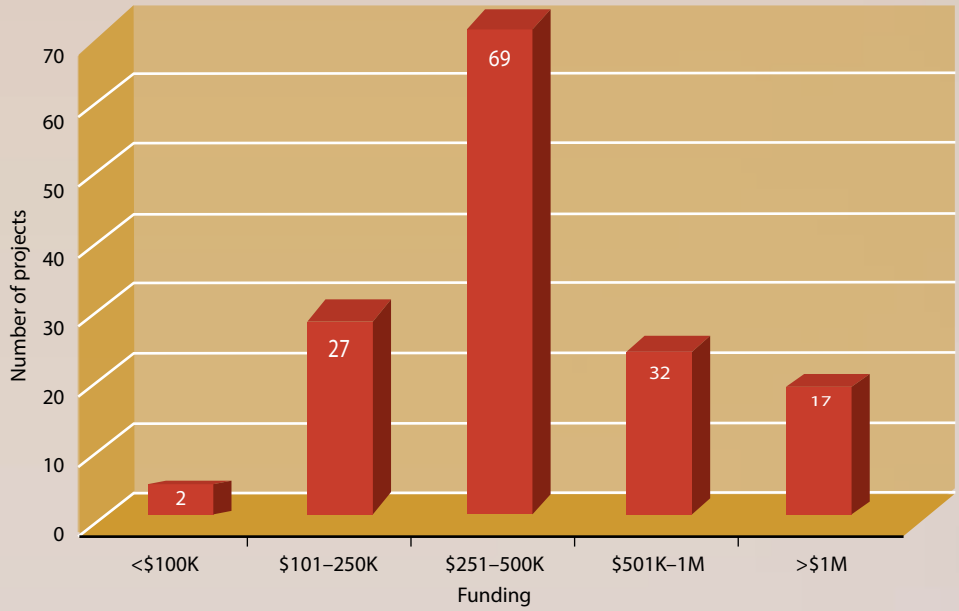
The LDRD Program funded 115 ER projects for FY14. The largest project category, ERs accounted for over 78% of the number of LDRD projects and over 68% of the budget for the fiscal year. Projects in this year's ER category were funded up to $1.6M.

Laboratory-Wide Competition

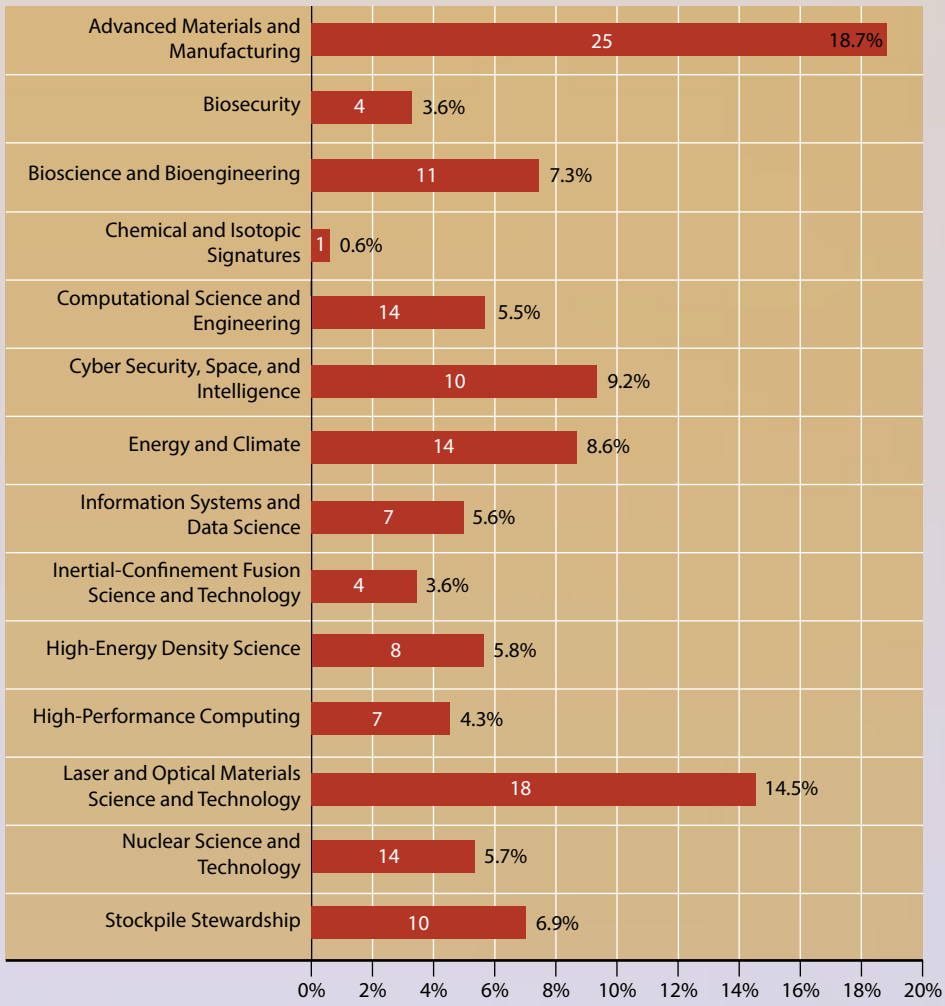
In FY14, 18 LW projects were funded, which represent slightly over 12% of the LDRD projects for the year and almost 6% of the budget. The LW projects for FY14 were funded up to $299K.

Feasibility Study

The LDRD Program funded only 1 FS project in FY14, which represents less than 1% of the LDRD projects for the year and less than 1% of the budget, with funding at $11K.



Number of projects and levels of funding. The average funding level for an LDRD project in FY14 was $532K.



Percentage of LDRD funding and number of projects in each research category in FY14.

Advanced Materials and Manufacturing

FY2014

Lawrence Livermore National Laboratory

Novel Rare Earth Permanent Magnets

Scott McCall (12-ERD-013)

Abstract

Recent restrictions by China on the export of rare earth elements have prompted concern about the impact a shortage would have on the world's economy, which is a significant national security concern. The physics of 4f-shell electrons found in rare earth elements make them peerless with respect to potential magnetic properties. Rare earth elements are essential components of the strong permanent magnets necessary for all technologies requiring a passive magnetic field, such as regenerative braking systems in hybrid automobiles, lightweight motor systems for compact hard-disk drives, and advanced megawatt windmills. We propose to create new rare-earth-element permanent magnets by developing a high-temperature synthesis capability and coupling it closely with the world-class capabilities already present at Lawrence Livermore in materials characterization, high-pressure physics, and quantum simulations to rapidly and systematically develop new materials.

The market for high-strength permanent magnets is so large that even a relatively modest improvement of a few percent in strength or a minor reduction in the quantity of rare earths required for the magnets could correspond to annual economic value in the hundreds of millions of dollars, intellectual property for the Laboratory, and opportunities to partner with industry. We expect that this project will establish LLNL's expertise in the area of rare earth materials synthesis and characterization, thereby positioning the Laboratory to make contributions to a problem with national security, environmental, and economic implications.

Mission Relevance

Our development of new high-strength magnets that use fewer (or cheaper) rare earth elements supports the Laboratory mission in energy and environmental security by providing a critical component for a clean, renewable energy source and fuel-efficient hybrid automobiles. In addition, research on rare earth elements can provide insight into the properties of actinide elements, which have similar properties to the rare earth elements and are important for stockpile stewardship science research. The synthesis of advanced materials is relevant to Lawrence Livermore's core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

In FY14 we (1) produced high-quality magnetic samples of lanthanum–cobalt and ytterbium–cobalt, which are structurally similar to samarium–cobalt but predicted to show volume collapses at pressures accessible with the designer diamond anvil cells; (2) measured the Hall effect (voltage difference across an electrical conductor) and anomalous Hall effect up to 15 GPa in lanthanum–cobalt and observed a change in the anomalous effect when we crossed the volume collapse at around 9 GPa; (3) produced samarium nanoparticles and evaluated their magnetic behavior on the nanoscale,

observing suppression of a magnetic phase at low temperatures; and (4) produced samarium and cobalt multilayer films and observed how the magnetic properties evolved as a function of annealing temperature as the metals reacted to form nanometer-scale particles. At higher temperatures, the evidence suggests grain growth and a reduction in ability to withstand an external magnetic electric field without becoming depolarized (coercivity).

Project Summary

The successful conclusion of this project resulted in development of an entirely new suite of sample synthesis capabilities at the Laboratory including (1) a quartz bench for sealing air-sensitive samples in vacuum or in an inert atmosphere, (2) a series of box and tube furnaces suitable to handle reactive gases as well as radioactive materials, (3) a glove box for work with lanthanide and actinide metals, and (4) a tetra-arc furnace able to heat samples above 2,500°C and produce large single crystals. In addition, we designed and constructed sample holders to work with air-sensitive samples, permitting extremely sensitive magnetic measurements to 800 K. We developed capabilities for making magnetic measurements in a piston cylinder clamp to 1 GPa and a Bridgeman pressure clamp capable of making resistivity measurements to 3 GPa. Our research provided the capabilities to develop new permanent magnets as part of the Critical Materials Institute, to produce actinide-containing samples in support of a new project as part of the nuclear counterterrorism effort, and to expand basic science work on actinides.

Publications and Presentations

Huang, P., et al., 2013. *Electronic structure and equation of state of Sm_2Co_{17} from first principles DFT+U. March Mtg*. American Physical Society, Baltimore, MD, Mar. 18–22, 2013. LLNL-PRES-627396.

Jeffries, J. R., et al., 2013. *Fe moments in the pressure-induced collapsed tetragonal phase of $(Ca_{0.67}Sr_{0.33})Fe_2As_2$*. March Mtg. American Physical Society, Baltimore, MD, Mar. 18–22, 2013. LLNL-ABS-600492.

Jeffries, J. R., et al., 2013. *Magnets at extreme conditions: Understanding and tuning atomic mechanisms for novel, high-strength permanent magnets*. Workshop New Directions for High-Pressure Neutron Research, Oak Ridge TN, June 3–5, 2013. LLNL-POST-638566.

Jeffries, J. R., et al., 2014. "Robust ferromagnetism in the compressed permanent magnet Sm_2Co_{17}." *Phys. Rev. B* **90**, 104408. LLNL-JRNL-652802.

Kundu, S., et al., 2013. "Shape-influenced magnetic properties of CoO nanoparticles." *J. Nanopart. Res.* **15**(5), 1587. LLNL-JRNL-625473.

McCall, S. K., et al., 2013. *Pressure as a tool for permanent magnet research*. 3rd EU–US–Japan Trilateral Conf. Critical Materials, Brussels, Belgium, May 29–30, 2013. LLNL-PRES-637472.

McCall, S. K., et al., 2013. *The effects of pressure on the structural and magnetic properties of Sm-based permanent magnets*. March Mtg. American Physical Society, Baltimore, MD, Mar. 18–22, 2013. LLNL-ABS-601452.

A Scalable Topological Quantum Device

George Chapline (12-ERD-027)

Abstract

We propose to take the initial steps toward practical quantum information storage, where for the first time it would be possible to control the entanglement of large numbers of degenerate surface states. Specifically, we will demonstrate the feasibility of storing computational information for quantum computing in the form of entangled (quantum mechanically correlated) states of the surface modes of a three-dimensional topological insulator with a topologically nontrivial surface. We hope to demonstrate experimentally that these surface modes are protected from dissipation, that they can be prepared and entangled by exciting them with a coherent source of terahertz radiation, and that the degeneracy of the surface modes is proportional to the topological genus of the surface.

We expect to demonstrate, using a scanning superconducting quantum interference device detector, the creation and measurement of entangled topological states on the surface of a topological insulator containing many quantum bits (the quantum analogue of the classical computer bit) using coherent terahertz radiation. This would lay the foundation for a variety of immediate applications exploiting quantum-annealing techniques to solve a wide variety of computational problems that are currently very difficult or intractable.

Mission Relevance

Lawrence Livermore has a long history of using its superior computer facilities for national security applications, and it is consistent with this mission to develop novel approaches to data processing for national security needs. If successful, our approach to quantum information storage and processing could have a major impact on this mission. Using three-dimensional topological insulators could be an enabling technology for intelligence and surveillance applications, where because of massive volumes of data, it is difficult or impossible to analyze data in real time. In addition, exploring topological-insulator quantum states of matter supports the Laboratory's core competency in advanced materials and manufacturing.

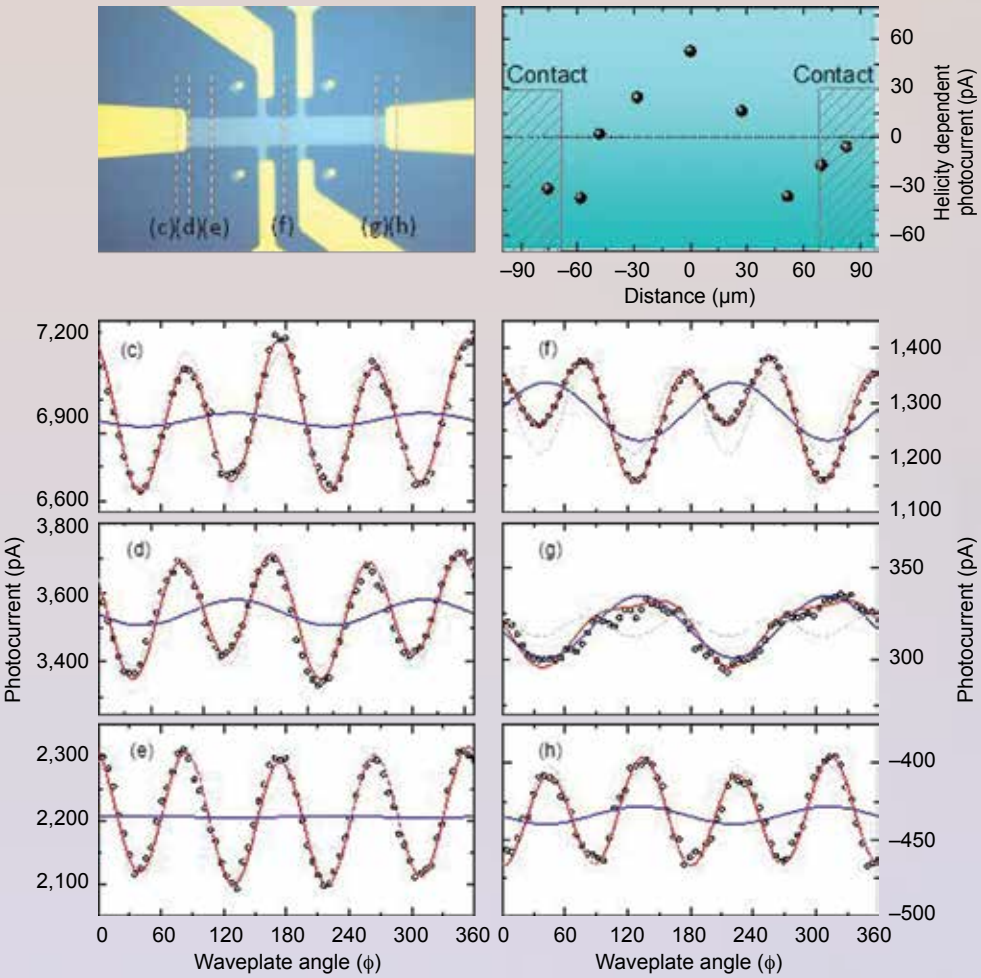
FY14 Accomplishments and Results

In FY14 we (1) augmented our optical cryostat scanning-photocurrent measurement system to include a variety of visible and mid-infrared lasers, including a continuous-wave laser with a 2-μm wavelength, which is expected to highlight the optical properties

of the surface states of topological insulators; (2) designed and developed a stable high-resolution system, allowing investigation of nanometer-scale local carrier dynamics in our devices; (3) observed a photon helicity-dependent photocurrent, which appears to originate from two types of states: the topological insulator states and spin-splitting quantum well states; and (4) observed that circularly polarized light generates spin-polarized photocurrents, which reverse their direction when approaching electrodes, and showed that the laser-induced photocurrent near a metal electrode is almost 100% spin polarized. Because of insufficient resources, we were not able to pursue experiments involving irradiation of our samples with terahertz radiation.

Project Summary

The remarkable nature of surface states in topological insulators is expected to give rise to a unique polarization-dependent response to electromagnetic radiation. This project has greatly improved our understanding of the exotic optoelectronic properties of novel topological-insulator quantum states of matter and will provide a foundation for the future implementation of topological insulators in emerging spintronic devices for computational applications. Our experiments contributed to the understanding of how to generate the unique topologically



Photocurrents induced on the surface of a 10-nm topological insulator film using an infrared laser. An optical image of topological insulator devices is shown top left. Photon polarization-dependent photocurrent as a function of laser spot position is seen top right. The measured photocurrent versus quarter-waveplate angle is shown in the six charts. The blue curve depicts polarization-dependent photocurrent.

protected electric and spin currents flowing on the surface of a topological insulator and how to detect these states via voltage measurements. Further research is needed to investigate using spin currents in hybrid topological-insulator and metallic film devices to generate terahertz radiation and to explore how topological insulator materials respond to terahertz radiation. Such research could lead to optoelectronic devices on a chip operating at terahertz frequencies. Longer term, we envision that the transient quantum coherence of surface states of topological insulators can be used for quantum information processing. We are exploring these possibilities and hope that within the next few years our understanding of how to build useful topological insulator devices will be sufficiently advanced to be of interest to the Defense Advanced Research Projects Agency and the Intelligence Advanced Research Projects Activity.

Publications and Presentations

Qu, D., Y. Hor, and R. J. Cava, 2012. "Quantum oscillations in magnetothermopower measurements of topological insulator Bi_2Te_3." *Phys. Rev. Lett.* **109**(24), 246602. LLNL-JRNL-594632.

Qu, D., S. K. Roberts, and G. F. Chapline, 2013. "Observation of huge surface hole mobility in topological insulator $Bi_{0.91}Sb_{0.09}$ (111)." *Phys. Rev. Lett.* **111**(17),176801. LLNL-JRNL-634412.

Qu, D., et al., 2014. *Photogalvanic effect in topological insulators towards scalable quantum devices*. LLNL-POST-656620.

Dynamically Tunable Nanometer-Scale Materials: From Atomic-Scale Processes to Macroscopic Properties

Juergen Biener (12-ERD-035)

Abstract

The future of sustainable energy strongly depends on scientific advances in materials for energy storage and conversion. Material interfaces are of particular importance, because all processes relevant to energy storage, whether physical or chemical, occur here. In classical bulk materials, only a negligible fraction of atoms are surface atoms; thus the interfacial area in these materials is quite small. By contrast, nanometer-scale porous materials (materials with pores smaller than 100 nm) have surface areas so large that the majority of atoms are part of a surface. Because of their highly accessible interfacial area, the properties of these materials are determined by surface interactions. We propose to establish a fundamental understanding of interfacial charge-transfer phenomena on interface-controlled materials by using a combination of experimental and theoretical tools, and use this information to develop a new class of dynamically tunable three-dimensional nanometer-scale materials for the next generation of energy-storage technologies.

We expect to develop a fundamental understanding of interfacial phenomena related to electrical energy storage that is needed to develop the next generation of energy-storage and harvesting devices. In addition, we will develop novel three-dimensional graphene-based materials (a honeycomb crystal film of graphitic carbon) with improved electrical energy-storage performance. Ultimately, the project will lead to the development of dynamically tunable bulk materials whose mechanical, chemical, and physical properties can be controlled by interfacial electric fields.

Mission Relevance

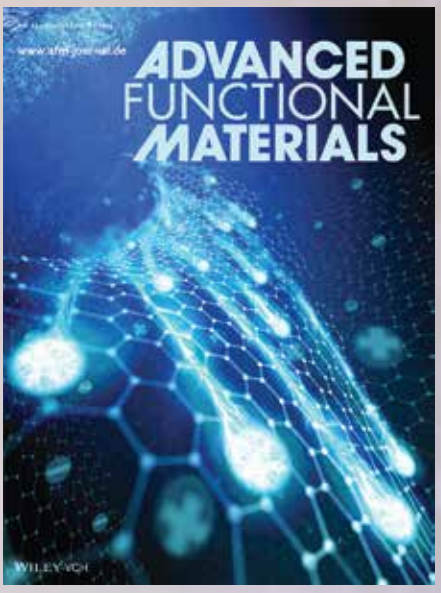
We will develop a fundamental understanding of interfacial charge-transfer phenomena, and use it to develop the next generation of energy-storage materials in support of the Laboratory's energy and environmental security mission to develop technologies to enable a carbon-free energy future. Furthermore, developing novel materials with improved performance for specific applications supports the core competency in advanced materials and manufacturing. Our project is also relevant to several cross-cutting research directions identified in a recent DOE report on *Basic Research Needs for Electrical Energy Storage*, including advanced in situ characterization capabilities and theoretical studies of charge-transfer processes at interfaces.

FY14 Accomplishments and Results

In FY14, using a combination of experimental and theoretical tools, we continued to develop a better understanding of interfacial charge-transfer phenomena on interface-controlled materials and developed the next generation of energy-storage materials. Specifically, we (1) studied the fundamental mechanism of electrochemically gated resistivity changes in graphene-based electrodes, (2) evaluated the performance of selected nonaqueous electrolytes, (3) explicitly included potential in atomic-scale modeling, (4) explored bottom-up and top-down nitrogen-doping approaches, and (5) further pushed the limit of energy density by optimizing morphology and surface functionalization.

Project Summary

The successful conclusion of this project resulted in the development of new design rules for next-generation supercapacitor materials. Specifically, we developed robust three-dimensional graphene bulk materials that combine ultrahigh surface areas (up to 3,000 m^2/g) with high electrical conductivity (~10^2 S/m, 100 times higher than previously reported). Our high-density supercapacitor electrodes match the system energy density (4–10 Wh/kg) of commercially available, organic electrolyte supercapacitors but use less-toxic aqueous electrolytes, which also enable 10 to100 times higher power densities. We discovered that electrochemical interface polarization triggers macroscopic strain effects (up to 2.2% strain) and electronic transport modulation phenomena (300% conductivity change), which opened the door to new low-voltage (<1 V) actuator and carbon-based transistor applications. Over the lifetime of the project, we generated four granted patents. This work opened the door to a future collaboration with the Automotive Fuel Cell Corporation, Canada.



Cover art for the *Advanced Functional Materials* journal in June 2014 illustrates dynamic control over electronic transport in ultrahigh-surface-area bulk materials with a three-dimensional open-porous network structure consisting of single- and double-layer grapheme (carbon) nanometer-scale platelets. Macroscopic conductance variations of up to several hundred percent are triggered by electrochemically induced charge-carrier accumulation or depletion accompanied by large variations in carrier mobility.

In addition, our proprietary materials will be offered to industrial partners for integration into products.

Publications and Presentations

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Dasgupta, S., et al., 2014. "Dynamic control over electronic transport in 3D bulk nanographene via interfacial charging." *Adv. Funct. Mater.* **24**(23), 3494. LLNL-JRNL-645196. *http://dx.doi.org/10.1002/adfm.201303534*

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Worsley, M. A., et al., 2012. "Mechanically robust 3D graphene macroassembly with high surface area." *Chem. Comm.* **48**, 8428. LLNL-JRNL-55239.

Worsley, M. A., et al., 2013. "Thick, binder-free carbon-nanotube-based electrodes for high power applications." *J. Electrochem. Soc.* **2**(10), M3140. LLNL-JRNL-641568.

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Accelerated Certification for Additively Manufactured Metals

Wayne King (13-SI-002)

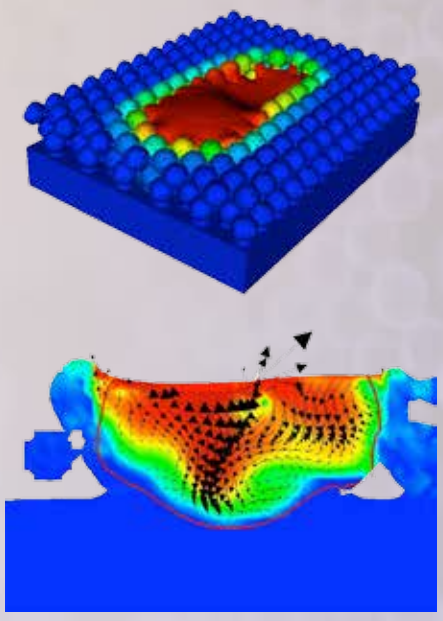
Abstract

Stockpile stewardship would benefit greatly from a new manufacturing approach that can speed development and certification of parts, reduce the manufacturing costs for a shrinking stockpile, reduce manufacturing waste, avoid the costs and delays associated with manufacturing processes, and reduce energy consumption in part fabrication. We propose to employ modeling, simulation, process optimization, in situ sensing, and uncertainty quantification for the accelerated certification of metals made by additive manufacturing, with the goal of guiding the process to yield optimized properties and performance. We will employ an effective medium simulation that models the process at the scale of the part and a powder simulation that models the process at the scale of the material powders used in parts manufacturing. Microstructure will be predicted using phase field models, and properties will be predicted using crystal plasticity and dislocation dynamics. The results of simulations will be validated against measured material properties and data acquired from real-time, in situ process monitors.

This project will advance the field of additive manufacturing by creating (1) a multiscale modeling and inverse-design methodology to assist in navigating complex process, structure, and property relationships; (2) a method for integrating the predictive process, structure, and property relationships into the additive manufacturing process; (3) a thorough understanding of the basic physics of additive manufacturing processes to capture the complexity in multiple interacting physical phenomena; (4) the ability to prescribe processing conditions and achieve the desired properties in only one or two attempts; and (5) a strategy to accelerate the certification of critical parts.

Mission Relevance

This project furthers the Laboratory's mission in stockpile certification and core competency in advanced materials and manufacturing by developing a substantially improved, science-based approach to develop mature manufacturing technologies and systems in a rapid and cost-effective manner.



Simulation laser melting of a powder layer containing a hexagonal array of 27-µm-diameter stainless-steel powder particles shortly after the laser was turned off, showing consolidation by surface tension and gravity. The color represents temperature. The molten pool is visible at top and the recirculation of the liquid metal is represented by the arrows in the cross section at the bottom.

FY14 Accomplishments and Results

In FY14 we (1) demonstrated the mitigation of overhang defects, computed residual stresses in a part that will be produced, and validated residual stress predictions with experiments for the effective medium model; (2) demonstrated a single-track modeling capability and incorporated surface tension, demonstrated powder melt and gas bubble migration, carried out the first multiple-track simulations on a single layer, and measured powder-bed thermal properties for the powder model; and (3) developed sampling strategies to identify feasible points for process parameters, identified relevant features in simulations, and developed algorithms to extract these features for both simulations and experiments for data mining and uncertainty quantification.

Proposed Work for FY15

In FY15 we will (1) deploy an adaptive mesh-refinement capability, evaluate alternative spatial discretizations providing greater efficiency, and develop homogenized support structure representation for the thermal and mechanical influences of support structures on part configuration and properties for the effective medium model; (2) add ablation pressure to the approach, simulate the overhang configuration to optimize parameters for surface finish, and perform experiments to validate the transition model and Marangoni convection flow for the powder model; and (3) develop initial codes for prediction with uncertainty incorporated for data mining and uncertainty quantification.

Publications and Presentations

Barth, W. E., et al., 2014. "Observation of keyhole-mode laser melting in laser powder-bed fusion additive manufacturing." *J. Mater. Process. Tech.* **214**, 2915. LLNL-JRNL-50285. *http://dx.doi.org/10.1016/j.jmatprotec.2014.06.005*

Boley, C. D., S. A. Khairallah, and A. M. Rubenchik, 2014. *Modeling of powder absorption in additive manufacturing*. CLEO: Applications and Technology, San Jose, CA, June 8–13, 2014. LLNL-PRES-648823.

Hodge, N., and R. Ferencz, 2013. *Part-level finite element simulation of selective laser melting*. 12th U.S. National Congress Computational Mechanics, Raleigh, NC, July 22–25, 2013. LLNL-ABS-618093.

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Wu, A. S., et al., 2013. *Effect of laser scanning pattern and build direction in additive manufacturing on anisotropy, porosity and residual stress*. TMS 2013 142nd Ann. Mtg. and Exhibition, San Antonio, TX, Mar. 3–7, 2013. LLNL-ABS-640994.

A Three-Dimensional Radioisotope Battery

Rebecca Nikolic (13-ERD-004)

Abstract

Spacecraft require an energy source that operates reliably and predictably for extended periods in harsh environments. For many years, the solution has been radioisotope power systems. Our objective with this project is to develop an electrical power-generation device for space-based applications that combines fissile material with a three-dimensional semiconductor to enable the use of a large volume of the semiconductor for power generation for dramatically increased power density instead of limiting it to the surface, which provides only microwatt output power. We also intend to investigate the use of wide band-gap, radiation-tolerant semiconductors and amorphous semiconductors to greatly extend the lifetime of the battery. We will carry out fundamental design work to determine material selection, fabrication, and architecture for a proof-of-concept battery. That will be followed by assessments of power scaling and device lifetime.

We plan to demonstrate current generation on the microcurie-to-millicurie scale by coating three-dimensional semiconductor structures with a radioisotope for high-volume radioisotope batteries with high power density. We expect to determine the alpha particle-to-electron conversion efficiency as well as a suitable structure for obtaining high currents with high amounts of activity. This is an attractive concept because all of the semiconductor processing work could be completed in a clean-room environment and then activated and subsequently packaged to shield the fissile

material and minimize external radiation. Small nuclear batteries, depending on the application and power, could become an off-the-shelf power-supply item, enabling long-term use of micro-powered devices and sensors capable of uninterrupted operation for decades. In addition, higher-power batteries will enable deep-space probes to operate with a smaller size and weight budget than conventional nuclear power supplies. These power-generation devices have applications for the NNSA, Department of Defense, and intelligence community.

Mission Relevance

The proposed technology enables development of long-term emplaced sensors in a wide variety of missions of interest to NNSA and other federal agencies. Our research is also closely aligned with Laboratory missions in advanced materials and manufacturing and actinide sciences, as well as enabling new energy sources for military applications.

FY14 Accomplishments and Results

In FY14 we (1) carried out chemical vapor-deposition growth of icosahedral boron phosphide thin films on planar silicon carbide substrates; (2) determined carrier concentration, degree of structural order, and electrical conduction; and (3) etched silicon carbide pillared platforms for crystal growth of icosahedral boron phosphide.

Proposed Work for FY15

For FY15 we will (1) determine defect mechanism for alpha damage in alpha-irradiated silicon carbide and icosahedral boron phosphide; (2) perform chemical vapor

Monitoring chemical vapor deposition of icosahedral boron phosphide for use in a radioisotope battery.

deposition of single-crystal icosahedral boron phosphide on flat and pillar platforms; (3) characterize the resultant films' degree of structural order, defect concentration, and electrical characteristics; and (4) fabricate and characterize a radioisotope battery comprised of icosahedral boron phosphide coated with uranium oxide.

Publications and Presentations

Frye, C. D., et al., 2014. *Cr/Pt and Ni/Au ohmic contacts to $B_{12}P_2$*. Electronic Materials Conf., Santa Barbara, CA, June 25–27, 2014. LLNL-PRES-656119.

Stoyer, M. A., et al., 2014. *Simulations of novel nuclear battery concepts*. 247th ACS National Mtg. and Exposition, Dallas, TX, Mar. 16–20, 2014. LLNL-PRES-651894.

Micro-Reflector Array for High-Speed Directed-Light-Field Projection

Robert Panas (13-ERD-009)

Abstract

We propose to create a new micro-mirror array to simultaneously direct multiple high-powered beams of light in various directions at high speeds. This device, known as a light-field directing array, is an independently controlled and scalable mirror array designed to achieve independent continuous control with speeds, ranges, array sizes, and light fill factors that are not currently achievable. The array will enable advanced applications for rapid steering of multiple high-powered laser beams for igniting targets for inertial fusion, true auto-stereoscopic images, multiple-material nanometer-scale fabrication using laser beams focused by a high-quality microscope objective (optical tweezers), and ultrarapid multiple-focal-point optical remote sensing or confocal microscopy. We will fabricate and demonstrate a large-area array of controlled high-performance micro-mirrors via fabrication research carried out at LLNL and in development with commercial fabrication experts.

We expect to be able to demonstrate ultrarapid spatial light modulation over an entire 50-by-50 mirror array to track a rapidly moving object over an arbitrary path. This technology will be capable of meeting the demanding performance requirements for applications that require the precise targeting of multiple-kilowatt-scale energies on microsecond timescales over wide fields of view. We intend to determine the fundamental performance limits of individual micro-mirrors and integrate micron-scale additive fabrication processes with traditional lithography to fabricate and test the performance of the micro-mirrors when assembled into arrays.

Mission Relevance

A mirror array design that can be used for rapid high-power laser target tracking supports the LLNL mission focus area in inertial-confinement fusion energy.

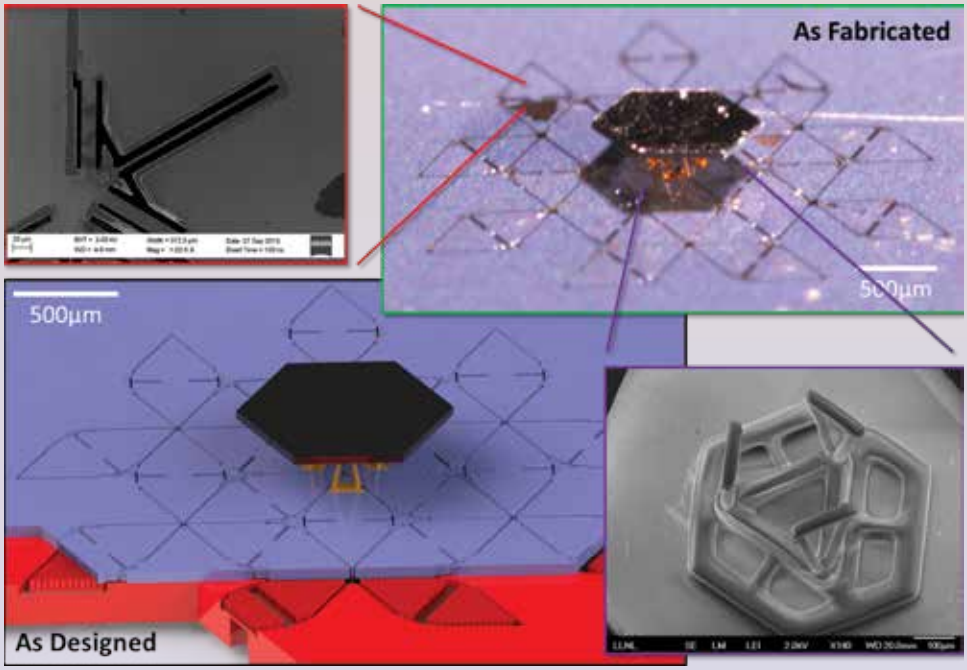
Furthermore, this mirror array could be used to guide a massive array of optical-tweezer laser beams in an effort to move and sinter in place millions of nanometer-scale particles simultaneously, which would be a fundamental advance in metamaterial additive manufacturing, in support of LLNL core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

In FY14 we achieved results on the three research fronts of micro-fabrication, additive fabrication and assembly, and design. Specifically, we (1) completed fabrication and testing of the first version of our micro-fabricated prototype, altering fabrication to account for observed issues such as epitaxial growth and wafer bonding; (2) produced a second version that resolved these issues; (3) produced and tested high-aspect-ratio flexure transmission structures in micro-stereolithography; (4) developed and demonstrated a pick-and-place process; (5) built hardware to assemble the transmission structures onto the micro-fabricated structures to create the full prototype; (6) completed the full-system model and validated it with finite-element analysis; (7) finalized the design theory used to develop the structure (an advance over the previous state-of-the-art methods); and (8) finalized the structural design, which provides further performance improvements in speed.

Proposed Work for FY15

For FY15 we propose to (1) test the performance of the full-system model of the second version of our micro-fabricated prototype, (2) produce a third version if warranted, (3) improve the additive fabrication capabilities to generate more elastic structures, (4) automate the assembly process to improve its robustness, and (5) demonstrate a functional mirror element at the conclusion of the project.



The present state of the micro-mirror project is illustrated for both the design and fabrication work. The device design with several internal features revealed is shown bottom left. A fabricated single-mirror device with scanning electron microscope closeups of the flexures (right).

Publications and Presentations

Hopkins, J. B., and R. M. Panas, 2014. *Flexure design for a high-speed large-range tip-tilt-piston micro-mirror array*. 29th Ann. Mtg. American Society for Precision Engineering, Boston, MA, Nov. 9–14, 2014. LLNL-ABS-658144.

Panas, R. M., and J. B. Hopkins, 2014. *Actuator design for a high-speed large-range tip-tilt-piston micro-mirror array*. 29th Ann. Mtg. American Society for Precision Engineering, Boston, MA, Nov. 9–14, 2014. LLNL-ABS-657938.

Panas, R. M., and J. B. Hopkins, 2014. *Lightfield directing array*. LLNL-POST-655689.

Rapid Synthesis, Functionalization, and Assembly of Nanometer-Scale Particles for Designer Materials

Thomas Han (13-ERD-022)

Abstract

One of the grand challenges of materials science is to create designer materials with pre-programmed building blocks that can assemble predictively or as directed into structures with unique functions and properties and with the ability to self-regulate or adapt their electrical conductivity or elasticity or their magnetic, optical, or mechanical properties. We propose to understand the governing principles of nanometer-scale particle assembly to create multiple-component three-dimensional composite materials. Specifically, we will synthesize nanoparticles with diverse chemical compositions, shapes, sizes, and structures that can be used as "artificial atoms" to construct nanoscale, mesoscale, and macroscopic "molecules" of nanoparticles with increasing complexity and function. The synthesis of nanoparticles and their assembly will be performed in custom-built combinatorial micro-reactors coupled with in situ characterization tools for real-time feedback with rapid synthesis and assembly. If individual nanoparticles with different chemical composition can be assembled systematically in any structure or order, then it may be possible to create new materials predictively and control and manipulate the emerging properties, providing a paradigm shift in materials science.

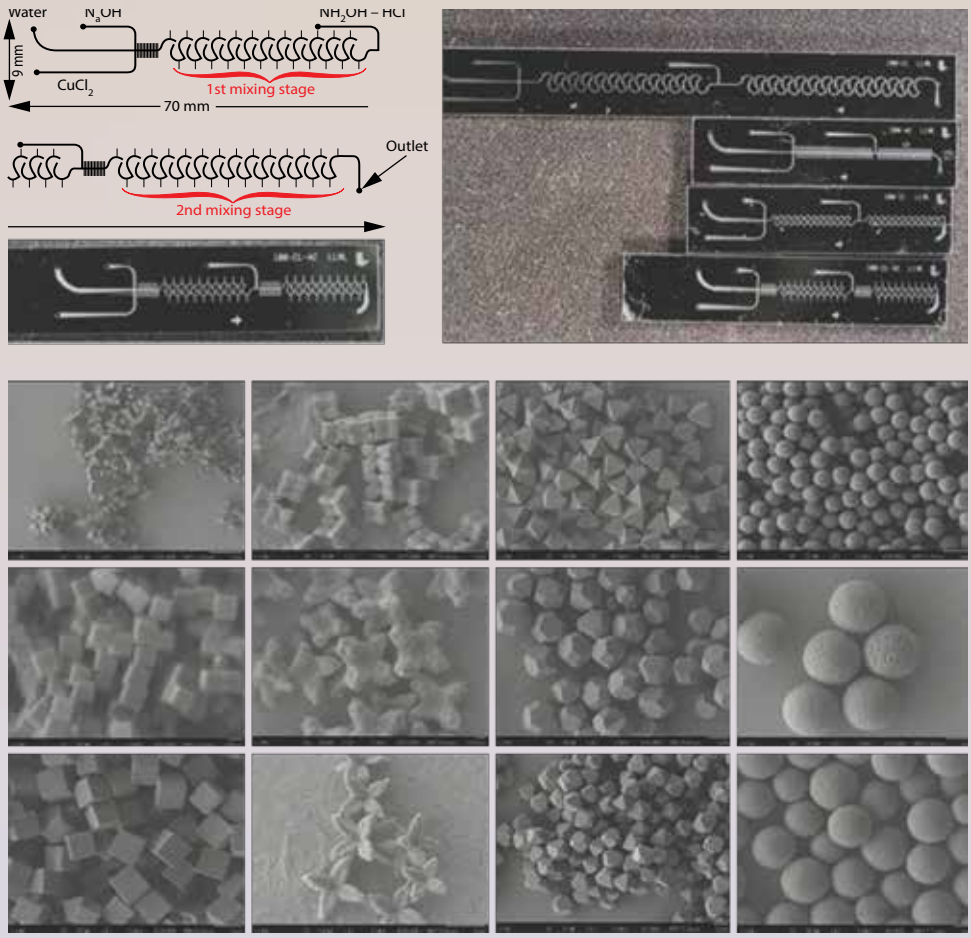
We expect the advances in materials science we propose will result in the rapid synthesis, functional capability, and assembly of nanoparticles, with precise control over each step. This will ultimately lead to understanding the fundamental principles of nanoparticle assembly. With this knowledge, we will be one step closer to making designer materials that can be utilized as weapon-critical components, and also fulfill nonproliferation and national security needs for radiation-detection materials, chemical-agent sensors and neutralizers, and high-explosives detectors. We expect to design and fabricate several sets of micro-reactors, which we will use in nanoparticle synthesis. We will start with the synthesis of copper oxide nanoparticles, known to exhibit a diverse morphology, and systematically evaluate how they assemble as a function of their sizes and shapes.

Mission Relevance

Our research directly supports the Laboratory's core competency in advanced materials and manufacturing to predict the behavior of and synthesize novel materials, as well as validate the predictions. Our project will explore length scales that can enhance and complement additive manufacturing efforts at LLNL. In addition, the synthesis of novel materials benefits a variety of applications in energy and climate, nonproliferation, and national security.

FY14 Accomplishments and Results

In FY14 we (1) developed and tested a fully functional continuous-flow synthesis platform that incorporates a pressure-driven reagent delivery system, LLNL-produced advanced chemical mixer chips, an aging chamber with temperature control, and an



Schematic and images of our micro-mixers used for advanced material synthesis (top) and scanning electron microscopy images of copper oxide nanometer-scale particles with a diverse morphology used to test the feasibility of these particles as feed materials for additive manufacturing (bottom).

in situ characterization module; (2) using the platform, synthesized gold nanometer-scale particles and moved towards synthesizing copper oxide particles as well as other inorganic solids for various applications, including feedstock for additive manufacturing processes; and (3) performed ligand exchanges on nanoparticles, including gold and copper oxide particles for directed self-assembly.

Proposed Work for FY15

We will (1) continue developing the continuous-flow synthesis platform to include advanced features such as a high-temperature synthesis capability, (2) identify and integrate the advanced in situ characterization tool, (3) perform directed assembly of materials synthesized for mesoscale assembly, and (4) continue to synthesize potential feedstock materials for additive manufacturing processes.

Publications and Presentations

Han, T. Y., et al., 2014. *Development of rapid synthesis and characterization platform for advanced materials discovery*. TechConnect World, Innovation Conf. and Expo, Washington, DC, June 15–18, 2014. LLNL-ABS-651531.

Theoretical and Computational Studies of Rare Earth Substitutes: A Test Bed for Accelerated Materials Development

Lorin Benedict (13-ERD-044)

Abstract

The price of rare earth elements, which are used in the manufacture of hard permanent magnets and phosphors, is rising dramatically, with China enjoying a near monopoly in production. We propose to advance the search for substitutes for rare earth elements, using a first-principles computational effort that will couple with existing Laboratory experimental efforts on rare earth substitutes and a computational and statistical effort aimed at material properties optimization. We will study the figures of merit for candidate permanent magnets with both higher-fidelity/high-cost methods and lower-fidelity/low-cost methods. Magneto-crystalline anisotropy and temperature-dependent magnetic excitations will be studied in detail, setting the stage for eventual multiscale modeling of the coercivity (magnetic intensity) properties of materials.

We expect to make major advances that are critical for finding substitutes for rare earth elements, and we will identify key gaps in the predictive capability for figures of merit for hard permanent magnets. We will improve the theoretical and computational fidelity for a subset of atomic-level properties most in need of improvement, such as finite-temperature magnetic excitations. In addition, we will produce theoretical and computational prescriptions of varying degrees of fidelity and associated cost, to be

fed into the Optimal Management Framework materials design effort. Finally, we will initiate the integration of our atomistic predictions into a multiscale model that addresses the full scope of the figures of merit (total magnetization, Curie-temperature, and coercivity) for real permanent magnets.

Mission Relevance

This project advances the Laboratory's strategic focus area of energy and climate and the core competencies in advanced materials and manufacturing and computational science and engineering. It also aligns with the DOE's initiative focusing on the search for rare earth element substitutes.

FY14 Accomplishments and Results

In FY14 we (1) completed our study of the pressure dependence of the magnetic and structural properties of lanthanum–cobalt-5 and yttrium–cobolt-5 under pressure; (2) solidified our understanding and modeling of ab initio phonon dynamics coupled to non-collinear spin excitations in shocked iron; (3) completed a comprehensive study of magneto-crystalline directional dependence (anisotropy) in the iron–cobolt–boron system as a function of cobalt concentration, which led to a study of the effect of substitutional disorder and atomic relaxations on anisotropy relevant to material defects; (4) developed a Monte Carlo code to compute Curie temperatures and temperature-dependent magnetic moments for generalized models of elementary particle spin interactions; and (5) initiated work on the development of a cluster expansion (lattice-model-based technique for alloy energetics) for anisotropy.

Proposed Work for FY15

In FY15 we will (1) apply our coupled phonon and quantized spin wave technique to permanent magnet materials and use constrained-spin density functional theory to compute magnetic domain-wall energies, (2) use a self-interaction technique to see if it is possible to better predict the anisotropy for materials like lanthanum–cobalt-5 and yttrium–cobolt-5 possessing localized electrons near the Fermi level (total chemical potential for electrons), and (3) study temperature-dependent anisotropy in iron-2–boron and cobalt-2–boron as well as anisotropy in the presence of defects and grain boundaries, thus setting the stage for more accurate parameters to be ultimately fed into a multiscale model of coercivity.

High-Explosive Components Using Advanced Manufacturing Methods

Alexander Gash (13-ERD-051)

Abstract

We propose to develop methodology and expertise for the additive manufacturing of plastic-bonded explosive (PBX) components. The PBX material is a composite of

predominately explosive crystals bonded together with a small amount of polymer. This project will enable manufacturing of PBX by a totally new method that will allow exquisite spatial density and compositional control within a single monolithic component, which is not possible with current methods. We will use experimental and predictive methods to understand the science behind particle synthesis in micro-reactors that enable continuous synthesis operations in micro-machined channels. We will also evaluate development of high-solids, high-density colloids as candidate materials for PBX formulation. Novel materials synthesis techniques will be utilized, along with crystallization and processing methods that include micro-reactors and direct ink writing for extruding explosive samples.

We expect to gain the scientific understanding and develop the knowledge base necessary to bring about a paradigm shift in PBX manufacturing. This will facilitate a transition from a subtractive manufacturing process, which is used currently, to an additive manufacturing process for PBX. Key to this project is the expertise to synthesize energetic materials through a continuous process with simultaneous control over size and morphology. This new approach will accelerate the rate of development of new energetics for the stockpile, bypassing the complications of scaling batch materials. Successful completion of this research in advanced manufacturing methodology will enable unprecedented control of spatial density and composition in a PBX component. This type of control is necessary to impart enhanced functionality (high-reliability and high-energy output) to meet future needs in national security applications.

Mission Relevance

Our proposed plan is well aligned with the Laboratory's strategic thrust in stockpile stewardship science. Aspects of this proposed work will increase our level of confidence in the safety and reliability of nuclear weapons by providing PBX components with enhanced functionality for future stockpile-life extension programs, and also supports our core competency in advanced materials and manufacturing with the development of novel material synthesis techniques.

FY14 Accomplishments and Results

In FY14 we (1) continuously synthesized the explosive LLM105 with 51% yield using a commercial micro-reactor, (2) designed a custom chip that enables stepwise nitration reactions in corrosive media to improve yield and throughput, (3) developed the automated direct ink printing of high-solids-loaded high-explosive ink (94% high-explosive loaded), (4) produced detonable parts with direct-ink-writing high explosive, (5) demonstrated the ink writing of 80% high-explosive-loaded formulation to a resolution of 200 µm, (6) demonstrated a yield comparable to the batch approach, (7) demonstrated single microchip production of high-purity LLM105 at a rate of approximately 1 g/h, (8) demonstrated controlled morphology crystallization of LLM105 in the micro-reactor, and (9) demonstrated the first instance of direct ink writing of full-density detonable structures of the explosive LX-20.

Proposed Work for FY15

In FY15 we will (1) implement a polymersome-based approach to continuous crystallization of high explosives; (2) integrate this new approach to achieve continuous crystallization and formulation of additive-manufactured high explosives; (3) expand the use of synthesis micro-reactors to other important DOE high-explosives materials (e.g., LLM-172, 175, and 200); and (4) prepare high-explosives components using direct ink writing with spatially varying density and composition.

Optimized Three-Dimensional Electrodes for Energy Storage

Eric Duoss (13-ERD-057)

Abstract

Emerging autonomous microelectronics found in micro-electromechanical systems such as miniaturized reactors, sensors, biomedical devices, and wireless communication devices will require on-chip, micrometer-scale power sources. New three-dimensional battery configurations must be developed that maintain short transport distances yet provide enough material to ensure adequate energy output for an extended time to power devices with a limited footprint. We propose to demonstrate the first-ever use of additive manufacturing to fabricate novel, high-performance, three-dimensional electrode architectures for energy storage by integrating mesoscale fabrication with tailored nanometer-scale structure synthesis, predictive modeling, and advanced characterization. By integrating these efforts and reducing iteration cycles, we will accelerate the discovery, fundamental understanding, and product realization of advanced battery materials. Furthermore, we will realize heretofore-unachievable improvements in performance by extending battery designs into the third dimension. This project will enhance the basic scientific understanding of battery mechanisms while increasing the energy density for applied micro-battery systems.

We expect to achieve large areal and volumetric energy capacities without sacrificing power density primarily by accelerating interfacial kinetics with high ratios of electrode surface area to volume and by reducing resistance losses by minimizing transport distances. This work will be the first demonstration of combining rapid material synthesis of tailored porous nanoscale particles with streamlined manufacturing, and it will integrate design, modeling, and characterization to rapidly produce novel battery architectures with improved performance.

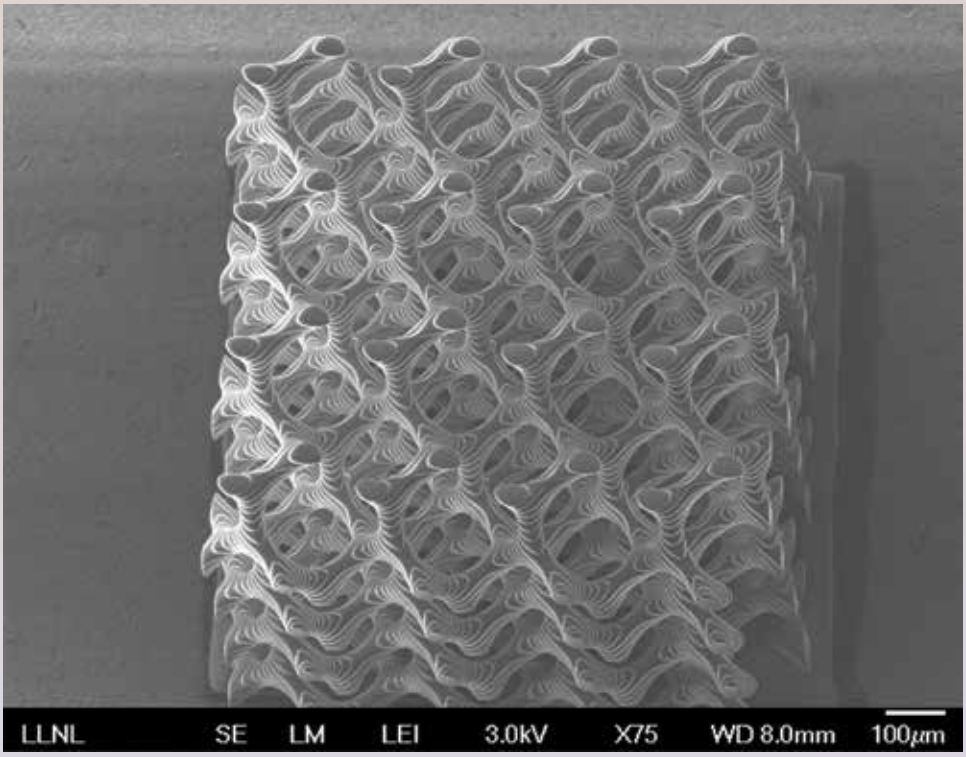
Mission Relevance

Our project supports the Laboratory's mission thrust area of enhancing national energy security and a core competency in advanced materials and manufacturing. We will develop new materials for energy storage, along with a transferable electrode design relevant to all solid battery types. Our design can leverage other battery

technology advances and will have an impact on the fabrication of next-generation batteries. These advances will help position the U.S. for continued competitiveness in the manufacturing and energy sectors, in furtherance of the Laboratory's mission to enhance the nation's energy security and economic competitiveness.

FY14 Accomplishments and Results

In FY14 we (1) developed synthetic routes to create large quantities of active nanoparticles with tailored properties for battery electrodes; (2) formulated functional inks, using these nanoscale active materials, for printing electrodes with microscale features; (3) developed a novel solid-state lithium-ion electrolyte material that can be printed three dimensionally and has ionic conductivities on par with other polymer gel electrolytes (see figure); (4) designed and printed three-dimensional battery architectures using the additive micro-manufacturing approach known as projection micro-stereolithography; (5) assembled and tested all of our battery materials to confirm performance; and (6) developed simulation codes for two- and three-dimensional battery designs that verify our designs have improved power density for a given energy density.



Scanning electron microscopy image of a three-dimensional printed micro-battery architecture. The architecture material is a lithium-ion solid-state electrolyte. The architecture design consists of two discrete but interpenetrating channels designed to reduce transport distances between the active material phases (i.e., anode and cathode).

Proposed Work for FY15

In FY15 we will (1) develop new materials and routes to pattern battery micro-architectures with direct ink writing and projection micro-stereolithography, (2) develop a simulation capability for a new three-dimensional interpenetrating battery architecture known as the gyroidal structure, (3) perform shape optimization to computationally create new two- and three-dimensional battery designs that increase power density, and (4) fabricate and test these new designs to experimentally validate their performance and prove that they increase power density.

Quantum Monte Carlo Benchmarks for Materials on Demand

Randolph Hood (13-ERD-067)

Abstract

The current challenge in condensed matter and chemical physics for development of a predictive foundation for materials on demand is the accurate solution of the Schrödinger equation that describes how the quantum state of a physical system changes with time. Despite decades of effort invested in its solution, there are still major difficulties in predicting and explaining many phenomena. With the advent of petascale computing, we are finally in a position to move beyond mean-field treatments such as density functional theory and employ more accurate quantum Monte Carlo approaches (Monte Carlo algorithms rely on repeated random sampling to compute their results) for understanding the behavior of materials at the level of the electrons that bind atoms together. We propose to carry out validating benchmark quantum Monte Carlo calculations for a wide range of materials that has been previously studied. Given the favorable scaling of quantum Monte Carlo and its inherent ability to obtain systematically improved results, we believe that it will play a leading role in future electronic structure calculations.

We expect to perform benchmark calculations of the fundamental electronic properties of selected materials across the periodic table using quantum Monte Carlo. Our objective is to establish the power of the method by demonstrating the level of accuracy achievable for materials properties associated with both equation-of-state and energy applications such as excited-state properties. Obtaining this data will enable us to establish the fidelity of the method and to pinpoint areas where quantum Monte Carlo still requires development. This achievement will lay the groundwork for the second phase in which we will extend the method to treatment of lanthanides and actinides, materials that are difficult to compute accurately yet promise to play an increasingly strategic role in technology and national security.

Mission Relevance

Developing a core competency in advanced materials and manufacturing at LLNL by validating quantum Monte Carlo as a high-fidelity approach and enhancing its accuracy where necessary by computing equation of state and determining excited-state data for

a broad range of materials will impact many programs. Equation-of-state models form the core of theoretical high-energy-density science and stockpile stewardship science at the Laboratory, particularly for actinides. Quantum Monte Carlo can also be used for band gap engineering in the development of cost-effective detectors for radiation detection and discrimination for use in nuclear threat reduction and to develop improved solar electricity generation for renewable energy applications.

FY14 Accomplishments and Results

In FY14 we (1) performed additional quantum Monte Carlo calculations of equation-of-state properties to determine optimal methods of choosing effective potentials (used as an approximation for the simplified description of complex atomic systems) for use in quantum Monte Carlo; (2) performed calculations of band gaps and spectral data of sodium chloride, magnesium hydride, silicon carbide, gallium phosphide, and indium phosphide solids; (3) determined the quantum Monte Carlo predicted phase stability of tin at high pressure; and (4) obtained an optimized pseudopotential for plutonium and have begun quantum Monte Carlo calculations of plutonium's equation of state.

Proposed Work for FY15

In FY15 we will carry out quantum Monte Carlo calculations of equation-of-state properties of one or more lanthanides and actinides, including plutonium. In addition, we will document the use of quantum Monte Carlo in treating highly correlated lanthanide and actinide materials.

Strength and Phase Transformation Kinetics Under Dynamic Compression

Joel Bernier (13-ERD-078)

Abstract

Enhancing our understanding of material behavior under the thermal and mechanical extremes of high pressure, stress, and strain rate is critically important to a wide range of applications, from energy generation and storage to stockpile stewardship science. We propose to study strength and phase transitions, including their kinetic dependencies, and develop a capability for performing ultrafast synchrotron x-ray diffraction measurements under dynamic loading, focusing on advanced laser loading techniques and Laue diffraction configurations (incident waves diffracted by a crystal lattice). We will develop the diffraction techniques in close collaboration with Los Alamos National Laboratory researchers using an instrument at the Advanced Photon Source at Argonne National Laboratory, and develop a laser compression drive at Lawrence Livermore.

We expect to develop a capability for ultrafast synchrotron x-ray diffraction measurements for materials subject to extremes of pressure and strain rate. We will also develop an arbitrary waveform laser compression drive capable of dynamically loading samples over a wide

range of pressures and strain rates with good repeatability and high throughput. We will then perform a first-of-its-kind scientific study of the kinetic dependence of strength and phase transitions in a suite of iron–nickel alloys having geophysical significance. This research will enable Livermore to take on a leadership role in developing and commissioning the Dynamic Compression Sector beam line at the Advanced Photon Source.

Mission Relevance

High-fidelity material models are a critical piece of the multiphysics simulation codes employed in stockpile stewardship. This project will enable the rigorous verification and validation of cutting-edge predictive models for strength and the phase diagrams of metals under extremes of pressure and strain rate, which strengthens the Laboratory's core competency in advanced materials and manufacturing relevant to the fundamental understanding of material behavior under dynamic conditions.

FY14 Accomplishments and Results

In FY14 we (1) executed a suite of experiments on an iron–nickel material system at Livermore's Titan laser and the Trident laser platform at Los Alamos National Laboratory at a series of pressures and initial temperatures, (2) acquired the focusing optics necessary for executing single-crystal experiments, (3) developed the infrastructure for executing dynamic Laue diffraction experiments on the iron and iron–nickel systems, (4) experimented with titanium and magnesium for target packages and for heating and cooling stages and multiplexed detector platforms with collaborators at Washington State University and Los Alamos National Laboratory, and (5) developed and released open-source diffraction pattern analysis software.

Proposed Work for FY15

In FY15 we will (1) use the new capabilities we helped establish at the Dynamic Compression Sector beam line to execute single-crystal measurements on iron and iron–nickel using dynamic Laue diffraction, (2) characterize stress relaxation in the base alpha phase in the first 20 ns after impact, and (3) characterize the orientation relationships between the alpha and epsilon phases of iron and iron–nickel as a function of initial orientation, and repeat at elevated temperature.

Nanometer-Scale Porous Designer Materials

Monika Biener (13-LW-031)

Abstract

The future of many sustainable energy technologies such as green catalysis, energy conversion, and energy storage strongly depends on the availability of functional cellular bulk materials with precisely controlled architectures, compositions, and densities. Such materials are also of interest to the Laboratory's condensed-matter, high-energy-density physics program. However, realization of specific combinations of

properties by traditional self-organization and synthetic approaches based on self-assembly continues to be challenging and time consuming despite the enormous progress made in recent years. We propose to use atomic-layer deposition for rapid, on-demand development of nanometer-scale porous bulk materials with tailored properties by applying the principles of surface engineering and templates to material systems for which robust synthesis strategies have previously been developed.

We recently discovered that nanometer-thick atomic-layer-deposition surface coatings drastically improve the mechanical and thermal stability as well as the catalytic activity of nanoporous gold. We therefore expect to establish atomic-layer deposition as a general tool for rapid on-demand development of tailored nanoporous bulk materials that, for example, are urgently needed for the next generation of energy-storage and harvesting devices. In addition, we expect to develop a new class of ultralow-density bulk materials with precisely controlled density, composition, and morphology.

Mission Relevance

This project is closely aligned with the Laboratory's missions in energy security, high-energy-density physics, and the accelerated synthesis of tailored materials for advanced materials and manufacturing. Functional high-surface-area nanoscale materials have been identified as a focus area in a recent DOE Office of Science report on Basic Research Needs for Electrical Energy Storage. Ultimately, the project will strengthen the Laboratory's role in the development of nanoporous high-surface-area designer materials.

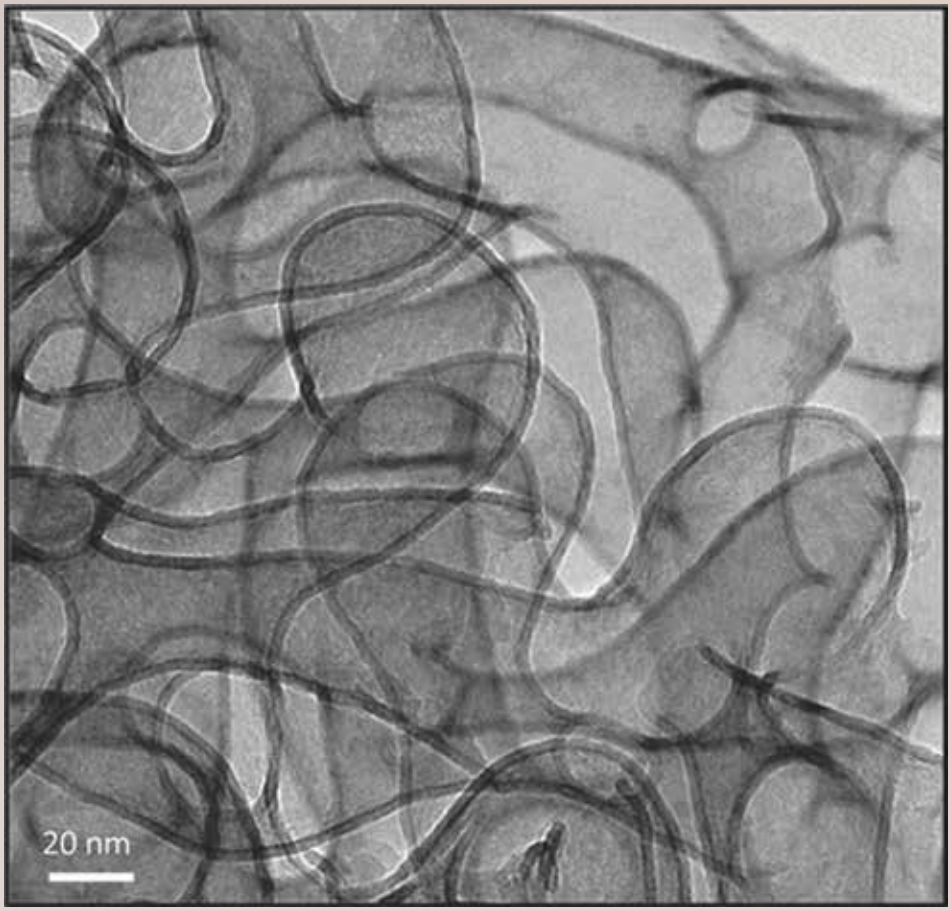
FY14 Accomplishments and Results

In FY14 we (1) studied the temperature-dependent catalytic activity of the atomic-layer-deposition functionalized material for carbon monoxide conversion; (2) investigated length scale effects of the composite material for energy storage applications (such as in lithium ion batteries); (3) developed a reliable etch process for removing the template material; (4) explored alternative template structures such as polystyrene beads, engineered microtrusses, and carbon nanotube scaffolds; and (5) submitted a record of invention.

Project Summary

The successful conclusion of this project resulted in the development of a new class of low-density, ultrastrong nanotubular materials with tunable feature sizes and deterministic composition (see figure). Our material can be used as a highly efficient back-lighter material for ultrashort multi-kiloelectronvolt x-ray pulses reaching conversion efficiencies 17 times higher than pervious aerogels. These x-ray sources are of great importance for applications such as radiography for inertial fusion plasmas or high-energy-density experiments. Exploiting the unique morphology of our nanotubular material with two independent pore systems separated by a nanometer-thick three-dimensional membrane, we also developed a new concept for high-throughput gas separation and filtration applications and submitted a record of invention for this concept. We also generated a patent application based on results from this project. Our results on lithium-ion-battery electrodes provide fundamental insight for the rational design of the three-dimensional architecture of such electrodes for

This image shows an ultralow-density (>5 mg/cm^3) bulk material with interconnected nanotubular morphology and deterministic, fully tunable feature size, composition, and density. A thin-walled nanotubular design, realized by employing templating based on atomic layer deposition, makes the material about 10 times stronger and stiffer than aerogels of the same density.



batteries that yield supercapacitor-like power performance. The work attracted international attention and established the expertise to build bright x-ray sources for diagnostics in high-energy-density physics experiments. We have identified Laboratory programmatic interest in using the materials developed in this LDRD project for future high-energy-density physics experiments. Our activities in the catalysis field will continue through our membership in the Integrated Mesoscale Architectures for Sustainable Catalysis at Harvard University, which is a new DOE-funded Energy Frontier Research Center on catalysis.

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Manipulation of Surface Plasmon Resonance by Programmable Nanometer-Scale Particle Assemblies

Tammy Olson (13-LW-066)

Abstract

Sensing devices are critical in a wide variety of national security applications. These devices often use spectroscopic methods because of their high sensitivity, relatively low cost, and abilities for remote detection. In particular, Raman spectroscopy has demonstrated tremendous potential for detection applications because of its molecular specificity. Surface plasmons (oscillation of electromagnetic waves from a metal surface) have been

used to enhance the surface sensitivity of several spectroscopic measurements including fluorescence and Raman scattering. For surface-enhanced Raman spectroscopy, we propose a new method of constructing nanometer-scale particle assemblies, which will allow controlled optimization and significantly improved sensitivity. The assemblies will be built with nanoparticle-to-nanoparticle precision for manipulating the surface plasmon resonance of the overall nanostructure. This capability will enable pre-programming of nanoparticle assemblies with the desired optical characteristics. After fabrication of the assemblies, their surface plasmon resonance will be analyzed using point spectroscopy. Our proposed method of nanostructure fabrication is not limited to sensing applications. Programmable, scalable, fast assembly of nanostructures allows for the controlled fabrication of materials to interact with electromagnetic fields in a precise manner, which would be attractive for super-lenses, waveguides, antennas, and cloaking for scientific, commercial, and defense applications.

We expect to demonstrate a method for assembling metal nanoparticles into precise, pre-designed configurations using electrophoretic deposition. The metal nanoparticles used in surface-enhanced Raman spectroscopy have highly sensitive optical properties. We expect to discover new and significant correlations of structural-to-optical properties where very minor changes to the nanoparticle assembly can shift or change the optical response significantly. Incremental and controlled modification of the structures will allow precise tuning of their optical properties. Such nanostructures can be pre-programmed and fabricated for any particular application. For example, nanostructures with specific optical characteristics can optimize a molecule's surface-enhanced Raman signal. The proposed method can also be applied to other types of materials, such as magnetic and semiconducting nanoparticles.

Mission Relevance

This project is closely aligned with the Laboratory's biosecurity mission of rapidly mitigating the threat of terrorist use of bioagents, chemicals, or explosives. Optimizing the surface-enhanced Raman signal by pre-programming and fabricating nanostructures of optimal characteristics will greatly improve the ability to detect explosives and chemical and biological warfare agents as well as environmental contaminants, biomedical markers, and more. Our proposed new methodology of constructing nanometer-scale particle assemblies with specific characteristics also supports the Laboratory's core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

In FY14 we (1) used polystyrene beads, as a model particle system to successfully demonstrate programmable particle assembly using patterned electrodes and electrophoretic deposition, because of difficulty in stabilizing the gold nanoparticles in a nonaqueous solvent (gold was the original particle system of interest because of its tunable surface plasmon resonance—polyvinylpyrrolidone was eventually found to be an effective method of stabilizing the gold particles); (2) fabricated electrodes with hole patterns, where each hole pattern accompanied one particle deposition, allowing particle assembly down to the single-particle level; (3) determined the sensitivity of the pattern's

physical dimensions for a particle deposition event to occur—a particle deposition only occurred when a certain electric field threshold was obtained for a specific hole size, which can enable multi-material assembly; and (4) performed numerical simulations on the electric field emanating from the hole patterns to understand the discriminatory behavior of particle deposition with respect to hole size.

Project Summary

Our project has resulted in a system capable of particle assembly with single-particle precision. Electrophoretic deposition was utilized to drive particles to a patterned electrode, where dimers, trimers, and higher-ordered compound assemblies were fabricated with controllable interparticle distance. Tuning of shape-dependent properties can now be achieved. Furthermore, patterned electrodes with variable hole sizes have led to the discovery of location-selective, on-demand particle deposition, where discriminatory particle deposition events were observed based on the voltage and particle-to-hole-size ratio. With decreasing patterned hole size, a larger electric field was required for a particle deposition event to occur. The simple dialing of the voltage to selectively control when (via voltage) and where (via hole size) a particle deposition took place avoids time-consuming surface modification or subsequent re-patterning of the electrode. The ability to selectively perform particle assembly on specific sites on a substrate enables multiple-material fabrication where enhanced materials properties can be realized. Discussions with external sponsors and internal Laboratory programs are ongoing to further develop the technique towards nanometer-sized particles and multi-material deposition.

Publications

Qian, F., et al., 2014. "On-demand and location selective particle assembly via electrophoretic deposition for fabricating structures with particle-to-particle precision." *Langmuir* 10.1021/la502724n. LLNL-JRNL-663405.

Transformative Catalysts for Nonconventional Feedstocks

Marcus Worsley (13-LW-099)

Abstract

A cost-effective and environmentally sound method to convert heavy hydrocarbon feedstock to liquid fuel will allow the U.S. to utilize bio-feedstock and nonconventional hydrocarbon resources, including its vast coal reserves, for energy production. However, current methods for converting coal to liquid are inefficient, requiring high temperatures that lead to high carbon dioxide emissions. We propose a new class of electrically conductive, high-surface-area catalysts that can dynamically tune catalytic reactions using an applied electrical potential to realize the environmentally sound conversion of nonconventional feedstock. The novel catalysts will consist of a mechanically and thermally robust, high-surface-area carbon-based scaffold that is directly coated with nanometer-

scale particle metal sulfide catalysts. The high electrical conductivity of a carbon support structure combined with the superior activity of the metal sulfide catalyst allows the use of an applied potential to dynamically control the catalytic process. The three-dimensional, highly porous structures combine two recently demonstrated low-temperature synthesis techniques, both of which have a high manufacturing potential because of their scalability and tuning capability. The tenfold improvement in surface area and potential fivefold increase in catalytic activity via electrochemical means should drastically increase efficiency and decrease operating temperatures for feedstock conversion.

We expect to develop a deeper understanding of catalytic reactions for feedstock widely used in conventional petroleum upgrading. In addition, we anticipate that the new class of catalysts we develop will allow for a considerable reduction in catalyst reactor loading, thus eliminating expensive separation and catalyst recovery steps in slurry operations or significantly increasing reactor productivity in fixed bed applications. More active catalysts will also allow process temperatures to be lowered, thereby reducing carbon dioxide emissions. A technology that lowers the process temperature will be significant for biofuels and coal liquefaction. If successful, this project will enable domestic-based renewable and nonrenewable fuel for transportation.

Mission Relevance

This project is closely aligned with a core LLNL mission in energy security and core competency in advanced materials and manufacturing. It would enhance the economic and energy security of the nation by reducing imports of energy from foreign sources.

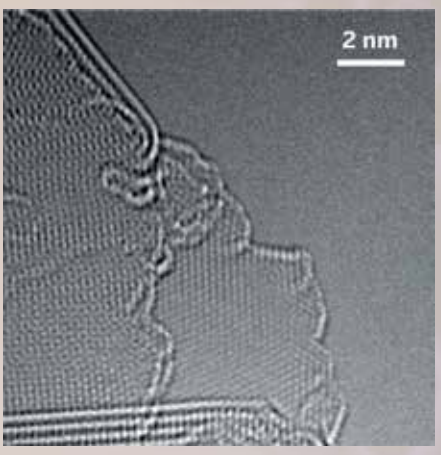
FY14 Accomplishments and Results

In FY14 we tailored the properties of a carbon scaffold and developed and tested the performance of catalysts for hydrogen evolution reaction and hydrogen sensing. Specifically, we (1) controlled the defect density of graphene (a honeycomb crystal film of graphitic carbon) in aerogel by using different annealing temperatures, which allowed us to tailor properties such as surface area, electrical conductivity, elastic modulus, oxidation resistance, and pore size; (2) developed a method to print the graphene aerogel in three dimensions, which allowed control over the macroscopic pores in the material and control of mass transport and mechanical properties; (3) determined that the printed graphene showed a tenfold higher elastic modulus than standard bulk graphene aerogel; (4) determined that the hydrogen evolution catalyst (a composite aerogel consisting of molybdenum disulfide coated onto graphene aerogel) showed a less than 100-mA overpotential (a potential difference relevant to voltage efficiency) for hydrogen evolution and was stable over many cycles; and (5) determined that the hydrogen sensing catalyst (a platinum nanoscale particle-decorated graphene aerogel) operated using extremely low powers (2.2 mW), was highly sensitive (65 ppm) and selective, and had very fast response and recovery (<1 s).

Project Summary

The successful conclusion of this project resulted in a number of technical accomplishments. Several novel carbon-based scaffold materials were developed

including (1) the first highly crystalline graphene aerogel with significantly higher electrical and mechanical properties and thermal stability (see figure); (2) a high-density bulk graphene with properties approaching that of a single graphene sheet; and (3) a high-density carbon-nanotube xerogel that maintained the large surface area of the much-lower-density aerogel. In addition, we developed novel catalyst materials such as the first dichalcogenide aerogels that show good catalytic activity for hydrogen evolution reaction, and a molybdenum-disulfide and graphene hybrid aerogel that exhibited high surface area, good conductivity, and good catalytic performance for hydrogen evolution reaction. We also developed metal nanoparticle-loaded graphene and carbon-nanotube aerogels that showed good catalytic performance for hydrogen evolution reaction and hydrogen sensing. We are in the process of establishing a cooperative research and development agreement with an industrial partner that will develop the metal nanoparticle-loaded carbon scaffolds for separation applications.



A high-resolution transmission electron micrograph reveals highly ordered carbon atoms within the layers of a graphene aerogel.

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Deterministic Multifunctional Materials and Manufacturing Initiative

Christopher Spadaccini (14-SI-004)

Abstract

Additive manufacturing—a breakthrough technology that is revolutionizing domestic and global manufacturing—creates the potential to engineer materials possessing desired structural, thermal, electrical, chemical, and photonic properties in a single package. While many types of additive manufacturing tools are commercially available, ranging from simple desktop thermoplastic printers to state-of-the-art laser powder-bed metal systems, there are still significant limitations. Some of these limitations include the ability to create components with mixtures of materials ranging from polymers to ceramics, metals, and semiconductors. Additionally, commercial platforms do not have the resolution to access unique and powerful physics at the microscale and the nanoscale. The goal of this project is to develop and thoroughly understand new design methodologies and manufacturing techniques that will enable us to produce unique multifunctional materials with mission-relevant applications ranging from stockpile stewardship to global security to nuclear fusion ignition. These material properties will be combinations of normally disparate physics such as unique structural properties combined with photonic functionality. To execute this ambitious goal, we will undertake two closely linked thrusts: inventing and developing new additive manufacturing processes and designing and characterizing multifunctional materials.

We expect to develop an understanding of the invention, development, and physical and chemical properties of new additive manufacturing processes, as well as advance the state of the art in design of material architectures. We will develop four new additive manufacturing tools: direct metal writing, diode-driven additive manufacturing, three-dimensional multiple-beam lithography, and massively parallel optical tweezing using a highly focused laser beam to hold and move microscopic dielectric objects. In addition, we will develop two methods for designing multifunctional materials: freedom-and-constraint topologies design and topology-optimized inverse design. Finally, we will design new multifunctional materials and produce them using our new additive manufacturing tools as well as tools developed previously at LLNL. The Laboratory's extensive capabilities in microscopy, nondestructive evaluation, chemical composition measurements, and mechanical testing will provide characterization of the material's performance, which will be fed back into the design process.

Mission Relevance

This project supports Livermore's missions in national security and energy security by developing techniques for manufacturing materials with applications in stockpile stewardship, energetic materials for defense uses, and materials for high-energy-density targets for fusion energy research. This project also aligns with LLNL's core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

In FY14 we (1) completed initial proof-of-concept experiments for diode-based additive manufacturing, which uses laser diodes to melt powders of polymer and metal, successfully fabricated a structure with nylon, and demonstrated the ability to melt stainless steel; (2) made progress with the direct metal write concept, identifying several alloys that can be used to extrude a semisolid form and completing initial system designs, as well as initial testing and hardware acquisition; (3) completed the initial design phase of the parallel optical-tweezer nanoscale manufacturing concept and have begun building the system; and (4) fabricated a multifunctional soft material with designed compression properties combined with electrical properties. In addition, we have established a new laboratory for housing the laser-based processes described here.

Proposed Work for FY15

In FY15 we will (1) print a small component using diode-based additive manufacturing, (2) extrude a semisolid metal using the direct metal write method, (3) assemble a working optical-tweezer system that can move and fuse multiple nanometer-scale particles, (4) establish a holographic lithography capability, and (5) print and characterize a multifunctional material using these fabrication methods.

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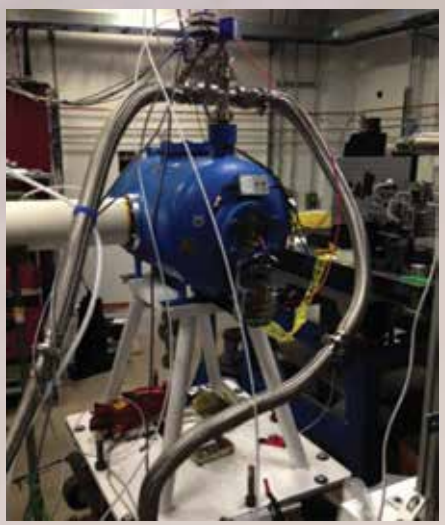
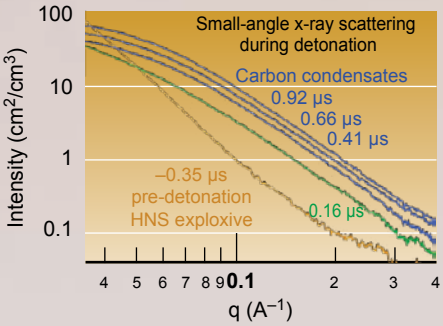
Time-Dependent Measurement of Carbon Condensation and Void Collapse in Detonating High Explosives

Trevor Willey (14-ERD-018)

Abstract

A major hindrance to developing more accurate models for high-explosive detonation is the lack of experimental data for processes occurring at sub-micron length scales (over a hundred times smaller than a grain of sand) and on timescales ranging from nanoseconds to microseconds (a millionth to a billionth of a second). We propose to address these issues with measurements of time-dependent carbon condensation and void collapse during





Detonation chamber we commissioned at the Dynamic Compression Sector at the Advanced Photon Source at Argonne National Laboratory (top). First small-angle x-ray scattering results from detonating HNS (hexanitrostilbene) is shown on the bottom. The carbon soot condensation occurs much more rapidly than expected (here, less than 0.4 μs) and the particles, once formed, do not appear to continue to grow in size over microseconds, as previously asserted for composition B, TATB (triaminotrinitrobenzene), and other explosives.

detonation. These phenomena are not amenable to measurement because of the optical opacity at the detonation front. We plan, therefore, to measure carbon condensation and void collapse using single-pulse, small-angle x-ray scattering. Experiments will begin with small amounts of about 100 mg of the carbon-rich high-explosive HNS (hexanitrostilbene), then we will use approximately 2 g of composition B (a mixture of TNT and RDX explosives), and finally transition to approximately 10 g of TATB (triaminotrinitrobenzene) and LLM-105-based materials. For HNS and composition B, we will use existing beam lines at the Advanced Photon Source at Argonne National Laboratory. For the TATB and LLM-105 experiments, we will use the Dynamic Compression Sector at the Advanced Photon Source, which is scheduled to begin operations in early FY15.

A key deliverable will be the ability to characterize the temporal evolution of void collapse and the kinetics of carbon nanometer-scale particle condensation during detonation of carbon-rich high explosives. We will derive nanoparticle-size distributions from x-ray scattering collected with 100-ps x-ray pulses over several microseconds. Initially, with HNS and composition B, we expect to observe graphitic and diamond nanoparticle growth, respectively. Subsequently, we will measure soot condensation and void collapse in LLM-105 and TATB, obtaining data that are crucial for validating predictive computational models of detonation.

Mission Relevance

Stockpile stewardship, counterterrorism efforts, and the design of advanced conventional munitions will directly benefit from our efforts to understand the fundamental processes of dynamic compression and collapse of voids for hot-spot formation and the carbon condensation and associated energy release behind explosive detonation fronts. Exploring the behavior of energetic materials during detonation also aligns with the Laboratory's core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

In FY14 we performed two major experiments: imaging exploding bridge foil initiators and measuring ultrafast small-angle x-ray scattering from detonating explosives. Specifically, we (1) obtained static images of a 0.64- by 0.64- by 0.06-mm flyer traveling at 2.5 km/s, at distances of 0.14 mm, 0.53 mm, and 0.91 mm above the surface for exploding bridge foil initiators; (2) used advanced computed tomography reconstruction algorithms to reconstruct the three-dimensional shape of the flyer; (3) determined in detail, for the first time, how the curvature and morphology of the flyer evolves with time; (4) designed, fabricated, tested, and commissioned a chamber for performing ultrafast small-angle x-ray scattering from detonating explosives at the Dynamic Compression Sector at the Advanced Photon Source (see figure); (5) measured scattering from detonating HNS, providing new insight into carbon condensation, as well as late-time energy release during detonation; and (6) determined that our data is promising for validation of LLNL detonation modeling codes.

Proposed Work for FY15

In FY15 we propose to measure time-dependent small-angle x-ray scattering during detonation of HNS and composition B explosives to dynamically measure soot condensation rates and size distributions behind detonation fronts. These processes are thought to occur over a few microseconds. The HNS explosives produce graphitic soots, while composition B produces nanometer-scale diamond soot, and we expect to see unique signatures for each. These experiments will be performed at the Dynamic Compression Sector as it becomes fully operational in FY15.

Structural Free-Standing Films with Atomic-Scale Thickness

Michael Stadermann (14-ERD-025)

Abstract

Ultrathin, tough, and defect-free polymer films are of interest to a variety of applications such as coating, packaging, separation membranes for desalination or carbon sequestration, masking layers for lithography, and sensor films. They also serve an important function in inertial-confinement fusion targets, where they support the fuel target in the center of the hohlraum target capsule. We propose to push the limits of thin-film synthesis and thin-film mechanical properties by producing, characterizing, and modeling freestanding films of atomic-scale thickness with sufficient strength to support macroscale objects. We will study the mechanical properties of ultrathin freestanding polymer films using both experiment and modeling with a special emphasis on yield and failure strain, two properties that remain virtually unstudied. Our study will include a thorough investigation of the effect of fabrication conditions on film properties, and it will link chemical and mechanical properties.

With the successful completion of this project, we expect to produce and characterize the strongest ultrathin freestanding polymer films ever made. We will gain insight into the chemical structures that give rise to this strength and develop an improved understanding of viscoelastic behavior of polymers, which will potentially transfer into bulk polymer behavior. The results can directly be utilized to produce thinner films for use in advanced laser fusion targets, where very thin, yet mechanically strong and stretchable films are desirable. Support films have to be as thin as possible to avoid perturbing the implosion, because even films as thin as 45 nm (only about 6 times thicker than a cell membrane) have been shown to have an effect on implosion. We expect to leverage LLNL's freestanding thin-film production capability that produces polyvinyl formal films for the National Ignition Facility targets by spin-coating or meniscus coating. These films are produced with nanometer uniformity on the wafer scale. Hence, the films are large enough to be tested with methods that directly measure mechanical properties, such as ball indentation or burst pressure tests.

Mission Relevance

Improved understanding of thin-film strength and advanced composite materials that will result from our research supports the Laboratory's advanced materials and manufacturing core competency, and will be useful to guide material selection for target capsule support, which is relevant to ignition-class laser fusion experiments in support of the stockpile stewardship mission and for fusion energy science and technology applications.

FY14 Accomplishments and Results

In FY14 we (1) studied the thin-film fabrication parameters and determined that more control over the environment is required to obtain reproducible results; (2) designed and began construction of a new deposition setup; (3) determined that the dynamic properties of the film are important, began efforts to alter these properties to see which values were desirable for stronger films, and developed new tests to measure these properties; (5) expanded the range of parameters studied per film from one to five to increase our ability to detect changes in the material as a function of deposition conditions; (6) used x-ray scattering, transmission electron microscopy, and ellipsometry to determine structure and crystallinity of the film as well as residual solvents; and (7) began finite-element modeling to describe burst pressure and indentation behavior.

Proposed Work for FY15

In FY15 we plan to (1) perform molecular dynamics modeling of thin-film material to elucidate the connection between chemical structure and mechanical properties—some of the characterization to establish initial film conditions for the model have already been completed, but more will be done as necessary; (2) perform chemical modifications such as cross-linking and side-chain variants, to verify results of trends found in our molecular dynamics modeling—we have created materials of varying molecular weights

A 2-mm steel ball supported by a 30-nm-thick plastic film, mounted on a holder. Ultrathin films can support up to 40,000 times their own weight.

to assist the modeling; and (3) begin work on graphene (a honeycomb crystal film of graphitic carbon) and polymer composites to determine if higher performance can be achieved with such a material.

Publications and Presentations

Stadermann, M., 2014. "Enhanced delamination of ultrathin free-standing polymer films via self-limiting surface modification." *Langmuir*. **30**(18), 5126. LLNL-JRNL-644912. *http://dx.doi.org/10.1021/la5011665*

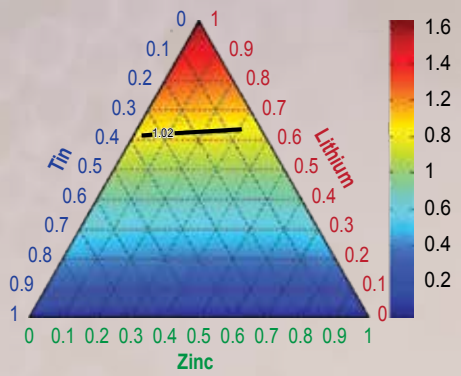
Ternary Alloy Development for Enhanced Safety and Performance of Fusion Systems

Wayne Meier (14-ERD-035)

Abstract

Pure lithium metal generally offers a balance of thermal, mechanical, and nuclear performance not found with conventional breeder materials in inertial-confinement or magnetic fusion systems. Liquid lithium advantages include its low density and resulting low hydrostatic pressures and stresses along with good heat-transfer properties and excellent corrosion properties with low impurity concentration. However, pure lithium's chemical reactivity with air, water, and other compounds and associated fire hazard remain a significant engineering challenge. Historically, the fusion community has developed advanced tritium breeding materials to compete with pure lithium. Binary alloys like lithium–lead and lithium–tin show strong promise in meeting tritium breeding ratio and energy multiplication requirements for an operational power plant. However, these alloys fall short in a number of other aspects including corrosion, weight, and operating thermal regime. We propose to study substitute ternary alloys that retain lithium's benefits, while reducing its chemical reactivity. Early modeling suggests that dilution of lithium–tin with zinc shows promise and could improve the value of fusion power plants. We will model appropriate phase diagrams and nuclear performance and study candidate alloys using standard calorimetric techniques. In addition, we will measure the hydrogen (or deuterium surrogate) solubility in these alloys and conduct air and water reaction studies for fire-safety improvements.

Our primary goal with this proposal is to quantitatively determine if a substitute alloy can be developed with the beneficial properties of pure lithium while mitigating its chemical reactivity issues. We expect to develop data on new lithium-based ternary alloys for fusion and high-temperature heat-transfer applications, perform first measurements of hydrogen solubility in that alloy, and measure fire-safety performance pertinent to fusion energy. A ternary alloy could replace the current pure lithium metal used as a heat transfer fluid for fusion energy applications. The corresponding reduction in fire hazard would significantly improve the practicality of laser inertial fusion energy and other fusion power plant designs.



This figure illustrates the effectiveness of tritium breeding for lithium–tin–zinc alloys with different fractions of each. Regions toward red have a high tritium breeding ratio while those in blue have a low tritium breeding ratio. The nearly horizontal line labeled 1.02 indicates combinations with the minimum acceptable tritium breeding ratio of 1.02—acceptable combinations of the three elements lie above this line.

Mission Relevance

Developing new underpinning technologies for fusion energy science and technology is one of the Laboratory's strategic priorities, and primary coolant reactivity has been identified as a substantial technology risk. Our proposal specifically addresses concerns in fusion safety and performances by striving to mitigate this risk by developing and studying a lithium-alloy tritium breeder that maintains performance while reducing chemical reactivity of direct lithium metal. Developing new alloys is also relevant to the Laboratory's core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

In FY14 we (1) began ternary alloy development with neutron transport and thermodynamics simulations—both sets of simulations have offered insight into the lithium–tin–zinc system, as well as led to other candidates; (2) procured the differential scanning calorimeter and installed it in a glove box; (3) began experiments on different alloy formations; and (4) began hydrogen solubility experiments.

Proposed Work for FY15

In FY15 we will (1) continue thermodynamics and neutron transport modeling for the next phase, which will include exploration of other lithium-based alloys; (2) perform optimization of the alloy formulations with collaborators at the University of California, Berkeley; (3) complete the differential scanning calorimetry task for measuring how physical properties of a sample change along with temperature over time; and (4) complete our hydrogen solubility experiments, as well as select candidates for further chemical reactivity study. These experiments will be performed with university and industrial collaborators, and results will be published in the open literature for inclusion in materials databases.

Publications

Turchi, P. E., 2014. *Thermodynamic assessment of the ternary alloy system Li–Sn–Zn*. LLNL-TR-653675.

From Topological Surfaces to Magnetic Collapse of f-Shell Electron Quantum Materials

Jason Jeffries (14-ERD-041)

Abstract

Quantum materials occupy a position at the leading edge of research in condensed-matter physics and offer tantalizing prospects for next-generation applications owing to exotic phenomena like unconventional high-temperature superconductivity, charge density modulations, and topologically protected surface states. Harnessing these

technological advantages is hampered by an incomplete understanding of their genesis, which ultimately arises from multiple quantum mechanical effects of the spin and orbit interaction and strong electronic correlations. These quantum mechanical perturbations can affect applications ranging from permanent magnets to nuclear fuel forms to quantum computing, and they can drastically alter the formation energies and phase stability of actinide alloys. We propose an experimental program combining sample synthesis and characterization, as well as high-pressure and chemical tuning. This effort will focus on select 4f-shell and 5f-shell electron systems where the electronic structure manifests topologically protected surface states, valence-induced emergent magnetism, and magneto-structural volume collapse. The measurements we perform will provide valuable benchmarks of the structure–property relationships in actinide systems, which can be simulated using state-of-the-art theories under development at LLNL.

We propose to synthesize, characterize, and tune a series of interesting f-electron materials to examine the roles of spin–orbit coupling and strong electronic correlations on the electronic, structural, and magnetic degrees of freedom. Particularly, we aim to examine topological surface states, emergent magnetism, and magneto-elastic volume collapse. We expect to reveal the first experimental evidence for conducting, topological surface states in an actinide compound; illuminate the origin of emergent magnetism in a rare earth Kondo insulator (a material with strongly correlated electrons and temperature-dependent resistivity); and discover the role that f-electron hybridization plays in magnetically driven volume collapse transformations. We will begin our studies by synthesizing single crystals using a molten metal flux technique, which permits the growth of crystals at a much lower and accessible temperature. Crystal quality will be assessed using conventional x-ray diffraction as well as electrical transport measurement.

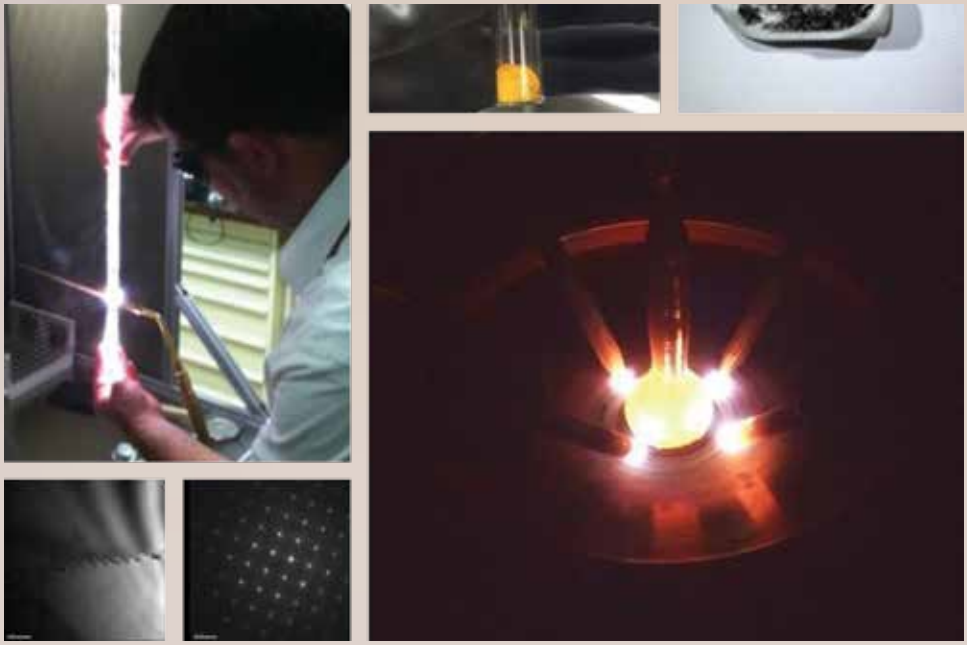
Mission Relevance

Results of a successful project will readily translate into benchmarks for predictive capabilities in actinide systems in support of LLNL's core competency in advanced materials and manufacturing, with ramifications for stockpile stewardship and weapons program activities in support of the central Laboratory mission in national security.

FY14 Accomplishments and Results

In FY14 we (1) selected and ordered an upgraded furnace for actinide synthesis that will be installed in a glove box at the Laboratory; (2) performed high-pressure, synchrotron x-ray characterization of the mixed-valent topological Kondo insulator samarium hexaboride and found that the it proceeds smoothly towards a 3+ valence state and that the x-ray emission sensitive to the f-orbital states changes little with pressure; (3) synthesized a uranium-based single crystal and provided samples to collaborators, who performed neutron scattering experiments examining the magnetic order; and (4) completed a study on a strong spin–orbit coupled system of bismuth, antimony, tellurium, and selenium, discovering superconductivity under pressure.

We have established a broadly equipped, singe-crystal synthesis laboratory for the preparation of high-quality, tailored material specimens.



Proposed Work for FY15

A postdoctoral researcher hired in FY14 will contribute substantially to FY15 efforts. Specifically, we will (1) take delivery of the actinide synthesis upgrade furnace and have it installed early in FY15, (2) begin synthesis of the plutonium compounds plutonium–tellurium or plutonium selenide, and (3) continue our rare earth and uranium-based synthesis and characterization, exploring two uranium-based alloys and a praseodymium–boron alloy. These systems provide interesting platforms that should be tunable by pressure, allowing us to examine the physics driving their magnetic and structural properties.

Real-Time Adaptive X-Ray Optics

Lisa Poyneer (14-ERD-056)

Abstract

All optical systems have distortions, which can be externally or internally generated. For astronomy applications, adaptive optics corrects these distortions by measuring the atmospheric distortions in the incoming light from a star or other object and sending electronic signals to a deformable mirror that can change its shape rapidly to correct for the distortions. In the last two decades, adaptive optics systems in astronomy have advanced from concept demonstrations to scientific mainstays, opening entirely new scientific frontiers, such as the first ground-based images and spectra of faint exoplanets orbiting other stars. Adaptive optics also have the potential to correct internal distortions and provide new, adaptive modes of operation for scientific use when applied to bright,

nearly coherent x-ray light sources. We propose to build and evaluate the performance of the first real-time adaptive x-ray optics system, using Livermore's best-in-class, 45-actuator x-ray deformable mirror with a wavefront sensor in experiments conducted at Lawrence Berkeley's Advanced Light Source. Success will enable improved beam lines, production of customized science instruments, and design and construction of new light sources. Concurrently, we will refine our simulation tools and theoretical understanding of components and system-level performance. We also hope to develop sufficient understanding about its performance to enable design of a science-specific instrument.

The development of new x-ray instruments and their subsequent use for programmatic and fundamental science investigations is a mainstay of Lawrence Livermore research. We expect to build and understand the performance of the world's first real-time adaptive x-ray optics system. Our first real-time demonstration of closed-loop adaptive x-ray optics will incorporate self-correction of our deformable mirror's surface errors at the 1-nm level. We will demonstrate improved focusing through correction of polishing errors and aspheric issues on a focusing Kirkpatrick–Baez active optic. We intend to use detailed theory and simulations to verify system performance. This proof of concept is a key step in moving adaptive x-ray optics towards regular use in x-ray light sources. The four U.S. x-ray light sources are all extremely interested in adaptive x-ray optics, and our research will provide Livermore with the opportunity to lead development of this capability.

Mission Relevance

Development of the first U.S. capability for adaptive x-ray optics can address broad DOE and Laboratory needs. Advanced x-ray optics may significantly improve beam quality at DOE x-ray light source user facilities such as the Linac Coherent Light Source at the SLAC National Accelerator Laboratory in Menlo Park and advanced diagnostics for laser-based user facilities such as Livermore's National Ignition Facility, in support of the Laboratory's core competency in lasers and optical materials science and technology, as well as the core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

In FY14 we (1) flattened the surface figure of the x-ray deformable mirror to 0.7 nm across its full 45-cm length, (2) rigorously characterized its actuator response and internal metrology sensors, (3) determined how the mirror shape changes with temperature and how to compensate for this, (4) characterized the strain gauges and ability to shape the mirror surface in a single command step, (5) integrated our detailed physics simulations into the MATLAB technical computing language code that controls our x-ray deformable mirror experiments, (6) completed the experimental design for the Advanced Light Source, and (7) successfully fielded our x-ray deformable mirror at the Advanced Light Source and commenced x-ray experiments (see figure).

Proposed Work for FY15

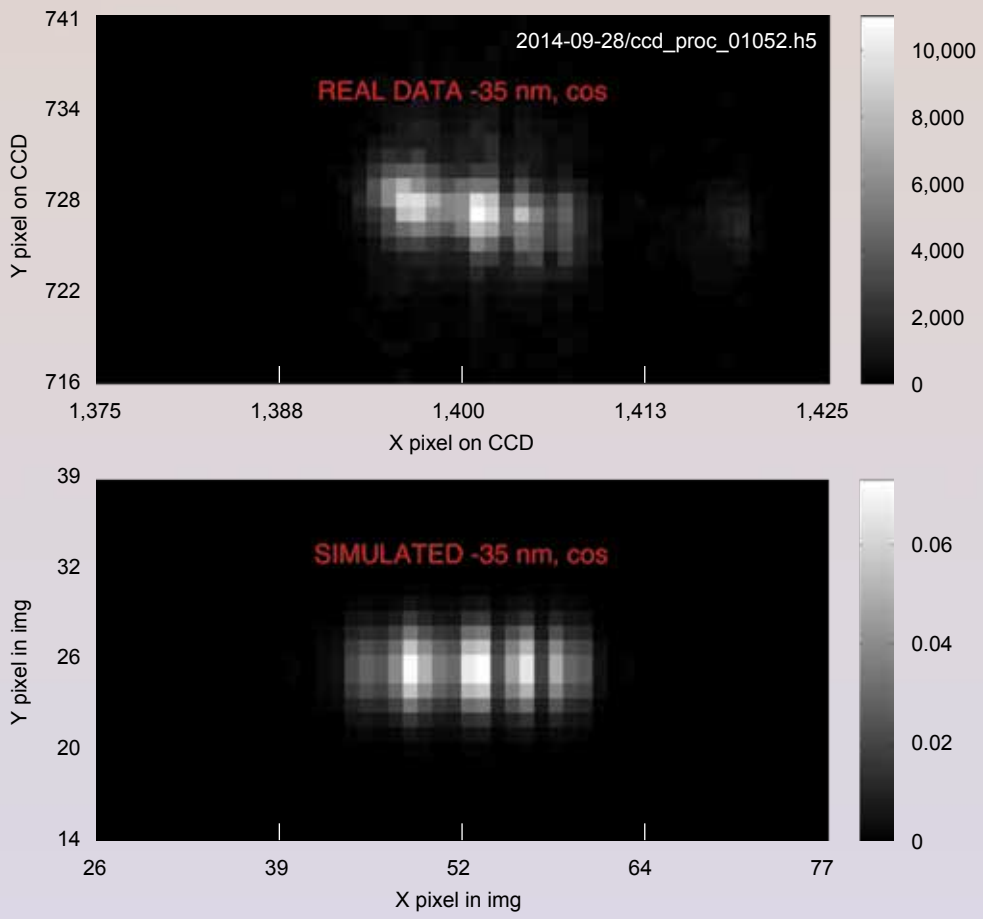
In FY15 we will (1) align the mirror and any additional hardware in the beam line at the Advanced Light Source and establish science-based metrics of performance, (2) develop

and implement algorithms to estimate correction quality from the scientific image, (3) verify that the mirror at optimal flatness improves beam quality from an un-powered state, (4) compare the beam performance to our detailed simulation of the beam line, (5) install at least one wave-front sensor and characterize it, (6) determine calibration protocol and characterize sensor performance by measuring known changes on the mirror, (7) determine impact of aliasing and noise on measurement quality, and (8) use sensor measurements to control surface height of the mirror.

Publications and Presentations

Poyneer, L. A., et al., 2014. "Control of a 45-cm long x-ray deformable mirror with either external or internal metrology." *Proc. SPIE, Adaptive X-Ray Optics III*, vol. 9208, p. 92080F-1. LLNL-CONF-659470.

Poyneer, L. A., et al., 2014. "Sub-nanometer flattening of 45 cm long, 45 actuator x-ray deformable mirror." *Appl. Optic. 53*, **3404**. LLNL-JRNL-649073. *http://dx.doi.org/10.1364/AO.53.003404*



Our x-ray deformable mirror has been deployed at Lawrence Berkeley's Advanced Light Source and is reflecting 7.5-keV x rays. At top is an experimental measurement of the x-ray spot with a 35 nm-amplitude cosine ripple placed on the height of the mirror. The height ripple has 5 cycles across the 45-cm mirror diameter. At bottom is the output of our Fourier optics simulation of this experiment.

Multifunctional Metamaterials

Mark Converse (14-ERD-064)

Abstract

We can create artificial materials with properties not found in nature whose performance is based on tightly coupled electromagnetic and mechanical microstructures. For electromagnetic materials, properties could include negative values of permittivity, permeability, or index of refraction. Mechanical properties of interest include low density, high stiffness, fracture strength versus stiffness, or even a specified stress-versus-strain curve. We propose to create one such material class, so-called multifunctional metamaterials, by combining electromagnetic and mechanical properties in novel ways that have interesting electromagnetic properties in the low-gigahertz frequency spectrum. The metamaterial will be optimized for negative index of refraction and low loss over a broad bandwidth, as well as high stiffness and low weight. A negative index metamaterial causes light to refract, or bend differently than in more-common positive refractive index materials. We will accomplish our objective by combining theory, design by computational simulation and numerical optimization, and the Laboratory's additive manufacturing expertise and capabilities. This coupling of functionality has never been done before with additive manufacturing. Such capability will greatly expand the potential of user-specified multifunctional materials.

If the project is successful, we will have created a new material class of electromagnetic and mechanical, negative-index-of-refraction multifunctional metamaterials. Such class of materials will be the first three-dimensional isotropic negative-index materials in the microwave regime. This work could affect radar and communications system design, device miniaturization, advanced sensor design, and the practical realization of many negative-index devices that are currently just a theoretical or lab-based novelty. Examples of benefits are the potential for microwave invisibility, advanced antenna design with a significantly greater range of achievable sizes and form factors, and large lightweight components, such as 10-ft microwave lenses that weigh just a few pounds. This technology could also enable sensors in which stress and strain response is coupled to the electromagnetic response to maximize sensitivity or accuracy.

Mission Relevance

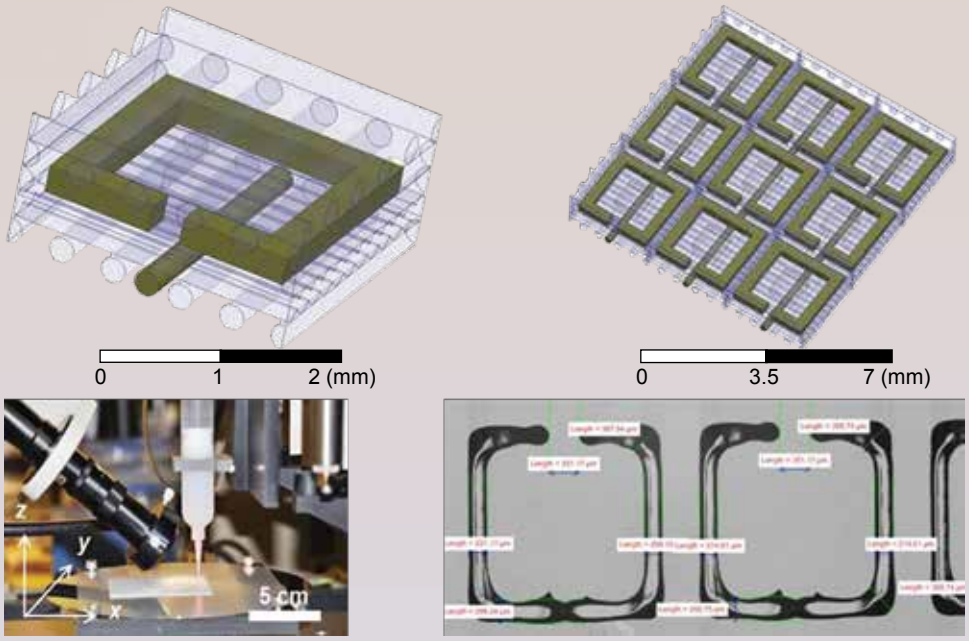
This work is important for the Laboratory's cyber security, space, and intelligence strategic focus area. It could lead to smaller microwave radar and communication components and systems, advanced microwave sensors, and greater design capabilities for any type of microwave system with user-specified materials. Creating a new material class of metamaterials also is well aligned with Livermore's core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

In FY14 we (1) determined the procedure to measure the microwave conductivity of different types and fill factors of conducting inks, (2) designed an anisotropic structurally sound material with a negative index of refraction (shown in figure), (3) attempted to develop new inks that will reliably allow smaller feature sizes while remaining flexible, and (4) worked to complete the design of an appropriate test fixture to measure new metamaterials, which is significant because we are pushing the boundaries of manufacturing techniques.

Proposed Work for FY15

In FY15 we will (1) design, build, and test a metamaterial that is isotropic, with a negative index of refraction, and built on a pre-optimized mechanically stiff and lightweight substrate; (2) expand manufacturing technique development (process and feedstock) and advanced simulation and modeling for this material, with substrate constraints, and perhaps modify the test fixture; and (3) continue conductivity measurements as needed to advance the state of the art in ink development.



A three-dimensional printed anisotropic negative-index-of-refraction metamaterial, which is our first design of a material with electromagnetic properties not found in nature that can be created with a three-dimensional printer. A single cell of design is shown top left. Silver-filled silicone (conducting) is shown in green-gold color, with clear representing silicone filaments for structural support. Multiple cells are shown top right, illustrating how the cells fit together. The material printing process is shown bottom left, and the printed split-ring components of the material are seen bottom right.

Advanced Synthesis and Characterization Techniques for Ultrahard Film Growth

Anthony van Buuren (14-ERD-067)

Abstract

Ultrahard materials are relevant to the National Ignition Facility for ignition targets, the Department of Defense for wear-resistant coatings, and the Defense Advance Research Projects Agency for low-temperature diamond growth. Chemical vapor deposition has attracted significant interest for the preparation of ultrahard coatings, yet characterization has largely been conducted post-synthesis, which restricts understanding of growth mechanisms because the surface environment changes during deposition. Developing a means to deposit ultrahard coatings with high spatial resolution and at low temperature on three-dimensional structures would represent a completely new capability that can be readily integrated into the additive manufacturing effort at Lawrence Livermore. Our proposal combines unique Laboratory-based, laser-assisted chemical vapor-deposition synthesis with x-ray-based in situ characterization to tackle this problem. Our synthesis method provides a versatile approach for ultrahard film synthesis with precise control over the location of deposition, and the in situ method enables a comprehensive characterization of the film structure at all stages of growth.

We expect to establish an x-ray-based in situ diagnostic for coating process control and rapid prototyping of ultrahard coatings. By rapidly exploring chemical and structural properties of candidate ultrahard materials with our laser-assisted diagnostic cell based on chemical vapor deposition, we expect to achieve a better understanding of these materials over short time periods. We will develop a predictive understanding of coating growth and resulting film structure and robustness, and will produce spatially selective coatings of ultrahard materials. We are confident that the research will attract the interest of the oil and gas service sector in the ability to produce designer coatings of materials such as diamond, boron carbide, and boron carbonitride for diverse applications such as high-wear parts in pumps, drill bits, and other mechanical surfaces. Materials that experience extreme chemical or thermal environments, as well as Laboratory mission-critical parts, will also benefit from our proposed synthesis method.

Mission Relevance

Our new laser-assisted chemical vapor-deposition capability and in situ diagnostics expertise supports the Laboratory's additive manufacturing efforts to produce unique materials. Research into ultrahard, fracture- and corrosion-resistant materials falls squarely in LLNL's advanced materials and manufacturing core competency, especially with regard to development of new materials and partnership building.

FY14 Accomplishments and Results

In FY14 we (1) commissioned parts for and assembled a compact portable laser-assisted chemical vapor-deposition reactor chamber with a design optimized for in situ diagnostics to be employed at a DOE x-ray user facility, (2) performed initial diamond laser-assisted chemical vapor-deposition experiments, and (3) measured the size, shape, and density of carbon films grown via flame synthesis using in situ x-ray small-angle x-ray scattering. Because of a late start and reduced budget, some of our proposed milestones for FY14 have been shifted to FY15.

Proposed Work for FY15

We plan to (1) establish laser-assisted chemical vapor-deposition growth of diamond-like carbon or boron carbide on silicon substrates with in situ small-angle x-ray scattering experiments and diffraction characterization feedback, (2) evaluate density functional theory models of growth on silicon substrates, and (3) achieve optimal diamond or boron carbide coating toughness and adhesion through full-parameter characterization of laser-assisted chemical vapor-deposition growth conditions with in situ feedback.

Optimal Fabrication Methodologies for Additive Manufacturing

Todd Weisgraber (14-ERD-087)

Abstract

Additive manufacturing is a revolutionary technology that offers the ability to fabricate, one layer at a time, the complex three-dimensional structures in a digital model through photochemical, electronic, or thermal manipulation of a feeder stock of material such as a resin or metallic powder. By tightly coupling design and manufacturing, it is spurring innovation across diverse applications in aerospace, biomedicine, and high explosives. The efficacy of each process is governed by several parameters, which traditionally have been adjusted in the laboratory to achieve robust and repeatable structures. This experimental approach towards optimization is time intensive, so there is a need for a more rigorous approach to determine if a given structure can be built, and if so, select the fabrication parameters that will produce repeatable parts with the desired material properties and functionality. Therefore, we propose to develop an optimization framework for our electrophoretic deposition and stereolithography models and apply this computational capability to improve our understanding of and streamline the manufacturing processes.

We expect to develop a new computational capability by incorporating our improved and experimentally validated process models for stereolithography optical additive manufacturing and electrophoretic coating deposition into a highly parallel optimization framework. By exercising this framework, we will provide a systematic and robust approach to determine the best processing parameters to manufacture a

variety of materials and structures that could serve as a set of standards and guidelines previously unobtainable by experiments alone. These optimization capabilities would enable more reliable builds, shorter build times, finer control over the microstructure, and aid in the design of scaled-up implementations of these processes. The outcome of our work will help establish an engineering competency in fabrication optimization.

Mission Relevance

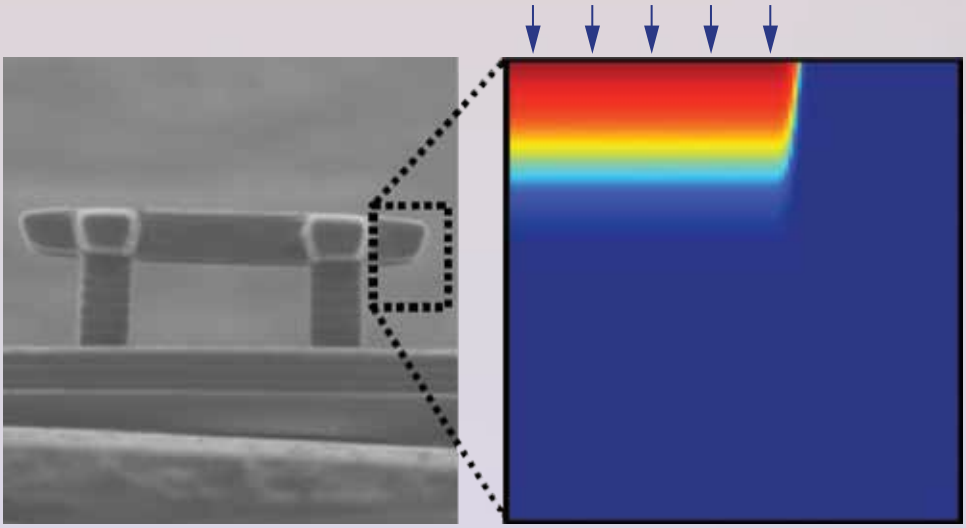
Our effort is directly aligned with the Laboratory's strategic core competencies in advanced materials and manufacturing and computational science and engineering to address the scientific and engineering challenges of accelerating the design, fundamental understanding, and development of new materials and manufacturing processes with the aid of a new computational capability. If successful, improvements in lithography and electrophoretic deposition processes enabled by this project would impact other Livermore efforts currently utilizing those fabrication methods for high-explosives, target development, and other NNSA and defense applications.

FY14 Accomplishments and Results

With a reduced budget for FY14, we adjusted our milestones and accomplished the (1) incorporation of additional chemistry for the stereolithography model, (2) development of a finite-volume version of the code within the mesh-refinement framework of Chombo software for solutions of partial differential equations with adaptive mesh refinement, (3) completion of a preliminary particle transport model for electrophoretic deposition, and (4) creation of optimization frameworks for both stereolithography and electrophoretic deposition.

Proposed Work for FY15

In FY15 we propose to (1) develop poly-disperse and multiple-material particle models with varying electrode geometries for electrophoretic deposition, (2) explore the



Comparison between a bridge structure overhang fabricated with stereolithography to the computed profile from the process model. Colors in the model indicate the amount of polymerization (blue, which is uncured, to red, which is fully polymerized).

feasibility of an electrophoretic deposition reduced-order model, (3) incorporate three-dimensional and light-scattering effects into the stereolithography model, and (4) exercise our new optimization infrastructure to improve the manufacturing processes. For example, in stereolithography, we plan to explore how to reduce the minimum feature size by perturbing the mask geometry.

Extending Atomistic Simulation to Mesoscale in Time and Length

Tomas Oppelstrup (14-ERD-094)

Abstract

Our goal is to develop algorithms for simulating materials on the mesoscale, using massively parallel kinetic Monte Carlo simulations in which results are calculated from random sampling. Current efforts are in material engineering, device design, and manufacturing target length and growth time where multiple atomic mechanisms interact to define material structure, properties, and performance. Yet existing simulation methods fall far short of the relevant length and time scales. We propose to fill this capability gap by building a code based on the ROSS (Rensselaer's Optimistic Simulation System) discrete-event simulator, which has proven to be scalable to the full Sequoia supercomputer at Lawrence Livermore. Two selected test-bed applications are crystal film growth by deposition and grain boundary network dynamics. To enable fast development, the physical models will be imported into the ROSS simulator from existing Monte Carlo codes.

We will develop, benchmark, and optimize algorithms and software for parallel kinetic Monte Carlo simulations of materials at the mesoscale with atomistic resolution. We expect to be able to simulate $10 \times 10 \times 10$ µm of material (~10^{13} atoms) grown at a rate of about 1 µm per minute at realistic physical deposition rates. This capability will enable simulation support for current efforts in additive manufacturing, rechargeable batteries, and high-powered magnets. We also expect to be able to simulate grain growth and coarsening kinetics in statistically representative (~10^3 grains), three-dimensional fragments of grain boundary networks over timescales identical to conditions used in laboratories to engineer novel high-performance materials resistant to thermal coarsening and irradiation. Successful completion of this project will aid in the development of several important mesoscale materials systems at the Laboratory across multiple programmatic areas.

Mission Relevance

Development of mesoscale atomistic simulation will enable LLNL to meet important mission needs for stockpile stewardship and energy security with growth of thick beryllium films for fusion ignition capsules, grain boundary engineering for enhanced radiation tolerance, fabrication of unconventional material architectures by additive manufacturing, and highly ramified (branch-like) material interfaces for energy storage. In addition, understanding material behavior at the mesoscale through advanced

simulations on high-performance supercomputers supports the Laboratory's core competencies in advanced materials and manufacturing and computational science and engineering, and was identified as a national priority in a recent DOE Basic Energy Sciences advisory committee report, *From Quanta to the Continuum: Opportunities for Mesoscale Science*.

FY14 Accomplishments and Results

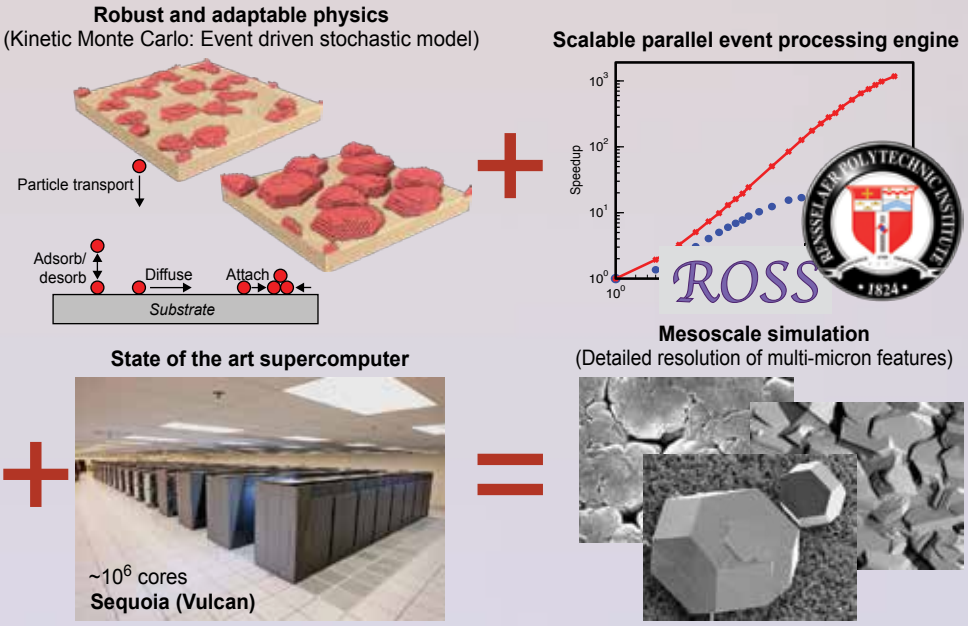
In FY14 we (1) implemented a mapping of a kinetic Monte-Carlo model onto the ROSS parallel discrete-event simulation framework; (2) tested the model on two- and three-dimensional lattice gases, which showed that even for fairly small simulations we can achieve thousandfold speedup with our parallel implementation compared to serial execution; (3) determined that the code scales to tens of thousands of processors; (4) implemented the energetics of the ADEPT (A Device Emulation Program and Tool) crystal growth code in our model, to allow realistic and scalable crystal growth simulations; and (5) developed, to support grain growth simulations, an algorithm in the appropriate form for ROSS parallel discrete-event simulation for Monte Carlo simulations of the kinetics of a general Potts spin model, which is used to study interacting electron spins on a crystalline lattice.

Proposed Work for FY15

In FY15 we will (1) develop the physics capabilities necessary for beryllium film simulation, (2) continue to tune the ROSS framework to allow increasingly large and long simulations, and (3) run the first multiple-micrometer simulation of beryllium film growth.

Publications and Presentations

Oppelstrup, T., et al., 2014. *Scalable parallel kinetic Monte-Carlo without approximation*.



We will be able to track trillions of atoms and simulate atom-by-atom growth of crystal films large enough to be visible using the supercomputing power available at Lawrence Livermore. We are combining kinetic Monte-Carlo simulation, which offers robust and time-adaptive physics modeling, with ROSS, a state-of-the-art scalable parallel discrete-event simulator.

MMM 2014, 7th Intl. Conf. Multiscale Materials Modeling, Berkeley, CA, Oct. 6–10, 2014. LLNL-ABS-652101.

Modeling Materials Under Strongly Driven Conditions

Alfredo Correa Tedesco (14-ERD-103)

Abstract

We propose to advance the field of quantum molecular simulations by developing a framework for nonadiabatic electron–nuclear dynamics (processes with energy transfer), which is a fundamental mechanism of state and phase changes in various dynamical processes of physics. Specifically, we propose to develop and apply a first-principles, parameter-free computational technique that is appropriate for describing materials under strongly driven conditions, such as those induced by electromagnetic radiation in intense laser fields; radiation of alpha and beta particles, protons, and swift ions; and nonlocal thermodynamic-equilibrium effects of different electron and ion temperatures. The Born–Oppenheimer approximation and the assumption of thermal equilibrium between ionic and electronic subsystems are ubiquitous in the field of quantum molecular dynamics. However, there are many instances, particularly for matter under extreme conditions, where these basic approximations are not appropriate. We propose to develop a new predictive simulation framework for nonadiabatic molecular dynamics by building upon our current implementation of coupled ion and electron dynamics (Ehrenfest nonadiabatic interactions between electrons and ions), where the time-dependent dynamics of electrons is taken into account explicitly.

With this project we expect to (1) develop a framework for systematic calculation of electronic and ionic excitations caused by nonadiabatic effects (e.g., electronic stopping power and collisions and ion ranges in solids of arbitrary complex materials); (2) develop fully ab initio simulations of a collision cascade including friction in the electronic environment; (3) implement electromagnetic coupling during simulated laser interaction (and after effects) for metal surfaces or damage in optical materials; (4) perform direct computation of ion–electron coupling for modeling of two-temperature systems in the electronvolt regime; (5) develop a general-purpose computational code to perform Ehrenfest dynamics; and (6) improve the scalability of existing Born–Oppenheimer dynamics via Ehrenfest dynamics.

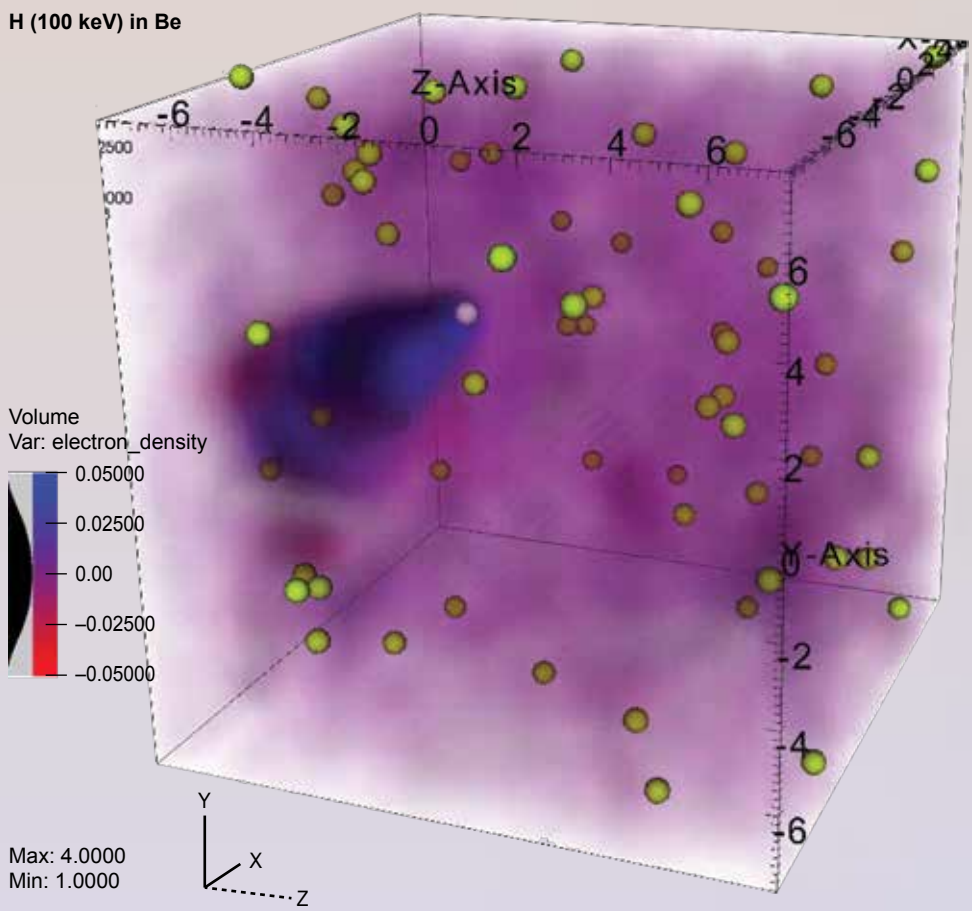
Mission Relevance

Our research into electromagnetic coupling, swift ions, and two-temperature systems has direct relevance to both NNSA and LLNL missions. The outcome of this project will result in the ability to carry out quantitative predictions of the effects of radiation in nuclear materials (as in new reactor designs and fuel containment) and strong laser

excitations (as in inertial-confinement fusion and optics), in support of Livermore core missions in national and energy security. The core competency in advanced materials and manufacturing is supported through our modeling of materials under strongly driven conditions. In addition, the code can make efficient use of the existing high-performance computing platforms at LLNL, and can be readily used by other researchers for various programmatic applications.

FY14 Accomplishments and Results

In FY14 we (1) developed the simulation framework, which is now in place and open to future extensions; (2) calculated stopping power of representative metals and insulators, which is in excellent agreement with experiments on gold, aluminum, and silica; (3) demonstrated proof of principle for electromagnetic coupling in the case of variable and static electric fields; and (4) demonstrated parallel capability of the code, showing scalability of up to a million cores in the supercomputer Sequoia and 200,000 cores in the BlueWaters supercomputer at the University of Illinois, achieving a considerable fraction of peak performance.



Electronic wake produced by a high-energy proton in liquid beryllium.

Proposed Work for FY15

In FY15 we propose to (1) apply electromagnetic coupling in real systems, such as ablation of surfaces and photo-fragmentation of molecules; (2) calculate nonlinear electric transport in this way for representative systems; (3) demonstrate Ehrenfest ion–electron coupling for both excited systems and equilibrium (but nonadiabatic) systems; (4) explore dynamics that go beyond the Ehrenfest approximation; and (5) investigate improvements in peak performance (currently at 10% in Sequoia).

Bioscience and Bioengineering

FY2014

Lawrence Livermore National Laboratory

Dynamical Imaging of Biomolecular Interactions

Matthias Frank (12-ERD-031)

Abstract

We propose to study the function of large biomolecules by obtaining molecular structures with high-resolution, coherent x-ray diffraction imaging using ultrabright x-ray pulses produced by the Linac Coherent Light Source (LCLS) at the Stanford Linear Accelerator Laboratory. The light source should allow high-resolution dynamic studies of conformational changes and interactions between molecules (e.g., between a membrane protein and a small drug molecule) on timescales ranging from sub-picoseconds to milliseconds. Our focus will be on membrane protein complexes and lipoproteins that have proven intractable to traditional structure determination efforts and that are also relevant to biosecurity, bioenergy, and human health. We intend to develop novel sample-delivery techniques that will drastically reduce sample consumption compared to current injection techniques, design x-ray imaging and pump–probe experiments to determine molecular structures with high resolution, and enable molecular movies of conformational changes and interactions.

Many of the proteins performing critical cellular functions such as nutrient uptake, signal transduction, photosynthesis, and secretion are membrane proteins, whose structure cannot be determined by traditional x-ray crystallography—this creates a major bottleneck in structural biology. Consequently, the structure of most membrane proteins remains unknown. We expect to help demonstrate the potential of coherent x-ray diffraction imaging to enable structural determination of membrane proteins and other macromolecules or complexes. If successful, our work will demonstrate broad applications for this imaging technology and provide new sample preparation and delivery methods. The work will also generate high-impact protein complex structures and dynamics with near-atomic resolution, which would greatly aid our understanding of protein function and is applicable to a wide range of fields.

Mission Relevance

Our proposed research is well aligned with the missions of both the NNSA and the Laboratory. Structure determination of virulence factors from select-agent pathogens will provide new fundamental knowledge of infectious diseases and enable new medical countermeasure development, in support of Laboratory efforts in biosecurity and core competency in bioscience and engineering. In addition, a greater understanding of biofuel synthesis proteins could facilitate the engineering of new biofuel production processes, in support of the energy security mission.

FY14 Accomplishments and Results

In FY14 we (1) led six LCLS beam-time windows where we measured two-dimensional protein crystals and three-dimensional protein micrometer- and nanometer-scale crystals using time-resolved optical pump and x-ray probe experiments on bacteriorhodpsin and

rhodopsin retinal-binding proteins as well as photoactive yellow protein, exploring structural changes on the picosecond to microsecond timescales; (2) created a three-frame molecular movie showing structural changes in photoactive yellow protein after photo-excitation at approximately 2-Å resolution; and (3) continued data analysis of results obtained at LCLS, including structural changes after photo-excitation of photosystem II (a protein unit involved in photosynthesis), resulting in the first two frames of a molecular movie, a new structure of a bacterial virulence factor from the tularemia-causing bacterium *Francisella tularensis*, and the first results from two-dimensional protein crystallography.

Project Summary

In this three-year project, we led and participated in a dozen LCLS beam-time windows. We obtained high-resolution diffraction patterns and structural information from a range of two- and three-dimensional protein microscopic- and nanometer-scale crystal samples and other biological nanoparticles. Samples included bacterial virulence factors, photosystems, coupled receptors such as rhodopsins, and other biomedically relevant proteins. We obtained the first frames of molecular movies from photosystem II and photoactive yellow protein and visualized time-resolved structural changes of other proteins. We also developed new methods of biological sample introduction including fixed-target approaches that reduce sample consumption and enable novel types of time-resolved pump–probe experiments. We are applying to the National Institutes of Health for continuing support of x-ray imaging related work through individual investigator-driven grants. Also, the DOE's Biological and Environmental Research program has begun funding work related to biological imaging with the x-ray free electron laser, which could provide us with additional support in the future.

Publications and Presentations

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Park, H. J., et al., 2013. "Toward unsupervised single-shot diffractive imaging of heterogeneous particles using x-ray free-electron lasers." *Optic. Express* **21**(23), 28729. LLNL-JRNL-648222.

Pedersoli, E., et al., 2013. "Mesoscale morphology of airborne core-shell nanoparticle clusters: X-ray laser coherent diffraction imaging." *J. Phys. B: At. Mol. Opt. Phys.* **46**, 164033. LLNL-JRNL-663494. *http://dx.doi.org/10.1088/0953-4075/46/16/164033*

Pedrini, B., et al., 2014. "7 Å resolution in protein two-dimensional-crystal x-ray diffraction at Linac Coherent Light Source." *Phil. Trans. R. Soc. B* **369**, 20130500. LLNL-JRNL-648039. *http://dx.doi.org/10.1098/rstb.2013.0500*

Redecke, L., et al., 2013. "Natively inhibited *Trypanosoma brucei* cathepsin B structure determined by using an x-ray laser." *Science* **339**(6116), 227. LLNL-JRNL-663462. *http://dx.doi.org/10.1126/science.1229663*

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Carbon Nanometer-Scale Membrane Channels

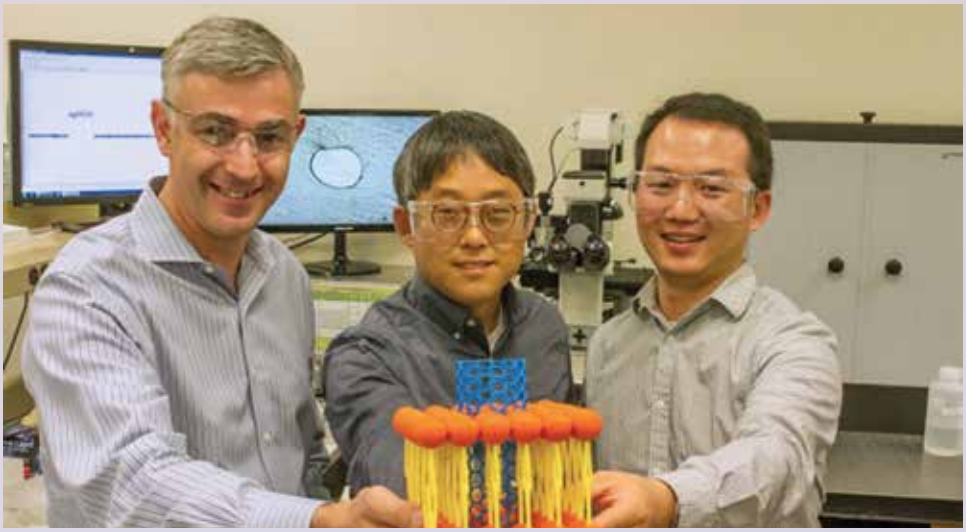
Aleksandr Noy (12-ERD-073)

Abstract

Living cells depend upon the flow of molecules across membranes for essential processes such as sensing, signaling, and energy production. Yet the cell membrane presents a formidable barrier to the transport of these molecules because they cannot cross the membrane unaided. As a result, living systems have evolved highly efficient trans-membrane protein channels that rapidly and selectively transport ions and molecules and play a key role in nutrient uptake, osmotic regulation, signal transduction, muscle contraction, and hormone secretion.

We propose to create the first artificial inorganic ion channel using short barrels of carbon nanometer-scale tubes. The inner channel of a carbon nanotube is narrow, hydrophobic, and very smooth, which has a remarkable similarity to the properties of natural biological pores. We plan to cut carbon nanotubes in short pieces that match the thickness of a lipid bilayer, insert the nanotube barrel into the lipid bilayer membrane to form a pore that permits ion transport across the bilayer, and use chemical modification to alter channel selectivity. Creating a functional abiotic mimic for these protein channels can produce new therapeutic agents, biosensors, and pore-forming antibiotic agents, as well as a versatile model system for studying design rules for transport efficiency and selectivity in membrane channels.

We expect to demonstrate a functional scaffold of a membrane channel that replicates the membrane affinity and transport properties of biological ion channels. We propose to build a family of transporters that will be based on a common structural element—a carbon nanotube membrane channel. A short segment of a cut carbon nanotube will span a membrane and form a pore that mimics a biological ion channel. We will



Lawrence Livermore National Laboratory scientists Aleksandr Noy, Kyunghoon Kim, and Jia Geng hold up a model of a carbon nanometer-scale tube trans-membrane protein that can be inserted into lipid membranes and live cell walls.

characterize transport efficiency and selectivity of these structures, as well as demonstrate specific targeting of these ion channels to bacterial membranes. The project also aims to characterize initial antibiotic activity of these structures using model bacterial systems.

Mission Relevance

Our research is well-aligned with the Laboratory's strategic thrust in biosecurity and core competency in bioscience and bioengineering through development of a membrane-penetrating structure that uses a completely different paradigm from existing membrane agents. Successful demonstration of this inorganic channel scaffold could lead to the emergence of a new class of potent antibiotic agents that would bolster resistance to pathogens and also be extremely resistant to environmental degradation. Such agents would make an important contribution to science and the development of biological countermeasures.

FY14 Accomplishments and Results

In FY14 we (1) synthesized carbon nanotube trans-membrane proteins, or porins (model shown in photo), and characterized them with cryogenic electron microscopy, measured carbon nanotube porin size selectivity, measured ion transport selectivity, and characterized selectivity mechanisms; (2) measured transport through a single carbon-nanotube porin channel, measured single-channel conductance, and demonstrated and characterized individual DNA molecule transport through carbon nanotube porins; (3) observed stochastic gating in carbon nanotube porins; (4) measured proton permeability in the porins; and (5) assembled a patch-clamp system for working with live cells.

Proposed Work for FY15

In FY15 we will (1) demonstrate protocols for chemical modification of carbon nanotube porins; (2) integrate the porins with biological organisms and demonstrate carbon-nanotube porin insertion in live carbon–hydrogen–oxygen cells, assay cell viability in bulk assays, characterize carbon-nanotube porin interactions with bacteria, develop a functional capability that targets porins to bacterial membranes, and assay antibacterial properties; and (3) prove that carbon-nanotube porins are a biocompatible and versatile mimic of a membrane channel with potential applications ranging from targeted drug delivery, to cell transfection, to antibacterial activity.

Publications and Presentations

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Kim, K., et al., 2014. "Osmotically-driven transport in carbon nanotube porins." *Nano Lett*. **14**(12), 7051. LLNL-POST-649913. *http://pubs.acs.org/doi/pdf/10.1021/nl5034446*

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Comprehensive Study and Treatment of Major Depressive Disorder Using Electrical and Chemical Methods

Vanessa Tolosa (12-LW-008)

Abstract

In 2009, suicide was the 10th leading cause of death in the U.S., with veterans accounting for 20% of all such deaths each year. Suicide rates among active-duty military personnel reached record highs in 2010, and the rate among 18- to 29-year-old veterans increased by 26% between 2005 and 2007 alone. A majority of suicide cases are linked to major depressive disorder (MDD), the underlying causes of which are largely a mystery. Consequently, many treatments remain ineffective or inefficient, and 20% of all MDD sufferers are deemed resistant to treatment. Deep brain stimulation has emerged as a promising tool to combat MDD, although its mechanisms are unknown and its parameters not optimal. We propose to develop a comprehensive method for treating and studying MDD and, in so doing, advance our understanding of both MDD and deep brain stimulation. We will combine a unique animal behavior model for depression with a novel multifunctional device to determine more-effective deep brain stimulation and pharmacological treatments as well as shed light on the pathology of MDD and other anxiety disorders.

We expect to develop a multifunctional array capable of monitoring and affecting discrete regions of the brain using more modalities than any single currently available tool of its kind. The array will be developed using technologies pioneered at LLNL that make unique use of micro-fluidic and chemical sensor expertise. We will determine whether discrete locations in the cortex are optimal for stimulation to treat depression and identify the possible role of specific receptors in the dopamine system in MDD treatment, including whether the receptors would be suitable pharmaceutical targets. Our success would lead to effective electrical and chemical treatments, including an implantable MDD treatment and monitoring device.

Mission Relevance

This project is directed toward improving military readiness in support of the Laboratory's national security mission and also contributes to the Laboratory's bioscience and bioengineering core competency. The Congressionally Directed Medical Research Program of the Department of Defense specifically identifies development of methods that will lead to improved prevention, detection, and treatment of psychological health as a research priority. Our project will enable a unique multifunctional array capable of in vivo measurements for diagnosis and drug-delivery treatment of MDD—a condition of particular interest to the Department of Defense because it afflicts a growing number of soldiers.

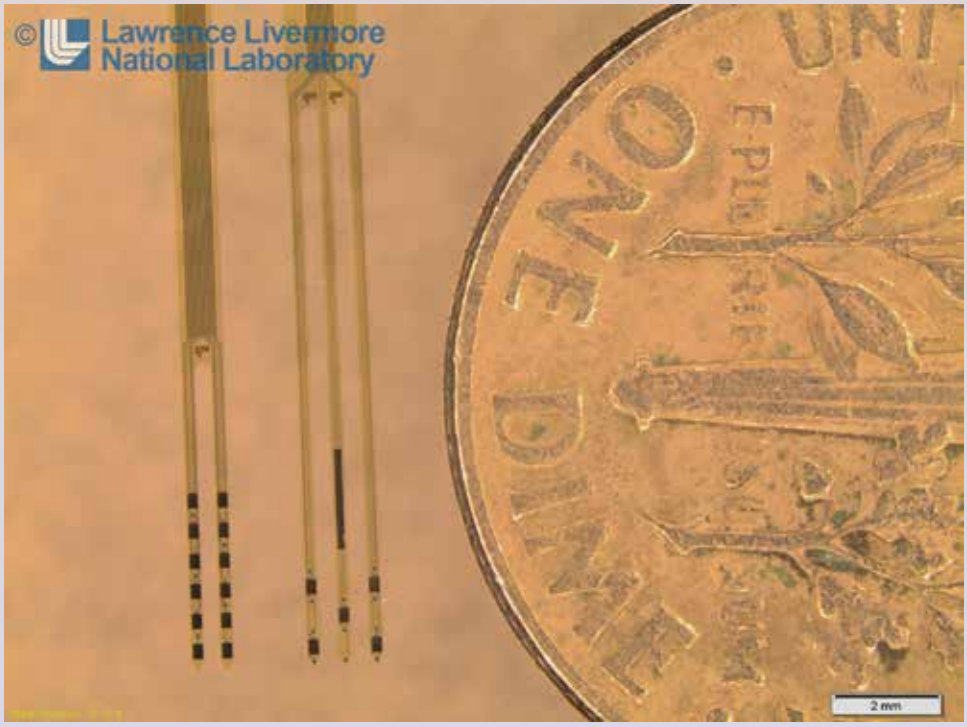
FY14 Accomplishments and Results

In FY14 we (1) designed and fabricated a neural interface customized for the sheep brain; (2) developed a novel method to modify this microfabricated polymer device into

a form factor suitable for implantation into a large animal brain; (3) prepared a patent for submission on this device modification method; (4) demonstrated that our dopamine sensor fabrication method could be modified to work on a different electrode array configuration, suggesting that our method can be used for sensors for different applications; and (5) collected in vivo recordings from animals implanted with LLNL multielectrode probe devices for up to several months, indicating that the quality and in vivo durability of these devices are very high.

Project Summary

We have successfully designed and developed a microfabricated neural interface appropriate for stimulation and recording brain activity in a large animal model (see figure). In addition, we have developed a method to take a thin-film flexible planar device and modify it to a tubular form factor with a 1.2-mm diameter. The microelectrode arrays maintain full electrical functionality even after modification. The device is designed to be about 10-cm long, able to reach the deep brain regions of a sheep. In addition, we continued to collect data with devices designed for small-animal models. An advantage of using the LLNL probes is the ability to stimulate discrete areas of the brain target in a highly specific way. In the clinical use of deep-brain stimulation, particularly for psychiatric illnesses, the effect of different frequencies for stimulation remains an open question. One of our aims was to test stimulation at frequencies from 2 to 130 Hz and to evaluate the effects on the amygdala (set of neurons located in the brain's medial temporal lobe). Preliminary data suggests that low-frequency stimulation may provide some therapeutic benefits. This effect is not seen when stimulating the medial prefrontal cortex with



Our microelectrode arrays are designed to record and stimulate brain activity.

a commercially available monopolar stimulating electrode. We are collaborating with the biomedical company Medtronic to pursue funding opportunities for expected Brain Research through Advancing Innovative Neurotechnologies Initiative grants through the Defense Advanced Research Projects Agency.

Publications and Presentations

Tooker, A. C., et al., 2013. *Microfabricated polymer-based neural interface for electrical stimulation/recording, drug delivery, and chemical sensing—Development*. 35th Ann. Intl. Conf. IEEE Engineering in Medicine and Biology Society, Osaka, Japan, July 3–7, 2013. LLNL-PRES-639756.

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Unraveling the Physics of Nanometer-Scale Fluidic Phenomena at the Single-Molecule Level

Francesco Fornasiero (13-ERD-030)

Abstract

Carbon nanotubes hold the potential to provide superior platforms for elucidating novel, poorly understood nanometer-scale fluidic phenomena, the mastery of which could impact several fields including ultrasensitive detection, protective materials, and energy harvesting and storage. To advance the understanding of these phenomena, we will develop a Coulter counter platform for investigating single-molecule transport using a single, narrow carbon nanotube as a flow channel. With this device and synergistic multiscale simulations, we will (1) decouple transport modes in carbon nanotubes, (2) unravel the relationship between molecular chemical and physical properties and transport under confinement, (3) demonstrate the detection of sub-nanometer single molecules, (4) control ionic flow with carbon-nanotube ionic diodes, and (5) understand and control the selectivity of single-molecule translocation through local carbon-nanotube function.

By coupling experiments with simulations of the electric field and pressure-driven transport of model analytes in a well-defined synthetic nanometer-scale pore, we will provide the first in-depth investigation of ionic and single-molecule flow in nanometer-scale confinement. In particular, we expect to explore the physics of biomolecular translocation in narrow pores as a function of molecular properties and driving forces. We will also establish criteria for controlling molecular flow (to provide selectivity for separation and sensing applications, for instance) and for controlling ionic flow in nanopores (to maximize efficiency in energy-harvesting and storage devices). In addition, this work may lead to a deeper understanding of the mechanisms of ion conduction and selectivity through similar-sized biological channels.

Mission Relevance

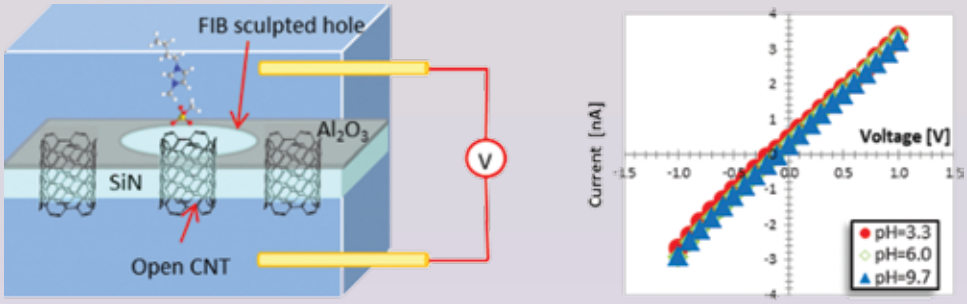
This project will further the Laboratory's core competency in bioscience and bioengineering by providing design criteria for ultrasensitive detection systems and for chemical and biological protective membranes, and will further the energy security mission by improving energy harvesting and separation. Our efforts in advanced functional nanometer-scale materials will also bolster LLNL's core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

In FY14 we (1) demonstrated that the current–voltage response of a nanometer-wide carbon-nanotube flow channel filled with ionic aqueous solutions (experimental setup shown in figure) can be linear, super-linear, or diode-like depending on the scanned voltage window and the device—unusual behavior that may be determined by a transition from partial to complete wetting of the carbon nanotube pore; (2) performed continuum calculations to examine the transport of aqueous electrolyte solutions in carbon nanotubes with symmetric and asymmetric charges; (3) conducted experiments aimed at decoupling different transport modes in carbon nanotubes, which gave clear evidence of strong electro-osmotic flow in these pores; and (4) investigated ionic flow in a carbon nanotube as a function of solution conditions and field strength, demonstrating giant ionic currents through carbon-nanotube channels and a reproducible sub-linear, power-law dependence of the conductance of a carbon-nanotube pore on concentration.

Proposed Work for FY15

In FY15 we propose to (1) investigate single-molecule translocation in a carbon-nanotube nanopore to elucidate how the Coulter-counter current signal for single-molecule translocation depends on molecular chemical and physical features such as size, charge, and shape, by employing model molecules differing by only one of these characteristics; (2) perform coupled molecular dynamics and continuum simulations for the same molecules used in the experiments to obtain fundamental insights into single-molecule motion in a carbon nanotube; and (3) investigate electrostatic and steric (atom arrangement) gating of single-molecule transport in a carbon nanotube.



Schematic representation of the experimental setup used to study electric-field-driven molecule translocation through a single carbon-nanotube channel (left). Example of a linear current–voltage response of a carbon-nanotube fluidic channel filled with ionic solutions at three different pHs (right).

Publications and Presentations

Guo, S., et al., 2014. *A simple, single-carbon-nanotube nanofluidic platform for fundamental transport studies.* 2014 Materials Research Society Spring Mtg., San Francisco, CA, Apr. 21–25, 2014. LLNL-PRES-653667.

Guo, S., et al., 2014. *A simple, single-carbon-nanotube nanofluidic platform for fundamental transport studies*. Biophysical Society Mtg., San Francisco, CA, Feb. 15–19, 2014. LLNL-POST-649578.

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Optimizing Drug Efficacy through Pharmacogenomics-Driven Personalized Therapy

Gabriela Loots (13-ERD-042)

Abstract

Patients respond differently to the same medication, which leaves a major unresolved challenge: balancing drug efficacy with toxicity to optimize drug treatments. It has been estimated that genetic makeup could account for as much as 90% of variability in drug disposition and effects. In this project, we aim to develop new methodologies for correlating genetic variation in humans with drug response. This project merges technologies in biological accelerator mass spectrometry and pharmacokinetics with genetics and genomics of bone disease in genetically modified mice. This combination, which has never been attempted, will be applied to develop novel methods for customized drug treatments based on genetic makeup. This kind of information is important because variation in drug response not only causes problems with efficacy, but has also been blamed for serious adverse events in certain therapies, including death, in up to 5% of the population.

We expect to establish new capabilities for biological accelerator mass spectrometry in the use of calcium isotopes and in bone disease and damage. This work is aimed at building capabilities for measuring metabolic and biological endpoints important for understanding and predicting variation in drug response and characterizing the magnitude of the variation. Also, with new capabilities for building and validating computational tools, we will be able to more rapidly develop and optimize therapeutic countermeasures for chemical, biological, and radiological threats. Finally, we will be able to better characterize the mechanisms responsible for bone disease and to develop therapies to treat bone damage.

Mission Relevance

This project is closely aligned with the Laboratory's national security mission in support of the military and in preparation for a chemical, biological, or radiological terror attack.

Lawrence Livermore is world leader in developing new capabilities for biological accelerator mass spectrometry, and this project will add the study of bone disease to the portfolio for this spectrometry technology, which is relevant to LLNL's core competency in bioscience and bioengineering.

FY14 Accomplishments and Results

In FY14 we (1) identified gene targets of Wnt signaling, in which proteins pass signals from outside a cell through cell surface receptors to the inside of a cell; (2) isolated bone cells from three strains of wild-type mice and treated these cells with five Wnt signal proteins (Wnt3A, 4, 5A, 7B, and 11); (3) generated 160 ribonucleic-acid sequenced samples; (4) sequenced and analyzed over 40 samples; (5) completed the analysis for Wnt3A in two strains of mouse cells; (6) found 622 genes to be differentially expressed; and (7) initiated the analysis of Wnt7B samples and found only 75 genes to be shared by Wnt3A and Wnt7B.

Proposed Work for FY15

In FY15 we will (1) conclude analysis of all cells treated with Wnt signal proteins, (2) complete sample generation and submit samples from two strains of mice for sequencing, (3) determine changes in bone and drug metabolism as a function of genetic makeup, (4) predict new genes that contribute to bone metabolism and parathyroid hormone treatment failure, and (5) initiate in vitro testing of new candidates to determine if some of these genes can promote bone formation, de novo.

Wonder Bugs and the Carbon Cycle: Characterizing the Carbon Metabolism of Thaumarchaeota

Anne Dekas (13-LW-032)

Abstract

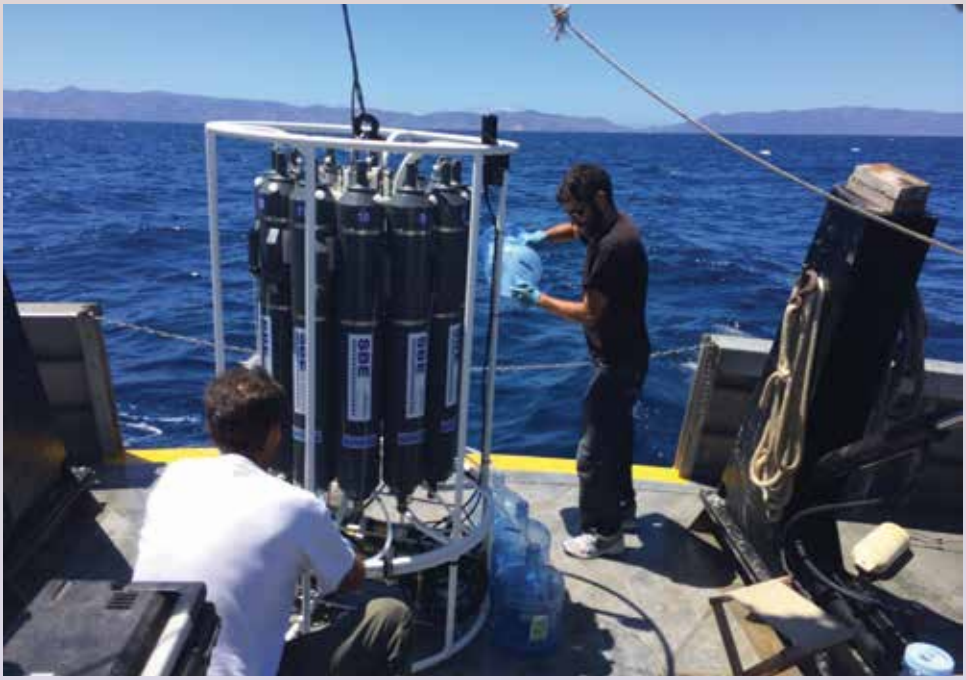
Microbial life is central to the global carbon and nitrogen cycles, but its major players and metabolic diversity are still being identified and characterized. Using experiments and technological development, we will use soil and marine samples to determine the carbon and nitrogen metabolism of Thaumarchaeota, a major yet only recently discovered group of microorganisms that are thought to constitute a significant carbon dioxide sink. They may utilize carbon to build biomass and additionally have been shown to contribute significantly to atmospheric nitrous oxide, a greenhouse gas. Previous attempts to investigate their metabolic capabilities have largely depended on their isolation and individual physiological characterization, a slow process that yields an incomplete view of the activity of these microorganisms in situ. We will apply the Laboratory's chip stable-isotope probing (Chip-SIP) technology for use with mRNA to directly link Thaumarchaeal carbon uptake to ammonia oxidation and to create a broadly useful measure of turnover times of different cellular nucleic acid pools. The Chip-SIP

is a combination of a microarray slide (the chip) and an analytical method commonly used by microbial ecologists called SIP, which is a high-throughput, high-sensitivity technique for linking the activities of microbes to their identity. We will quantitatively assess the carbon assimilation capabilities of Thaumarchaeota and the environmental factors that control them across a broad range of systems.

By comparing the uptake of nitrogen-15 and carbon-13 in the mRNA, rRNA, and DNA of microbial cultures, we will determine the turnover times of different types of nucleic acids and calculate the quantity of RNA synthesized from new material per cell division. This information represents a fundamental contribution to the field of cellular biochemistry. Comparing the magnitude of Thaumarchaea carbon-13 uptake into RNA from organic carbon substrates versus carbon dioxide will determine if these globally significant organisms are autotrophic or heterotrophic or able to use a mix of different sources of energy and carbon. Examining this uptake in the amoA-gene transcripts will reveal which carbon substrates the amoA-encoding Thaumarchaeota assimilate. This information, combined with previously collected data on their distribution, will establish their role in global carbon cycling and carbon dioxide sequestration.

Mission Relevance

This project is closely aligned with LLNL missions in climate and energy security because of its potential to contribute to our understanding of global carbon cycling and modeling of the global carbon cycle. Determining carbon and nitrogen metabolism of new microorganisms aligns directly with the Laboratory's core competency in bioscience and bioengineering.



Collecting marine water on a ship in the San Pedro Basin, Pacific Ocean for studies to determine the carbon and nitrogen metabolism of Thaumarchaeota, a major yet only recently discovered group of microorganisms that are thought to constitute a significant carbon dioxide sink.

FY14 Accomplishments and Results

In FY14 we (1) conducted 3 field expeditions, in total collecting over 800 L of Pacific Ocean water (see figure); (2) carried out 119 isotope-labeling experiments with 9 potential microbial food sources labeled with carbon-13 and nitrogen-15; (3) extracted RNA and DNA and obtained sequences to determine the diversity, identity, and activity of microorganisms present in the experiments; (4) measured assimilation of the isotopically labeled food sources by both bulk isotope-ratio mass spectrometry and single-cell nanoscale secondary ion-mass spectrometry (>2,500 cells analyzed); and (5) demonstrated that individual marine microbes, likely including Thaumarchaea, live autotrophically, heterotrophically, and with a mix of both.

Proposed Work for FY15

In FY15 we will explore the genetic basis of the activity observed during our current isotope-tracer experiments. We will collect three sets of data complementary to our current analysis, using the same samples we are studying: (1) single-cell amplified genomes of uncultured Thaumarchaea, selecting the individual cells to represent the archaeal phylotypes demonstrating interesting patterns of activity; (2) meta-transcriptomes at two marine and two terrestrial sites, indicating which phylotypes mediate which metabolisms in situ; and (3) archaeal populations in each of our organic-carbon-amended incubation experiments, performing iTag polymerase sequencing to determine which phylotypes are enriched under what circumstances.

Publications and Presentations

Dekas, A. E., 2013. *Investigating climatically-relevant archaeal and bacterial metabolisms*. LLNL-POST-639316.

Simulated Opening of the Glutamate Receptor for Enabling Alzheimer's Treatment

Timothy Carpenter (13-LW-085)

Abstract

Glutamate receptors are one of the brain's most prevalent and important neurological receptors. Upon activation, the receptor channel opens and conducts cations, which depolarize the cell and propagate the nerve impulse. Over-stimulation of glutamate receptors leads to nerve cell damage and death, which is linked to neurodegenerative diseases such as Alzheimer's, Huntington's, and Parkinson's diseases. Thus, prevention of nerve cell damage and death arising from over-stimulation of these glutamate receptors can be achieved through their inhibition. Glutamate receptor inhibitors that are partial open-channel blockers may be the best bet for treating chronic forms of dementia. However, any development of future potential therapeutics based on the structure of an open-channel blocker-binding site is hindered, because conformation of the glutamate receptor is only known in the closed state. We propose to generate

an open-channel model of the N-methyl-D-aspartate glutamate receptor (NMDAR), by combining pioneering molecular dynamics techniques, which will enable us to characterize both its opening mechanism and the open-channel blocker-binding sites. This detailed atomistic understanding will provide a valuable tool for designing safe and effective inhibitors of nerve cell damage and death.

We expect to develop an NMDAR open-state model, which will represent the first accurate model of the open state for this glutamate receptor. By combining the pioneering techniques of steered and targeted molecular dynamics and the nudged elastic-band method (used to identify reaction pathways in biological systems when both the initial and final states are known and linearly interpolating a set of images between the known states), we will observe the opening mechanism of the NMDAR and provide a biologically relevant conformational path for the opening mechanism. In addition, we will provide the first atomistic-scale understanding of the binding site and mechanism that will lead to advancement of ligand binding design. The simulation of this system (500,000 atoms) represents quite a technical challenge, necessitating the high-performance and massively parallel computing facilities available at LLNL. Even a standard molecular dynamics simulation of this system would represent one of the largest explicit simulations of membrane protein ever performed.

Mission Relevance

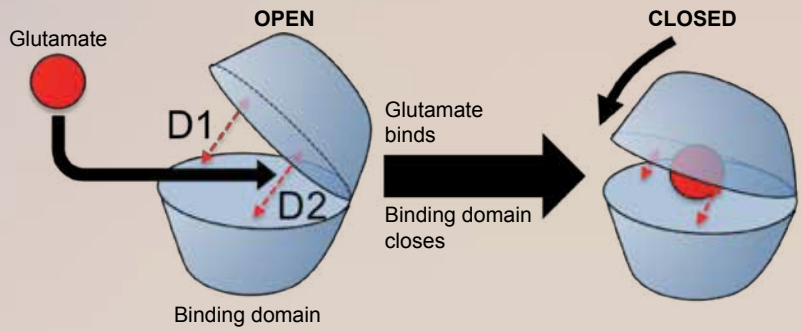
In addition to addressing the advance of medical therapeutics, the project also has applications to medical countermeasures in the fight against chemical terrorism in support of the Laboratory's mission in national security. Upon nerve agent exposure, a neurotransmitter release cascade occurs in the brain (of which NMDARs are involved), leading to an uncontrollable spiral towards seizure. Our research will provide vital information for structure-based drug design by providing a detailed model of the NMDAR open state, which is relevant to the Laboratory's core competency in bioscience and bioengineering.

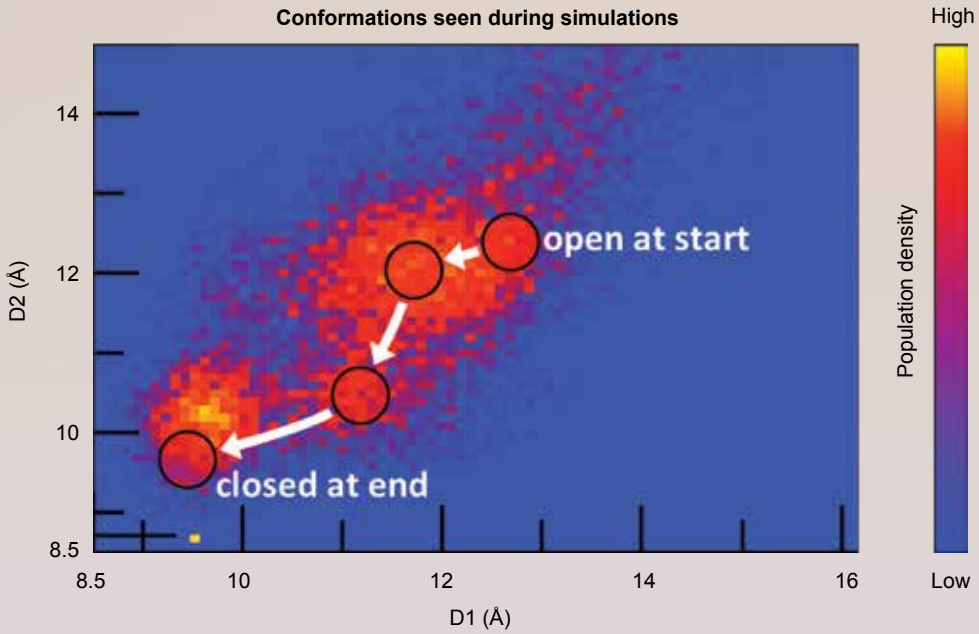
FY14 Accomplishments and Results

In FY14 we (1) thoroughly completed the nudged elastic-band analysis to discern the closing pathways for the "clamshell" ligand-binding domains and showed complete, unbiased closure of the ligand-binding domain upon glutamate binding (as shown in the figure)—these pathways proved to be more complex than originally anticipated, progressing in a stepwise fashion via several intermediates; (2) identified the intermediates involved in these complex pathways; and (3) re-evaluated the scope of the project to concentrate on the ligand-binding domain to ensure that the behavior was correct.

Project Summary

The rescoped project was focused on the detailed, successful evaluation of the ligand-binding domain closing pathway. The protocols and simulations needed to be validated in several ways, including work on other well-known proteins. We were able to show complete closure of the ligand-binding domain upon glutamate binding, something that has not been previously demonstrated. This closing pathway also





An illustrative diagram of the "clamshell" binding domain (top). From analyzing the population density of observed conformations, we are able to construct a closing pathway for the domain upon glutamate binding (bottom).

revealed hitherto unidentified intermediates of the domain closure. These intermediates necessitated further investigation to understand the nuanced, subtle mechanism by which closure occurs. This investigation was carried out using the nudged elastic-band advanced molecular dynamics method, as implemented within the AMBER molecular dynamics package. The package is now successfully installed and running efficiently on the Livermore Computing Center machine Sierra, in collaboration with the San Diego Super Computer Center. The methodologies and protocols developed in this study will be further implemented in upcoming projects and the Livermore-led Biological Applications of Advanced Strategic Computing Initiative. It has also lead to the establishment of an industrial collaboration with AnaBios Corporation in San Diego, which focuses on drug discovery and safety.

Publications and Presentations

Carpenter, T. S., and F. C. Lightstone, 2014. *Simulated closing of the glutamate receptor ligand-binding domain*. Biophysical Society 59th Ann. Mtg., Baltimore, MD, Feb. 7–11, 2015. LLNL-ABS-662135.

Zhao, C., et al., 2014. "GABA(A) receptor target of tetramethylenedisulfotetramine." *Proc. Natl. Acad. Unit. States Am*. **111**(23), 8607. LLNL-JRNL-649601. h*ttp://dx.doi.org/10.1073/pnas.1407379111*

In Vitro Chip-Based Human Investigational Platform

Satinderpall Pannu (14-SI-001)

Abstract

Building upon the success of a developmental platform for investigating primary human dorsal root ganglion cells, we propose to integrate human organ systems into an instrumented, micro-fluidic platform. Our ultimate objective is to create a highly integrated, multiple-organ, human-relevant in vitro platform to reproduce in vivo physiological response. We intend to develop tissue systems including dorsal root ganglia, central nervous system neurons, blood brain barrier, and cardiac tissue. We envision that this platform can be used to rapidly assess and predict the toxicity, safety, and efficacy of countermeasures against chemical and biological agents. Our research will reduce preclinical testing and improve relevance to clinical outcomes with technologies that utilize in vitro platforms with primary human cells organized in a physiologically relevant manner. The platform will also enable investigation of the mechanisms of infection for emerging threats, and it will be used to understand the evolution of threats in physiologically relevant human tissue.

We expect to deliver a validated human investigational platform with four human tissue systems of dorsal root ganglia, central nervous system neurons, blood brain barrier, and cardiac tissue. We will validate this platform against known chemical agents to determine its performance compared to in vivo toxicity data. Our five main milestones are (1) determine protocols and techniques to harvest, digest, and culture tissue systems; (2) determine the appropriate assays to quantify cell viability and response to exposures; (3) develop the platform with integrated fluidics and electrodes; (4) demonstrate the in vitro platform with tissue systems; and (5) validate the in vitro platform with known chemical agents.

Mission Relevance

Our development of the first in vitro platform for testing and characterizing toxins on neural and cardiac tissues supports the Laboratory bioscience and bioengineering core competency and addresses the biosecurity mission in the area of medical countermeasures for rapid mitigation of emerging and unknown threats. Our research will enable timely medical countermeasures that treat disease or toxic effects from a terrorist attack that employs chemical or biological agents.

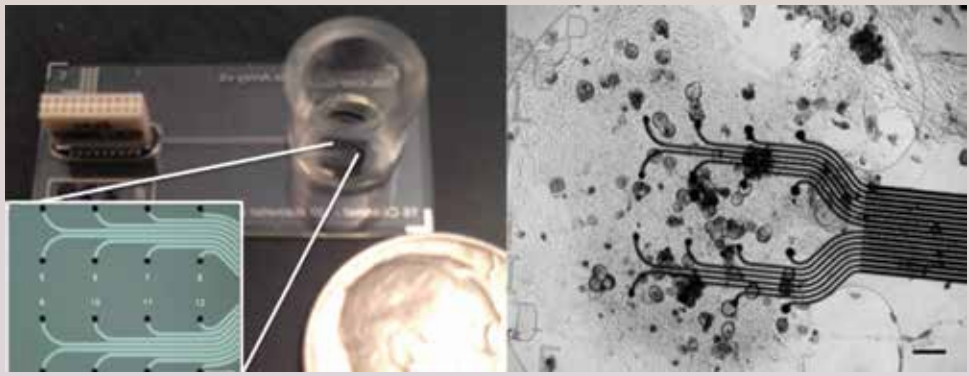
FY14 Accomplishments and Results

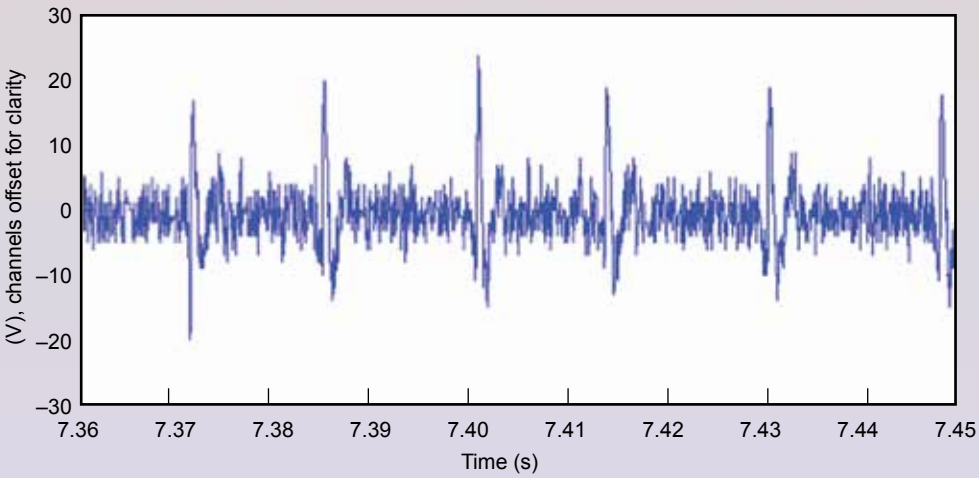
In FY14 we focused on the validation of our novel peripheral nervous system. Specific accomplishments included (1) demonstration of the first primary human cell response

to a surrogate chemical exposure on the human investigational platform at LLNL, as shown in the figure; (2) maintenance and demonstration of human primary dorsal root ganglia neurons to be viable on the platform for one month in vitro—not only were the cells viable, but upon exposure to the irritant capsaicin, action potentials were recorded with the embedded electrode array; (3) incorporation of an automated fluidic delivery system into the investigational platform in preparation for exposure of these cells to additional chemical agents—the fluidic system is gentler on the cells; (4) the first correlated simultaneous optical and electrical recordings in response to chemical exposures; and (5) maintenance of nonhuman neurons on the iCHIP platform—the neurons were viable for more than three months.

Proposed Work for FY15

In FY15 we will (1) add a heart tissue system to the human investigational platform; (2) develop and validate the platform as well as the protocols to sustain cardiac cells—the cardiac platform will include both mechanical and electrical sensors for determination of cell health; (3) validate the heart tissue system by exposing the cardiac cells to chlorpyrifos (an organophosphate), which binds to the muscarinic acetylcholine protein receptors that regulate cardiac function; and (4) begin development of the blood brain barrier system.





Multiple-electrode arrays are embedded in automated fluidic device (upper left). After human dorsal root ganglion neurons have been seeded on the device, neurons are challenged with chemicals and both their optical and electrophysiological responses are recorded (upper right). A section of an electrophysiological spike train from a single neuron after a chemical challenge is shown at bottom.

Publications and Presentations

McNerney, M. W., et al., 2014. *Calcium imaging of cultured DRGs exposed to an organophosphate pesticide*. LLNL-POST-645723.

Biological Printing of Vasculature for Artificially Grown Tissue

Elizabeth Wheeler (14-ERD-005)

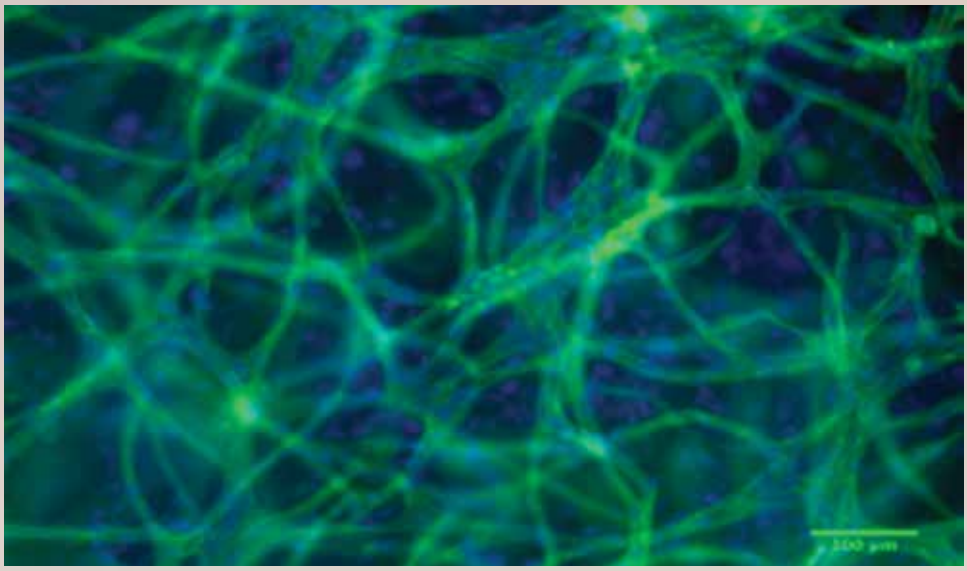
Abstract

The development of engineered tissue that represents human physiology will help increase the quality and predictability of therapies that move through the Food and Drug Administration approval pipeline and into clinical use. In vitro tissue models to date feature engineered organs that survive for only a few weeks and grow, at most, to a few hundred microns thick. Based on our understanding of human physiology, a single monolayer of tissue is not representative of an entire organ. In addition, tissue found in vivo is within 200 µm of the closest capillary network. Beyond this distance, nutrients and oxygen diffusion is limited and tissue does not survive. The primary reason that thicker tissue constructs have not been widely demonstrated is because of the difficulty of integrating vascular networks into artificially grown tissue. We propose to address this challenge by using biological printing methods to assemble sophisticated capillary networks that can deliver nutrients through thick human tissue. Our four specific goals for this project include (1) development of printable biological ink, (2) coaxial printing and extrusion of a biological vessel, (3) vascular-membrane tissue characterization, and (4) perfusion studies for homogenous distribution of nutrients through multiple layers of thick tissue.

Our research could enable the first sustainable platform of networks capable of distributing oxygen and nutrients homogenously through several layers of tissue, with the use of only natural biological materials. We propose to address this challenge by implementing a more advanced printing technique to assemble biological material into sophisticated capillary networks. The goals of this project will be achieved by developing a strong team of biologists and engineers at LLNL as well as by forming strategic partnerships outside of the Laboratory. The proposed research will develop capabilities in tissue engineering of interest to the Defense Advanced Research Projects Agency, Defense Threat Reduction Agency, National Institutes of Health, and Food and Drug Administration, all of which have expressed interest in developing organ-on-a-chip technologies to accelerate research of host–pathogen, therapeutic, and vaccine interactions with human tissue.

Mission Relevance

Integrating complex microvascular networks in thick living tissue will generate relevant tissue models that measure the therapeutic response to unknown chemical and biological agents, in support of a central Laboratory mission in national security, specifically in the area of biosecurity to develop platforms and tools for rapid medical countermeasures to emerging threats, which also supports the core competency in bioscience and bioengineering.



Human microscopic vessels printed with Lawrence Livermore's biological printer.

FY14 Accomplishments and Results

We have been able to hire a biomedical engineer with an emphasis in tissue engineering for our research, which has allowed us to gain much experience in a short time. Major accomplishments for the year include (1) performing a biological printing of a vasculature network in the presence of support cells (microscopic vessels shown in figure), (2) performing a comparison of free-form versus structured networks in three dimensions, (3) characterizing the material properties of both the tissue scaffold and the biological ink, and (4) printing a functional three-dimensional configuration of cells by optimizing the structural support, organization, and stability of the biological ink and extracellular matrix.

Proposed Work for FY15

Goals for FY15 include (1) characterizing the vasculature network by measuring the tight junctions of the endothelial barrier (cellular lining of blood vessels), (2) introducing additional tissue types, (3) perfusing the vasculature network and monitoring cell health, and (4) building more complicated (larger and thicker) structures.

Analysis of a Metabolically Engineered Microbial Consortium for Optimal Production of Biofuels

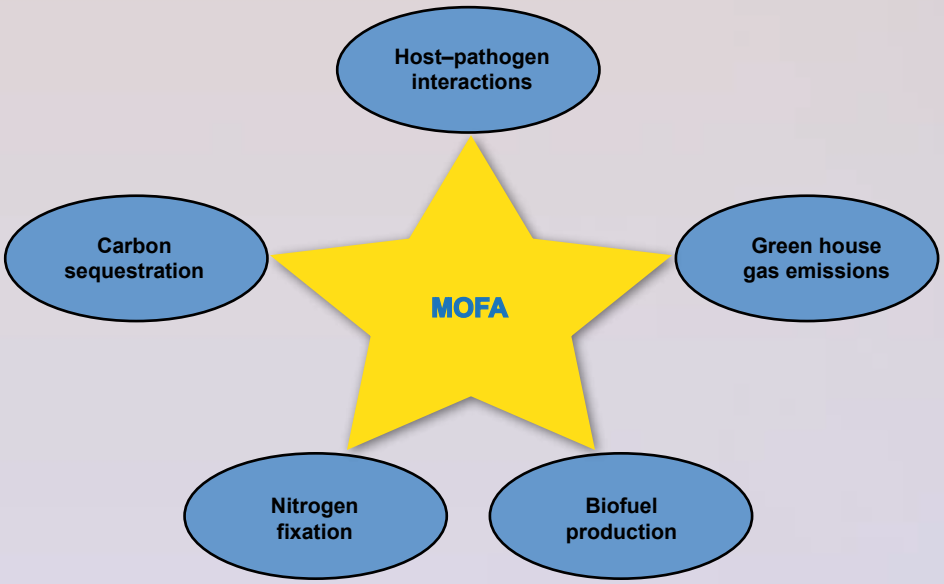
Ali Navid (14-ERD-091)

Abstract

Engineering of microbial consortia is a new frontier in synthetic biology. By programming the conduct and performance of select microbial communities, we can

force these organisms to coordinate their efforts to achieve a specific goal such as production of compounds of interest like biofuels or drugs. Engineering multicellular communities to achieve a specific goal requires system-level understanding of the workings and capabilities of each organism in the community and their interactions. Computational models are usually used to conduct such system-level analyses. Unfortunately, available modeling tools and methods are limited to examining only one objective of the system, while analysis of multicellular communities requires developmental models that conduct multiple-objective flux analysis of the system (depicted in figure). Our primary goal with this project is to develop an algorithm for generating multiple-objective flux-analysis models for interactions among cells in multicellular communities. We will use this method to optimize biofuel production in a synthetic co-culture of mutant strains of *Clostridium phytofermentans*, an anaerobic, rod-shaped bacterium capable of producing ethanol and hydrogen gas.

The success of this project will result in (1) development of a computational tool for generation of genome-scale multiple-objective flux-analysis models that can be run using the Laboratory's high-performance computers, (2) added insight into central carbon metabolism of biofuel-producing organisms, (3) establishment of a novel metabolically engineered consortium of multiple strains of *C. phytofermentans* that have been optimized for peak production of ethanol, and (4) a system-level analysis of multicellular communities that greatly benefits microbial consortia engineering efforts. Developing a tool for automatic generation of genome-scale multiple-objective flux analysis models of metabolism will be of great utility for systems biology studies of multicellular systems. Coupling this progress to the Laboratory's extensive computational capabilities will place us at the forefront of examining and engineering complex multicellular systems.



A multiple-objective flux analysis (MOFA) of multicellular communities allows for a system-level examination of the metabolic capabilities of these groupings. This new insight can be used to engineer microbial consortia that can help in achieving a number of critical environmental, industrial, and biomedical objectives.

Mission Relevance

This project aligns well with LLNL's strategic focus area in biosecurity and core competency in bioscience and bioengineering. Results of our development of an algorithm for automatic generation of models that would account for different metabolic objectives of diverse members of a microbial community can be used for the development of new countermeasures against biosecurity threats and examination of interactions between immune system and pathogens. In addition, it will allow development of novel systems for generation of renewable fuels, in support of the strategic focus area in energy and climate.

FY14 Accomplishments and Results

For FY14 we (1) established multiple cultures of *C. phytofermentans*, including one wild type and three adapted strains that use xylan, cellobiose, and cellulose as preferred carbon sources; (2) employed quantitative metabolic techniques to experimentally characterize the growth rate and metabolism of these organisms under a variety of different nutritional conditions; (3) acquired an automatically generated draft flux-balance analysis model of *C. phytofermentans*; (4) corrected and added constraints to the draft flux-balance analysis model using results from our experimental characterizations; and (5) conducted system-level computational analyses of metabolism in the different strains of *C. phytofermentans*—in particular, we identified genes that should be inactivated to generate single-sugar-consuming mutant strains.

Proposed Work for FY15

During FY15 we will (1) complete curation of the *C. phytofermentans* flux balance analysis model and identification of genes that should be inactivated to generate single-sugar-consuming mutant strains, (2) engineer mutant strains for metabolic analysis, (3) verify model predictions and establishment of desired phenotypes, (4) assess metabolic and growth characteristics of mutants growing alone and in a community using quantitative study of the chemical processes involving metabolites, and (5) develop an algorithm for generation of models of an engineered community using mopA genes linked to protein folding and associated processing used in metabolic engineering, and analyze them using high-performance computing.

New Steady-State Viral Culturing Platform for Infectious-Disease Therapeutics

Maxim Shusteff (14-LW-077)

Abstract

Recent decades have seen enormous leaps forward in DNA sequencing, bioinformatics, and viral genetics. Despite these advances, viral infectious diseases continue to present major public health threats worldwide, as well as to U.S. national security. A major reason that effective therapies and countermeasures remain extraordinarily challenging to

develop is that viruses are dynamic systems, while most tools for studying them are static. Studying viruses in vitro significantly distorts their infection patterns, evolutionary parameters, and replication dynamics. Where animal models exist, experimental flexibility is significantly more limited than in vitro. A bridging capability is needed that can combine the control and flexibility of in vitro environments with more realistic in vivo system dynamics. Our main goal with this project is to apply microfluidic acoustic filtering to build a viral culture platform that more realistically mimics the dynamic equilibrium of in vivo infection, compared with standard in vitro culture methods. The proposed system will use acoustic separation to clear free virus particles and re-circulate infected and uninfected host cells back into the culture, establishing the first quasi-steady-state viral culture. In collaboration with world-class virologists, we will demonstrate the utility of the system for investigating different host–virus systems.

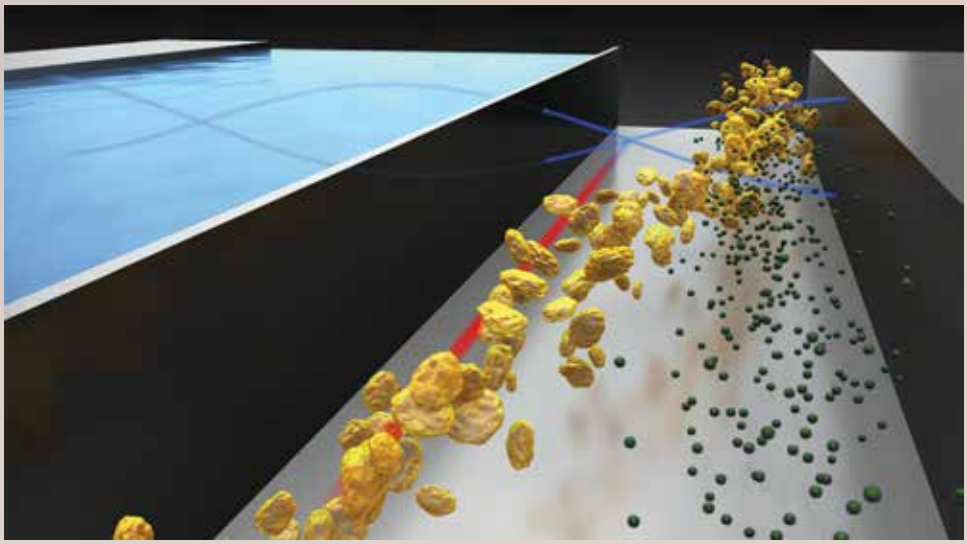
Successful completion of this project will result in a bridging technology, filling the capability gap between static viral culture (simple and inaccurate) and the use of animal models or clinical studies of human patients (complex and costly). We will create a microfluidic device for development of an equilibrium culture system. To accomplish the influx of fresh cells required simultaneously with clearance of viral particles, we will couple the microfluidic filter to a commercial mammalian cell-culture bioreactor that will deliver uninfected lymphocytes to the culture to replenish cells killed by viral infection. The new viral culturing platform, together with the associated mathematical tools, will allow entirely new measurements of evolutionary selection parameters, host-cell response, drug effectiveness, and the emergence of drug resistance in viral infections. Most broadly, the success of this work will enable a radically new paradigm in studying infectious disease, and enhance the development of effective and safe therapies.

Mission Relevance

This work is relevant to LLNL's strategic biosecurity mission to rapidly mitigate evolving and unknown biological threats, of which diagnostic platforms and understanding of host–pathogen interactions are key enabling capabilities. We anticipate establishing new capabilities in the realms of pathogen detection and characterization and host–pathogen interactions that will impact development of countermeasures to biological threats, which is also relevant to the core competency in bioscience and bioengineering. Federal agencies such as the Defense Advanced Research Agency and National Institutes of Health have likewise identified the development of new platforms for drug development as a key mission area.

FY14 Accomplishments and Results

In FY14 we (1) demonstrated the required cell–virus separation performance of the microfluidic device with 2 virus types and 3 cell types, at flow velocities of 0.1 to 0.5 mL/min (shown in figure); (2) built, configured, and programmed the automation of a recirculated culture system, with several successful 3-day experiments with microspheres; (3) developed a model of long-term particle dynamics, a critical component of future experiments; (4) performed small-scale experiments using hybrid cells produced by the fusion of an antibody-producing lymphocyte with a cancer cell; and (5) integrated the additional hardware required for long-term biological cell culture in our recirculated system.



A rendering of cells (yellow) and viruses (green) being separated by an acoustic field (blue lines). Fluid flow is toward the viewer, with a stream of mixed particles entering the device side-by-side with a clean fluid stream. The acoustic field moves the cells toward the pressure node (red line) and into the clean fluid, while the undeflected viruses continue straight, exiting the device through a separate outlet (not shown).

Proposed Work for FY15

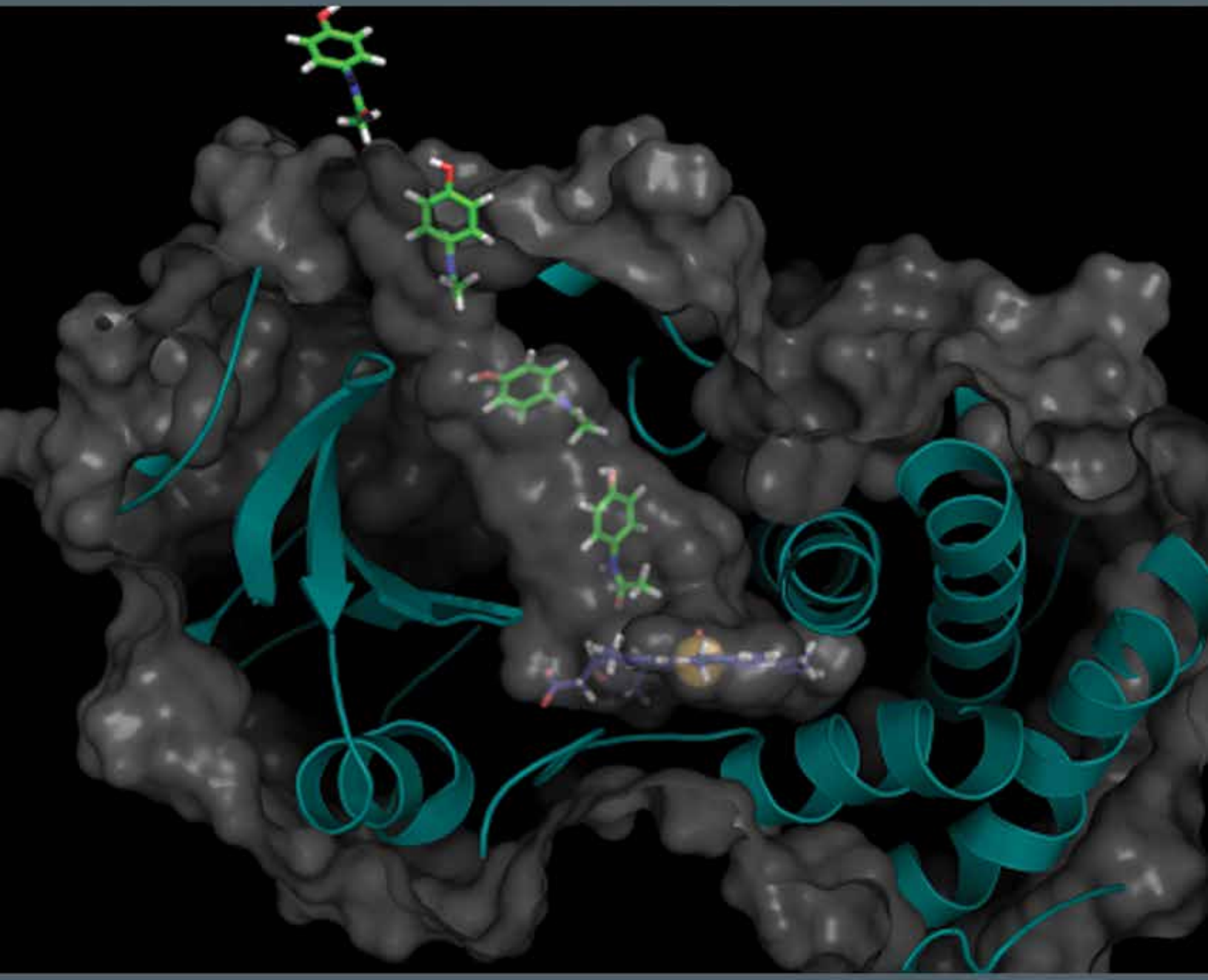
In FY15 we propose to (1) produce long-term cultures of the mosquito-transmitted dengue virus; (2) expand interactions with our University of California, San Francisco collaborators, using the model parameters they derive from viral infection kinetics to inform operation of the recirculating culture system; (3) begin collaborative running of long-term cultures of cells infected with HIV (human immunodeficiency virus) incorporating LLNL's acoustic microfluidic device; and (4) initiate work with our Mount Sinai School of Medicine collaborator, running separations of influenza A virus from its host cells, leading to steady-state influenza virus culturing at Mount Sinai.

Publications and Presentations

Fong, E. J., et al., 2014. "Acoustic focusing with engineered node locations for high-performance microfluidic particle separation." *Analyst* **139**(5), 1192. LLNL-JRNL-640796. *http://dx.doi.org/10.1039/c4an00034j*

Fong, E. J., et al., 2014. *Continuously variable node position in a high-throughput acoustofluidic device*. 18th Intl. Conf. Miniaturized Systems for Chemistry and Life Sciences (MicroTAS 2014), San Antonio, TX, Oct. 26–30, 2014. LLNL-POST-662327.

Biosecurity



FY2014

Lawrence Livermore National Laboratory

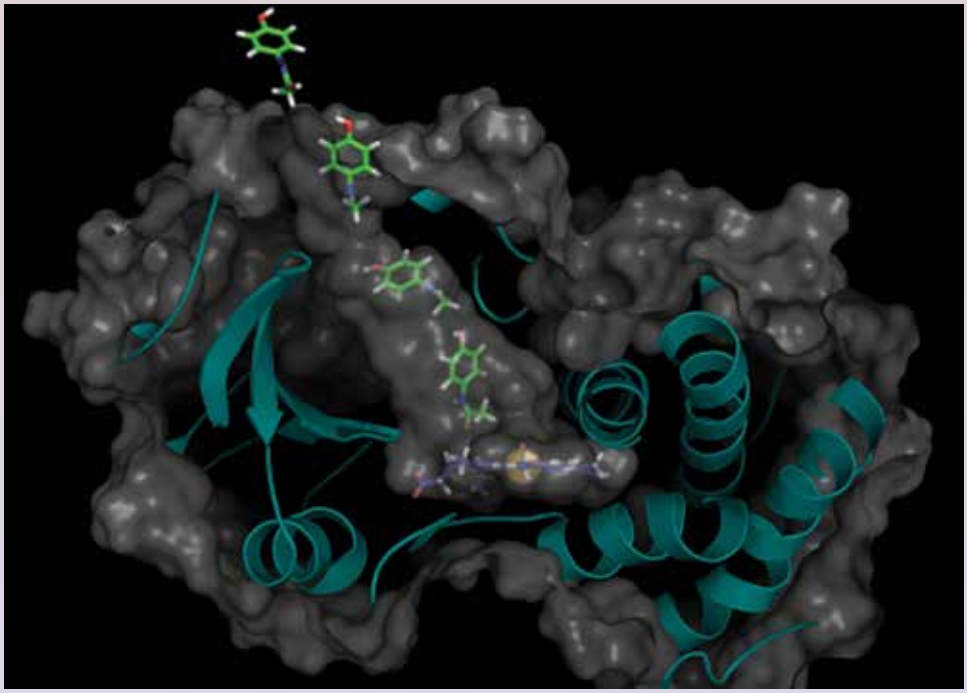
Computational Advancements in Countermeasures for Emerging Bio-Threats

Felice Lightstone (12-SI-004)

Abstract

To meet the national need to develop medical countermeasures against emerging bio-threats, we must accelerate the drug development process. With this project, we will develop capabilities to predict pharmacokinetic properties and adverse side effects in the initial optimization stage to enable successful clinical outcomes for drug candidates. Our system will combine systems biology, physiologically based pharmacokinetics modeling, biophysics, computational chemistry, and informatics to create a predictive pharmacokinetic capability based on a drug candidate's chemical structure. The project utilizes the Laboratory's text-mining capability and world-class expertise in high-performance computing.

The successful outcome of this project will result in a new state-of-the-art capability at Lawrence Livermore that will drastically accelerate medical countermeasure development and position Livermore as a world-class facility in the area of computational pharmacology. This new capability will be the first of its kind to predict the pharmacokinetics and adverse side effects of a drug candidate from its chemical structure, using highly accurate physics-based methods coupled with informatics. Once accurate predictions are made, the drug development process can be shortened, and more drug candidates will succeed in clinical trials. Accurately predicting



Predicting the binding of the common drug acetaminophen into the active site of cytochrome P450, a common enzyme that metabolizes drugs.

the pharmacokinetics of a drug candidate will dramatically reduce the time to approval by the Federal Drug Administration and will have a profound impact on therapeutics for human health.

Mission Relevance

One of the missions of the Laboratory is to rapidly mitigate evolving and unknown bio-threats. Success in this effort will provide an advanced technology and expertise to better predict the human outcome of small-molecule therapeutics so that rational drug design approaches will become more successful and prohibitive risks will be mitigated.

FY14 Accomplishments and Results

In FY15 we (1) continued development of each of the modules for the prediction of pharmacokinetic properties of drugs and expanded the pharmacokinetic modeling beyond the common drug acetaminophen; (2) examined interactions between the antibiotic ciproflaxin and caffeine; (3) linked adverse side effects to virtual drug interactions with a panel of human proteins—input into the statistical model was the result of high-performance computing calculations, which require no experimental input; (4) established and automated our ligand binding pipeline and published the associated results; and (5) established a protocol to compute kinetic binding rate constants from molecular dynamics simulations, which paves the way to predict pharmacokinetics of novel drugs.

Project Summary

The successful conclusion of this project resulted in establishing a credible capability for an all-computational prediction of drug side effects, drug interactions with off-targets, linking molecular interactions to pharmacokinetics, predicting penetration of the blood–brain barrier, and establishing in-house expertise in this field. The software we created is now an in-house resource that Laboratory researchers are routinely using, and is also available to external investigators via a download page. We determined our approach can outperform, for particular adverse drug reaction classes, the best competing model that uses freely available experimental data. We have established the foundation for rapid and cheaper development of medical countermeasures using high-performance computing, and have been invited to submit a grant to the National Institutes of General Medicine Sciences to continue using high-performance computing to predict adverse drug reactions of drugs and drug candidates.

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Detection of Novel Infectious Agents from Clinical Samples Through Immunoglobulin M and Toll-Like Receptor Capture

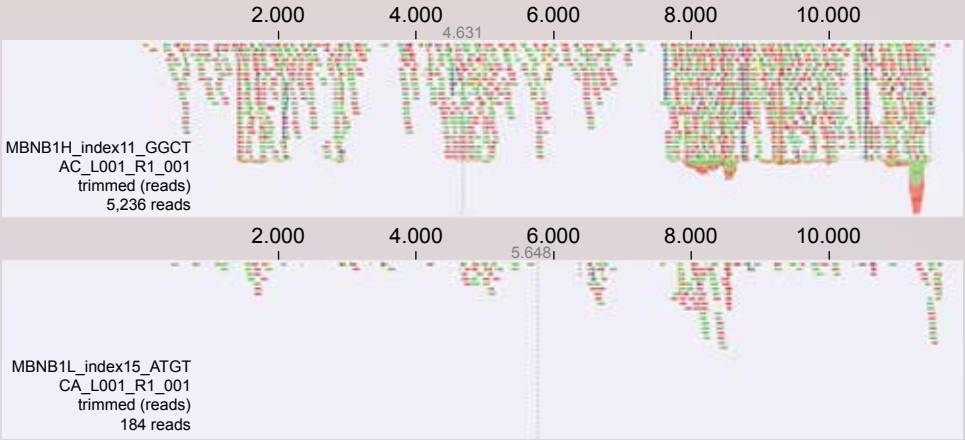
Monica Borucki (13-ERD-020)

Abstract

Novel pathogens may circulate in a population for years prior to detection, severely hampering prompt and appropriate treatment and containment. Recent advances in characterizing microbes include microarrays, meta-genomics, and next-generation sequencing. These techniques rely on detection of the microbe genome, which exists in much smaller amounts than the host genome. This project aims to develop rapid techniques that separate pathogen nucleic acid from host genetic material to enable detection of novel pathogens from complex samples. Methods will be developed to

rapidly isolate pathogen genetic material captured by the host's Toll-like receptors, which are a class of proteins that play a key role in the innate immune system, and immunoglobulin M, which is a basic antibody. The captured genomes will be characterized using meta-genomics. In addition to pre-symptomatically detecting pathogens before an outbreak, this technique could be used to screen archived blood samples from all over the world to identify the genotypes circulating before the outbreak and pinpoint the genetic changes that led to the outbreak. These data could also be used to generate a database of early-stage infections for determining which viruses routinely infect humans and the geographical location of each virus, as well as providing insight into interactions between a host and pathogen.

We expect to produce assays in which the extraction and purification of viruses or viral ribonucleic acid bound to immunoglobulin M and Toll-like receptors, respectively, from pathogen-infected mice allows microbial nucleic acid to be concentrated away from host nucleic acid. Once the pathogen genome is isolated in this way, it will permit efficient and in-depth meta-genomic characterization of the pathogen genome. We will focus on detecting ribonucleic acid viruses, because these are the most challenging group of pathogens to detect. Once tested and optimized on control samples, the methods we develop in this project can then be tested in human biological-surveillance efforts such as testing serum and nasal samples from military personnel returning from overseas deployment for the presence of exotic microbes.



Meta-genomic data obtained via immunoprecipitation of the TLR-3 pathogen recognition receptor and mapped to the Sindbis virus genome. The top row shows sequencing reads recovered from ribonucleic acid bound to the TLR-3 protein and mapped to the viral genome. Reads span virtually all of the genome with most regions covered by multiple reads. The bottom row shows sequencing reads recovered from unbound ribonucleic acid and mapped to the viral genome. Much of the genome is not covered by reads, which yields incomplete genetic information on the viral genome. Only 3.5% of the reads were recovered as compared to the bound-fraction yield.

Mission Relevance

In support of the Laboratory's strategic focus area of biosecurity, this project will enable the potentially pre-symptomatic detection of emerging and bio-engineered viruses, thereby providing precious time for the rapid development of therapeutics against these threats. This method could also be used to screen archived samples to create a genomic database for identifying the genetic changes that lead to outbreaks and for other insights into host–pathogen interactions, supportive of the core competency in bioscience and bioengineering.

FY14 Accomplishments and Results

In FY14 we (1) developed immunoprecipitation assays for the pathogen-recognition receptors TLR-3, MDA5, and RIG-I and tested these assays using BW5147 cells (derived from mouse lymphoma) that were infected with Sindbis virus (a mosquito-borne virus that causes joint pain and rash in humans); (2) determined, from the meta-genomic data, that the assay removed over 90% of host nucleic acid; (3) obtained, using multiple receptors in the assay, sequence data for all regions of the viral genome—we modified the protocol to significantly increase recovery of the viral sequence; (4) obtained and tested clinical samples from subjects infected with a virus to further test the assay; (5) tested assays for isolating viruses bound with immunoglobulin M in mice (the first antibody to appear in response to initial exposure to an antigen); (6) determined, from the resulting data, that excess circulating immunoglobulin M saturated all available binding sites, minimizing recovery of viruses bound with the immunoglobulin; and (7) tested samples from a modified immunoglobulin-M protocol via meta-genomic analysis.

Proposed Work for FY15

In FY15 we will (1) optimize each of the assays tested in FY14 to increase speed, sensitivity, and specificity, using virus-infected human and primate samples; (2) test assays using clinical samples, viruses from multiple families, and different routes of infection; (3) develop antibodies that bind only to immunoglobulin-M antibodies that are bound to antigens (immunoglobulin M shifts shape when bound to antigens), increasing the sensitivity of the immunoglobulin M assay; and (4) test antibodies to immunoglobulin M and immunoglobulin A (an antibody that plays a critical role in mucosal immunity) for retrieval of a virus from nasal secretions.

Cyclodextrin-Based Nanometer-Scale Scaffolds for Capture and Catalytic Degradation of Chemical Warfare Agents

Carlos Valdez (14-ERD-048)

Abstract

New technologies for capturing and catalytically degrading chemical weapon agents would fill a critical national security need. We propose the use of molecular-complex

scaffolds known as cyclodextrins that possess chemical and physical characteristics suitable for detection and analysis, decontamination, and medical countermeasures, where the demand for broad-spectrum solutions is urgent. Our research project focuses on efforts for two classes of chemical warfare agents: the organophosphorus-based nerve agents and a series of incapacitating agents known as fentanyls. Both are of high priority because of their ease of production and availability, as well as their toxicity at low doses. We intend to develop and validate an integrated experimental and computational approach whose goal is the development of cyclodextrin-based nanometer-scale scaffolds for the capture and catalytic destruction of organophosphorus nerve agents and fentanyls. Although successful technologies have been developed to mitigate individual issues for known chemical threats, establishing broadly applicable and rapidly responsive methodologies targeted at known and, more importantly, emerging threats remains critical. Although this proposed research addresses the development of designer cyclodextrin molecules for two classes of agents, the foundation of our work is an integrated experimental and computational strategy for the discovery of efficacious cyclodextrins for broad-spectrum application.

We expect to develop an understanding of mechanisms for the action of fentanyl on protein receptors, which is key to the development of cyclodextrin for sequestration of these toxic chemicals. Elucidating conditions under which zinc-based organometallic catalysts most efficiently degrade organophosphorus agents will provide insight into the development of effective metallocyclodextrins for decontamination. While the cyclodextrins themselves are important for mitigating organophosphorus- and fentanyl-specific threats, it is the development, demonstration, and validation of an integrated design approach capable of optimizing arbitrary host–guest complexes that will be the central accomplishment of our proposed work.

Mission Relevance

Our strategy focuses on the development of physical and medical countermeasures against fentanyls and organophosphorus nerve agents, which supports the Laboratory's strategic thrust in biosecurity for rapid mitigation of evolving and unknown threats. In addition, this project serves to strengthen computational and analytical methods critical to existing LLNL missions. By evolving an integrated computational and experimental capability for the rapid discovery of highly effective, broadly applicable scaffolds, we are providing a foundation for future progress in environmental remediation technologies, exposure detection capabilities, and the production of advanced materials for use against known and emerging threats.

FY14 Accomplishments and Results

In FY14 we (1) synthesized the fentanyl library and designer cyclodextrins, (2) determined baseline binding between fentanyls and native and modified cyclodextrins, (3) completed synthesis of the zinc catalyst library, (4) completed a catalytic degradation study of paragon (an organophosphorous pesticide) and began another for G-series non-persistent nerve agents, (5) developed an analytical technique to monitor organophosphate degradation in situ, (6) performed molecular

dynamics simulations to investigate conformation and dynamics of complexes between fentanyl and a variety of cyclodextrins, and (7) performed nuclear magnetic resonance studies on a paragon and cyclodextrin inclusion complex formation. In addition, we submitted two records of invention.

Proposed Work for FY15

In FY15 we will (1) develop designer cyclodextrins for fentanyl capture based on current data, while being strongly informed by further computational work; (2) continue molecular dynamics studies into ground-state conformations of zinc catalysts for organophosphate degradation; (3) begin molecular dynamics simulations on current zinc catalysts and their ability to degrade G-series agents and V-series persistent nerve agents, and perform tandem nuclear magnetic resonance experiments; (4) synthesize and characterize catalytically active metallo-cyclodextrins for enhanced organophosphate degradation; and (5) develop an experimentally validated computational approach for facile molecular dynamics identification of more effective cyclodextrin candidates for both organophosphate degradation and fentanyl sequestration.

Publications and Presentations

Mayer, B. P., et al., 2014. *Nuclear magnetic resonance and computational study of inclusion complexes between cyclodextrins and fentanyls.* LLNL-POST-657940.

Rapid Detection and Characterization of Emerging Foreign Animal Disease Pathogens

Crystal Jaing (14-ERD-081)

Abstract

The identification and control of foreign animal diseases such as foot and mouth disease and swine fever is critical to the protection of U.S. agriculture and food systems. Past outbreaks have occurred in Europe, Central and South America, Asia, and Africa. A swine fever outbreak in Netherlands in 1997 cost about $2.3 billion to eradicate. Swine fever is also a disease of concern for biological warfare. Early detection of disease outbreaks is crucial to the effective surveillance of emerging infectious diseases to safeguard animal and human health. Our goal is to develop new and effective surveillance technologies for early detection of known, emerging, and new foreign animal disease outbreaks; to enable biomarker discovery; and to transition the technologies to national agricultural laboratories. In collaboration with Kansas State University, we will not only enhance the scientific foundation for advanced research in agricultural and animal disease pathogens, but also increase our engagement with global agricultural pathogen surveillance efforts.

We expect to develop at least two novel foreign animal disease and agricultural pathogen surveillance panels—a multiplexed Luminex bead assay to detect swine disease and near-neighbor species, as well as a broad-spectrum livestock pathogen

detection microarray. We will collaborate with researchers at Kansas State University in agricultural pathogen surveillance and detection, and position the Laboratory to be more broadly engaged in developing techniques for detecting global animal disease outbreaks. We expect to transition both assays to Kansas State University as well as to international collaborators. Additionally, we will identify genetic markers that could distinguish virulence capability for African and classical swine fever viral isolates. We also will identify potential diagnostic biomarkers for classical swine fever infection in pigs.

Mission Relevance

A comprehensive suite of novel technology platforms to rapidly detect and characterize foreign animal disease pathogens will help protect the agricultural economy, food supply, public health, and biosecurity of the nation. This effort aligns with the Laboratory's strategic biosecurity strategic focus area to safeguard the nation against emerging biological threats, and enhances our core competency in bioscience and bioengineering.

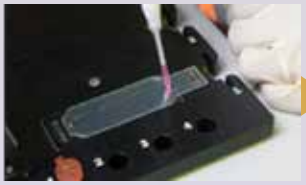
FY14 Accomplishments and Results

In FY14 we (1) made significant progress towards building a long-term strategic partnership with the National Bio and Agro-Defense Facility at Kansas State as well as other federal stakeholders—we established a national advisory group with representatives from the Department of Homeland Security, U.S. Department of Agriculture, and the pork industry to provide quarterly advice; (2) developed the swine respiratory disease multiplex panel with embedded foreign animal disease pathogens (African and classical swine flu); (3) completed the bioinformatics analysis, received swine flu samples, and developed assays for transition to Kansas State; and (4) tested swine blood, oral fluids, and tissue samples from diseased animals and tested them on the LLNL microarray for sensitivity and specificity.

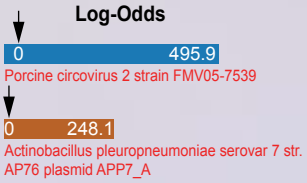
Proposed Work for FY15

In FY15 we will (1) complete the transition of swine multiplex assays and the microarray to Kansas State; (2) perform gene expression analysis of pigs infected with African swine flu virus and identify diagnostic host biomarkers correlated with the disease—there are currently no vaccines to counter it and our goal is to use infection models to understand the acute disease, chronic infection, and persistence of African swine flu; (3) infect pigs with African swine flu virus, collect blood and tissues, and perform phenotypic analysis; (4) perform transcriptomic analysis of infected pigs with the RNA-seq approach to transcriptome profiling that uses shotgun-sequencing

Pig oral fluid samples, extract DNA and RNA



Hybridize samples to Microbial Detection Array



Automated data analysis to identify pathogens and co-infections

The Lawrence Livermore Microbial Detection Array has been evaluated to detect viral and bacterial pathogens from swine oral fluid samples.

technologies, and identify gene markers that correlate with African swine flu virus infection phenotypes; and (5) confirm RNA-seq findings using a variation on the DNA microarray from NanoString Technologies in Seattle, Washington, as well as quantitative polymerase chain reaction analysis, and identify the most informative markers that will predict outcomes of African swine flu infection.

Publications and Presentations

Jaing, C., et al., 2014. *Microarray detection of pig pathogens.* LLNL-JRNL-659677.

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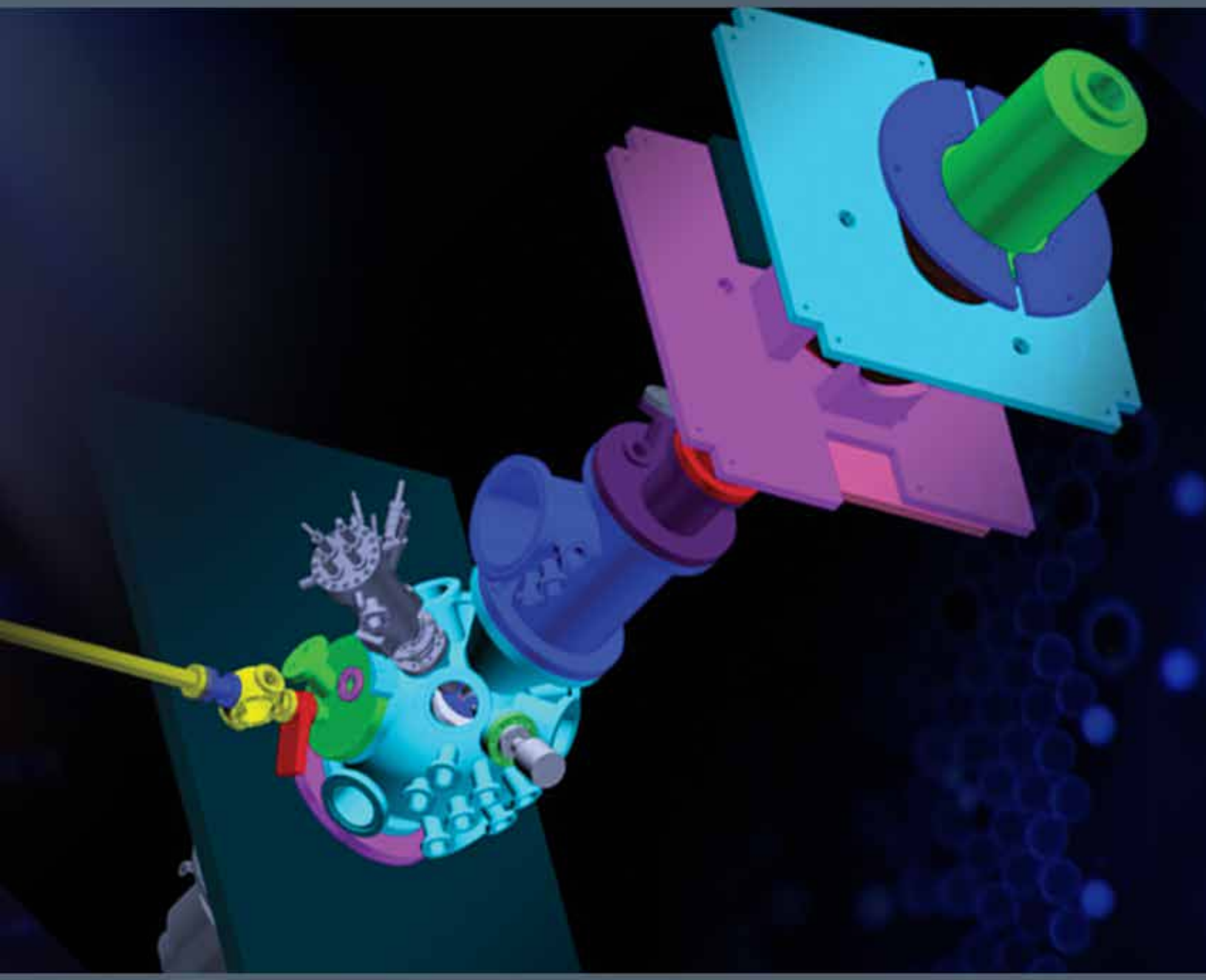
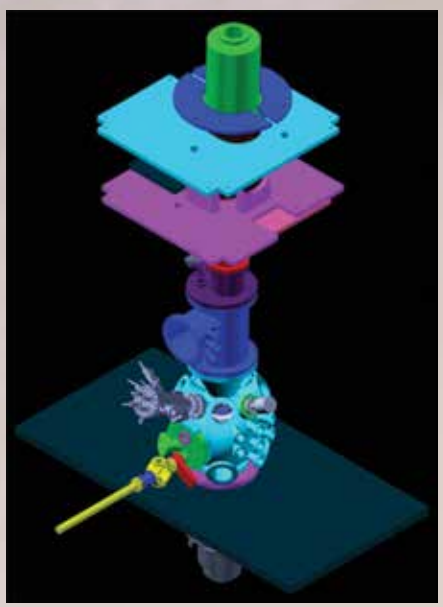
Chemical and Isotopic Signatures



FY2014

Lawrence Livermore National Laboratory



The new LION (laser ionization of neutrals) instrument, currently under construction at LLNL to accelerate the pace of development for applying resonance ionization mass spectrometry to characterization of nuclear materials. The LION is a 2-m time-of-flight mass spectrometer, specifically designed to measure ions created by laser ionization of secondary neutral atoms.

Improving Resonance Ionization Mass Spectrometry for Next-Generation Nuclear Forensics

Brett Isselhardt (14-ERD-082)

Abstract

Our aim is to address research issues related to the isotopic analysis of low-abundance materials, such as early solar system materials or nuclear fallout. Today these issues limit our ability to answer fundamental chemistry questions about the genesis of our solar system or to rapidly quantify actinide isotope ratios in fallout. We will use resonance ionization mass spectrometry to rapidly and accurately quantify, in situ, isotope ratios for materials including plutonium, uranium, magnesium, beryllium, and lithium.
The technique is a high-sensitivity, elementally selective, laser-based form of mass spectrometry that offers the potential to determine isotopic composition of materials without sample preparation and isobaric interference. Resonance ionization uses pulsed laser light that is resonant with a electronic excited state to excite and eventually remove an electron from the atom or molecule to create positively charged ions, which are then collected and accelerated into a mass analyzer. Our work will enable a method for answering long-standing questions in cosmochemistry that deal with the chemical composition of changes in the universe. This work will also lay the foundation for development of a resonance ionization mass spectrometry capability for characterizing debris from a radiological or nuclear event.

Our goal is to address a set of outstanding technical and scientific problems that have hindered the application of resonance ionization mass spectrometry to the analysis of materials ranging from post-detonation debris to meteoritic inclusions. The potential to quickly quantify relative isotope abundances of actinides and other diagnostic elements in post-detonation debris will enhance our ability to draw conclusions regarding device design and performance from debris analysis. Resonance ionization mass spectrometry can provide definitive isotope-specific information on fuel composition in post-detonation scenarios within hours of sample receipt. Successful completion of this research has the potential to address prominent needs identified by both the U.S. nuclear forensics and cosmochemical communities.

Mission Relevance

Developing a state-of-the-art resonance ionization mass spectrometry capability at the Laboratory will ensure continued leadership in the core competency of chemical and isotopic signatures, in support of national security, specifically nuclear threat reduction via nuclear detection and forensics. It will also meet the goal of developing technological capabilities that advance scientific frontiers and address next-generation basic scientific issues related to the evolution of our solar system.

FY14 Accomplishments and Results

In FY14 we (1) performed spectroscopic measurements of the hyperfine structure of plutonium to improve our understanding of its resonance ionization probability

and included that data in the predictive model of plutonium ionization currently under development, (2) studied the atomization of plutonium from tantalum and titanium substrates to understand the neutral atomic yield of sputtering and laser ablation from these surfaces, and (3) co-located and installed a multiple-wavelength, femtosecond-laser adjacent to the nanosecond resonance ionization lasers to be used for studying femtosecond-laser desorption.

Proposed Work for FY15

In FY15 we will (1) complete the plutonium ionization modeling effort; (2) extend our modeling approach to more isotope systems of interest such as americium, cesium, and barium; (3) continue to explore the in situ reduction of oxidized samples by titanium overcoating to increase the neutral atomic yield; and (4) study femtosecond-laser desorption of uranium or plutonium from oxide and silicate matrices.

Publications and Presentations

Eppich, G. R., et al., 2014. "Constraints on fallout melt glass formation from a near-surface nuclear test." *J. Radioanal. Nucl. Chem.* **302**(1), 593. LLNL-JRNL-650394.

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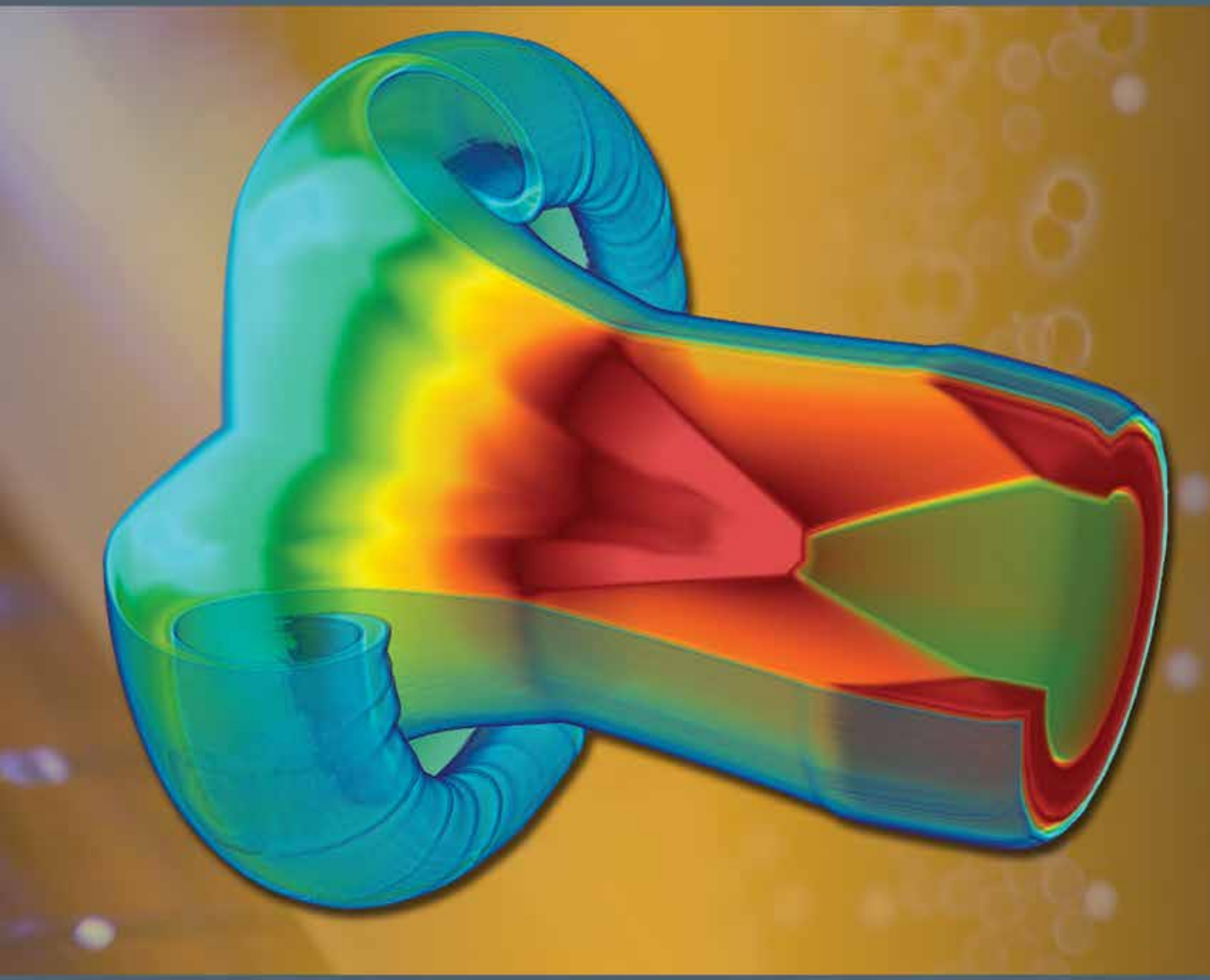
Computational Science and Engineering



FY2014

Lawrence Livermore National Laboratory

Computational Gyro-Landau Fluid Model for Tokamak Edge Plasmas

Xueqao Xu (12-ERD-022)

Abstract

The edge plasma is one of the most important regions in magnetized fusion reactors for having predictive models, yet the edge plasma is also one of the most challenging regions to simulate because of its complex physics and geometry. Our objective with this project is to develop a predictive capability for tokamak edge-plasma transport through a gyro-Landau fluid extension of the BOUT++ code, which is a framework for parallel plasma fluid simulations. This fills a critical gap between the fluid models currently in use (which are intrinsically limited) and full gyro-kinetic models (which have practical computing limitations). We will develop advanced physics models and novel numerical techniques in a massively parallel computational environment, and will validate the models against data from the two largest superconducting tokamaks: the Experimental Advanced Superconducting Tokamak (EAST) in China and the Korea Superconducting Tokamak Advanced Research (KSTAR) in Korea, as well as from General Atomics' DIII-D in San Diego. We will collaborate with the Institute of Plasma Physics at the Chinese Academy of Sciences, Peking University, and the Korean National Fusion Research Institute.

If successful, the edge-plasma model created and validated in this project will generate a theoretical and simulation capability far beyond the present state of the art. An accurate gyro-Landau fluid simulation model may offer orders-of-magnitude savings in computational resources compared to a gyro-kinetic simulation, and makes a gyro-Landau fluid code very attractive as a component in an integrated, whole-device simulation. Moreover, a gyro-Landau fluid code could potentially be extended to treat core physics and enable a global model of nonlinear plasma dynamics for the entire tokamak. This work will contribute to the validation and application of gyro-Landau fluid models that will be needed to design experiments at the ITER international experimental fusion reactor in the south of France, as well as future reactors.

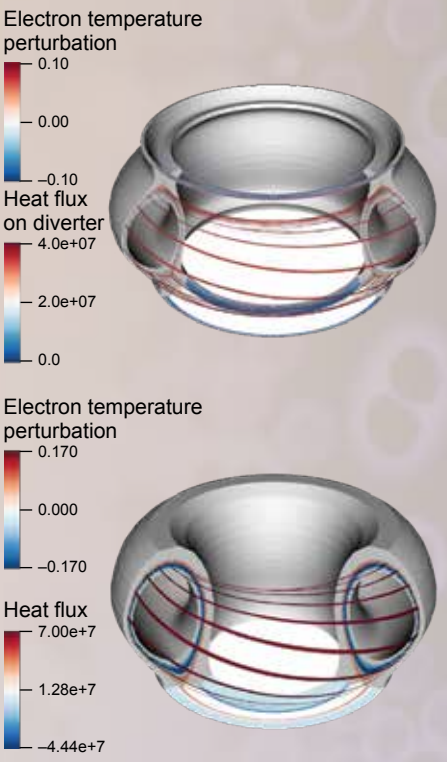
Mission Relevance

This research will fill critical gaps in theoretical understanding and simulation capability of interest to the international fusion research community and other scientific fields such as astrophysical and space plasmas. The project supports the Laboratory's energy and national security missions in the core competencies of high-energy-density science and computational science and engineering, and it offers an excellent opportunity to forge alliances with emerging Asian fusion programs.

FY14 Accomplishments and Results

In FY14 we (1) analyzed the accuracy of nonlinear wave–particle and wave–wave coupling; (2) investigated nonlinear kinetic effects at large perturbation; (3) identified computational bottlenecks and numerical strategies that efficiently project to a

massively parallel computational environment; (4) implemented a 3 + 1 electromagnetic model and began implementation of second-order closures; (5) investigated advanced numerical techniques for gyro-Landau fluid closures such as a generalized three-dimensional Poisson solver; (6) began extension of the 3 + 0 electromagnetic gyro-Landau fluid model from core to edge; (7) implemented a radio-frequency sheath physics model; (8) validated turbulence, edge-localized modes, and the impact of radio-frequency on plasma turbulence with data from EAST in China, KSTAR in Korea, and DIII-D in San Diego; and (9) benchmarked collisional closures for parallel electron transport against a multiple-mode kinetic code and the Fokker–Planck code, in which approximations are used to derive manageable particle collision terms.



The BOUT++ simulations reveal dynamics of filamentary eruptions of hot plasma from the edge in a tokamak fusion experiment. Accurate predictions of thermal conduction need to incorporate kinetic physics in the model. The only code offering this capability is BOUT++. A double-null divertor configuration provides a larger wall interaction area and more shaping flexibilities (bottom) than the single-null diverter configuration at the top.

Project Summary

The successful conclusion of this project resulted in significant improvement of predictive capability for tokamak edge-plasma transport through a gyro-Landau fluid extension of the BOUT++ code. We developed an efficient and versatile non-Fourier method for the computation of Landau-fluid closure operators, based on an approximation by a sum of modified-Helmholtz equation solves in configuration applications. We uncovered a new nonlinear criterion for fast-edge relaxation (crashes) events in high-confinement tokamak plasmas. In addition, our nonlinear edge-localized mode simulations, including both standard and moment models of gyro-Landau fluid, were compared with fast measurements from the DIII-D tokamak, high-mode plasmas (a strong and sudden change in plasma characteristics) for EAST, and electron cyclotron emission imaging for KSTAR. Of special note is that our nonlinear simulations were the first to obtain a collision scaling of edge-localized mode energy losses consistent with the International Tokamak Physics Activity database. In summary, our project has enabled a capability to fill a critical gap between the fluid models currently in use and full gyro-kinetic models under development. The DOE office of Fusion Energy Sciences will provide support to continue investigation of physics predictions of these new gyro-Landau fluid models and to continue implementation of the advanced models that were developed. We have established an international collaboration with Chinese partners, and plan to fully develop this technology for use in integrated edge-plasma simulations and validation.

Publications and Presentations

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Dimits, A. M., et al., 2013. *Efficient non-Fourier implementation of Landau-fluid operators in the BOUT++ code*. 54th Ann. Mtg. APS Division of Plasma Physics, Providence, RI, Oct. 29–Nov. 2, 2012. LLNL-POST-595935.

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Guo, H. Y., et al., 2014. "Recent advances in long-pulse high-confinement plasma operations in Experimental Advanced Superconducting Tokamak." *Phys. Plasma.* **21**, 056107. LLNL-JRNL-663433. *http://dx.doi.org/10.1063/1.4872195*

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High-Order Curvilinear Arbitrary Lagrangian–Eulerian Hydrodynamics

Tzanio Kolev (12-ERD-030)

Abstract

The framework of arbitrary Lagrangian–Eulerian (ALE) large-deformation shock physics codes forms the core of large-scale hydrodynamics codes used at LLNL for stockpile stewardship and other mission-relevant work. Current ALE schemes are an improvement over pure Lagrangian methods that summarize the dynamics of a system, but they also introduce numerical problems such as lack of energy conservation and artificial material breakup. Recent advances in high-order curvilinear finite elements that are used in solving differential equations, where simple-element equations over many small curved geometric sub-domains are connected to approximate a more complex equation over a larger domain, have shown significant benefits for the Lagrange phase of ALE. We propose to apply these curvilinear finite elements to the ALE advection phase. We will develop new and more robust high-order ALE algorithms, while preserving the accuracy of the high-order Lagrange step. To this end, we will research and develop new methods for optimizing curvilinear mesh geometry representations, conservative monotonic high-order field remapping, and handling multiple-material curvilinear zones.

This project will produce the first high-order curvilinear method for ALE hydrodynamics, enabling higher-quality simulations of multiple-material ALE hydrodynamics. These algorithms can potentially eliminate the need for adjusting mesh-motion parameters and for manual intervention by users, minimize diffusive

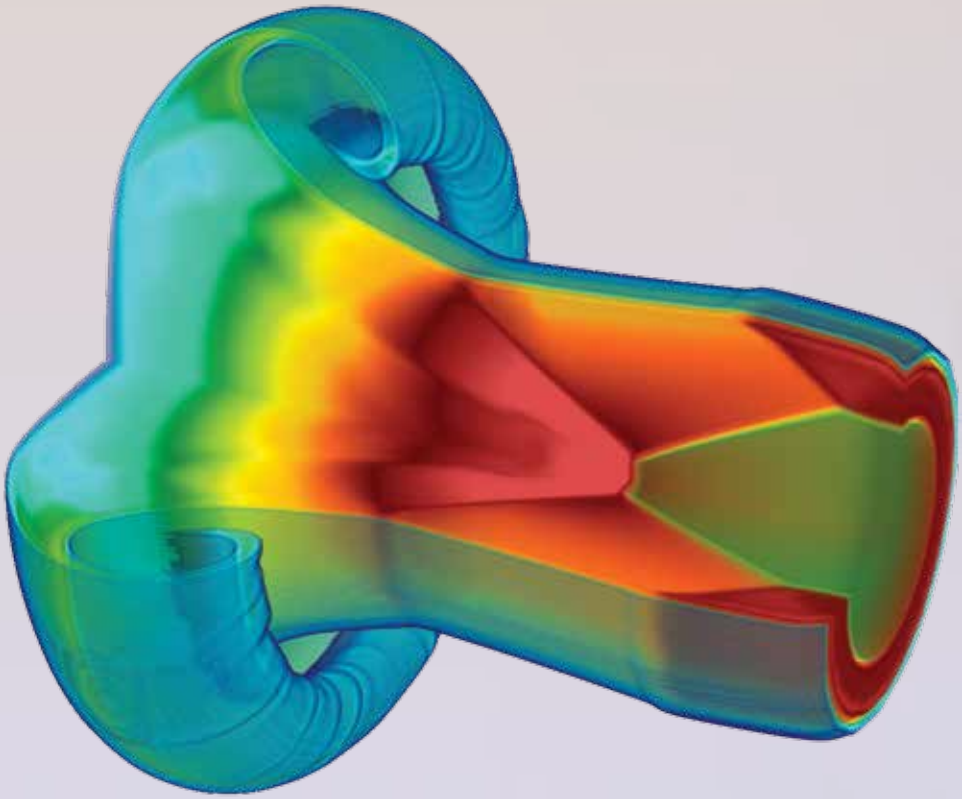
errors by running longer in a Lagrange mode, improve accuracy by diminishing mesh imprinting and improving symmetry preservation, and more effectively utilize future multiple-core and graphics processing unit architectures because of the algorithms' local intensity in floating-point operations.

Mission Relevance

Success with this proposed research will improve the predictive capability of hydrodynamics simulations while requiring fewer user-adjustable parameters. These simulations are of importance for stockpile stewardship and inertial-confinement fusion in support of the Laboratory's missions in national and energy security and relevant to the core competency in computational science and engineering.

FY14 Accomplishments and Results

In FY14 we (1) completed, implemented (in the BLAST high-order finite-element hydrodynamics software), and demonstrated the scalability of the high-order, single-material curvilinear ALE algorithm; (2) developed and implemented a high-order multiple-material Lagrangian algorithm and high-order generalizations of closure models; (3) completed a multiple-material ALE algorithm using high-order material indicator functions and applied it to two- and three-dimensional ALE benchmarks; and (4) investigated code transformations and new high-order finite-element algorithms to address performance gaps, leading up to a twelvefold speedup in the Lagrangian phase.



Our new high-order finite-element algorithms for arbitrary Lagrangian–Eulerian hydrodynamics have very good parallel scaling and symmetry-preservation properties on highly unstructured three-dimensional meshes. Shown is a volume plot from a single-material run on 16,000 cores of Livermore's Vulcan supercomputer using third-order spatial and time discretizations.

Project Summary

The successful conclusion of this project resulted in the first high-order multiple-material method for ALE hydrodynamics on high-order curvilinear grids, paving the way to higher-quality simulations with improved predictive capability and better performance on modern computer architectures. These simulations are of importance for stockpile stewardship and inertial-confinement fusion in support of the Laboratory's missions in national and energy security. We specifically developed novel methods for robust curvilinear mesh optimization, high-order conservative and monotonic discontinuous Galerkin (a numerical method for solving differential equations) advection-based field remapping, high-order representation and evolution of multiple materials through material indicator functions, high-order closure models, and synchronized multiple-material remapping. We also implemented the new algorithms in our research code BLAST, where we performed large-scale numerical simulations of two- and three-dimensional ALE benchmarks to demonstrate their parallel scalability. In summary, our project demonstrated that the benefits of the high-order finite-element approach that we observed in the purely Lagrangian case can also be extended to full multiple-material ALE hydrodynamic settings, making a strong case that these algorithms can form the foundation of next-generation simulation capabilities at LLNL. The hydrodynamics algorithms we developed are being extended to multiple-physics radiation-hydrodynamic problems, where we can also evaluate their feasibility as a next-generation simulation capability at LLNL.

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Automatic Complexity Reduction for Electromagnetic Effects Simulation

Daniel White (12-ERD-038)

Abstract

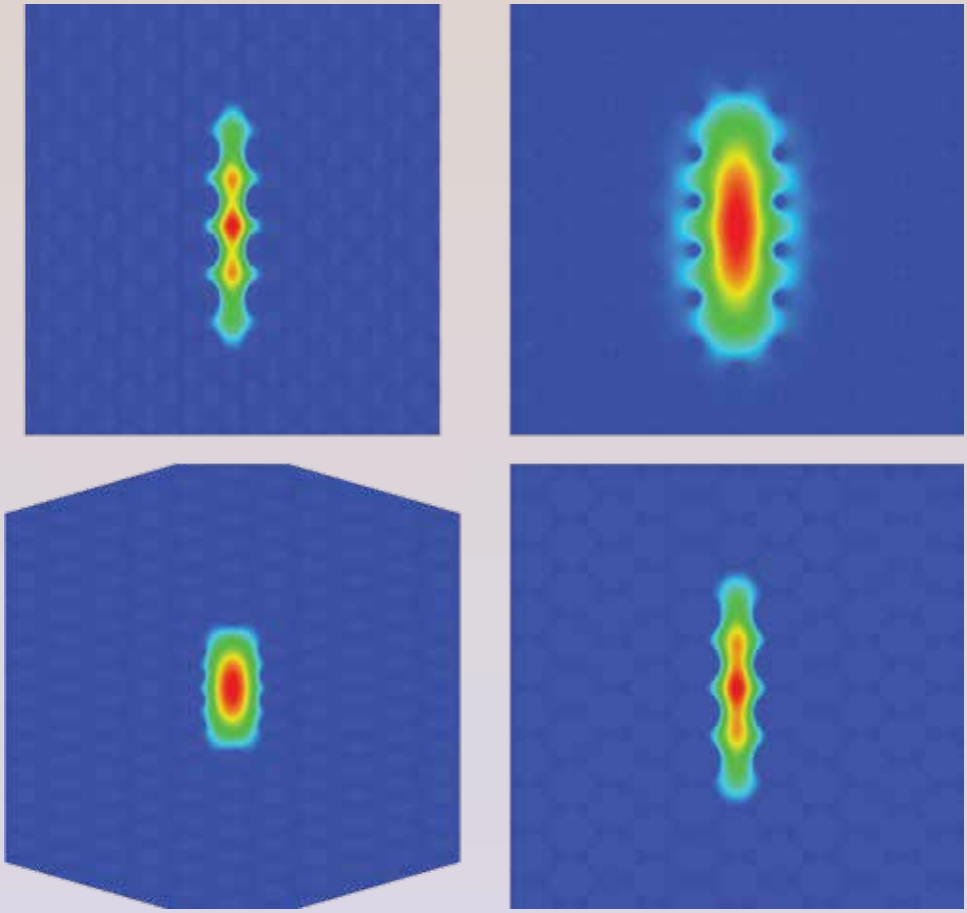
Our goal is to develop a new method for reducing the complexity of electromagnetic effects simulations on circuits. While documented experimental evidence shows that an electromagnetic wave of modest power can temporarily shut down an electronic circuit, not all circuits are affected and not under all conditions. Simulation is required to better understand the electromagnetic effect. Lawrence Livermore has sophisticated massively parallel finite-element and boundary-element codes for solving Maxwells equations, which describe the fundamentals of electricity and magnetism. However, trillions of simulations are required to understand how electromagnetic effects vary with circuit

layout, frequency, and location of the circuit. Using a combination of reduced-order models, radial basis functions for interpolating matrix triple products, and parameter adaptivity, we propose to develop an automatic complexity-reduction algorithm for simulating electromagnetic effects, test it on supercomputers, and validate the results using experimental electromagnetic effects data.

If successful, this new method for reducing the complexity of exploring parameters via simulation has many applications, including heat transfer, elasticity, and related partial differential equations. We expect to publish the results and license the software.

Mission Relevance

Electromagnetic effects can disrupt any device that contains an electronic circuit, from improvised explosive devices in war zones, to cell phones, information technology equipment, the electrical grid, and industrial and military control systems. By creating the capability to predict electromagnetic effects, this research will support Lawrence Livermore strategic missions, including national and international security and energy security, and supports the core competency in computational science and engineering.



Application of reduced-order modeling to electromagnetic waveguide design for improved understanding of the effects of electromagnetic waves on electronic circuits. The images show electric-field intensity for various combinations of design parameters. Our approach is over 800 times faster than a standard finite-element approach for this particular simulation.

FY14 Accomplishments and Results

For FY14 we (1) developed a prototype order-reduction algorithm for accelerating electromagnetics simulations, which consists of the two steps of applying a change of basis to the discretized problem (also known as a truncated proper orthogonal decomposition) and interpolation of reduced dimensional matrices; (2) researched and implemented two different methods for the second step—interpolation using radial basis functions and using the empirical interpolation method; (3) determined that the advantage of radial basis function is that it is a black-box method, and does not require any modification of the electromagnetic field solver—however, the error in the interpolation using radial basis functions proved difficult to estimate and control; and (4) determined that the empirical interpolation method was invasive and required modification of the electromagnetic field solver to compute selected matrix elements for arbitrary values of parameters. The advantage of the method is that it is a numerically stable method, and we were able to achieve arbitrary interpolation accuracy as the number of samples was increased.

Project Summary

The primary product of our research effort is a suite of software tools for exploring the applicability of a reduced-order model to electromagnetic simulations. It is perhaps easiest to explain its benefit by an example. Consider designing a photonic crystal-fiber waveguide, and there are three parameters to be varied: the frequency, the hole radius, and the ellipticity. A brute force finite-element simulation using 100 different frequencies, 20 different radii, and 20 different elliptic ratios would require 7 years of computer time. Instead, we compute the full finite-element solution for only 260 specially selected combinations of parameters. These solutions are referred to as snapshots. From these snapshots we construct a simplified model: the reduced-order model. This model approximates the full finite-element solution to within 0.3% for any combination of parameters, but is over 800 times faster to evaluate than the full finite element. The key research issues were how to choose the snapshots, fast methods for constructing the reduced-order model, and estimating the error of the model. We will pursue follow-on work with the Department of Defense.

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Multiscale Capabilities for Exploring Transport Phenomena in Batteries

Brandon Wood (12-ERD-053)

Abstract

We propose to build state-of-the-art multiscale capabilities for modeling transport phenomena in batteries. Such capabilities will overcome limitations of the traditional macroscopic approach to enable accurate predictions of the performance of novel nanometer-scale structured battery electrodes. Once available, they will provide much-needed support and guidance to the optimization of next-generation battery architectures at different length scales. We will develop mesoscale and atomistic modeling capabilities for simulating different transport processes in batteries and integrate them to perform comprehensive transport simulations for battery operation, which will be supported by in situ characterization experiments for critical validation.

If successful, we expect to produce comprehensive multiscale capabilities for modeling charge-transport processes in batteries at the frontier of high-performance computing relevant to energy storage technology. We will create a high-performance code for efficiently performing large-scale battery cell-level simulations, and a mesoscale model

that quantitatively explains and predicts the important coupling phenomena in nanoscale structured electrodes and their influence on battery performance. This research will produce an improved scientific understanding of the charge-transfer kinetics at the electrolyte and electrode interfaces and implications for electrode microstructure optimization.

Mission Relevance

The central goal of this project, to cultivate unique multiscale capabilities for investigating transport phenomena in batteries and energy storage systems in general, is closely aligned with Lawrence Livermore's core mission to meet national energy security challenges. The new capabilities will also help LLNL develop strategic partnerships with the energy industry through the LLNL Livermore Valley Open Campus Initiatives to address urgent problems in energy-related applications, and our methodology supports the core competency in computational science and engineering.

FY14 Accomplishments and Results

In FY14 we (1) implemented the phase-field models for electrolyte and electrode transport, including basic interface kinetics to mimic electrochemical rate activity at the electrode–electrolyte interface; (2) completed synthesis and extensive testing of carbon anode and hybrid materials based on carbon and metal oxides, and collected x-ray spectroscopy and electrochemical cycling data for each sample; (3) performed first-principles calculations of storage capacity and diffusion kinetics in carbon-based systems; (4) completed our multiscale modeling efforts on the electrochemical lithiation of silicon, and on lithium storage and transport in graphene anode derivatives; and (5) applied our modeling descriptor developed for battery electrodes to explore additional applications in the electrocatalysis community.

Project Summary

We have completed implementation of a multiscale modeling framework for simulating transport processes in battery electrode particles, based on the phase-field approach and informed by ab initio calculations and experimental observations. We discovered several previously unreported phenomena in battery cathode materials, including key explanations for experimentally observed microstructural features. We developed a new statistical approach to examining entropic effects at phase boundaries. We also introduced ab initio models for understanding the fundamental origins of lithium capacity in carbon anode materials—these models were used to propose a universal descriptor for charge-transfer binding systems. This same descriptor was subsequently adapted to examine electrocatalysts, resulting in the discovery of a new electrocatalytic material and an associated record of invention. Next, we outlined a general approach for predicting surface diffusion and redistribution kinetics, applying the approach to lithium and hydrogen redistribution on graphene. Finally, specific suggestions for electrode performance improvement were offered, some of which were directly verified experimentally. Several of the multiscale methodologies developed here were directly leveraged in a proposal to examine transport and phase-transformation kinetics in hydrogen storage materials, which was recently awarded by DOE's Office of Energy

Efficiency and Renewable Energy. As part of the project, we also established close collaborations with Rice University and Arizona State, as well as a working relationship with Bosch, LLC. We are currently pursuing the possibility of establishing a cooperative research and development agreement with Bosch to explore solid-state transport in battery materials.

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Predictive Models for Target Response During Penetration

Tarabay Antoun (12-ERD-064)

Abstract

Hardened and deeply buried targets, used by potential adversaries to protect strategic assets, are increasing in number and hardness, making them largely invulnerable to today's conventional weapons. The objective of our proposed research is to develop new, high-fidelity, three-dimensional modeling capabilities for predicting conventional penetrator performance against such targets. To develop this modeling capability, we will use a physics-based approach that makes use of mesoscale simulations to account for material heterogeneitie s and deformation mechanisms such as fracture, fragmentation, pulverization, and granular mechanics.

Successful execution of this project will result in a new capability with unprecedented fidelity for modeling the response of frictional materials to extreme dynamic loading environments such as those encountered during the interaction of an earth penetrator with a geologic target or the interaction of a bullet or a shaped charge with ceramic armor. This modeling framework will support the design of advanced penetrating weapons that are smaller, lighter, faster, and more effective against hardened and deeply buried targets. Also, this work will make it possible to design more efficient transparent ceramic armor capable of providing superior protection against a wide range of threats, including shaped charges and improvised explosive devices.

Mission Relevance

We will build on state-of-the art modeling capabilities to support the Laboratory's mission in international and domestic security, with specific emphasis on defense applications to enhance U.S. military effectiveness and better protect military and domestic targets against attack. Developing new modeling capabilities

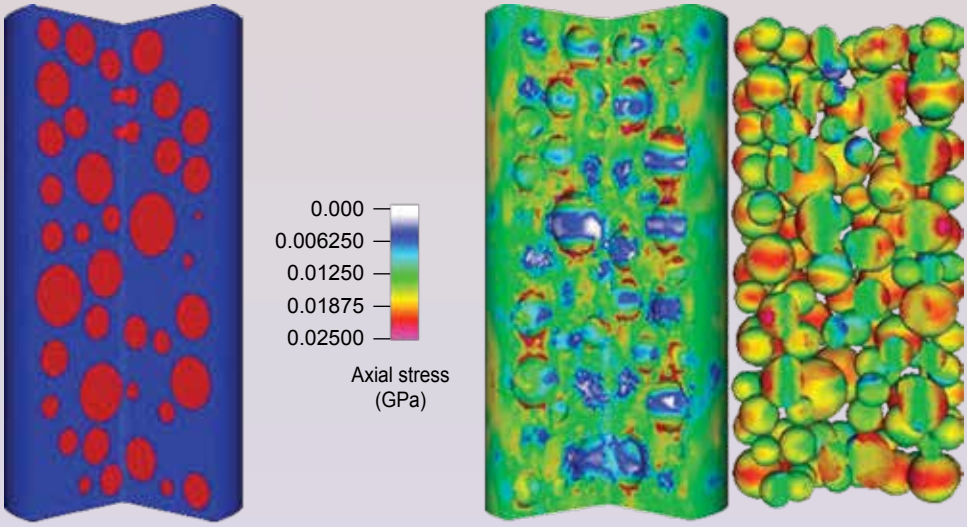
is directly relevant to the Laboratory's core competency in computational science and engineering.

FY14 Accomplishments and Results

During the last year of the project, we have (1) refined and optimized our mesoscale computational algorithms, thereby enabling the most robust, accurate, and efficient mesoscale simulations ever performed of fracture and fragmentation in brittle materials; (2) developed a continuum model for concrete for which parameters can be developed using the mesoscale simulation results; (3) implemented the model in the modular material model library GEODYNLib, thus making the model accessible by several LLNL codes, including ALE3D (three-dimensional arbitrary Lagrangian–Eulerian multiple-physics numerical simulation software); and (4) utilized the model to perform simulations of penetration events.

Project Summary

Our project has resulted in the development of a new capability for modeling the response of frictional materials to extreme dynamic loading environments such as those encountered during the interaction of an earth penetrator with a geologic target, or the interaction of a bullet or a shaped-charge with ceramic armor. A major scientific achievement of our research effort was the development of massively parallel computational algorithms in a Lagrangian framework for simulating discrete fracture and fragmentation. Complementary capabilities for extracting effective continuum properties from mesoscale simulations were also developed and demonstrated. These capabilities enabled mesoscale simulations of unprecedented details to examine the microstructural processes that govern deformation and failure in concrete (see figure), leading to the development of a constitutive model at the macroscale suitable for performing simulations of penetration into concrete and other geologic targets. We



Slice of RVE before loading

Red = Aggregate
Blue = mortar

Mortar

Aggregates

Slice of RVE after unconfined uniaxial compression for a strain of 0.28%

Mesoscale simulations of a cylindrical representative volume element (RVE) of concrete are being used to elucidate deformation and failure mechanisms. The results are used to inform a physically motivated constitutive model for the response of concrete to a penetrating object.

are arranging for a new project for the joint DOE and Department of Defense Munitions Technology Development Program in FY16 that will focus on multiscale modeling of concrete for penetration applications.

Publications and Presentations

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Illuminating the Dark Universe with the Sequoia Supercomputer

Pavlos Vranas (13-ERD-023)

Abstract

We propose to use the Sequoia supercomputer at Livermore, along with lattice gauge theory, to simulate theories that can explain the nature of dark matter and lead to experiments that will detect it. Approximately 83% of all matter in the universe does not interact directly with the electromagnetic or strong nuclear force—light does not bounce off it and ordinary matter goes through it with only the feeblest of interactions. Essentially invisible, it has been termed dark matter, yet its interactions with gravity produce striking effects on the movement of galaxies, leaving little doubt of its existence. In the time it takes to read this page, an astonishing amount of this material, about a billion particles, can pass through the human body. Our project involves numerical simulations of models using the methods of lattice gauge theory to convert continuous space–time into a regular four-dimensional grid of points called the lattice, which maps naturally onto the grid of compute nodes of a massively parallel supercomputer such as LLNL's Sequoia. We plan to begin with higher-precision measurements on existing lattice configurations and follow with new configuration generation and measurements based on our findings and experimental results that become available.

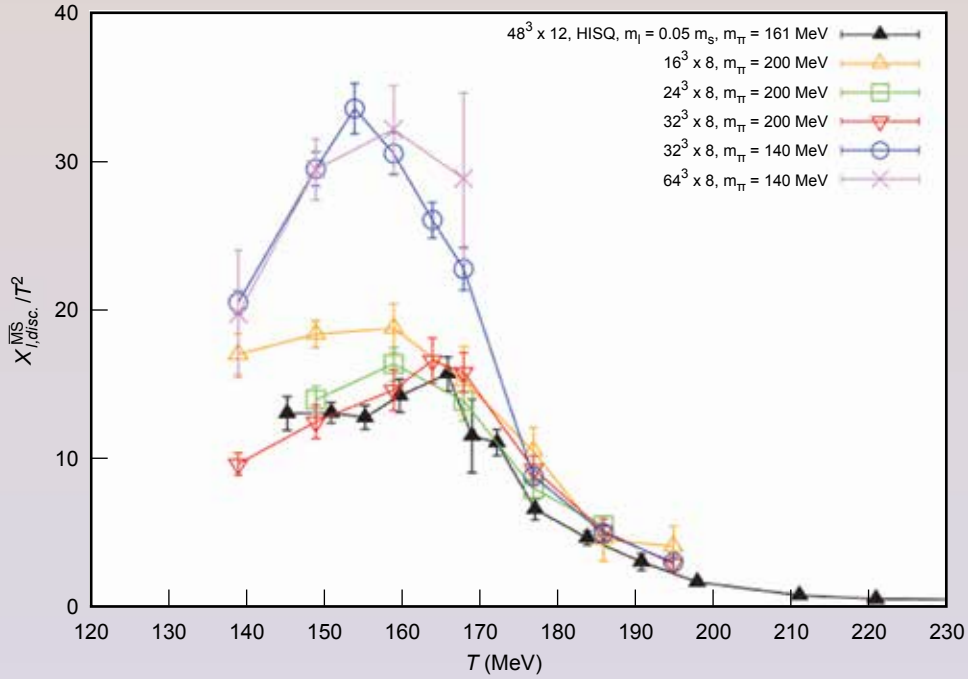
Strong-force dynamics is a prime candidate theory for understanding dark matter physics. It predicts that new composite particles thousands of times smaller than nuclear particles exist as dark matter. We will calculate cross sections of the composite strong-force dynamics particles with various detector materials using Sequoia, which will enable new experiments for direct detection of dark matter. We expect to calculate (1) the charge distributions and radii, dipole and magnetic moments, and polarization capabilities for strong-force dynamics theories; (2) nuclear physics quantum chromodynamics form factors, interactions, and effective theories to identify detector materials sensitive to strong-force dynamics dark matter; and (3) the thermodynamic phase transition of strong-force dynamics dark matter in the early universe.

Mission Relevance

Our research will add a strong theoretical component to LLNL's premier experimental dark matter detection program and establish LLNL as a leader in dark matter theory. The project supports the Laboratory's strategic mission focus area in stockpile stewardship science with study of the structure and interactions of nuclear particles, which is directly relevant to the physics of light–ion reactions that occur in high-energy-density environments. Our lattice simulations using Livermore supercomputers is relevant to the Laboratory's core competency in computational science and engineering.

FY14 Accomplishments and Results

In FY14 we (1) completed simulation of the quenched four-color gauge theory ("color" is the analog of electric charge in quantum chromodynamics) and measured particle spectrum and interaction of dark matter candidate particles with detectors, (2) simulated the dynamical two-color theory and measured the mass of the state of a "glueball" (a composite particle consisting solely of gluon particles), (3) completed generation of a large ensemble of zero-temperature quantum chromodynamics configurations (64 sites per dimension) and 300-MeV pion subatomic particles, (4) calculated the cross section of the four-color dark matter particle with the Large Underground Xenon detector in South Dakota via Higgs exchange, (5) completed a calculation of the three-color thermal transition with chiral fermion and physical pion particles, (6) completed calculation of four-color gauge theory polarization capability of the subatomic baryon composite particle, and (7) calculated the s- and p-wave nucleon–nucleon scattering.



The quantum chromodynamics thermal transition took place microseconds after the Big Bang. This is the first faithful calculation of the transition critical temperature (location of peak) performed using lattice numerical simulations on Livermore's Vulcan supercomputer.

Proposed Work for FY15

In FY15 we will (1) calculate the polarizing capability of the four-color composite dark matter candidate particle; (2) calculate its cross section with the latest dark matter detector via polarizing interaction; (3) simulate the two-color theory and calculate its dark matter particle properties; (4) perform a two- and three-color theory search for a composite light scalar particle that can be a Higgs particle candidate, continuing the two-color work for different vacuum alignment angles and measuring effects on the mass; (5) begin generation of an ultimate ensemble of lattice configurations with physical or near-physical pion mass to be used for a wealth of zero-temperature quantum chromodynamics observables of interest to nuclear physics; and (6) measure charge fluctuations of a quark–gluon plasma near transition to determine temperature at chemical freeze-out.

Publications and Presentations

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Fast Running Codes via High-Fidelity Reduced-Order Models

Kyle Chand (13-ERD-031)

Abstract

We propose to develop efficient techniques for construction of composite adaptive reduced-order models from the high-fidelity and multiple-parameter simulation tools characteristic of LLNL applications. These models will be orders of magnitude more efficient (for both the central processing unit and computer memory) than their corresponding high-fidelity models, making them invaluable in applications that require large numbers of simulations such as design optimization, parameter sampling, or rapid evaluation of an engineering model. We will develop new time-, space-, and parameter-adaptive reduced-order model techniques that are incrementally built from high-fidelity simulations. To support this development, we will research new locally adaptive approaches for development of the composite adaptive reduced-order models, as well as error estimation for the models.

At the successful conclusion of this project, we will have developed the mathematical theory for, and practical implementation of, effective model-reduction techniques for the highly dynamic and feature-rich simulations performed at LLNL. Our composite reduced-order modeling approach should be able to construct reduced-order models that are adapted to the local time, space, and parameter features of high-fidelity simulations. As part of these new techniques, effective error-estimation procedures will be developed that allow users to determine when the models are applicable. This technology will be demonstrated on both model problems (initially) and mission-relevant high-fidelity simulations relevant to applications in renewable energy and building energy efficiency.

Mission Relevance

Combining LLNL's existing simulation expertise based on partial differential equations with a sophisticated reduced-order modeling infrastructure will position the Laboratory to expand into new avenues of research and development critical to its mission in energy security. Our proposed technology will be useful for designing advanced building control systems and optimal sensor placement, as well as for generating large-eddy simulations that have great potential in optimization problems, such as placement of wind-farm turbines and enhancing phenomenological models for turbine wake aerodynamics.

FY14 Accomplishments and Results

In FY14 we (1) developed a parallel implementation based on C++ computer programming language of our incremental and adaptive orthogonal decomposition algorithm; (2) tested the new incremental algorithm in parallel as well as implemented reduced-order models based on simulation codes from the Overture and SAMRAI code frameworks for solving partial differential equations; (3) further developed a reduced-order-model generation framework based on the MATLAB technical computing language and interactive environment designed to rapidly test new snapshot, error estimation, and construction algorithms for reduced-order models; and (4) developed new theoretical error estimates for reduced-order models.

Proposed Work for FY15

In FY15 we will (1) use theory to guide the improvement of our algorithms for selecting snapshots adaptively that reduce errors of the downstream reduced-order model; (2) test new theoretical error estimates, developed in FY14, in model applications to examine their usefulness in real-world type problems; (3) harden, document, and release for more general use the C++ and MATLAB software infrastructures; and (4) test our algorithms on laboratory-scale applications on high-performance computing systems to demonstrate the utility of our new reduced-order modeling techniques.

Simulation of Engineering Fracture and Fragmentation

Jessica Sanders (13-ERD-047)

Abstract

The fracture of structural materials has many national security applications, from stockpile stewardship science to penetrating munitions. However, the modern history of solid mechanics is littered with unrealized claims of a silver bullet that will make fracture simulation as easy as standard structural finite-element analysis. Despite many publications and much interest in the field, predictive methods for complex problems remain elusive. Furthermore, current numerical fracture methods at LLNL tend to be either only loosely based on the underlying physics or highly tailored to specific applications, in most cases requiring careful and specific calibration, and so are not suitable for mission-critical applications. We propose to develop a robust numerical framework for representing fracture and fragmentation in finite-element models for solid mechanics. Our focus is on developing a fundamental, mathematically discrete technology that can accommodate the broad range of physical and material phenomena. One common approach to failure modeling represents material separation implicitly through homogenized or distributed damage. We instead seek to explicitly model fracture as the creation of new free surfaces within a previously continuous body. To this end, we propose to develop a numerical crack description that is independent of mesh design.

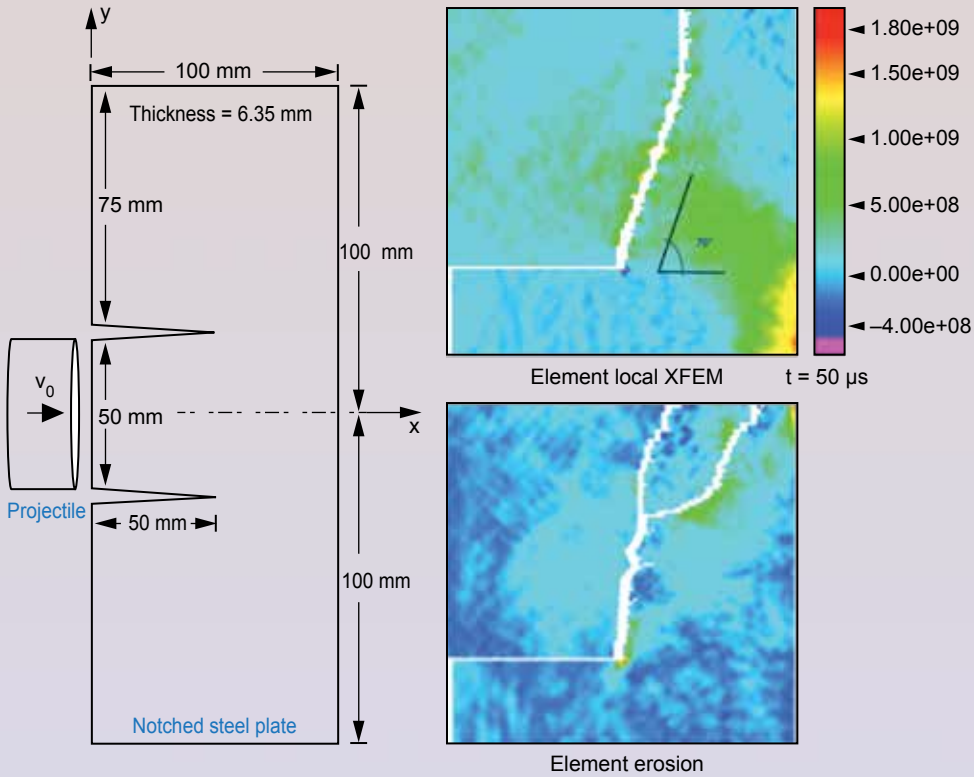
We expect to develop a numerical method that propagates the topological changes attending fracture in continuum bodies. Our focus will be a computational scheme that offers a uniform interface and can accommodate different physical models for material separation. Such approach would form a sound foundation for hosting advanced physics models of material damage that span the range of LLNL problem classes. We intend to provide advanced algorithm specification and develop a prototype along with a corresponding test suite. An implementation in Livermore's DYNA3D mechanical deformation code will enable Laboratory engineering analysts to exercise the method on their problem classes.

Mission Relevance

Representing fracture and fragmentation in finite-element models is supportive of the laboratory's core competency in computational science and engineering. The fracture of structural materials is relevant to many national security applications. Defense applications include prediction of structural survivability for transport security and blast protection of armored vehicles, as well as bullet impact on propellants, impact-driven fragmentation in high explosives, and structural spall during high-pressure deflagration events.

FY14 Accomplishments and Results

In FY14 we (1) fully extended our numerical method to three dimensions and developed several test problems that demonstrate the capability; (2) populated the test suite with simulations that are representative of problems with experimental data available to



In a well-known experiment conducted by Kalthoff and Winkler, a projectile impacts a notched steel plate. At low impact velocities, the steel fails in a brittle way. The notches develop cracks at a 70° angle. Simulations with the extended finite-element numerical technique (upper right) capture this failure behavior better than the standard alternative (lower right) for the same mesh.

validate our approach; (3) created a capability to handle crack branching at an elemental level, which facilitates global crack branching and coalescence; (4) demonstrated the method for brittle fracture models; and (5) began extending the method to ductile failure models.

Proposed Work for FY15

In FY15 we will develop a computational test suite for ductile failure models that includes validation problems with known experimental data. In addition, we will develop the modeling capability to capture the recontact of surfaces after fracturing.

Publications and Presentations

Sanders, J. D., and M. A. Puso, 2013. *A phantom node approach to dynamic crack propagation*. 12th U.S. Natl. Congress on Computational Mechanics, Raleigh, NC, July 22–25, 2013. LLNL-PRES-641179.

Measuring Dark Energy with the Large Synoptic Survey Telescope

Michael Schneider (13-ERD-063)

Abstract

It is well established that the expansion of the universe is accelerating, in contradiction to the expectation for a universe filled with matter as we know it. This discovery, for which the 2011 Nobel Prize in physics was awarded, has led astronomers to postulate a new cosmological energy component dubbed dark energy. While the mean energy density and equation of state of dark energy have been reasonably constrained with current observations, determining its nature—whether a cosmological constant, new particle, or modification to gravity—requires a difficult measurement requiring mapping the cosmological expansion and matter perturbation growth rates over much of the volume of the universe. The Large Synoptic Survey Telescope (LSST) is the most comprehensive wide-field survey proposed to study dark energy to date. We propose to combine atmospheric and cosmological simulations and observations to enable the discrimination of dark energy models with the gravitational lensing observations planned with the LSST. We will combine LLNL expertise in image simulation and LSST engineering that will be superior to performance of competing machine-learning methods in the community.

We expect to combine unique LLNL engineering, simulation, and astrophysics expertise to develop image simulations and analysis algorithms incorporating our physical knowledge of the atmosphere and the LSST instrument to establish gravitational lensing as a viable and robust astronomical measurement for the LSST and other future surveys. Our simulations will be unique in including optical wave-front propagation through the atmosphere and telescope, as well as integrated optical

and astrophysical image analyses. We can make a large scientific contribution by significantly reducing the risk in measuring galaxy shapes with LSST, thereby enabling unbiased gravitational lensing observations to distinguish between a cosmological constant (or quantum vacuum energy) and a new dynamical component (or fundamental particle) of the universe as explanations for dark energy. The project could establish LLNL as a leader in image and cosmological simulation efforts for LSST, and thereby enhance recruiting efforts as well as provide newsworthy demonstrations of high-performance computing resources applied to cosmology.

Mission Relevance

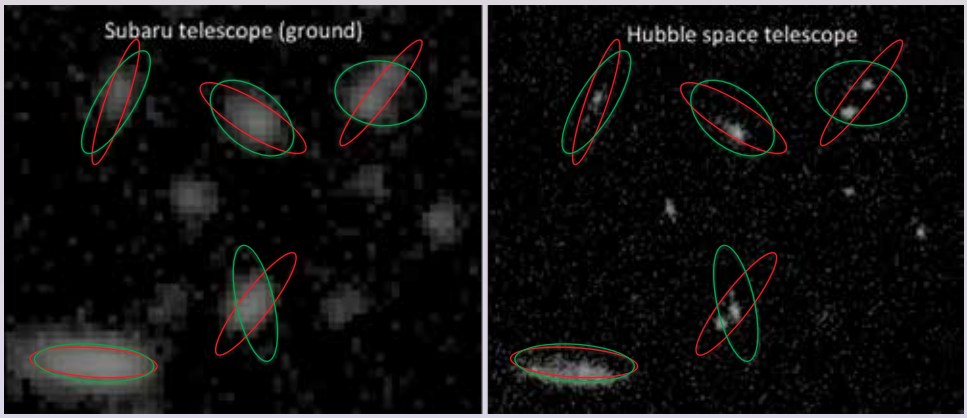
The simulations and algorithms that we intend to develop will directly contribute to the creation of robust simulation codes that are a key element of LLNL's core competency in computational science and engineering, as outlined in the Laboratory's strategic plan. Our research may also have applications to Livermore programs such as space situational awareness when using ground-based optical observations.

FY14 Accomplishments and Results

In FY14 we (1) performed simulation-based forecasts of the active optics wave-front control for LSST on galaxy shape-measurement systematic errors; (2) developed an analytic model and statistical framework to correct perturbed wave fronts from wave-front sensor measurements; (3) validated simulations of light propagation through atmospheric turbulence phase screens of arbitrary size, including common simulation approximations and atmospheric model parameter variations; (4) created software to apply modern machine-learning methods to the galaxy shape-measurement problem for LSST and related astronomical surveys; and (5) identified object blending from atmospheric image blurring as a major new systematic error for future dark energy measurements from the comparison of galaxy shape estimation in ground- and space-based observations.

Proposed Work for FY15

In FY15 we will (1) study effects of the outer scale of atmospheric turbulence and frozen-flow approximation to the evolution of large-scale structures in the universe on



Comparing galaxy images from ground (left) and space (right) shows that galaxy ellipticities for gravitational lensing measurements are biased by blending of distinct galaxies because of atmospheric turbulence. Red ellipses are the ellitpicities seen from the ground while green are those seen from space. Our algorithms are designed to use available space-based observations to reduce the bias in ellipticity measurements from the ground.

the predicted and modeled point-spread functions, (2) determine active optics control impacts on gravitational lensing measurements and mitigation strategies for object-blending impacts on galaxy shape-measurement systematics, and (3) apply improved point-spread functions modeling and interpolation and galaxy shape-measurement algorithms to existing astronomical data sets to demonstrate improved gravitational lensing inferences.

Publications and Presentations

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Search for Metallic Hydrogen: An Advanced First-Principles Study

Miguel Morales-Silva (13-LW-004)

Abstract

Hydrogen is one of the most important elements in the periodic table, and an accurate understanding of its properties is crucial to many fields of science, including high-pressure physics, astrophysics, planetary physics, inertial-confinement fusion, and energy production. In addition, hydrogen offers many unique and exciting features in its

high-pressure phase diagram, including the possibility of a zero-temperature quantum liquid. Despite decades of intense efforts from many experimental high-pressure groups, little concrete information is known about details of the phase diagram for pressures above 300 GPa and temperatures above a few hundred Kelvin. Not even structures of the ordered phases have been obtained experimentally. Over the last decade, first-principles computer simulations have been crucial in the correct interpretation of experimental results as well as resolving conflicting experimental results in the field of high-pressure hydrogen. We propose to perform breakthrough calculations of the fundamental, electronic, and optical properties of hydrogen at high pressure using quantum Monte Carlo methods, which rely on repeated random sampling to obtain numerical results. This work will generate the most accurate theoretical description of hydrogen in the metallization and dissociation regime of the phase diagram, directly including nuclear quantum effects and employing next-generation, first-principles methods based on quantum Monte Carlo.

We intend to elucidate the phase diagram of hydrogen in the region of pressure where molecules in the molecular crystal break under the effect of pressure and the system transforms into an atomic crystal. We also want to trace the melting lines of these crystal structures of hydrogen, providing a phase diagram purely from first-principles simulations. We will examine the stability of a possible low-temperature quantum liquid in hydrogen as well as the characteristics of dissociation and metallization in the solid. Finally, we will perform accurate calculations in the regime of the phase diagram where the liquid–liquid transition meets the melting lines of both molecular and atomic solids.

Mission Relevance

This project is closely aligned with Livermore missions in stockpile stewardship and core competencies in high-energy-density science and computational science and engineering. Developing one of the leading alternatives for next-generation, first-principles simulation methods will not only place the Laboratory in a leading role in the development of electronic structure methods, but will also reinforce Livermore's position as one of the leading centers for the study of high-pressure materials.

FY14 Accomplishments and Results

In FY14 we (1) completed the most accurate study of the zero-temperature phase diagram of high-pressure hydrogen in the regime of molecular dissociation and metallization; (2) used state-of-the-art quantum Monte Carlo calculations to perform first-principles calculations without any mean-field approximations, routinely used in other calculations in the field; (3) obtained results that, for the first time, agree with experimental extrapolations of the location of the phase transition from metal to insulator; (4) performed a detailed study of the melting of solid metallic hydrogen at pressures beyond 450 GPa and its relation to the liquid–liquid transition, and incorporated nuclear quantum effects in the protons to properly account for zero-point effects; and (5) implemented a semi-classical quantum dynamical method based on path integrals and applied it to the problem of vibrational spectroscopy on

hydrogen, which provides an alternate method to locate phase transitions in the solid at finite temperature and can be compared directly with experimental measurements.

Project Summary

The successful conclusion of this project resulted in the most accurate study of the low-temperature phase diagram of high-pressure hydrogen to date. We used advanced, state-of-the-art electronic structure methods to study the various structural and electronic phase transitions in hydrogen without relying on mean-field approximations that have limited the predictive capabilities of theoretical work in the past. We have made a detailed and accurate account of the influence of nuclear quantum effects in the electronic, structural, and optical properties of hydrogen, which lead to dramatic influences like a decrease in the melting line of the atomic solid by about 100 to 200 K (compared to classical simulations), and a reduction in the band gap of molecular solids by more than 1 to 1.5 eV. We made predictions on the location and character of the metal–insulator transition in the solid phase that will stimulate further experimental study in the community. We hope to expand the methods developed by this project to other materials in the periodic table, including hydrogen–helium mixtures and high-pressure phases of lithium.

Publications and Presentations

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A Coupled Seismic and Acoustic Simulation Capability

Arthur Rodgers (14-ERD-001)

Abstract

Many events of interest occur near or above the interface between the air, water, and solid earth. Currently, we cannot jointly model acoustic and seismic signatures from these events with existing tools—therefore, we propose to develop a coupled seismic and acoustic simulation capability. We will extend our current capability of the WPP (Wave Propagation Program) and SW4 (Seismic Waves, 4th order) codes for seismic motions in solid earth to model energetic sources and motions in fluid regions of the Earth (air and water) and properly account for the flow of energy between the solid

and fluid regions. Energetic events such as explosions and earthquakes near the Earth's surface generate motions in the atmosphere and water as well as seismic motions in the solid earth. Simulation of these waves, including propagation biases from the intervening material, enables more accurate inference of source properties. We will accomplish this by extending recent work on elastic waves to acoustics.

Successful execution of this project will expand the variety of energetic phenomena we can model. We expect to produce a simulation capability for coupled seismic and acoustic motions that will be compatible with LLNL's hydrodynamic codes such as the GEODYN compressible solid and fluid dynamics code, and ultimately enable simulation of motions from detailed hydrodynamic source descriptions to far-field seismic and acoustic waves. We will be able to model sources and mechanical motions in the atmosphere and water as well as the solid earth. We intend to simulate seismic and overpressure data collected in near-surface explosion tests and account for measurable weather effects on overpressure, such as wind and sound-speed profiles, as well as sub-surface geologic structure. This will help us understand biases in source estimates that arise from predictable wave propagation effects and establish backwards-modeling capabilities for forensic analysis of explosions.

Mission Relevance

Our research will enable understanding of seismic and acoustic propagation effects that bias source estimates with the development of a simulation capability that is compatible with Laboratory hydrodynamic codes. This capability falls squarely within the Laboratory's core competency in computational science and engineering and high-performance computing. This foundational science is supportive of critical mission needs such as nuclear nonproliferation to monitor nuclear explosions, national security to determine the defeat of hard and deeply buried targets, and new ways to extract underground energy more efficiently while minimizing risk and environmental impact.

FY14 Accomplishments and Results

In FY14 we (1) formulated acoustic wave propagation based on the Euler equations of compressible flow to solve for the pressure, density, and three-component velocities allowing fully three-dimensional variations in background pressure, density, and wind velocity; (2) developed interface conditions for the coupled acoustic–elastic case and tested it in one dimension; (3) developed the first generation of our message-passing interface parallel code SAW4 for modeling the purely acoustic case—this code currently includes absorbing and reflecting boundary conditions to represent the far-field radiation and a flat Earth surface, respectively; and (4) continued development of the coupled seismic–acoustic case without topography or attenuation.

Proposed Work for FY15

In FY15 we will (1) complete the second generation of our modeling code SAW4 for simulating the coupled seismic–acoustic case without topography or attenuation; (2) develop, for this modeling code, a generalized methodology to allow for realistic topography and intrinsic attenuation for the purely acoustic case; (3) solve, for this

purpose, the acoustic wave equations on a curvilinear mesh, conforming to the topography of the Earth; (4) research an efficient and accurate approach for modeling intrinsic attenuation in the atmosphere; and (5) implement these modeling capabilities in the third generation of our modeling code, and verify it against canonical problems.

Publications and Presentations

Petersson, N. A., 2014. *Summation-by-parts methods for accurate seismo-acoustic wave simulations*. LLNL-PRES-651569.

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Petersson, N. A., B. Sjogreen, and A. J. Rodgers, 2014. *Two summation-by-parts finite difference codes for large scale simulations of seismic motion*. LLNL-POST-653713.

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Sjogreen, B., and N. A. Petersson, 2014. *SBP finite difference approximations for seismic and seismo-acoustic computations*. LLNL-PRES-655825.

Atmospheric Source Reconstruction with Uncertainty Quantification

Ronald Baskett (14-ERD-006)

Abstract

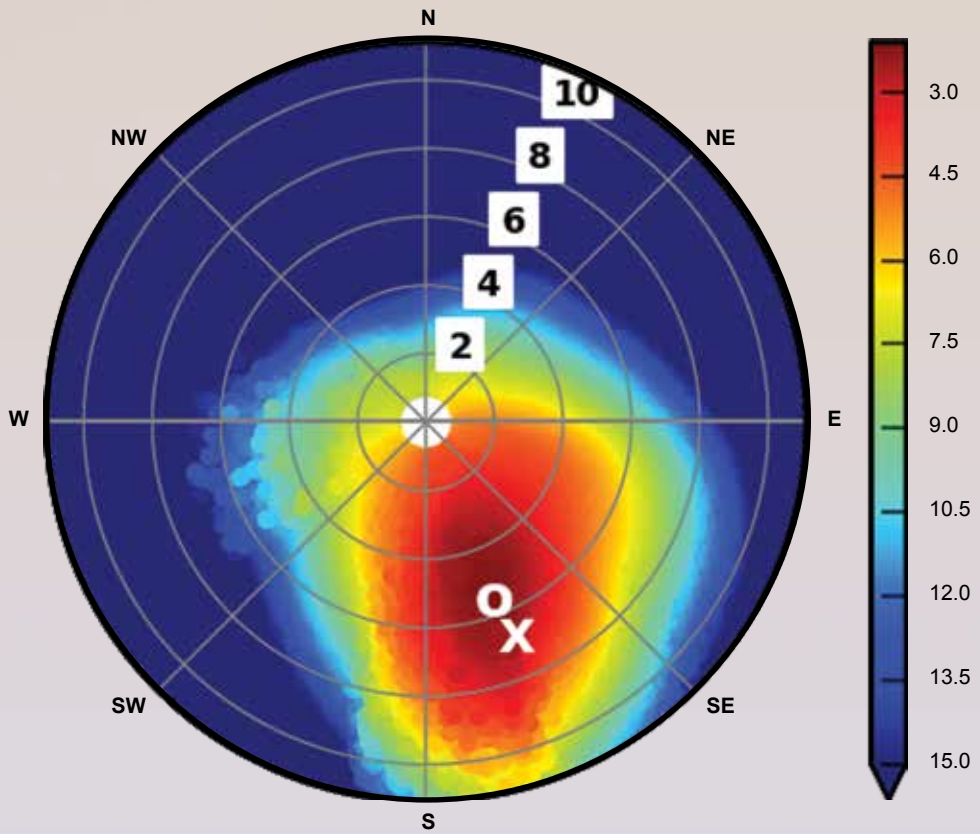
Accidental or terrorist releases of hazardous materials into the atmosphere can impact large populations and cause significant loss of life or property damage. Knowledge of the source properties for these releases is critical to understanding the event and predicting its consequences, yet this knowledge is often incomplete. We propose to address the need to reconstruct poorly characterized sources of atmospheric releases for events such as radioactivity released from the Fukushima Daiichi nuclear power plant in Japan in 2011, as well as for applications in attributing methane from landfills and wellheads. We will develop a Bayesian analysis system of statistical inference based on interpretation of probability to estimate source properties using available plume concentration or deposition measurements, in a way that accounts for prior uncertainties in the source properties, meteorological uncertainties, and unknown biases in models and observations. We will verify our capability against several key data sets.

We expect to develop a stand-alone set of computer scripts and codes that will provide an automated probabilistic estimate of the most likely source characteristics based on an ensemble of meteorological conditions. This project will address national

needs in emergency response, counterterrorism, and attribution programs with an advanced capability to identify unknown sources of atmospheric plumes with quantified uncertainty. This capability will also be relevant for determining point-source emissions for international monitoring efforts such as for the Comprehensive Test-Ban Treaty Organization and for the Organization for Economic Cooperation and Development measurement of the long-range transport of air pollutants, and enable source identification for odors or air pollutants, including methane leakage from wells used in hydraulic fracturing for release of oil or gas from rocks.

Mission Relevance

Our research in atmospheric source reconstruction is applicable to the monitoring, detection, analysis, and rapid response needs of multiple areas that have atmospheric components, including consequence management of nuclear power plant events,



Urban dispersion models based on computational fluid dynamics that can resolve flow and turbulence in and around buildings with 1-m resolution are very sensitive to initial boundary conditions. A change of a few degrees in the model's initial inflow can result in order-of-magnitude differences in concentrations downwind. The figure shows the log-likelihood (color scale) of inflow wind speed (radial values of 1–10 m/s) and direction (polar location) computed using an ensemble of Aeolus urban computational fluid-dynamics simulations. Determined from a Bayesian method to minimize the differences between modeled and measured tracer concentrations, the inferred maximum-likelihood inflow value (shown by the X at 6.8 m/s and 157°) is comparable to the measured value at an upstream location (shown by the O at 5.6 m/s and 159°).

nuclear nonproliferation, nuclear forensics, and greenhouse gas and air-quality emission compliance in support of Laboratory missions in both national and energy security. The project is relevant to the Laboratory's strategic focus area in energy and climate, which includes work on uncertainty quantification, model diagnosis and comparison, and managing large data sets for climate research. In addition, our research methodology aligns with the core competency in computational science and engineering.

FY14 Accomplishments and Results

In FY14 we (1) constructed an initial Wx-ATD (weather atmospheric transport and dispersion) model run stream, (2) implemented an initial Bayesian inversion tool set into the atmospheric source reconstruction system, (3) built Oklahoma City case study data sets, and (4) ran atmospheric source reconstruction against Oklahoma City case studies and analyzed the results as well as estimated sensor location uncertainties.

Proposed Work for FY15

In FY15 we will (1) update the Wx-ATD model run stream with various weather research and forecasting, particle dispersion, and regional modeling codes including WRF, AEOLUS, INPUFF, FLEXPART, and LODI; (2) extend the Bayesian inversion tool set to the atmospheric source reconstruction system to treat uncertainties in three-dimensional meteorology and uncertainties in measurements; (3) build the 1986 Diablo Canyon and 2011 Fukushima data sets; (4) run the atmospheric source reconstruction against one Diablo Canyon case study data set and analyze the results; (5) update atmospheric source reconstruction system documentation; and (6) provide the atmospheric source reconstruction software and documentation to the atmospheric modeling test bed.

Publications and Presentations

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Lucas, D, et al., 2014. "Designing optimal greenhouse gas observing networks that consider performance and cost." *Geosci. Instrum. Method. Data Syst.* **4**, 705. LLNL-JRNL-659224.

Advanced Discretization Techniques for Paraxial Laser Propagation

Jeffrey Banks (14-ERD-032)

Abstract

The efficiency of high-intensity lasers can be improved, and their use as experimental facilities expanded, through an improved understanding and optimization of laser components. For instance, understanding and controlling the deleterious edge-diffraction

effects caused by beam-line obstructions requires the resolution of small features in transmission masks. Furthermore, shot planning would benefit from the ability to perform whole-beam modeling of fratricide from optics defects. High-resolution simulations are a major contributor to such studies, but many problems remain to be addressed. In this project, considering the long-term research needs for high-fidelity beam analysis and simulation, we will investigate using the paraxial model of light propagation (i.e., at a small angle to the optical axis of the system) and adopting new discretization techniques (transforming continuous attributes into discrete ones) for the paraxial wave equation with high-order finite differences. In addition, we will examine eliminating the splitting error in Fourier discretization, using super-grid absorbing boundary conditions, and developing reduced-order models based on new, advanced discretization techniques.

The primary deliverables of this project are a series of technologies. For beam analysis, we will develop high-order discretization, nonuniform meshing, alternating-direction implicit methods, and deferred correction, as well as conduct preliminary investigations of reduced-order models—all novel capabilities. We will also investigate related applications that would benefit from this work. For instance, Schrödinger equations, of which the paraxial wave equation is a subset, have application in a variety of fundamental science scenarios, including atomic structure, the Bose–Einstein condensate state of matter, and ocean wave propagation.

Mission Relevance

This project advances the Laboratory's strategic focus area of lasers and optical materials and core competency in computational science and engineering by enhancing our ability to perform large-scale, high-fidelity simulations that will improve our understanding of laser components in the near term, enable whole-beam modeling of the effects of optic defects in the medium term, and advance short-pulse applications in the long term by predicting the effects on beam characteristics of phenomena such as spectral dispersion.

FY14 Accomplishments and Results

In FY14 we focused on explicitly integrated high-order discretization based on finite differences to create an initial capability from which further research can be based. Specifically, we (1) developed explicit methods of second- through tenth-order discretization, (2) investigated absorbing boundary conditions using a perfectly matched layer, and (3) began to understand the performance of these capabilities for the nonlinear Schrödinger equation model.

Proposed Work for FY15

In FY15 we will (1) continue work on the finite-difference discretization strategy including incorporating a defect-correction strategy for high-order time integration, (2) begin work on the reduced-order modeling components, (3) begin investigating creation of the reduced-order model basis using the explicit code constructed in FY14, and (4) begin examining the efficacy of the reduced-order modeling approach for high-intensity laser propagation.

Exploiting the Gemini Planet Imager: Revolutionary Exoplanet Science and Advanced Adaptive Optics

Stephen Ammons (14-ERD-076)

Abstract

Direct studies of exoplanets with current technology has plateaued—only the very youngest planets are visible, in the infrared with released gravitational potential energy. New capabilities are needed. The Gemini Planet Imager (GPI) is the most advanced astronomical adaptive optics system in the world, dedicated to imaging and characterizing extrasolar planets. Lawrence Livermore led the construction of GPI and will have unique access to it when it becomes operational. We will lead the first-light and campaign science observations, and propose to develop tools to maximize GPI scientific yield such as planetary signal extraction and optimal campaign simulators. The GPI is also a unique tool for probing atmospheric turbulence and adaptive optics control dynamics. Therefore, we will analyze its performance in detail to guide future high-performance adaptive optics system programs—in particular, algorithms that predict atmospheric turbulence a few milliseconds into the future.

Adaptive optics is a key Laboratory competency for a variety of programmatic and scientific applications, including remote sensing, laser beam control, astronomy, and microscopy. Astronomical adaptive optics has long been the engine that drives this, providing high-visibility projects to strengthen outside collaborations, allowing testing of innovative approaches, and enabling recruitment of the best young scientists and engineers in this field. The GPI will be an order of magnitude more sensitive than any existing facility, and it will probe solar systems that are very different from those accessed by the National Aeronautics and Space Administration Kepler mission. During first-light observations, we expect to characterize known planetary systems, determining the temperature and structure of young extrasolar giant planets and extrasolar asteroid and comet belts. We will produce a comprehensive performance budget for GPI. As the survey continues, we expect to discover 20 to 50 imaged extrasolar planets—enough to determine the frequency of solar systems resembling our own—with complete near-infrared spectra for 10 to 20 planets and estimates of their orbital eccentricity and period. In addition, we will perform on-sky testing of predictive control algorithms.

Mission Relevance

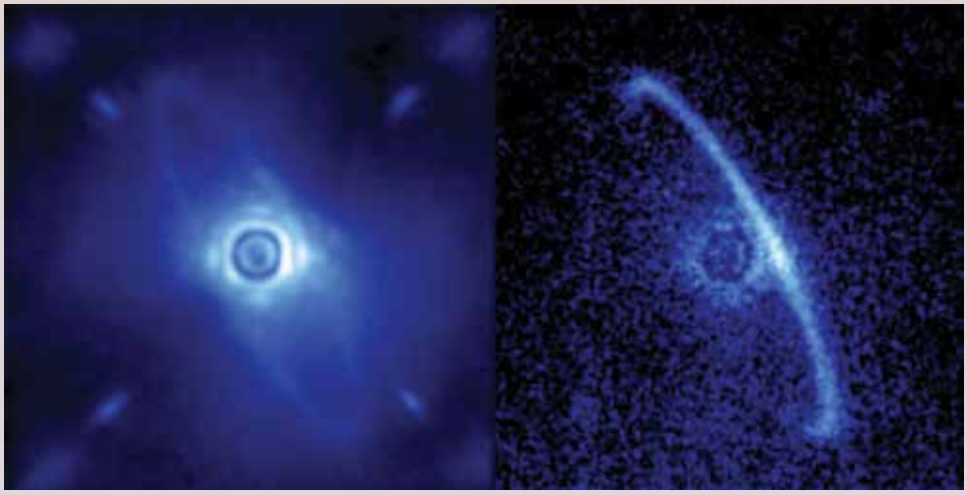
The high-contrast and high-performance techniques for adaptive optics developed here with the aid of predictive algorithms of atmospheric turbulence and GPI system simulations will be particularly applicable to space surveillance applications such as observations of targets requiring rapid telescope motion or faint sources close to bright targets. This work is relevant to the Laboratory's strategic focus area in cyber security, space, and intelligence to enhance situational awareness for space systems, as well as the core competency in computational science and engineering.

FY14 Accomplishments and Results

In FY14 we (1) participated in four extremely successful commissioning runs in November, January, March, and September; (2) identified targets for first light; (3) prioritized targets for GPI's science campaign; (4) analyzed GPI's telemetry, discovered telescope vibrations that limited performance, developed a controller fix for tip and tilt and focus, and performed on-sky testing; (5) completed an adaptive optics error budget validated from telemetry; and (6) studied the feasibility of predictive control for GPI.

Proposed Work for FY15

In FY15 we will (1) participate in GPI observations of planets and disks for the larger science campaign; (2) exploit the archive of GPI telemetry to refine adaptive optic-calibration sequences and optimize performance and contrast; (3) measure planetary masses of GPI discoveries with astrometric follow-up observations using Gemini multiple-conjugate adaptive optics, adaptive optics at Hawaii's Keck Observatory, and the European Space Agency's Gaia space observatory; (4) develop a maximum-likelihood algorithm for measuring planetary acceleration from poorly sampled astrometric data; (5) use the GPI adaptive optics simulator and realistic atmospheric screens to quantify the improvement in limiting guide star brightness and science reach expected in GPI with predictive adaptive optics control; and (6) develop a rigorous simulation of a GPI-like system for space situational-awareness applications.



Gemini Planet Imager's first-light image of a dust disk around the star HR4796A, in total infrared intensity (left) and polarized light (right). Image courtesy of Gemini Observatory, and image processing by Marshall Perrin, Space Telescope Science Institute.

Publications and Presentations

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Ammons, S. M., et al., 2014. *First light for high-contrast adaptive optics*. Advanced Maui Optical and Space Surveillance Technologies Conf., Maui, HI, Sept. 9–12, 2014. LLNL-PROC-660012.

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Savransky, D., et al., 2014. *Campaign scheduling and analysis for the Gemini Planet Imager*. IAUS 299, Exploring the Formation and Evolution of Planetary Systems, Victoria, Canada, June 2–7, 2014. LLNL-CONF-640216.

Cyber Security, Space, and Intelligence

FY2014

Lawrence Livermore National Laboratory

First-Principles Materials Characterization and Optimization for Ultralow-Noise Superconducting Qubits

Vincenzo Lordi (12-ERD-020)

Abstract

The application of superconducting quantum bits (qubits) in quantum information processing is currently limited by unidentified noise sources, which increases the speed of decoherence (loss of information from the system) to impractical levels. Whereas conventional computer bits come either in ones or twos, a qubit can be the equivalent of both a one and a two at the same time. In theory, this capability will allow the quantum computer to perform many different computations simultaneously. Our project aims to use first-principles atomic-scale simulations to develop a quantitative understanding of the microscopic origins of noise in superconducting qubits and devise strategies for reducing the concentration or impact of identified sources. We will characterize the sources of paramagnetic noise by developing atomistic models of defects in constituent materials and at the interfaces between them, focusing on niobium, rhenium, and aluminum superconductors; aluminum oxide tunnel junctions; and silicon–silicon dioxide substrates, based on the results of experimental superconducting qubit fabrication.

The main results of this project will be identifying and characterizing the electronic structure of defect and interface structures in superconducting qubit devices. Once we have identified the microscopic structures that generate paramagnetic noise through unpaired spins or fluctuating charge states, we can then develop passivation or purification strategies to reduce the concentration of noise sources in qubit devices. The strategies we develop to reduce this paramagnetic noise will enable superconducting qubit devices with decoherence times long enough for practical quantum computation.

Mission Relevance

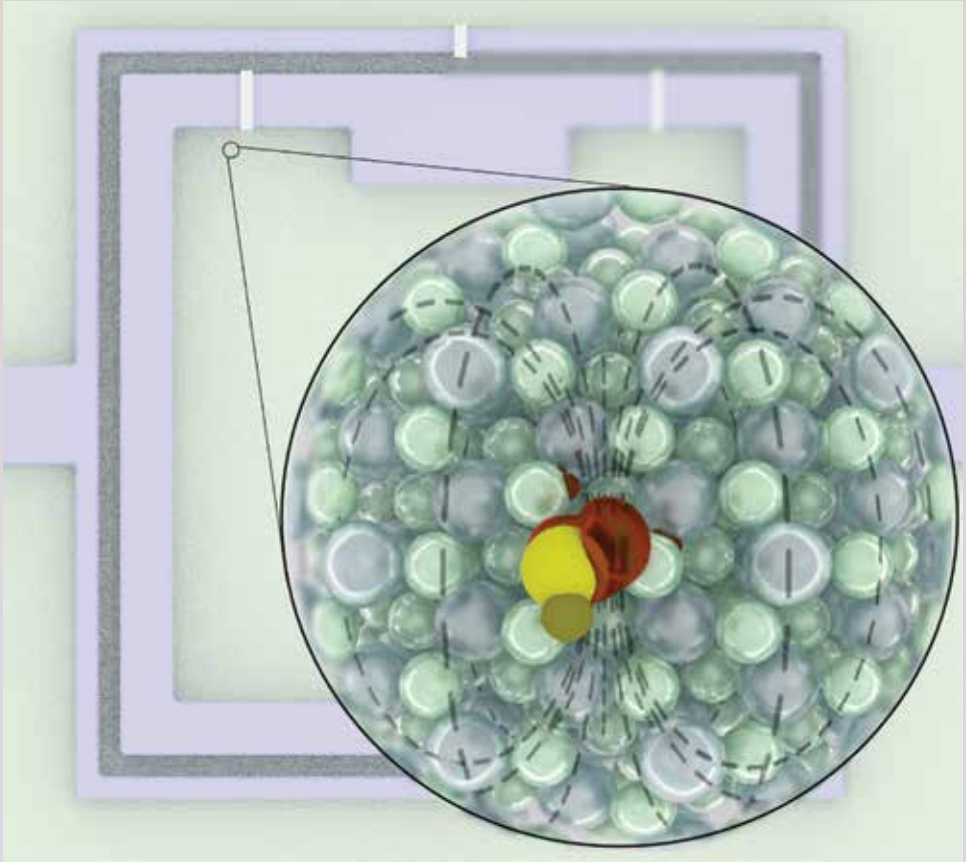
This project supports LLNL's cyber security mission by furthering the realization of solid-state quantum information processing systems that could play an important role in future cryptography technologies. By developing the science for fundamental computational materials, this project also bolsters the Laboratory's core competency in advanced materials and manufacturing and high-performance computing.

FY14 Accomplishments and Results

In FY14 we (1) completed an assessment of paramagnetic defects on amorphous silica surfaces, which are associated with certain native oxygen-deficiency centers; (2) modeled interactions among surface spin defects on the surface of sapphire and diamond, and defect-mediated surface spin–spin interactions; and (3) determined the mechanism of how distant spins may interact on surfaces through substrate mediation, relevant to observed spin clusters that can cause decoherence noise in qubits.

Project Summary

This project resulted in the identification of possible microscopic sources of paramagnetic decoherence noise on substrate surfaces used to fabricate superconducting qubits for two different technologically important substrates: sapphire and silica. On sapphire, we identified adsorbate impurities related to air (water vapor) exposure that create paramagnetic centers, as shown in the artist rendering. On amorphous silica, intrinsic structural defects (local oxygen deficiencies) were found that are paramagnetic. For sapphire, we performed a computational screen of over two dozen possible chemical passivants and found at least one (ammonia) that chemically passivates the surface and can reduce flux noise in qubits. We also found that an external electric field can passivate the surface. For silica, our results indicated only limited mitigation options, because paramagnetism is associated with intrinsic defects. We further studied how spins on sapphire may interact over long distances and discovered that strain can tune such interactions. In summary, our project elucidated possible microscopic origins of decoherence noise in qubits and provides rational paths forward to reduce noise toward practical levels in devices. In future work, we hope to expand these fundamental studies by coupling microscopic noise models to higher-level device models, enabling prediction of real-device responses to variations in material structure and processing conditions.



A hydroxyl adsorbate impurity formed from ambient water vapor interacting on the surface of sapphire (which is used to fabricate superconducting qubits) leads to a localized magnetic moment (indicated by the field lines). This phenomenon can introduce decoherence noise (information loss) through fluctuations.

Publications and Presentations

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Adelstein, N., et al., 2014. *Magnetic fluctuations from oxygen deficiency centers on the SiO_2 surface*. LLNL-JRNL-645056.

Adelstein, N., et al., 2014. *Magnetic noise in quantum systems due to paramagnetic electrons on the silica surface.* 2014 Intl. Workshop Silicon Quantum Electronics, Albuquerque, NM, Aug. 18–19, 2014. LLNL-POST-658854.

Lee, D., 2014. *First principles investigation of magnetic noise sources for superconducting qubits on alpha-Al_2O_3*. SPIE Optics and Photonics 2014, San Diego, CA, Aug. 17–21, 2014. LLNL-ABS-650192.

Lee, D., J. L. Dubois, and V. Lordi, 2012. *First principles investigation of magnetic noise sources for superconducting qubits on α-Al_2O_3*. 2012 MRS Fall Mtg. and Exhibit, Boston, MA, Nov. 25–30, 2012. LLNL-ABS-562793.

Lee, D., J. L. Dubois, and V. Lordi, 2014. "Identification of the local sources of paramagnetic noise in superconducting qubit devices fabricated on alpha-Al_2O_3 substrates using density-functional calculations." *Phys. Rev. Lett.* **112**, 017001. LLNL-JRNL-626953. *http://dx.doi.org/10.1103/PhysRevLett.112.017001*

Lee, D., J. L. Dubois, and V. Lordi, 2012. *Microscopic source of paramagnetic noise on Al_2O_3 substrates for superconducting qubits.* MRS Fall Mtg. and Exhibit, Boston, MA, Nov. 25–30, 2012. LLNL-PRES-604158.

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Network Simulation and Its Applications

Peter Barnes (12-ERD-024)

Abstract

Our proposed computer network simulation capability will contribute to Lawrence Livermore leadership in large-scale network simulations, with demonstrated applications in cyber security, global network situational awareness, performance modeling, and prediction. Predictive analysis of cyber mission risk and performance is one of the major gaps in our national cyber capability. What we propose is groundbreaking on several fronts. We will simulate realistic networks, derived from real and synthetic network maps, that incorporate real hardware and geographic constraints at the enterprise scale (at least 10^3 compute nodes); incorporate near-real-time updates from the global Internet; and generate traffic from realistic traffic models matched to observed data.

Achievement of real-time predictive models of complex enterprise, mission, and global networks will establish Lawrence Livermore as a national leader in cyber modeling and situational awareness. Completion of this effort will position the Laboratory to invent game-changing approaches to real-time cyber-situation awareness, as well as new approaches to intelligence analysis in the modern networked world.

Mission Relevance

The ultimate aim of this proposal is to meet the grand challenge of the Laboratory's cyber, space, and intelligence mission focus area—predictive models and simulations for complex information systems. In particular, we aim to build real-time models of the state and behavior of complex networks up to a global scale.

FY14 Accomplishments and Results

In FY14 we (1) developed a complete process for writing, pre-processing, and running extremely large network models, with up to one billion simulated network nodes; (2) successfully connected simulation output to the Laboratory's network mapper tools, enabling detailed visualization and analysis of simulation results; (3) completed development of the realistic application traffic model, specifically a statistical representation of real-world Websites used to drive simulated Web servers and browsers; and (4) completed studies of the mission execution model.

Project Summary

With this project, we dramatically improved the capabilities of the ns-3 network simulator, from a practical limit of less than one million nodes, to a one-billion node demonstration. We developed an XML description (Extensible Markup Language

text-based format used to share data on the World Wide Web) for simulation models, which is both compact and human readable, and developed automatic tools to prepare a model for parallel execution. In addition, we developed a faster parallel-scheduling implementation, which is now part of the released code, and improved the memory scaling by more than a factor of 1,000. Finally, we fixed or contributed to fixing 35 software bugs, and wrote or updated approximately 25,000 lines of software documentation. We expect to complete our one-billion node demonstration with support from the Department of Defense High Performance Computing Modernization Program and the U.S. Army Research Laboratory.

Publications and Presentations

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Continuous Network Cartography

Celeste Matarazzo (13-SI-004)

Abstract

Network technologies are changing at an extraordinarily fast pace and necessitate new mapping, or cartography, tools to maintain effective large-scale, security-focused network mapping in this challenging environment. Today, most organizations do not map their networks. They do not have up-to-date knowledge of the actual components and structure of the cyber domain they work in. The available tools are aimed at periodic compliance snapshots of a network. They are slow, intrusive on network operations, provide static views of the network, require the network to be operated in a reduced security posture, and do not handle the required mission scale. We propose to build continuous network cartography capabilities and analytics that apply machine-learning and statistical methods for understanding network activities.

This project also focuses on mapping and inferring hidden or obfuscated network components. These two focus areas directly address the gaps and limitations of today's network-mapping technologies and seek to provide analysts with a view of an activity or behavior, enhancing an analyst's ability to make timely decisions and effectively change the outcome of a cyber situation. This effort will leverage the Laboratory's existing Mapper framework to provide a structure for integrating prototype capabilities and testing in actual operational network environments.

We expect to establish the foundations of future infrastructures of network situational awareness by building on current Laboratory network-mapping expertise and capabilities. The technical approach will include a high-performance active and passive data-collection capability that is scalable to many months worth of mapping results for an enterprise network. In addition, it will include the application of machine-learning and statistical methods for processing continuous maps for understanding of network activities. Our endeavors will span two broad areas—continuous network mapping and analytics—for the continuous discovery of network components and activities, as well as techniques for mapping and situational awareness that are robust to noncooperative, complex, or adversarial intrusion, denial, or deception tactics.

Mission Relevance

This project is strategically aligned with the strategic focus area in cyber security, space, and intelligence—specifically, cyber situational awareness. It will enable LLNL to develop more sophisticated, robust techniques for national security and homeland security applications, such as critical infrastructure cyber programs and continuous network monitoring.

FY14 Accomplishments and Results

In FY14 we successfully accomplished all planned activities and conducted continuous mapping data collection and analysis throughout the year. Specifically, we (1) developed and evaluated metrics to determine continuous mapping frequency for prioritized mapping; (2) created our integrated dTrend change-detection framework and evaluated it with real data, as well as presented the dTrend output in an interactive multi-timescale visualization (shown in the figure here); (3) evaluated change-detection performance versus collected data on the mapping cluster; (4) developed an interactive visualization tool as a step toward incorporating human analysts in the loop; (5) implemented mission-identification algorithms and collected baseline mission data from multiple sources; (6) developed and applied role-based analysis to identify functions that are useful for assessing importance of hosts to a given mission; (7) evaluated the state of the art for mobile detection and collected data sources and analyzed them for indicators; (8) created an interactive continuous mapping interface for controlling mapping setup and execution; and (9) developed a network resilience taxonomy, and given the large scale of data collected for the year, measured performance and scalability of data-management infrastructure with real data.

Proposed Work for FY15

In FY15 we will (1) expand our efforts to complex and noncooperating networks where access may be limited, configuration knowledge is limited, and data and structure may be intentionally obfuscated or spoofed; (2) develop algorithms to characterize networks behind network address translation or firewall devices, and detection capabilities for virtual enclaves and overlay networks; (3) identify and develop network probes that are capable of automating the collection of a distributed array of data points that fuse together, to enrich our understanding of these networks; and (4) expand upon our mobile device and mission-mapping capabilities to demonstrate the ability to work with dynamic networks and infer the activities and actions that are being conducted.

Publications and Presentations

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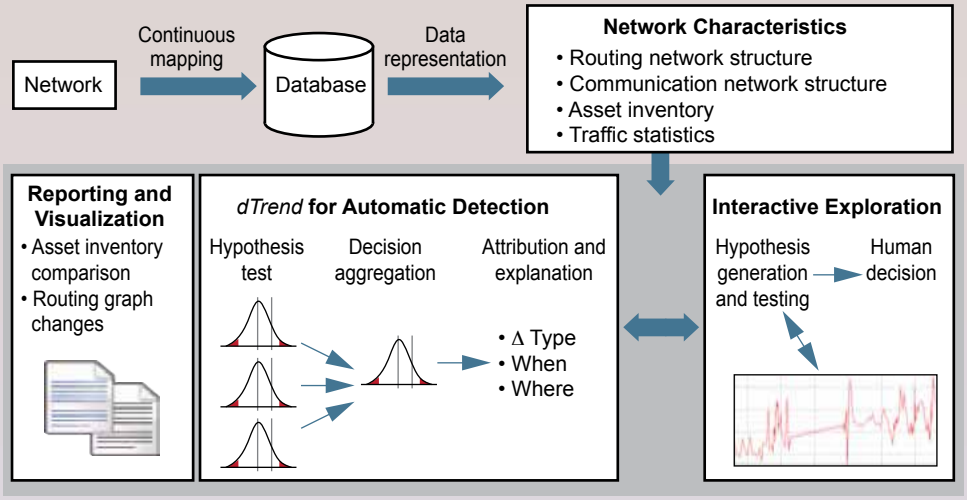
CNC demonstrated end-to-end change detection capability on a computer network



Change detection is a complex task that exercises all aspects of the mapping analytics pipeline. Continuous mapping data is required to observe changes over time. Important events potentially involve changes in many network characteristics, from network structure to traffic statistics. Continuous network cartography (CNC) takes several approaches to change detection. When we know the changes we are looking for, such as asset inventory differences, we show changes to a user as a report. When we don't know what has changed in the network, we explore two approaches: human-driven interactive visualization and machine-learning-based automated change detection and explanation.

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Radio-Frequency Noise in Superconducting Devices

Sergey Pereverzev (13-ERD-016)

Abstract

Detection of coherent neutrino scattering using weak neutral current interactions of subatomic particles has been predicted for many decades, but has never been realized despite extended efforts. We propose to develop an ultrasensitive detector to enable phonon detection produced by the low nuclear recoil from coherent neutrino scattering that will enable conclusive proof of this fundamental effect. If successful, this detector will find applications in infrared single-photon spectrometry for clandestine reactor detection as well as biosecurity and medical use, such as cell biochemical imaging or trace biochemical impurities detection. The detector technology will be based on a

superconducting sensor mounted on a large interaction crystal with a superconducting quantum interference device (SQUID) used to measure extremely subtle magnetic fields. The superconducting detector arrays we develop will not only improve neutron detection sensitivity by two orders of magnitude, but may also provide directional information for the neutrino, which is of scientific and programmatic significance.

We expect to fabricate superconducting phonon sensors and SQUID detectors and demonstrate single-event detection sensitivities of about 30 meV for single-pixel devices, two orders of magnitude higher than previously possible. The method we propose to develop uses a superconducting film on the surface of a crystal to absorb phonons that are created when the antineutrino or neutrino transfers small momentum to the atom in the crystal. The lower transition-temperature superconductor acts as a calorimeter to provide an electrical readout proportional to the total energy deposited by the absorbed phonons, and hence the energy deposited by the neutrino. We will also fabricate superconducting sensor arrays to demonstrate position and directional resolution for energies corresponding to typical neutrino recoil energies.

Mission Relevance

The phonon-sensitive detector proposed in this research will enable improved particle detection sensitivity as well as directional neutrino measurements. This will enhance capabilities to locate unknown nuclear reactors from a neutrino signature that cannot be shielded and will enable reactor tomography, in support of the Laboratory's central missions in national security and reduction of nuclear threats, as well as the strategic focus area in cyber security, space, and intelligence.

FY14 Accomplishments and Results

In FY14 we (1) developed technology to convey high voltages up to 100 kV into a dilution refrigerator at 15 mK, and filed an associated record of invention on compact and reconfigurable systems for high-voltage distribution in cryogenic and vacuum applications; (2) designed a 100-kV high-voltage cell to apply strong electric fields to the surfaces of superconducting samples with liquid helium as the insulator, and tested critical elements of the design at 4 K for leaks and high-voltage stability; (3) discovered several new mechanisms for noise and decoherence in superconducting devices associated with nuclear magnetic moments and localized electron magnetic moments, and proposed approaches to measure and suppress them; (4) filed a record of invention on isotopically pure materials for quantum computing and ultralow noise cryogenic devices; and (5) identified a magnetometer measurement on a stack of thin sapphire wafers to test recent theoretical predictions at LLNL about free-electron magnetic moments on sapphire surfaces—this technique can be applied to a large class of surfaces and interfaces relevant for quantum computing applications.

Project Summary

After consultation with Laboratory and outside experts, we identified radio-frequency noise in superconducting detectors as a technical hurdle that should be addressed prior

to applying these detectors to neutrino scattering detection as originally intended. As a result, we altered the scope of our project to study these fundamental noise mechanisms at radio-frequencies in superconducting devices in high-electric fields at ultralow temperatures. We obtained a 15-mK base temperature with our dilution refrigerator, developed technology to achieve up to 100 kV for an experimental setup at 15 mK, and analyzed the shielding and filtering of our refrigerator, which led to us rebuilding the system for quantum measurements. We developed an unique research program on noise mechanisms, with the potential to carve out a unique niche for LLNL in quantum computing and cryogenic sensor development, filing two records of invention in the process (*Isotopically Pure Materials for Ultra-Low Noise Cryogenic Devices and Q-Bits* and *High Voltage Distribution System for Cryogenic and Vacuum Applications*). We explored the possibility of fabricating resonators and SQUIDs out of single-crystals composed of isotopically pure zero-nuclear-spin materials to avoid nuclear spins in materials and localized electron spins on inter-grain boundaries in metal. This will also eliminate two-level systems associated with atoms jumping on near-defect sites. We envision low-temperature magnetometry as a method to study electron spins on surfaces and interfaces and their dependence on surface passivation and a high-electric field on the surface to minimize the number of surface electron spins and their effect on noise at low temperatures. Quantum computing, detector development, and material science would all benefit from the proposed improved SQUIDs. A future goal would be to identify and ultimately remove the microscopic origins of athermal noise, which would improve both quantum bit coherence time and SQUID performance.

Scalable, Revealing Factorizations of Directed Graphs and Hypergraphs

Van Henson (13-ERD-072)

Abstract

There has been an explosion of interest and activity in recent years in data mining and graph analysis for large relational data sets, commonly modeled with extremely large, scale-free graphs and "hypergraphs," where entities (or properties) of one type are connected by an edge to several entities of differing type. These graphs are used to show hierarchies that may represent computer networks, communications, the Internet, social networks, power grids, and a host of other applications. The principal challenges arise in the sheer size of the matrices because the graphs can become exceedingly large. Until recently, these sizes have posed extremely difficult computational challenges because of problems of both computational and algorithmic scalability, which has restricted analysts to using very simple approximations, precluding the discovery of deeply significant but subtle relationships. In this project, we will develop scalable multilevel factorizations for the deep analysis of data sets represented by nonsymmetric square and rectangular matrices, and new and effective affinity measures for partitioning, clustering, community

identification, and topological analysis of data. These new methods build on our prior work in scalable eigensolvers and multilevel methods to enable the effective deep analysis of complex data relationships.

We expect to deliver both theoretical advances and practical algorithms and codes that will enable the analysis of diverse relational data at a scale not previously possible. Our new tools will enable us to break the data sets into fundamental components that can be analyzed, filtered, and synthesized to highlight relevant connections. Moreover, the building blocks created in this project will be applicable to a range of problems and applications of interest to graph theorists, data miners, bioinformatics researchers, and numerical analysts working on extremely large data sets.

Mission Relevance

This project is closely aligned to the Laboratory's cyber security, space, and intelligence strategic focus area. Data mining and analysis using large-scale graphs and hypergraphs are fundamental to analysis of Internet traffic, network intrusions, financial networks, and flow of funding; email message traffic within and across communities; the power grid; information propagation; and information retrieval, to name a few. This research can be important to all these application areas and is also relevant to the core competency in information systems and data science.

FY14 Accomplishments and Results

In FY14 we (1) implemented serial codes to factor graphs and hypergraphs for clustering, community identification, and seed-set expansion; (2) implemented and tested several graph and hypergraph generators meeting diverse constraints, including degree and triangle distribution, correlation between compute nodes, and diverse communities; (3) developed a framework for extending factorization and generation techniques to directed graphs; (4) devised, implemented, and tested a novel affinity measure for ranking "closeness" among vertices; (5) implemented and tested an efficient multilevel hypergraph factorization technique that successfully performs overlapping co-clustering of feature matrices; and (6) implemented efficient symmetric linear solutions on difficult topologies.

Proposed Work for FY15

In FY15 we will (1) implement and test efficiency for parallel versions of our most promising factorization techniques for both directed graphs and hypergraphs; (2) improve the efficiency of our graph generators and continually improve their abilities to model diverse sets of graph classes; (3) extensively test and improve our multilevel factorization techniques for directed graphs and hypergraphs; (4) increase robustness for our linear solver technologies, including incorporating nonsymmetric systems; and (5) provide our resulting codes and algorithms for development and implementation into production codes by our Laboratory collaborators.

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Cooperative Constellations: Resilient, Persistent, and Flexible Satellite Systems

Michael Pivovaroff (14-SI-005)

Abstract

Space remains vital to our national security, but the evolving strategic environment increasingly challenges U.S. space advantages. The nation's space capabilities allow our military to see with clarity, communicate with certainty, navigate with accuracy, and operate with assurance. We propose to develop the concepts, technologies, and models to demonstrate the ability to use cooperative constellations of small satellites to perform space missions important for U.S. national security. We will utilize and improve existing physics-based modeling tools and preliminary technologies already at LLNL. We intend to operate two Pathfinder satellites launched in FY14, develop active and passive techniques to control milk-carton-sized satellites (nanosatellites) and learn how to fly and maintain formations of these low-mass satellites, and study optimal constellation configurations and develop enabling technologies for a wide variety of missions.

The U.S. is reliant on the advantages afforded by an active space program, but internal and external forces threaten the nation's ability to continue operations in this once-benign environment. We expect to apply the Laboratory's significant modeling and simulation capabilities and its nascent efforts in designing and building individual miniaturized satellites to develop the key and enabling technologies needed to demonstrate that cooperative constellations of small satellites can meet high-priority missions. We will validate our ability to accurately model instrument performance, create new imaging and sensing concepts, and show how we can synthesize data from multiple satellites.

Mission Relevance

Our research into the deployment and demonstration of nanosatellites for satellite collision-avoidance strategies and investigation of their utility for high-priority space security and fundamental science missions is directly aligned to the Laboratory's strategic focus area in cyber security, space, and intelligence. Our development of

miniaturized satellite constellations with improved sensing and imaging technologies is also relevant to LLNL's strategic focus area in energy and climate, through measurement of pollutants in the upper atmosphere.

FY14 Accomplishments and Results

In FY14 we (1) supported launch of a second Pathfinder nanosatellite—the satellite malfunctioned, and we used ground-based observations (shown in image) to assist in anomaly resolution; (2) implemented a six-degree-of-freedom propagator on a high-performance computing platform; (3) developed a ground-based test bed for optical tracking of satellites; (4) created new novel satellite structures via three-dimensional printing; (5) developed and optimized large-aperture monolithic telescope designs; and (6) formed an academic partnership with researchers at the Rochester Institute of Technology in New York for advanced image simulation and an academic partnership with researchers at the Naval Postgraduate School in Monterey, California, for environmental testing of miniaturized satellites.

Proposed Work for FY15

In FY15 we will (1) use nanosatellites to validate and improve physics-based models of sensors and satellite components and to conduct experiments to better understand the low-earth-orbit environment; (2) create and test models to control small satellites using active and passive techniques; (3) develop algorithms for synoptic and multistatic imaging missions; and (4) begin to explore, in partnership with researchers at the National Aeronautics and Space Administration's Goddard Space Flight Center, how best to use nanosatellites to perform earth-observing science missions such as measuring pollutants in the upper atmosphere and monitoring water and ice concentrations in clouds. This effort will include modeling and simulation studies, as well as fabrication and testing of prototype hardware.

Lawrence Livermore's ground-based observing capability for characterizing orbits of miniaturized satellites.

Publications and Presentations

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Improved Sensor Performance Using Innovative Algorithms

Milton Smith (14-ERD-039)

Abstract

Algorithms are needed that improve performance and extend capabilities of spectral laser sensors to detect scarce materials or gases for a wide variety of intelligence, surveillance, and reconnaissance applications. There exists a significant need for algorithms that improve performance of remote sensors already in service, as well as for next-generation sensor systems currently under development. The information-processing chain for sensors is assumed independent of the hardware, having relative minor impact on system performance. It is generally assumed that differences in performance are mostly because of hardware differences between systems and that the processing chain is generalized to yield optimal performance to all systems for all conditions. We propose to select two algorithm domains important to existing sensor systems that address two performance issues, namely coverage rates for active systems and structured noise in mercury–cadmium–telluride focal-plane imaging arrays. Changing the operation and processing paradigm of these two problems can realize a minimum of tenfold performance improvement for each sensor type, significantly increasing the utility of these sensors. These solutions will be experimentally validated in the laboratory and by using archived data sets. Algorithms will be integrated into operational systems and validated for performance.

We expect to enable a tenfold improvement in active sensor coverage rates and detection and identification performance, with an equivalent reduction in false alarms. These results will extend the range of useful applications for both active and passive sensors. Success will significantly aid in an operational sensor applied to nonproliferation objectives for remote detection and detection of solids. The benefits include increased sensitivity, reduction of false alarms, and improved cost benefit from higher coverage rates, all of which directly impact the end-to-end performance. These performance enhancements will lead to improvements in capabilities benefiting national security missions and will help maintain Livermore's role in sensor technology to the intelligence community.

Mission Relevance

Improvements in sensor performance are central to the Laboratory's mission focus area in cyber security, space, and intelligence. In addition, this research has applications in a wide range of national security missions.

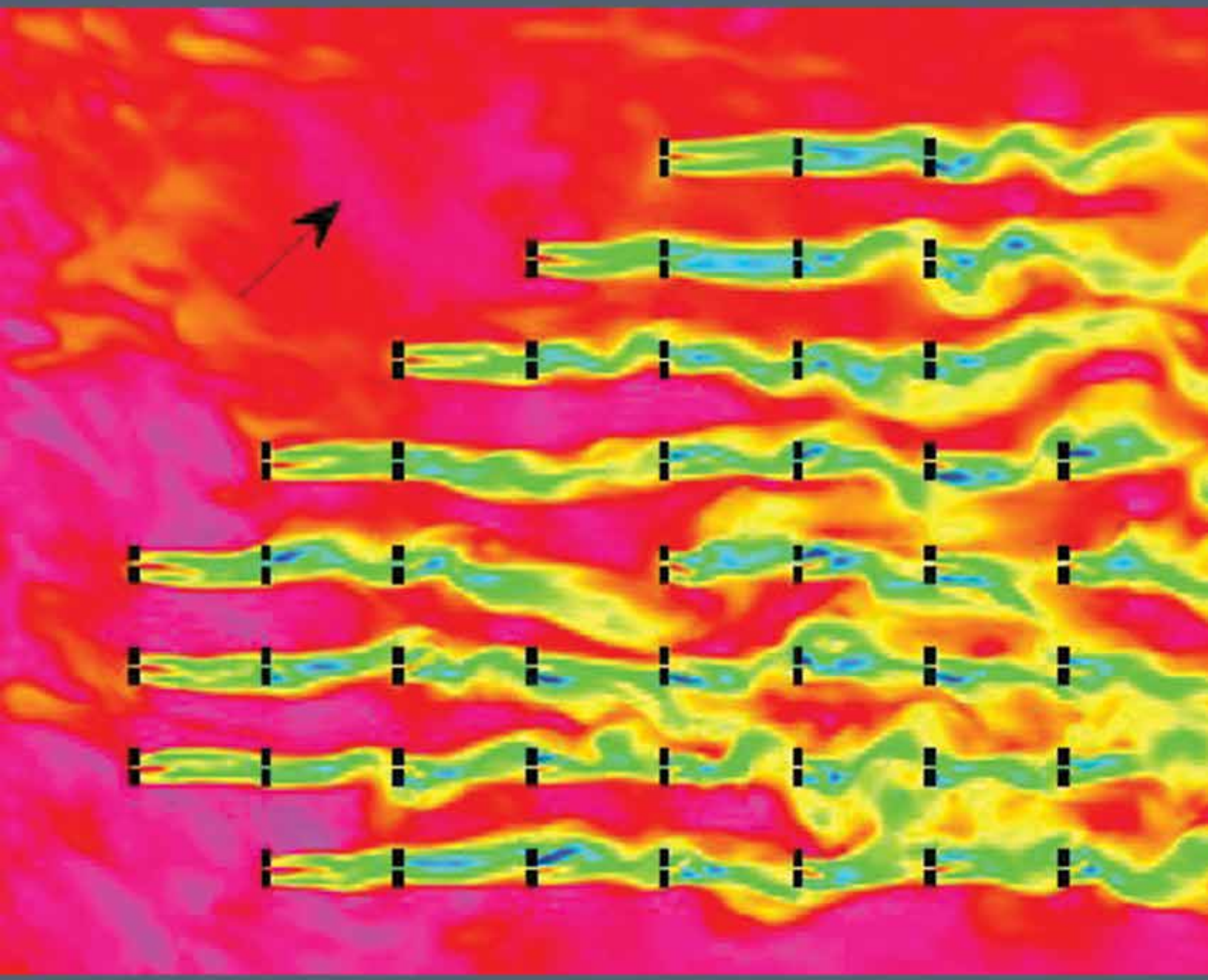
FY14 Accomplishments and Results

In FY14 we (1) implemented a near-real-time operational bad-pixel replacement algorithm to mitigate scene-dependent structured noise for sensors, (2) tested the symmetric monotonic regression algorithm using globally derived data sets from several sensors and various collections, (3) defined a new algorithm that uses sensor data to define the background subspace rather than using models and spectral library measurements, (4) validated a threefold to fivefold scene improvement in noise-equivalent spectral radiance for Livermore long-wave infrared sensors using the symmetric monotonic regression algorithm, (5) modeled a strategy demonstrating that a small number of spectral measurements (3–4 bands) can be strategically applied to improve coverage rate up to sixfold, (6) demonstrated a minimized number of wavelength measurements to include a small suite of targets, and (7) discovered optimizing target spectra in combination with wavelengths significantly improved performance.

Proposed Work for FY15

In FY15 we will (1) validate coverage-rate optimization algorithms using an active ground sensor, (2) formulate analytical expressions for real-time coverage-rate optimization of our sensor, (3) test and validate the coverage-rate algorithm with an operational sensor, (4) validate our bad-pixel replacement algorithm based on sensor-defined subspace, (5) create synthetic scenes and dynamic tests that cause structured noise in mercury–cadmium–telluride focal-plane arrays, (6) quantify the bad-pixel algorithm's performance using defined metrics on synthetic scenes, and (7) test applicability of bad-pixel replacement on mercury–cadmium–telluride focal plane arrays in the visible, near-infrared, and short-wave infrared regions of the electromagnetic spectrum.

Energy and Climate



FY2014

Lawrence Livermore National Laboratory

Creating Optimal Fracture Networks for Energy Extraction

Frederick Ryerson (11-SI-006)

Abstract

The technology required to extract the energy resources contained within the Earth`s crust is constantly evolving. Resources that were once considered unconventional become conventional through a combination of economic factors and improved technology. The key to developing unconventional subsurface energy resources is the creation of fracture permeability, which provides access for extracting hydrocarbons from tight formations and enhances the circulation of water for the more effective use of hydrothermal energy resources. However, our inability to predict the development of fracture networks and their performance limits our ability, for example, to develop resources such as geothermal resources at depths greater than 3 km. The amount of clean, carbon-free energy in such enhanced geothermal systems is virtually unlimited. We propose to develop the computational and observational capabilities needed to unlock these resources.

A major deliverable of this project will be a computational hydraulic fracturing simulation capability, GEOS, that allows the design of subsurface fracture networks in a variety of geologic settings to support the extraction of deep geothermal energy and natural gas from shales. This capability will describe the optimal fracture network, how to create this network, how to determine what has been created, and how this perturbed system evolves over time. This high-fidelity code will describe both hydraulic and explosive fracturing in the subsurface, and it will be linked to a wave propagation code for predicting seismicity associated with the hydraulic fracturing process.

Mission Relevance

Our research will help promote the development of unconventional energy resources such as carbon-free enhanced geothermal systems, which supports the Laboratory mission of energy security.

FY14 Accomplishments and Results

In FY14 we (1) completed the implementation of implicit solvers within the GEOS code to allow more computationally efficient simulations to be performed at time and length scales relevant to underground oil, gas, and geothermal reservoirs; (2) completed the addition of thermal diffusion and one-phase Darcy flow, which describes the flow of a fluid through a porous medium, to better simulate reservoir productivity; (3) completed the addition of a seismic wave propagation capability, which allows the construction of synthetic seismograms based on source mechanisms associated with induced fracture determined by the core GEOS capability and the overlying geology; and (4) developed methods for simulating surface deformation associated with reservoir stimulation.

Project Summary

The successful conclusion of this project resulted in the development of a three-dimensional multiple-scale, multiple-physics geomechanics code framework (GEOS), for simulation of hydraulic stimulation of unconventional oil and gas reservoirs and enhanced geothermal reservoirs. The code links fluid injection with bulk matrix response, fracture propagation and growth, seismic response, and enhancement of permeability and stimulated volume. Designed for a high-performance computing environment, the code framework supplies data handling and communication facilitating the addition of new physics solvers, such as those for thermal diffusion, Darcy flow, and seismic wave propagtion that have recently been added to the code. The GEOS code has also been applied to analysis of seismic risk associated with fluid injection, near-well-bore geomechanics, and geologic storage of carbon dioxide. Applications of the GEOS code will be supported by DOE's National Risk Assessment Partnership, and has resulted in four external partnerships, which consider various geomechanical issues associated with unconventional oil and gas reservoirs.

Publications and Presentations

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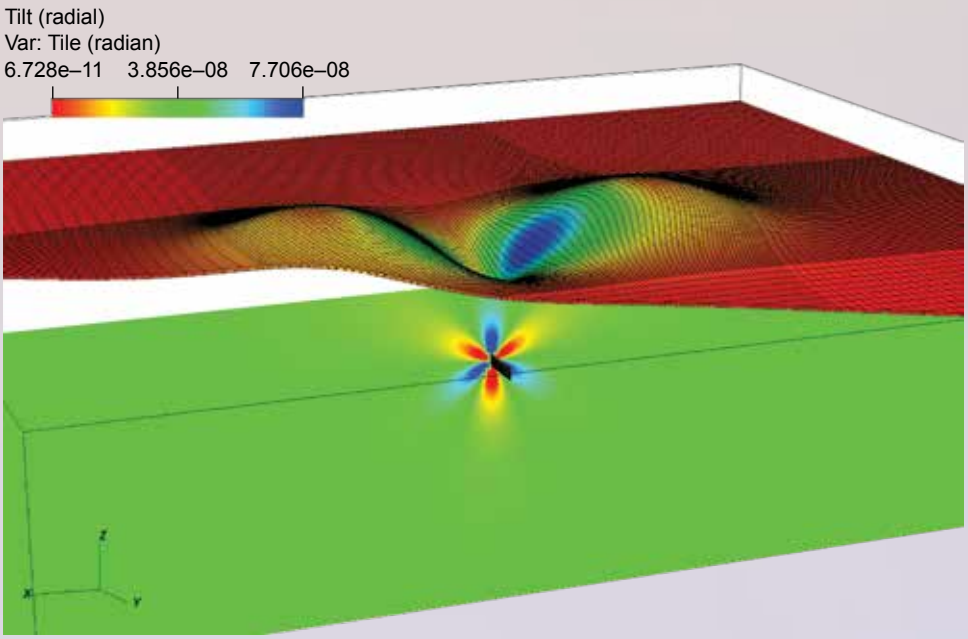

A GEOS hydraulic fracturing simulation capability of surface deformation (tilt) as a result of opening a 1,000- by 100-m crack caused by a net pressure increase of 1 MPa. Deformation is likely too small for detection by interferometric synthetic aperture radar used for measuring Earth's surface deformation, but should be observed in surface tiltmeter arrays.

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Large-Scale Energy System Models: Optimization Under Uncertainty

Thomas Edmunds (11-ERD-076)

Abstract

We propose to develop new models and algorithms tailored to high-performance computing platforms that address the challenges of optimizing large-scale energy systems under conditions of uncertainty. This work is motivated by the increasing complexity of operating the country's power grid, with large contributions from intermittent wind and solar resources. Planning and managing the grid requires solving large-scale, nonlinear optimization problems under uncertainty. Given the $360 billion the U.S. spends each year on electricity and the $800 billion capital investment, a small improvement in efficiency would significantly contribute to energy security and competitiveness. Working with our academic collaborators, we plan to scale up existing codes and develop new algorithms to address these challenges. We will build scalable grid models; apply uncertainty quantification methods to characterize the sensitivity of grid models with respect to input uncertainty and, if possible, reduce their influence; and develop and implement stochastic optimization methods for large-scale systems.

We expect to address energy system design and operations problems that impact critical issues identified by the power industry. Our primary goal is to develop large-scale optimization tools that enable better long-term, day-ahead, and real-time decisions for building and operating the electrical power system. Our research products will include optimization algorithms, code, and studies that show how to build electric power systems that accommodate large contributions from intermittent wind and solar generation. We will deploy these new optimization tools on Livermore's high-performance computing systems to provide solutions to large-scale energy benchmark problems for use by the academic community and industry and for determining the level of detail necessary for the energy grid model to accurately make long-term planning decisions.

Mission Relevance

The effort draws on Laboratory expertise and unique capabilities in developing complex simulation tools and uncertainty quantification. It leverages high-performance computing resources to support the Laboratory's mission in energy security.

FY14 Accomplishments and Results

In FY14 we (1) used statistical clustering techniques to identify a minimal set of forecast trajectories that capture the weather uncertainty—this parsimonious representation of uncertainty is necessary to ease the computational burden imposed

on the stochastic optimization algorithm used in later stages of the overall simulation of grid operations, (2) conducted joint research with the IBM Corporation to improve performance of their stochastic optimization algorithms for solving power grid optimization problems in the presence of high contributions from intermittent wind and solar generation, (3) demonstrated a factor of 20 increase in speed using LLNL's high-performance computing platforms, and (4) conducted simulation experiments to demonstrate the value of an LLNL patent for distributed control systems that can improve the stability of the power grid when shocked by component failures.

Project Summary

Our research developed new capabilities for stochastic modeling of the weather and large-scale optimization of a power grid under uncertainty. Our ensemble representation of renewable generation uncertainty for each day of the simulated year provides an improvement over current practice of sampling simple seasonal statistical models of wind speed and cloud cover, and our statistical research demonstrates how to condense thousands of possible weather trajectories into a representative set of 20 to 30 that sufficiently capture uncertainty in the system. Our collaborative work with the University of California, Berkeley and IBM Corporation resulted in stochastic optimization algorithms that determine optimal operation of the power grid given the uncertainty in renewable generation. Working with software vendor Energy Exemplar, we demonstrated solutions to problems of interest to the California Independent System Operator organization, which is struggling with mandates to increase the amount of renewable generation in the state. Coordination of renewables, conventional generation, energy storage, demand response, and geothermal generation can be improved using the techniques and codes we developed. In collaboration with our academic and industrial partners, we plan to use the techniques and codes developed to support the DOE's grid modernization efforts and California's efforts to reduce emissions while maintaining a cost-effective and reliable grid.

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A New Approach for Reducing Uncertainty in Biospheric Carbon Dioxide Flux

Sonia Wharton (12-ERD-043)

Abstract

Reducing uncertainty in biospheric carbon dioxide (CO_2) flux exchange at the site level is a critical first step for reducing uncertainty in global and regional terrestrial carbon sink and source estimates, which are essential components of climate research and modeling. We propose to develop a novel tool for reducing CO_2 flux uncertainty by integrating state-of-the-art ecosystem flux towers, soil respiration chambers, and boundary-layer observations with a multilayer ecosystem and atmosphere flux model, the Advanced Canopy Atmosphere Soil Algorithm (ACASA). We will collaborate with the University of California, Davis, and apply expertise in boundary-layer meteorology, the terrestrial carbon cycle, and land, surface, and atmospheric modeling to develop

a better understanding and quantification of the natural, background CO_2 exchange between vegetated land surface and the atmosphere.

By coupling research-grade instrumentation that measures the atmospheric profile with high spatial and temporal resolution with a sophisticated flux model that uses these data to simulate turbulent processes that otherwise are difficult to parameterize, we expect to demonstrate an improved technique for filling the gap of CO_2 flux tower data that will reduce the uncertainty of annual CO_2 source or sink estimates. We will accomplish this by developing an input-forced version of ACASA with high-resolution observations of boundary-layer profiles and soil fluxes. We will fully validate ACASA at two flux sites: Lawrence Livermore's Site 300 and the Wind River AmeriFlux tower in the Washington Cascades. Successful validation will produce a technique that can model CO_2 flux tower data with higher certainty than is currently available. In addition, this validated set of modeling and measurements tool can then be used to simulate and verify terrestrial CO_2 emissions.

Mission Relevance

Greenhouse gas emissions monitoring and verification on a national scale, such as proposed in Livermore's strategic investment plan, will require a better understanding and quantification of the natural CO_2 exchange between the vegetated land surface and the atmosphere. This project supports a central Livermore mission in energy and climate by developing strong research sites for boundary-layer CO_2 research and producing a modeling tool at 1-km resolution for biospheric CO_2 flux verification.

FY14 Accomplishments and Results

In FY14 we (1) completed the ACASA simulations for all sites, including running four newly modified versions of the model to handle biomass and hydrological seasonality; (2) completed the validation of all ACASA simulations against site-measured climatological and eddy covariance data; (3) completed analysis of above-canopy and subcanopy boundary layer flows and their impact on eddy covariance fluxes for all sites; (4) conducted quality assurance on Diablo and Wind River flux data and submitted the data to the AmeriFlux database; and (5) completed site-calibration campaigns at Wind River and Diablo with the Lawrence Berkeley National Laboratory's AmeriFlux Management Team.

Project Summary

The successful conclusion of this project resulted in advancing the ACASA model and in providing a better understanding of atmospheric drivers of nighttime fluxes at three AmeriFlux sites. Modifications to ACASA resulted in a more adaptable code that can now be used for modeling more diverse ecosystems, including those with major drought stress. In addition, we performed an in-depth verification and validation of the code. We determined which site conditions within the AmeriFlux network would benefit from having additional instrumentation (e.g., light detection and ranging at the flux tower).

Additional instrumentation at these sites will reduce the errors associated with measuring carbon fluxes, and using these instruments in combination with ACASA demonstrates a capability for increased accuracy in measuring the terrestrial carbon cycle and for reducing uncertainty in biospheric CO_2 fluxes. It also provides a tool for independently verifying biospheric CO_2 emissions. We have established promising ties with academia, national laboratories, AmeriFlux, and managers from the DOE's Office of Biological and Environmental Research to pursue support for continuing this research.

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Forecasting and Uncertainty Quantification of Power from Intermittent Renewable Energy Sources

Wayne Miller (12-ERD-069)

Abstract

We propose to address the scientific and technical challenge of the inherent intermittency, or variability, of wind in large-scale, integrated, renewable-energy systems. This challenge is a critical limitation to optimizing wind energy systems and their efficient integration with the energy grid. Our focus will be to understand the problem as a coupled transient system of intermittent natural energy resources and energy transduction with uncertainty quantification of the resulting power forecasts. Uncertainty quantification will be applied to evaluate sensitivities, errors, and uncertainties in real-time and short-term energy forecasts made from these renewable resources. The outcome will facilitate and inform the task of integrating high percentages of intermittent renewable energy sources into the grid. Forecast results will be verified by field data of meteorological measurements and actual wind-farm power data.

We expect to address the scientific and practical challenges of enhancing energy security by large-scale integration of intermittent renewable energy sources. The end result will be quantified error bounds on forecasts at all relevant timescales as well as sensitivity studies for the contributing geophysics parameters forcing the intermittency. Reducing forecast uncertainty for intermittent renewable power will have an immediate and tangible benefit for expanding renewable energy in the $300 billion U.S. power market, as well as for efficient utility management. The work will build upon the existing capabilities in wind resource characterization at LLNL. Our analysis will add to this by capturing the significant natural resource physics driving intermittency and the

transduction of natural energy into line power. Deliverables will be the best-practice process for performing validated forecasts and the suite of tools needed to perform these forecasts.

Mission Relevance

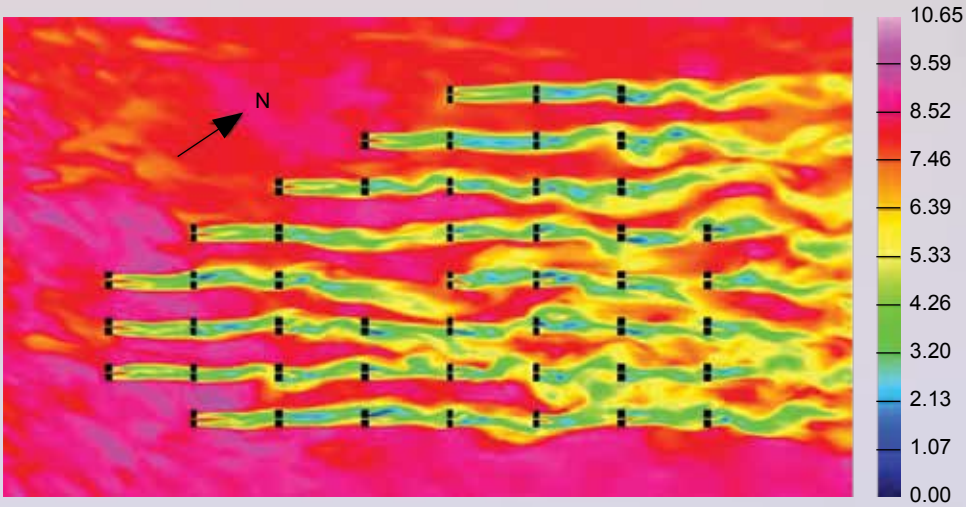
The challenge of power intermittency and predictability at large scales is recognized as a topic of national importance with significant technical obstacles. This topic is also in line with the LLNL strategic plan relating to energy security, climate change, and our research activities in renewable energy and utility power analysis.

FY14 Accomplishments and Results

In FY14 we (1) completed a multiphysics, multianalysis ensemble simulation for numerous wind ramp case studies for both flat and complex terrain; (2) verified ensemble forecasting results via statistical analysis with on-site observations; (3) completed an extended field campaign to characterize inflow wind characteristics approaching non-waked turbines with high accuracy across the rotor disk; (4) validated agreement of measurements taken from light detection and ranging sensors, a meteorological tower, and the housing of a wind turbine once instrumental errors were identified and corrected; (5) trained three statistical models and demonstrated that predicted power uncertainty provides a substantial improvement in wind forecasting results; (6) improved and added model input variables that are critical predictors of power output; (7) implemented and validated a mesoscale and microscale coupling framework to simulate multiple-turbine wind plants in realistic operating conditions; and (8) simulated wind farm power-generation fluctuations resulting from downstream wake interference in both flat and complex terrain.

Project Summary

Successful completion of the project has led to practical advances that allow the wind industry to forecast wind power with greater accuracy in the critical day-ahead window used for energy markets. For this project, we focused on four areas: ensemble forecasting,



Wind turbine wakes in turbulent atmospheric flow predicted by a new multiscale simulation tool using the community Weather Research and Forecasting Model for atmospheric winds and turbulence, combined with the Livermore code CGWIND for local wind farm and turbine modeling. Both atmospheric-scale and turbine-scale resolution are needed for accurate wind power forecasts.

complex terrain, multiscale modeling, and the dynamic power curve. In the area of ensemble forecasting, wind forecasts that use ensemble methods develop statistically relevant forecast accuracy. Thus, we developed advanced methods to sample the ensembles and to analyze the results, with an emphasis on wind ramp prediction. The presence of variable terrain, such as hills, create complexities in the wind flows that are difficult to predict. As a result, we developed unique observational and modeling techniques to elucidate the effects of terrain on wind power. Our multiscale modeling efforts included developing simulation techniques that allow mesoscale weather models to be downscaled to the turbine. These models span 10 orders of magnitude in scale, which is necessary to assess the winds at the turbine scale when they are driven at the planetary scale. Finally, we developed and validated a new statistical method to predict wind turbine power output for a given wind inflow. This improved model leads to an approximately 10% better prediction of power production, which is a highly significant improvement. We are receiving interest in our research outcomes from the wind industry, and anticipate that the DOE Wind Program will fund the dynamic power-curve modeling we developed as part of this project so it can be extended from one turbine to entire wind farms.

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Reactive Materials for Hydraulic Fracturing

Roger Aines (13-ERD-029)

Abstract

Water use in hydraulic fracturing for natural gas production is strongly affected by the need to drive fracture materials, known as proppants, which are typically rounded sand grains, into the created fracture to hold it open during gas production. The water must be viscous to move the dense sand particles, which results in a complex mixture of thickeners and friction reducers that makes it difficult to treat and reuse the water. If these agents were unnecessary, the fluid would be much easier to treat. However, removal of these thickeners would require that the proppant have a density similar to the water used to place it. We propose to address this need by creating neutral-density proppants composed of a reactive material encapsulated in a silicone shell, which reacts within the fracture to become very strong and expansive.

We expect our research will enable the Laboratory to build a strong presence in gas production and fracturing-based science by addressing the necessary second step in fracturing: keeping the fracture open once it has been optimally created. We will develop the materials science and engineering to allow transport and reaction under specific conditions, which could be applied in many other fields. We will also provide experimental support for the necessary engineering and theoretical science, allowing us to demonstrate the applicability of our new fracture material while developing a strong base of new knowledge about fracture flow of particulates and proppants. Our success will improve both the efficiency and environmental impact of natural gas production while developing a basic science understanding of proppant physics.

Mission Relevance

This work addresses a core LLNL strategic focus area in energy and climate with applications to natural gas production and geothermal energy production. We will collaborate with the National Energy Technology Laboratory in West Virginia, the dedicated fossil-energy laboratory, which will conduct the imaging of proppant placement in their x-ray tomography facilities. Developing proppant materials also aligns well with the Laboratory's core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

In FY14 we (1) created the first temperature-set proppants, which have a liquid core inside a polymer shell—during transport they are malleable and of neutral density, ensuring deep placement, and then set to solids when exposed to greater than 70°C; (2) obtained x-ray images of these proppants inside laboratory samples of Marcellus shale from the Appalachian Basin using the x-ray tomography system at the National Energy Technology Laboratory, as well as optical images using printed, transparent versions of the same fractures; and (3) demonstrated tracer units that move with the proppant pack and are identical in size, but upon heating release a tracer to provide information on pack placement.

Our new high-throughput system produces proppant fracture material capsules that are about 500 μm in diameter.

Proposed Work for FY15

In FY15 we will (1) create mineral filling for the proppant capsules that not only sets, but slowly expands upon curing; (2) complete the transport model and test it using x-ray facilities at the National Energy Technology Laboratory, and examine the effect of asperities and changes in fracture aperture on transport; (3) optimize the proppant design to maximize transport and strength with good permeability after the proppant hardens; (4) demonstrate scale-up of production to enable large-scale use of these materials; and (5) continue to work with partners in oil companies and service companies to maximize the utility of these proppants, and to license and utilize them in shale gas operations.

Selecting Better Models for Climate Change Detection and Attribution

Benjamin Santer (13-ERD-032)

Abstract

We intend to develop a rigorous, scientifically credible strategy for selecting models for use in climate change detection and attribution. This requires estimates of climate response to external forcing agents such as atmospheric trace gases, and climate "noise" for each model, both of which will be derived from archives containing output from two to three dozen models. Despite the importance of an accurate picture of noise, little work has been done to investigate the ability of different models to simulate observed spatial and temporal variability. We will therefore investigate the sensitivity of detection

and attribution results to model capability. To this end, we will develop novel measures of model performance, with an emphasis on metrics for spatial and temporal variability, feature identification and tracking, and multiscale analysis. This work will leverage the Laboratory's proven capabilities in climate change detection and attribution and in developing and applying climate model performance metrics.

We will develop a suite of metrics to evaluate spatial and temporal variability at different scales and use these metrics to evaluate the performance of the climate code CMIP-5 (Coupled Model Intercomparison Project, Phase 5). We will also use these measures to evaluate the sensitivity of existing detection and attribution results to model quality, generate fingerprints and leading noise modes as simulated by better models, develop codes for feature detection and tracking in climate model data, and ensure they are efficient, well-documented, and freely available to the wider community.

Mission Relevance

This project supports the Laboratory's strategic focus area of energy and climate by advancing the state of the art in computational models for detecting climate change and attributing its cause.

FY14 Accomplishments and Results

In FY14 we applied the analysis techniques developed in FY13 to modeled and observed trends in cloud cover. Specifically, we (1) performed the first formal detection and attribution study of cloud changes over the satellite era; (2) obtained model-derived fingerprints of externally forced changes to three cloud properties—by considering simultaneous changes in all three properties, we defined a coherent multivariate fingerprint of cloud response to external forcing and used cutting-edge climate models to calculate the average time to detect these changes; (3) determined, given perfect satellite cloud observations beginning in 1983, that the models indicate a detectable multivariate signal should have already emerged; (4) established that independent satellite observations show a poleward migration of the zonal cloud cover pattern that is incompatible with the current generation of models—nevertheless, a detectable multivariate signal is indeed present in one observational dataset; and (5) initiated work to explore the impact of human-induced (anthropogenic) forcing on the phase and amplitude of the annual cycle of atmospheric temperature.

Proposed Work for FY15

In FY15 we will (1) investigate whether anthropogenic forcing yields statistically identifiable changes in the phase and amplitude of the seasonal cycle—our preliminary research indicates that certain anthropogenic forcing, such as human-caused depletion of stratospheric ozone, perturb both the phase and amplitude of the seasonal cycle of stratospheric and tropospheric temperature; (2) determine whether such forced changes from human activities can be discriminated from the background noise of internal variability; and (3) determine whether such forced changes are distinguishable from the natural variability arising from changes in solar irradiance and volcanic aerosol loadings.

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Large-Scale Integrated Electric Transmission and Distribution Grid Dynamic Simulation

Liang Min (13-ERD-043)

Abstract

Tomorrow's electric grid will increasingly include renewable resources and electric energy storage in both transmission (delivering power from generation to distribution circuits) and distribution (delivering power from distribution circuits to consumers). This electric grid will require simulations that can ensure reliable long-term operation of an extremely complex system comprised of millions of distributed units. Simulation technology for electric grid operations has developed over the last several decades in a piecemeal fashion, within narrow functional areas and well before the development of modern computational capabilities. As such, current simulation technology is insufficient to address pending complex smart-grid systems simulation where modeling effects of renewables and energy storage in the distribution network will have significant impacts throughout the entire grid. We propose to develop the first platform in the power energy community supporting large-scale integrated transmission and distribution systems simulation. We will create a software framework for evaluating simulation methodologies for integrated transmission and distribution systems, and will develop efficient numerical methods and parallel algorithms enabling coupled simulations on high-speed, state-of-the-art computers.

If successful, our prototype software platform will allow commercial software vendors, researchers, and utilities to build on it to solve problems associated with dynamic

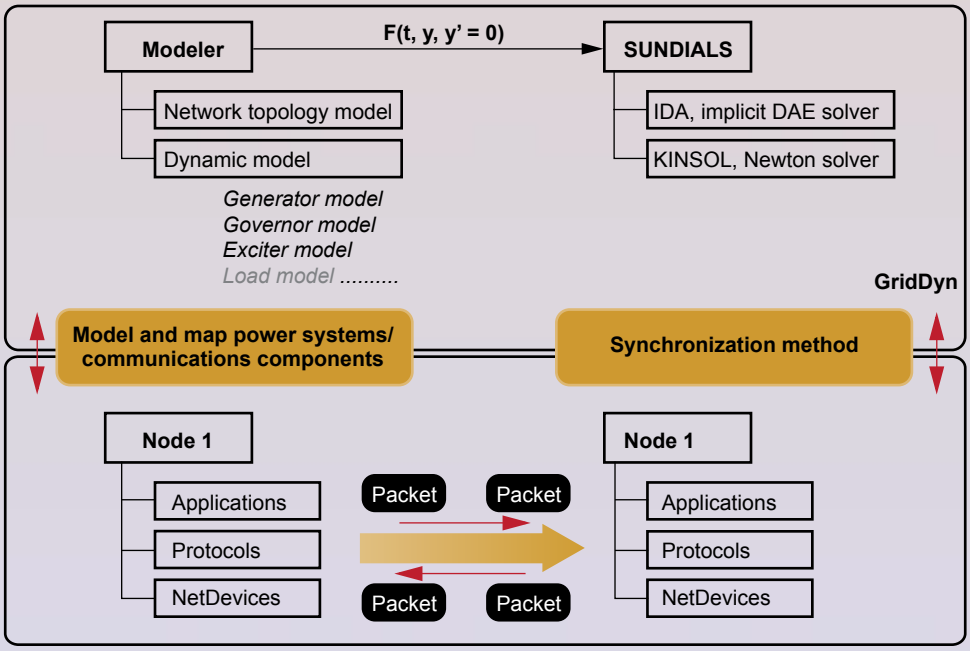
system interactions. This new capability will enable practitioners and researchers to identify not only potential reliability impacts of emerging technologies and control modes that cannot presently be fully evaluated, but also mitigation measures to ensure reliability. The current lack of such a software platform increases uncertainty not only about the viability of frequency regulation and voltage support resources, but also about emerging technologies, which could result in adverse economic, technical, and social impacts.

Mission Relevance

Energy security is a strategic thrust of the Laboratory in support of national priorities. Our research supports LLNL's efforts to support the national energy system by providing novel modeling and simulation approaches suitable for advanced computational architecture to reduce major power outages and ensure energy security through an optimized electric grid.

FY14 Accomplishments and Results

In FY14 we (1) constructed a transmission simulator prototype that uses the IDA and KINSOL solvers from LLNL's SUNDIALS (a suite of nonlinear and differential/algebraic equation solvers); (2) completed the initial parallel framework design and implementation for a power system simulation—at each time step a single transmission model was advanced, followed by parallel instances of the distribution software for each substation on the transmission network; (3) conducted a validation study of the scaled-up GridLAB-D power distribution system simulator, developed by the Pacific Northwest National Laboratory, using a test case provided by the California-based Pacific Gas and Electric utility company; and (4) investigated a federated system for electric grid and communication co-simulation and validated it on an IEEE (Institute



A federated system for electric grid and communication co-simulation.

of Electrical and Electronics Engineers) 39-bus system, which represents a greatly reduced model of the power system in New England and is commonly used to study both static and dynamic problems in power systems.

Proposed Work for FY15

In FY15 we will (1) develop parallel solution strategies for each sub-model of an integrated power transmission and distribution system simulation, (2) complete performance tests on a large system, and (3) explore and investigate the interaction between communication network and power systems.

Enzyme-Embedded, Microstructural Reactors for Industrial Biocatalysis

Sarah Baker (14-ERD-010)

Abstract

Most chemical reactions of interest for clean energy, such as the conversion of sunlight to chemical energy and the transfer of carbon dioxide into and out of solution, are routinely carried out in nature. Conventional industrial approaches to catalyze these reactions are either inefficient or have yet to be developed. Certain enzymes have been identified that carry out these reactions under mild conditions, which presents an opportunity for biocatalysis (the use of natural catalysts such as protein enzymes to chemically transform organic compounds) and biomimetics (the application of designs from nature to solve problems in engineering, materials science, medicine, and other fields) to fill the gap between available technologies and natural processes. Enzyme biocatalysis is currently limited primarily to low-volume, high-value products such as pharmaceuticals because of the narrow operating parameters required to preserve biocatalyst activity, slow rates of throughput from low catalyst loading, and limited mass transfer and susceptibility to contamination and poisoning. However, the ability to generate functional molecules or cells of interest now enables synthesis of engineered enzymes that are more amenable to industrial processes, widening the available operating parameters. These enzymes must be integrated with functional materials to fully realize their potential in large-scale chemical processes. Using cellular microstructural design strategies as inspiration, we will develop a new class of gas-to-liquid bioreactors based on enzyme-embedded, nanometer- and micrometer-structured polymeric materials.

The enzyme-embedded, high-permeability, polymer-based reactor components we propose will permit new uses of biocatalysis for energy applications. These tunable, modular reactor components will incorporate engineered properties at multiple size scales using advanced manufacturing techniques. Inspired by the hierarchical structures of living systems, the components will enable efficient use of enzymatic reactions without the drawbacks of living-cell fermentation. The ability to additively manufacture

high-throughput bioreactors will enable new large-scale systems such as those that will be required for creation of designer liquid fuels from methane. Successful implementation of this technology could offset the methane leaks associated with shale gas production, improving the case for shale gas as a major domestic energy source.

Mission Relevance

This project will further the Laboratory's expertise in the strategic focus area of energy and climate relevant to delivering innovative manufactured materials for gas-to-liquid conversion. The project will develop the capability to generate a new class of engineered and manufactured materials to support biocatalysis for energy applications.

FY14 Accomplishments and Results

We (1) implemented an assay to measure methane-to-methanol conversion; (2) made stable, active, 200-nm-sized liposomes of the particulate methane monooxygenase enzyme from crude cell-wall material by a simple extrusion process that promises to be industrially scaleable; (3) stabilized isolated particulate methane monooxygenase enzymes with nanometer-scale lipoprotein particles; (4) synthesized networks of copolymers that have both water-soluble and non-water-soluble portions in their structures; (5) modeled the nanoscale structure of these copolymers with mesoscale simulations; and (6) demonstrated enzyme activity in a single-phase cross-linked polymer.

Proposed Work for FY15

In FY15 we will place our stabilized enzyme constructs in the selected polymer systems—first in single-phase polymers, then in copolymer networks. In addition, we will refine the polymers and assembly techniques to create domain sizes and geometries suitable for the enzyme constructs to achieve and enhance activity in the polymer environment.

Enabling Multiscale Simulations of Atmospheric Flow over Complex Terrain in Earth System Models

Katherine Lundquist (14-ERD-024)

Abstract

High-resolution simulations of atmospheric flow are critical in applications such as wind energy, hydropower, power grid capacity, atmospheric transport and dispersion, and regional downscaling of climate predictions. Grid nesting, a global modeling technique using multiple telescoping grids at increasing resolution, provides the framework for multiscale simulations. However, in practice, there are limitations that prevent the application of this technique to simulations of flow over complex terrain, thus preventing current modeling tools from producing accurate multiscale high-resolution atmospheric simulations. Our objective is to create an atmospheric numerical modeling

framework that dynamically integrates simulations at multiple physical scales from the microscale of 1 to 1,000 m, to the mesoscale of 1 to 200 km. We will use the immersed boundary method, a computational fluid dynamics approach for simulating fluid and structure interactions, to enable this numerical tool to handle complex terrain and urban geometries at the microscale. This high-fidelity approach will allow integrated simulations at previously unapproachable scales and will facilitate state-of-the-art research in atmospheric sciences and environmental engineering. The capability we develop will open the door to new atmospheric applications that have not been possible because of model limitations, including forecasting for wind energy, predicting atmospheric transport and dispersion, and downscaling atmospheric simulations.

Our proposed numerical model will directly benefit wind energy development, addressing the critical need for multiscale models capable of handling complex terrain. We expect to develop our multiscale modeling framework within the Weather Research and Forecasting model, through the LLNL Atmospheric Modeling Testbed. Our framework will thus be made available to the community of over 10,000 Weather Research and Forecasting users and will accelerate research with national security relevance and will impact the larger atmospheric science community. Our modeling effort will expand atmospheric modeling capabilities, allowing for additional opportunities such as atmospheric modeling for hydropower applications and energy grid capacity.

Mission Relevance

Our project will directly benefit research on wind energy with a model capable of handling complex terrain, in support of the Laboratory's mission in energy security. It will benefit climate research through improved knowledge of downscaling, in support of the Laboratory's strategic focus area in energy and climate. In addition, it will provide high-fidelity research-grade simulations relevant to the core competency in computational science and engineering for improving tools for responding to atmospheric releases of hazardous materials, in support of the Laboratory's national security mission.

FY14 Accomplishments and Results

In FY14 we (1) made progress in developing an approach for coupling terrain-following and immersed boundary Weather Research and Forecasting domains—we developed an interpolation algorithm between domain types that allows for arbitrary vertical coordinates, eliminating the requirement that the same vertical grid be used on all domains; (2) developed routines for initializing an immersed boundary method domain within a terrain-following coordinate domain; (3) developed boundary conditions at the immersed boundary that parameterize surface fluxes of momentum; (4) validated the new boundary condition using theoretical results, and began work on validating the boundary condition using field observations; and (5) developed our Weather Research and Forecasting coupled with the immersed boundary method for use with meteorological data, and developed techniques for using the immersed boundary method with three-dimensional descriptions of geometries.

Proposed Work for FY15

In FY15 we will (1) continue the development of standard initializations for coupling the Weather Research and Forecasting and immersed boundary method models, (2) modify physics routines for use with the coupled models, (3) develop immersed boundary method models for higher-order turbulence closures, and (4) perform multiscale simulations to test the interface to the Weather Research and Forecasting model.

Publications and Presentations

Daniels, M. H., and K. A. Lundquist, 2014. *Development of the Weather Research and Forecasting (WRF) model with the immersed boundary method (IBM) for high-resolution simulations in complex terrain.* LLNL-POST-656661.

Lundquist, K. A., et al., 2014. *Enabling high-resolution simulations of atmospheric flow over complex terrain in the WRF model.* 21st Symp. Boundary Layers and Turbulence, Leeds, United Kingdom, June 9–13, 2014. LLNL-PRES-655456.

Wetlands as a Source of Atmospheric Methane: A Multiscale and Multidisciplinary Approach

Karis Mcfarlane (14-ERD-038)

Abstract

Understanding the interacting processes that govern net emissions of methane (a powerful greenhouse gas) from wetlands is critical for predicting biospheric feedbacks to climate change. We will investigate the response of these processes to warming and to elevated levels of carbon dioxide at the DOE Office of Biological and Environmental Research's Spruce and Peatland Responses Under Climatic and Environmental Change (SPRUCE) project site and, based on our results, assess how the processes interact to drive changes in net emissions. Specifically, we will (1) link underground processes to atmospheric fluxes using observations of carbon-13 and hydrogen-2 (measured with isotope-ratio mass spectrometry) and carbon-14 (measured with accelerator mass spectrometry), (2) identify key microbial species influencing methane production and consumption using stable-isotope probing with nanometer-scale secondary-ion mass spectroscopy, (3) constrain ebullition phase transition rates with measurements of noble gases in pore water using membrane-inlet mass spectrometry, and (4) integrate our findings into a box model that describes wetland response to global warming and elevated levels of carbon dioxide.

We expect to provide the first comprehensive study of the link among the methane-producing microorganisms methanogens and methane-consuming methanotrophs, the in situ production and consumption of methane, attributed carbon sources, and net emissions. Our results will be integrated to identify how individual processes respond to experimental treatments and how this response scales from the microscale

to the ecosystem, enabling testing of current working hypotheses for divergent responses of wetland emissions to future change. Our deliverables include the quantitative, process-level information required to evaluate the physiochemical parameters of methane and carbon dioxide fluxes from wetlands and to integrate microbial community function into these descriptors for improved prediction. Finally, our project will demonstrate the use in carbon cycle science of new capabilities—carbon-14 and hydrogen-2 measurements, nanometer-scale secondary-ion mass spectroscopy, and membrane-inlet mass spectrometry for detection of noble gases.

Mission Relevance

This project supports the Laboratory's mission in energy security by advancing capabilities in predicting and understanding climate change. It also directly supports the Laboratory's core competency in chemical and isotopic signatures by promoting the continued development and early application of specialized techniques in accelerator mass spectrometry and isotope-ratio mass spectrometry.

FY14 Accomplishments and Results

In FY14 we (1) validated protocols for collecting carbon-14 methane from surface flux chambers; (2) evaluated noble gas concentrations in peat columns in situ and added hydrogen-3 analysis; (3) upgraded our isotope-ratio mass spectrometry system for hydrogen-2 analyses; (4) conducted pre- and post-deep-peat warming field sampling; (5) completed measurements of carbon-14 in samples collected in 2014; (6) attained gene libraries from our external collaborator, who agreed to provide samples from their carbon-13 label incubations and archived samples; (7) designed chips for probing

Clockwise from upper left: an experimental plot at the DOE Office of Biological and Environmental Research's Spruce and Peatland Responses Under Climatic and Environmental Change project research site, a surface flux chamber (without lid) used to sample carbon dioxide and methane emitted from the bog surface, sampling chamber headspace gas, and a sample being readied for laboratory radiocarbon analysis.

of methanotrophs and methanogens using a stable isotope labeled with carbon-13; and (8) expanded our research team with a Lawrence scholar (Ph.D. student) for field and lab components of the project and an LLNL postdoctoral researcher to perform modeling work.

Proposed Work for FY15

In FY15 we will (1) complete chip-based stable-isotope probing using carbon-13 labels, air extraction, and chemical analyses for all samples collected in FY14 (pre-warming and deep-peat warming experiment); (2) use FY14 results to finalize plans for a whole-system warming experiment; (3) perform field sampling for chip-based stable-isotope probing using carbon-13 labels, pore-water noble gases, surface carbon-14 flux, and canopy carbon dioxide and methane isotopes to test whole-ecosystem warming and elevated carbon dioxide effects; (4) complete spring and early-summer air extractions and analyses and begin late-summer extractions and analyses; and (5) begin ecosystem carbon dioxide-methane modeling work.

Publications and Presentations

Guilderson, T. P., et al., 2014. *Sources of respired carbon in a northern Minnesota ombrotrophic spruce bog: Preliminary ^{14}C Results from the SPRUCE site.* AGU Fall Mtg., San Francisco, CA, Dec. 15–19, 2014. LLNL-ABS-658437.

McFarlane, K. J., et al., 2014. *Belowground functioning of a forested bog as a sink or source of atmospheric carbon.* Grand Challenges, Great Solutions—ASA, CSSA, and SSSA Intl. Ann. Mtg., Long Beach, CA, Nov. 2–5, 2014. LLNL-ABS-654667.

Pett-Ridge, J., et al., 2014. *Functioning of wetlands as a source of atmospheric methane: A multi-scale and multi-disciplinary approach at SPRUCE.* LLNL-ABS-663036.

Real-Time Microseismic Processing for Induced Seismicity Hazard Detection

Eric Matzel (14-ERD-051)

Abstract

Induced seismicity is an inherent issue associated with several underground energy technologies of strategic interest to Livermore including geologic carbon sequestration, enhanced geothermal systems, and shale gas development. If fracturing fluids from shale gas development, for example, are injected close to a pre-existing fault or fracture system, the resulting elevated pressures may lead to dynamic earthquake slip. We propose to develop an approach to predicting and avoiding induced earthquakes, with the central goal of deployment as an on-site, real-time tool for use by field operators in the energy sector. We will combine innovative techniques for analyzing microseismic data with a physics-based inversion model to forecast microseismic cloud evolution (a cloud of

spatially correlated individual microseismic events). Inversion models are used to convert observed measurements into information about objects or systems. Fast-running tools of the type we propose will allow operators to respond quickly to changing subsurface conditions and thus develop underground resources in a responsible manner.

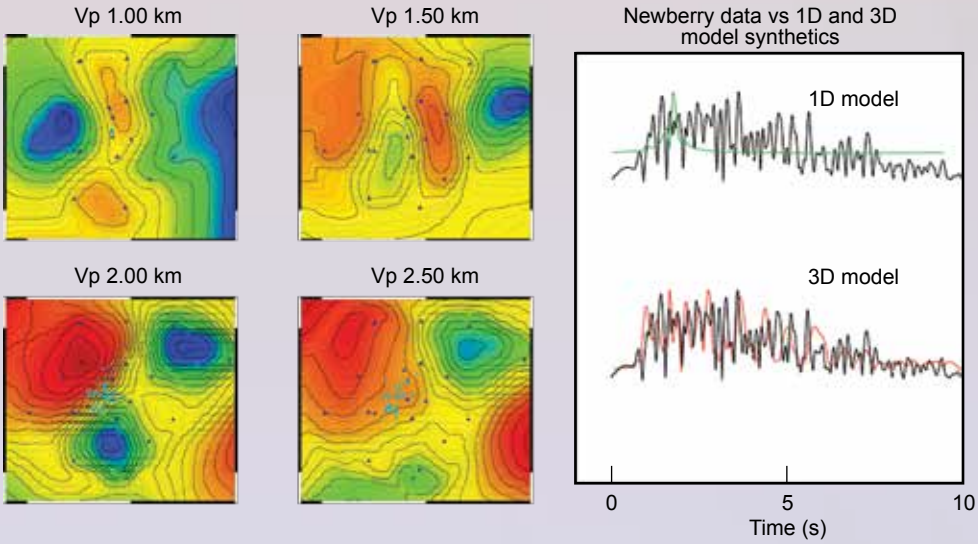
Current state-of-the-art methods in this field of energy exploration and development contain almost no insight into the equilibrium and motion of fluids and of solid-body processes leading to earthquakes. Our proposed research will improve the foundational science and practical capabilities necessary to develop predictive tools. The ultimate deliverable is a real-time methodology for continuously evaluating the likelihood of triggering a large earthquake at any given field site. This evaluation will be dynamically updated every few hours as new microseismic or injection-rate data becomes available, and it will allow operators to quickly react to changing subsurface conditions. A central goal is that these processing approaches be fast enough that they can be applied in real time and synchronized with data acquisition.

Mission Relevance

A rational strategy for assessing and mitigating seismic risk must be developed if large-scale fluid injection operations are to continue responsibly. Our proposed work will produce both foundational science and a practical capability in both advanced monitoring techniques and addressing public and regulatory concerns over seismic risk, supportive of a central Laboratory mission in energy to advance the nation's security through the production, development, and deployment of energy resources and technology.

FY14 Accomplishments and Results

In FY14 we (1) demonstrated the power of seismic interferometry for site characterization, dramatically improving the accuracy with which we can match the microseismic waveform—combining this with the Bayesloc multiple seismic-event locator procedure



The precision of the three-dimensional model of the Newberry geothermal project, obtained from ambient noise correlation, allows us to predict where microseismicity will occur and captures much of the complexity seen in the seismic records.

and matched-field processing techniques allows us to identify and precisely locate individual microseismic occurrences, and the virtual seismometer method is showing great promise for measuring seismic sources; (2) obtained complete data sets from the Newberry and Basel geothermal injection sites—the Newberry volcanic site in Oregon has been characterized and serves as a test bed for the methodologies; and (3) applied our techniques to the Basel geothermal field in Switzerland, which will allow us to apply and develop these techniques together with the inversion processes for hydromechanical phenomena for an area that has seen induced seismicity.

Proposed Work for FY15

For FY15 we will focus on identifying how well we can measure microseismicity, whether we can identify patterns of slip that are indicative of faults, and how well we can monitor the cloud of microseismicity and predict the evolution of the pressure plume. Precise measurements will be key. Using a hydromechanical inversion, we will create a model of the permeability field that is consistent with the data and seismic measurements. These tasks will require continued development of the seismic and hydromechanical inversion techniques, and particularly in tying the seismic and hydromechanical techniques together to improve prediction.

Publications and Presentations

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Matzel, E. M., et al., 2014. *Imaging the Newberry EGS Site using seismic interferometry.* Stanford Geothermal Workshop, Stanford, CA, Jan. 26–28, 2014. LLNL-PRES-650577.

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Statistical and Dynamical Approaches to Probabilistic Decadal Climate Prediction

Gardar Johannesson (14-ERD-095)

Abstract

The current state of the art in providing robust global climate predictions for the 21st century is based on mining the Coupled Model Intercomparison Project (CMIP) database of multiple-model ensembles of global climate simulations. The latest incarnation of the project (CMIP5) includes a new set of simulation experiments aimed at providing decadel climate simulations to explore the variation across models in predicting the climate one to three decades out. However, uncertainty in decadal climate simulations is mainly driven by structural uncertainty in global climate models, uncertainty in natural external forcing such as volcanic and solar activity, and natural intrinsic climate variability. We propose to provide improved probabilistic decadal regional-scale climate predictions that are suitable for general climate-change risk assessment and decision support. We will use our new set of simulations and develop advanced Bayesian multiple-model decadal prediction methods and use them to assess the impact of forcing uncertainty on climate predictions.

Our three main objectives with this project include providing (1) decadel climate predictions that reflect the capability of individual global climate models to produce decadel simulations, (2) additional regional-scale information to resolve important small-scale processes using targeted shorter-term high-resolution simulations, and (3) climate impact quantities of interest that are scaled to regional and local areas using statistical methods as well as global-scale predictions in combination with targeted high-resolution simulations. We expect that our project will yield statistical methods and algorithms to produce realistic probabilistic decadal regional climate predictions by leveraging the multiple-model CMIP5 database of climate simulations. The uncertainty in the simulation will reflect and account for the structural uncertainty across models, the natural variability in the climate, and uncertainty in natural forcing of the system. Through targeted short-term simulations, this work will provide deeper insight into the impact that uncertainty in past and near-future natural forcing will have on climate projections. We intend to deliver the capability to synthesize weather-related processes that are needed as inputs for other climate models such as hydrology that reflect large-scale near-term decadel variation and trends in the global climate process.

Mission Relevance

Providing regional-scale climate predictions on a timescale of several decades that are suitable for general climate-change risk assessment is well aligned with the Laboratory's strategic focus area in energy and climate to understand climate challenges and develop options for future adaption, including decadal climate projections at the regional scale. Mining data from climate simulations also aligns well with the core competency in information systems and data science.

FY14 Accomplishments and Results

The FY14 tasks were revised because of a late start and reduced budget. Specifically, we (1) constructed a local database of roughly 4,000 decadal CMIP5 simulations, along with their corresponding historical and forecast simulations; (2) defined climate quantities and regions of interest; (3) developed and applied Python tools for biological computation to post-process the needed climate simulations and observations; (4) began the development of a probabilistic, decadal climate-prediction model utilizing simulations from a single climate model; (5) developed an initial prototype of a statistical de-convolution model for a given climate signal time series and applied it to assess what role volcanic aerosol uncertainty and underestimation has in the recent warming hiatus in the latest CMIP5 simulations; and (6) hired a postdoctoral research scientist, who started in June 2014.

Proposed Work for FY15

In FY15 we propose to (1) develop an application of the Bayesian probability, decadal-climate prediction model that utilizes only a single climate model; (2) perform initial development and testing of a Bayesian probability, decadal climate-prediction method that uses an ensemble of climate models; and (3) develop an application of a statistical de-convolution model to external climate forcing uncertainty and the recent warming hiatus.

Detecting and Partitioning Carbon Dioxide Fluxes

Jessica Osuna (14-LW-079)

Abstract

Predicting future climate scenarios relies strongly on the understanding and quantification of the global carbon cycle, which in turn relies on understanding the tips and balances between land carbon sources and carbon sinks. While respiration and photosynthesis respond distinctly to environmental drivers, current techniques provide no specific information regarding the sensitivities of different carbon fluxes to a changing environment. We propose to quantify the photosynthesis and respiration partitioning of net ecosystem carbon dioxide (CO_2) exchange and collect measurements of carbon and oxygen isotopes at high spatial and temporal frequency with a miniature tunable diode laser sensor employing absorption spectroscopy developed at LLNL. Easily

deployable isotope sensors are needed to understand how fluxes of carbon will change with changing climate. Our miniature tunable diode laser reduces cost, infrastructure, and power compared to commercially available sensors and enables increased sampling frequency. The instrument will be fully tested and then deployed to LLNL's Site 300 and AmeriFlux Wind River Field Station in Washington to measure partitioned CO_2 fluxes.

We expect to refine a miniature tunable diode laser capable of measuring the stable isotopes of carbon and oxygen in CO_2 in situ at 10 Hz or greater at a precision great enough to detect small diurnal variations in varying CO_2 isotope compositions. We will demonstrate the instrument in situ alongside commercially available instruments. Most importantly, this project will reveal the contribution of photosynthesis and respiration to the net ecosystem CO_2 exchange via isotopes at a forest and an annual grassland, as well as how these contributions vary with respect to time, space, and environmental drivers. The mobility of the sensors allows for measurements of canopy storage and lateral advection of CO_2 fluxes, neither of which is currently measured at either site.

Mission Relevance

Terrestrial carbon science is widely thought to be an uncertainty in the global carbon budget. Success of the isotope sensor would provide a mechanism to quantifying the land carbon balance, and more importantly, to understanding vulnerabilities and sources of major greenhouse gases under climate change, supportive of the Laboratory's strategic focus area in energy and climate.

FY14 Accomplishments and Results

In FY14 we focused on measurements of CO_2 using a vertical-cavity surface-emitting laser. Specifically, we (1) reduced the limit of detection from 100- to 5-ppm CO_2; (2) determined it was necessary to use a 4,300-nm laser to detect $^{13}CO_2$ absorption peaks; (3) installed an environmental chamber to control temperature and humidity; (4) established a record of invention for our technology; (5) calibrated the CO_2 absorption peaks with the laser against the High Resolution Transmission database; (6) established the laser's stability, precision, and accuracy; (7) improved the sampling protocol to shorten the time step of measurements toward achieving 10-Hz sampling; (8) developed a calibration from the 2ω signal to CO_2 concentration in the sample gas; and (9) aligned our peaks of absorption at the wavelengths expected according to the High Resolution Transmission database.

Proposed Work for FY15

In FY15 we will (1) work with the 4,300-nm quantum cascade laser to define the accuracy, precision, and limit of detection of CO_2 molecules that differ in their isotopic composition; (2) develop a technique to measure $^{13}CO_2$ and $^{12}CO_2$ at 10 Hz continuously for flux measurements; (3) test sensor stability when exposed to the temperature and humidity experienced during deployment; (4) purchase and program data acquisition units to operate the sensor with less power; and (5) deploy, in collaboration with the AmeriFlux network, an array of sensors to measure CO_2 fluxes in northern California

to test performance relative to commercial instruments. We will also test the sensor alongside commercially available instruments that measure $^{13}CO_2$.

Testing Hypotheses of the Little Ice Age and Holocene Climate Change

Susan Zimmerman (14-LW-091)

Abstract

The climate of the last 1,000 years is dominated by the inception, persistence, and abrupt end of the Little Ice Age, which was a period of unusually cold conditions between 1250 and 1850 when glaciers in Europe were greatly extended and rivers such as the Thames froze solid during the winter. One of the most critical questions in climate science today is, was the abrupt end of the Little Ice Age a natural fluctuation of the climate back to a warmer state, or was it a result of the modification of the climate system by anthropogenic changes inflicted during the Industrial Revolution.

We expect to produce the first directly dated record of glacial advance in North America, which will be the first comparison of temperature-dominated glacial advance with precipitation-dominated tree-ring and lake records. This will also be the first time that the precipitation signal dominated by the El Niño and Southern Oscillation and the temperature signal dominated by the Intertropical Convergence Zone (commonly known as the doldrums) near the equator have ever been compared using glacier and lake records together. The expression of these climate changes over the last 1,000 years in climate models has become a key tool for understanding future changes, but no comparison of the El Niño and Southern Oscillation patterns that we plan to examine exists in the published literature.

Mission Relevance

Our work in integrating the glacial history of the Sierra Nevada into the regional records of climate, and using millennium runs of global climate models to examine the influence of the El Niño and Southern Oscillation, supports the Laboratory's core capabilities in energy and climate research. Researching the mechanisms of climate change is relevant to LLNL's strategic mission focus in understanding climate challenges and enabling climate projections at the regional scale.

FY14 Accomplishments and Results

In FY14 we (1) hired our postdoctoral candidate to assist with the research effort, (2) readied our sample preparation lab and determined fieldwork sites and strategies, (3) completed final plans and permitting for fieldwork, (4) mapped and collected samples at selected field sites, and (5) began processing samples and standards in our lab.

Proposed Work for FY15

In FY15 we will (1) produce detailed glacial maps of the field sites, (2) prepare and measure all beryllium-10 samples for the project, (3) date and interpret lake cores, (4) interpret the Holocene glacial history of the central Sierra Nevada and integrate that history with climate reconstructions at a specific point in the geological past that we have developed as well as those from the literature, and (5) compare glacial history of precipitation and temperature with results of model runs.

Publications and Presentations

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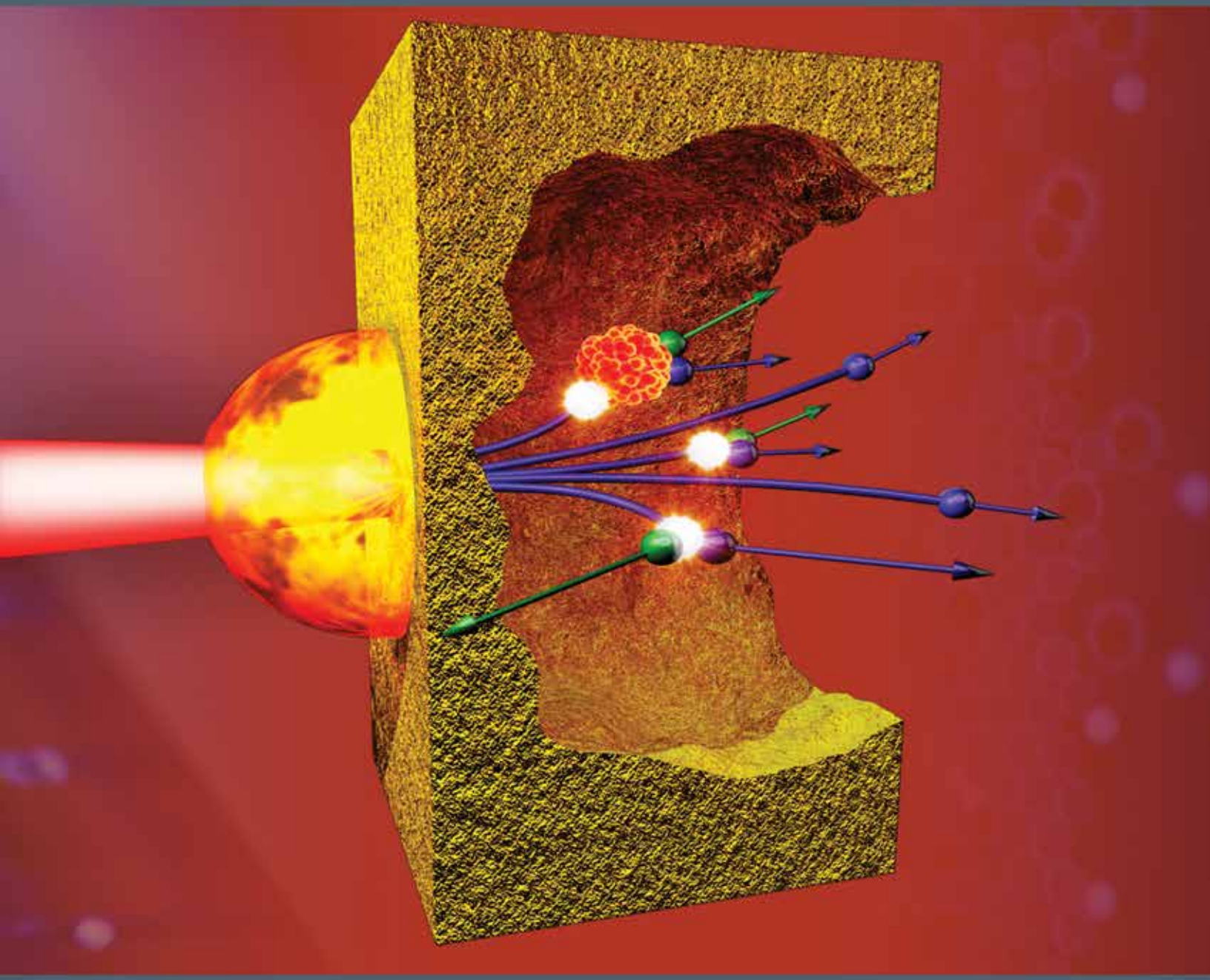
High-Energy-Density Science



FY2014

Lawrence Livermore National Laboratory

Our article in *Nature* demonstrates the high-impact nature of research at extreme compression to materials physics under planetary interior conditions.

Extreme Compression Science

Jon Eggert (12-SI-007)

Abstract

This effort will launch a new exploration of materials science at high-energy-density conditions using the National Ignition Facility (NIF) at Livermore and supporting facilities. We propose to enable several high-energy-density physics experiments that have been granted time on NIF, help guide the materials-concept development proposals, pioneer new directions in extreme matter physics, and build a community to grow this new extension of materials science. We will concentrate on three experimental thrusts. For hydrogen at extreme densities, the proposed experiments will document, for the first time, that it is possible to perform quantitative quasi-isentropic compression experiments on deuterium or hydrogen molecules pre-compressed from 1 to 5 GPa. For matter at atomic pressures, the principal goals include learning to compress matter 100 to 1,000 Mbar and developing diagnostic tools to map physics from the atomic to thermodynamic levels. We will perform x-ray diffraction and extended x-ray absorption fine structure at NIF and the OMEGA laser facility at the University of Rochester to produce the fidelity required to map the structure, phase, and thermodynamics for the complex states expected to exist at high pressures. For achieving gigabar pressures relevant to astrophysical objects, we will develop convergent compression techniques and radiography.

We expect to develop a major materials science effort built around NIF, which could alter our understanding of materials and condensed matter in profound ways. This research will extend the range of materials studies under pressure by at least three orders of magnitude and provide external pressures into the atomic regime for the first time. This could facilitate the identification of altogether new forms of matter on Earth, provide new insight to the periodic table and lead to creation of novel materials, provide enhanced understanding of dynamical processes in high-density materials and the nonequilibrium of matter at extreme pressures, enable a laboratory test bed for the wealth of new astronomical observations of planets outside the solar system, and provide enhanced routes to inertial fusion energy.

Mission Relevance

This research will contribute to strategic themes of the Laboratory's strategic plan in high-energy-density science and materials on demand, which are key to mission thrusts in stockpile stewardship science, nuclear threat reduction, laser inertial fusion energy science and technology, and advanced laser optical systems and applications.

FY14 Accomplishments and Results

In FY14 we (1) met our goal of establishing extreme-compression science as a legitimate field to be addressed using high-energy lasers; (2) performed collaborative experiments with collaborators from the University of California, Berkeley; Princeton; Stanford; Brown; and Harvard universities in the U.S. and Oxford University, University

of Edinburgh, University of Bayreuth, the Atomic Weapons Establishment, and the Atomic Energy Commission in Europe; (3) enabled development of the TARDIS (target diffraction in situ) powder diffraction diagnostic on NIF and performed one fundamental science TARDIS shot on diamond; and (4) are contributing to the planning and development of the following: the Matter in Extreme Conditions beam line at the Linac Coherent Light Source at the SLAC National Accelerator Laboratory at Stanford, the Dynamic Compression Sector at the Advanced Photon Source at Argonne National Laboratory, and the high-energy-density instrument at the European X-Ray Free Electron Laser facility in Hamburg, Germany.

Project Summary

With this project, we supported the development of several new diagnostic techniques for high-power laser-driven compression experiments such as a new broad-band reflectivity instrument, single-photon energy-dispersive x-ray diffraction, and a transverse-motion VISAR (velocity interferometer system for any reflector) at Livermore's Janus laser. At the Linac Coherent Light Source at the SLAC National Accelerator Laboratory, we collaborated on five and led two campaigns studying diamond, iron, titanium, bismuth, sodium chloride, aluminum oxide, and aerogels. We performed many experiments at the OMEGA laser in Rochester, New York, and measured the Hugoniot stress–density–temperature relationship and phase transitions in dense silicon dioxide (stishovite), nitrogen gas, and water. We continued the development of x-ray diffraction, observing the structure phase transition in magnesium oxide for the first time; phase transitions in iron, tin, molybdenum, and iron oxide; as well as the first-ever shock-melt refreeze diffraction experiment (in tin). We successfully measured the extended x-ray absorption fine structure in iron to 560 GPa, providing a new experimental probe of temperature. Using ramp-compression wave-profile equation-of-state experiments, we measured the stress density of cold iron and magnesium oxide. We performed three ramp-compression equation-of-state fundamental science shots on NIF (one on diamond, two on iron), and helped develop the gigabar and target-diffraction in-situ platforms at NIF and participated in four gigabar and one target-diffraction in-situ fundamental science shots. We are taking active roles in the development of laser-driven compression experiments at new beamlines at the Linac Coherent Light Source at the SLAC National Accelerator Laboratory, the Dynamic Compression Sector at the Advanced Photon Source at Argonne National Laboratory, and at the European X-Ray Free Electron Laser facility in Hamburg, Germany, as well as continuing our efforts at the OMEGA laser. The target-diffraction in-situ diagnostic on NIF, based on the work funded by this project, was awarded two campaigns in the latest round of discovery science proposals for NIF on the dynamically compressed carbon iron melting curve. Finally, as a direct result of our work, we will perform the first high-atomic-number shots on NIF using the target diffraction in-situ diagnostic in FY15.

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Strength in Metals at Ultrahigh Strain Rates

Jonathan Crowhurst (12-ERD-042)

Abstract

We intend to obtain accurate data on the dynamic strength of metals—such as aluminum, copper, vanadium, and tantalum—and the time-dependent evolution of shock waves at ultrahigh strain rates. Motivations for this work include the current lack of high-time-resolution data to allow direct comparison with molecular dynamics simulations and a lack

of directly measured strengths and related properties necessary to refine and test dynamic strength and multiscale models. In addition, currently we face an inability to fully test the validity, and thus account for, fundamental scaling laws—for example, the fourth power law, which describes energy radiated per surface area per unit time for a black body. We will use a proven tabletop laser-shock apparatus to obtain strain rates consistent with a time resolution of 1 ps. Data acquisition will be rapid and inexpensive, and results will be analyzed using state-of-the-art dynamic strength models.

We expect to obtain high-fidelity data on the dynamic strength of metals and the time-dependent evolution of shock waves at the highest strain rates to date. Strain rates would be consistent with our time resolution of approximately 1 ps, which would correspond to strain rates of up to 10^{12}/s—in aluminum this would be expected to correspond to peak stresses of approximately 100 GPa. We will fully integrate both experimental and theoretical approaches to obtain unprecedented insight into the physics of shock waves in metals. Our results will be expected to substantially increase the accuracy and validity of current multiscale models.

Mission Relevance

Shock waves, and the material data they provide, are of direct relevance to the Laboratory's strategic focus area of stockpile stewardship science, as well as lasers and optical materials science and technology and high-energy-density science core competencies. Obtaining a more detailed understanding of the detailed physics associated with high-strain-rate dynamic compression is also important for fundamental research in a range of disciplines.

FY14 Accomplishments and Results

In FY14 we (1) obtained a time scale for the alpha–epsilon transition in iron at very high strain rates (10^9/s) as well as the deviatoric stress (a measure of strength) before the transition takes place; (2) prepared, characterized, and shocked tantalum thin films, measuring very large precursor amplitudes that decay relatively rapidly with propagation distance—this implies an unexpectedly fast relaxation time that may be a consequence of the fine-grain nature of the samples; (3) investigated combining a tabletop shock wave system with an in-situ x-ray diffraction probe—such a system would permit investigations at the scale of the lattice and would directly reveal kinetics of high-strain-rate phenomena while maintaining all the advantages of a small-scale system; (4) studied aluminum using various drives (shock-like and ramp-like); and (5) collaborating with our colleagues at the Max Planck Institutes in Hamburg to perform in-situ electron diffraction on shocked metals.

Project Summary

We have shown that at approximately 10^9/s, the alpha-to-epsilon polymorphic transition in iron begins within 100 ps after an initial very large (~10 GPa) and mostly elastic compression and appears largely complete within a similar time frame thereafter. The corresponding deviatoric stress before the transition begins can exceed 3 GPa, while the transition stress itself is up to 25 GPa, nearly twice the value measured at low strain rates. Our study thus

provides fundamental insight into this famous shock-driven transition. We also acquired very high-strain-rate data from tantalum thin films (shown in the figure) that should allow us to obtain the dynamic strength of this programmatically relevant material. Additionally, we constructed a second-stage amplifier for our shock drive that produces laser pulses of energy in excess of 40 mJ—that is, hundreds of times the energy we have used in all our other experiments on metals, hydrogen, reactive liquids, and high-explosives materials. We expect multiple-megabar stress while still maintaining all the advantages of a small-scale system. We are actively seeking programmatic follow-on support to continue our work on metals and expect to be part of a larger-scale project to comprehensively study the kinetics of phase transitions in metals on both an experimental and theoretical basis. We are maintaining existing, and will forge new, collaborations with external institutions.

Publications and Presentations

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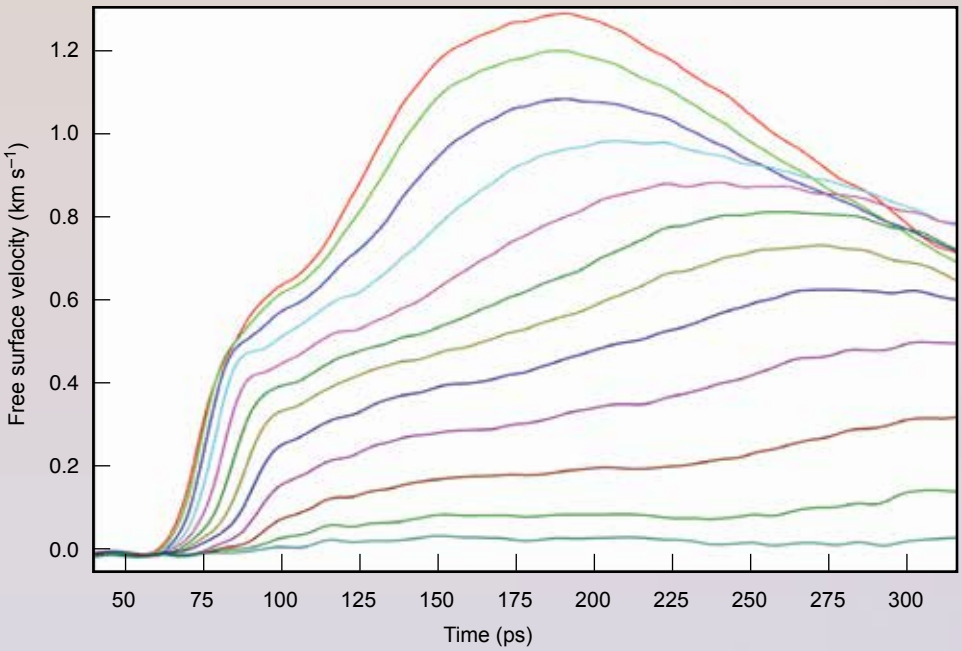

Velocity histories measured at the free surface of a body-centered-cubic tantalum film of nominal thickness 1.1 μm. The curves, corresponding to different parts of approximately one half of the breakout profile, show the evolution of the shock structure as position varies from near the edge (lowest energy, dark cyan) to near the center (highest energy, red). The elastic precursor grows with position and likely exceeds 20 GPa near the center of the breakout. The data is a stack of 10 individual shots.

Equation of State of Polymers Under Extreme Conditions with Quantum Accuracy

Nir Goldman (12-ERD-052)

Abstract

Accurate modeling of materials containing the bonded elements carbon, hydrogen, and oxygen (such as polymers and low-density foams) is essential for National Ignition Facility target capsule design and laser-driven ramp wave studies, in which chemical reactivity can lead to unexpected relationships of pressure to density, or Hugoniot states. We will create a predictive capability for materials containing carbon, hydrogen, and oxygen that are accurate at temperatures greater than 1 eV (11,605 K) and pressures greater than 10 Mbar for use in interpreting and designing experiments on Livermore's National Ignition Facility. This capability will include density-functional, tight-binding parameters that will increase computational efficiency by several orders of magnitude over standard quantum codes, while achieving comparable accuracy. Our models will help to fully elucidate the equations of state and chemical processes that occur in these experiments.

We will develop a high-efficiency quantum model for simulating carbon, hydrogen, and oxygen materials at high temperatures and pressures to circumvent the critical issues of timescale and system length presented by standard quantum codes, while achieving comparable accuracy. Our ultimate goal is to develop and constrain hydrodynamics code simulations by efficiently and accurately estimating equation-of-state data and the chemical kinetic effects of shock compression and release behavior in bonded carbon–hydrogen–oxygen materials. Our simulations will address critical needs for experiments on the National Ignition Facility by helping to interpret current and proposed ramp-compression experiments using carbon-based foams, and by making predictions for future National Ignition Facility capsule experimental design.

Mission Relevance

This project supports the Laboratory's high-energy-density science core competency and missions in national security and energy security by creating a predictive capability for essential materials in fusion ignition capsules used in stockpile stewardship and fusion energy research. This capability could also be extended to other fusion target materials, such as diamond and beryllium, to strongly shocked energetic materials used in national security efforts, and to planetary fluids such as hydrogen and water, in support of the Laboratory's fundamental science mission.

FY14 Accomplishments and Results

In FY14 we (1) concluded optimization of our new density-functional tight-binding models, thereby creating a general capability for simulating carbon–hydrogen–oxygen bonded systems under pressures up to 20 Mbar and temperatures up to 3 eV; (2) used our new models to elucidate the nanosecond-timescale formation of nanoscale diamond, graphene ribbons, and exotic phases such as carbyne (a chain of carbon atoms) during

adiabatic expansion from shock-compressed states; (3) investigated experimentally observed volume expansions during shock compression to approximately 30 GPa and 3,000 K; and (4) conducted simulations of foams with varying oxygen compositions to estimate the kinetics of carbon cluster versus carbon monoxide synthesis in ramp-compression experiments.

Project Summary

We have developed a high-efficiency quantum simulation model for carbon–hydrogen–oxygen bonded materials at over 10 Mbar pressures and multiple-elecronvolt temperatures that circumvents the critical timescale and system length issues with standard quantum codes while retaining most of their accuracy. Our simulations can efficiently provide accurate estimates of equation-of-state data and chemical kinetic effects of shock compression and release behavior in carbon–hydrogen–oxygen bonded materials (e.g., diamond and hydrocarbon foams) that can be used to develop and constrain hydrocode simulations. Our results address key needs for experiments on the National Ignition Facility by both helping interpret current and proposed ramp-compression experiments, as well as making predictions for future National Ignition Facility capsule experimental design. We have obtained support from the Department of Defense and DOE Joint Munitions Technology Development Program to use our models in the study of ammonium dinitramide under detonation conditions. We have also obtained programmatic funding to study polymers related to additive manufacturing. Furthermore, we have applied to the National Aeronautics and Space Administration's exobiology program to obtain support for studies of the synthesis of prebiotic compounds from simple starting materials under reactive conditions.

Publications and Presentations

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Pair-Plasma Creation Using the National Ignition Facility

Hui Chen (12-ERD-062)

Abstract

Relativistic electron and positron (antimatter) pair plasmas and jets are believed to exist in many astrophysical objects. Their presence would explain energetic phenomena related to gamma-ray bursts and black holes. Yet the ability to study these dense relativistic electron–positron plasmas in the laboratory has been elusive until now because no experimental platform has been capable of producing the high-temperature pair-plasma and high-flux pair jets required to simulate astrophysical positron conditions. Experimental scaling based on data from smaller, short-pulse laser experiments shows that, when completed, Livermore's Advanced Radiography Capability at the National

Ignition Facility will be the only facility where a pure pair plasma can be created. We propose to initially use short-pulse laser experiments and ultimately use the Advanced Radiography Capability to create and study electron–positron pair jets and plasmas and the consequent gamma-ray bursts that result from pair annihilation.

If successful, this project will build a new experimental platform that will create, for the first time ever in a laboratory, the high-temperature pair-plasma and high-flux pair jets required for understanding exotic, ubiquitous, yet little-understood astrophysical phenomena. The results will have an impact in the high-energy-density field and may establish the plasma physics of laser and antimatter interactions as a new sub-field in high-energy-density science.

Mission Relevance

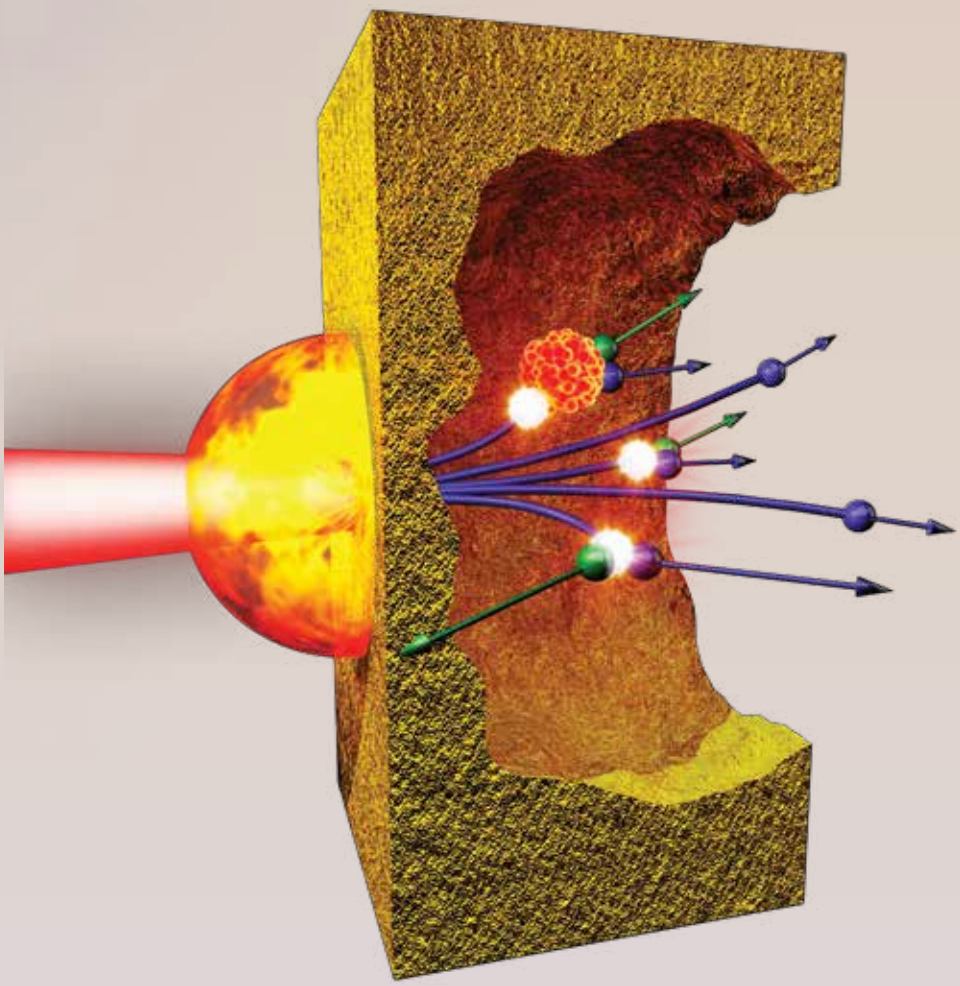
These novel high-energy-density laboratory experiments will aid in understanding astrophysical phenomena in support of fundamental and stockpile stewardship science. In addition, high-density, laser-produced positrons to be produced in this project will also enable new applications for the Laboratory's nuclear security mission. Such applications include diagnosing high-energy-density plasmas and providing a source of pulsed, mono-energetic gamma rays for radiography of dynamic processes in materials.

FY14 Accomplishments and Results

In FY14 we (1) conducted our first experiments on focusing pair jets with externally applied magnetic fields, demonstrating that such fields can effectively collimate the pair jets, increasing the pair density in the jets and reducing the electron–positron charge ratio—a critical step to achieving a charge-neutral plasma; (2) performed the initial pair jet–plasma interaction experiments using our FY13 simulation results; (3) completed our final pair-scaling experiments, (4) analyzed data produced by our experiments; (5) produced a initial design based on the scaling results for the first experiment to be conducted on the Advanced Radiography Capability laser; (6) studied the detailed physics of pair production process using various materials; and (7) analyzed the experimental data systematically and found conditions favorable to achieve pair-plasma interactions using lasers for understanding some of the most energetic events in astrophysics.

Project Summary

The successful conclusion of this project resulted in a number of significant results. We found that laser-produced positrons have several characteristics that may prove essential for creating a relativistic pair plasma. The first is that lasers can make a very large number of positrons (10^{10} to 10^{12} per shot) in a short time (10 to 100 ps). This feature, in combination with the small volume (on the scale of cubic millimeters) these positrons occupy, leads to a high density of positrons. The second characteristic is that the target sheath field can accelerate these positrons to tens of megaelectronvolts, enabling positrons to be created and accelerated to the relativistic regime in one integrated process. The third characteristic is that megaelectronvolt electrons and positrons produced from the laser–target interaction form overlapping jets behind the target,



Relativistic electron–positron pairs are produced when intense lasers interact with gold targets. The megaelectronvolt pairs form a plasma jet at the back of the target. Such jets may provide the first opportunity to study, in the laboratory, some of the most energetic cosmic events.

as depicted in the figure. In addition, we found that the emittance of laser-produced positrons is comparable to that obtained at the Stanford Linear Collider at the SLAC National Accelerator Laboratory. The nonlinear scaling of positrons as a function of laser energy is evident when the laser energy is greater than 1,000 J. Using plasma collimation, multiple-kilojoule, short-pulse laser systems, and more advanced target designs, we show it is possible to create the first relativistic high-density pair plasmas in the laboratory—a completely novel system enabling detailed study of some of the most exotic and energetic systems in the universe.

Publications and Presentations

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Williams, G. J., et al., 2014. "Calibration and equivalency analysis of image plate scanners." *Rev. Sci. Instrum.* **85**(11), 11E604. LLNL-POST-655590.

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Willingale, L., et al., 2013. "Surface waves and electron acceleration from high-power, kilojoule class laser interactions with underdense plasma." *New J. Phys.* **15**, 025023. LLNL-JRNL-618112.

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Generation and Characterization of Matter at Extreme Gigabar Pressures at the National Ignition Facility

Andrea Kritcher (13-ERD-073)

Abstract

Matter at extreme gigabar pressures occurs widely in astrophysical objects such as giant gas planets or highly evolved stars. The equation of state at these pressures—that is, the behavior of matter under a given set of physical conditions—has not been measured for any material. We propose to measure the equation of state of matter at greater than gigabar pressures (about a billion Earth atmospheres) for the first time at Livermore's National Ignition Facility (NIF). In these experiments, we will reach gigabar pressures by driving a strong shock wave through solid carbon–hydrogen or diamond spheres using a fielded and tuned NIF platform. Creating and probing material at gigabar pressures with x-ray radiography and Thomson scattering can only be done under the conditions available at NIF. Our results will provide data for benchmarking models that describe astrophysical bodies, inertial-confinement fusion experiments, and weapons physics. We also intend to develop a new x-ray scattering capability at NIF, including advanced spectrometers, and perform fusion target fabrication and design.

We expect to develop a new model to simultaneously determine the mass density and opacity profiles from radiographic measurements. In addition, we will create a platform to probe the equation of state for a variety of materials relevant to stockpile stewardship and fundamental science. The new x-ray scattering capability that will result from this project can be employed on NIF shots to provide more information on ablator implosion dynamics, which is important for stockpile stewardship.

Mission Relevance

Our high-energy-density experiments and development of a new x-ray scattering technique closely aligns with the Laboratory strategic focus areas of stockpile stewardship and high-energy-density science, as well as efforts to understand giant gas planets and highly evolved stars and in national energy security.

FY14 Accomplishments and Results

In FY14 we (1) completed radiation-hydrodynamics simulations of gigabar shock experiments in solid diamond and carbon–deuterium targets, as well as the design of a reduced preheat drive to investigate preheat conditions; (2) fabricated solid carbon–deuterium targets and coated solid diamond targets; (3) completed the

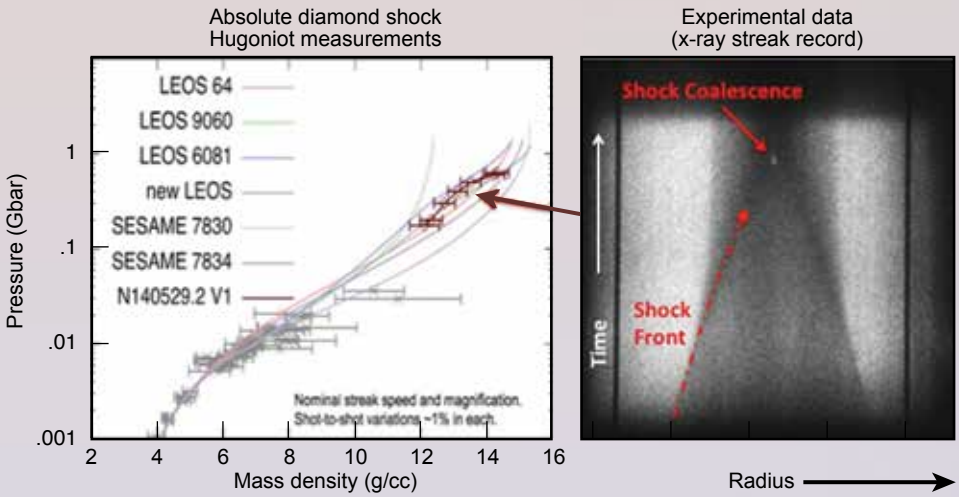
design, review, and experimental setup of three gigabar shots that were performed in FY14; (4) completed three NIF shots including a low-drive shot for platform validation, diamond equation-of-state shot at pressures greater than 500 Mbar, and a carbon–deuterium equation-of-state shot; (5) investigated alternate diagnostics to measure ion and electron temperature at shock stagnation; (6) completed the analysis of x-ray scattering data taken in FY13; and (7) completed radiography marker-layer analysis to constrain equation-of-state measurements.

Proposed Work for FY15

In FY15 we will (1) complete post-shot simulations of experiments performed in FY14, (2) field and analyze alternate diagnostics to measure electron and ion temperatures at stagnation and compare to simulations, (3) design and field experiments with radiographic marker layers to improve the simultaneous density and opacity evolution with shock-front movement from time-resolved radiographic images, (4) develop fabrication of buried marker-layer targets to be shot in FY15, and (5) complete analysis of experimental radiography and Thomson scattering data taken in FY14.

Publications and Presentations

Bachmann, B.-L., 2014. *Penumbral imaging of micrometer size plasma hot spots at shock stagnation in Gbar EOS experiments on the National Ignition Facility*. 56th Annual Meeting of the APS Division of Plasma Physics., New Orleans, LA, Oct. 27–31, 2014. LLNL-PRES-784430.



Absolute shock Hugoniot measurements (relationship of pressure to density) of diamond at pressures greater than 500 Mbar (red data points) plotted with previous equation-of-state measurements (grey data points) and Livermore equation-of-state tables (LEOS) and Sesame equation-of-state model (left). Measured transmission radiograph of 9-keV back-lighter x rays passing through a solid ball of diamond that has been shocked using a hohlraum ignition target capsule radiation drive (right).

Bachmann, B., 2014. *Self emission at stagnation of a converging spherical shock wave in Gbar EOS experiments on the NIF*. 20th Topical Conf. High Temperature Plasma Diagnostics, Atlanta, GA, June 1–5, 2014. LLNL-POST-649779.

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Hawreliak, J. A., T. Doeppner, and D. Swift, 2013. *Gbar equation of state measurements on the NIF*. LLNL-POST-617112.

Kraus, D., and T. Doeppner, 2014. *Probing matter at Gbar pressures at the NIF*. LLNL-PRES-653141.

Kraus, D., et al., 2014. *X-ray continuum emission from hot dense matter at Gbar pressures.* LLNL-POST-655188.

Kraus, D., et al., 2014. *X-ray continuum emission spectroscopy from hot dense matter at Gbar pressures*. 20th Topical Conf. High Temperature Plasma Diagnostics, Atlanta, GA, June 1–5, 2014. LLNL-PROC-654869.

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Swift, D., et al., 2012. *Gigabar material properties experiments on NIF and Omega*. AIP Conf. Proc. 1426, 477. LLNL-PROC-492261. *http://dx.doi.org/10.1063/1.3686321*

Physical States and Processes in Inertial-Confinement Fusion: Matter at Extreme Energy Density

Gilbert Collins (14-SI-003)

Abstract

A burning plasma provides a unique environment of extreme temperature and density conditions, as well as in terms of the flux of energy and particles. Currently, the modeling of burning plasma behavior relies on a fluid description. However, these models have known limitations, and a fuller kinetic description may allow a better understanding of current models and provide the opportunity to consider other burning scenarios. The demand for benchmark data and increasingly sophisticated microphysics models for high-energy-density matter is increasing as advanced laser systems approach hot-spot ignition. These laser systems manipulate high-energy-density matter to such extremes that state-of-the-art simulations are no longer predictive, and in many cases, experimental outcomes are difficult to predict. Predictive implosions at Livermore's National Ignition Facility, for example, require a new generation of microphysics models. We propose to collect benchmark data, modernize high-energy-density microphysics models, and engage a broad science community to enable predictive control of burning plasma on advanced laser systems. We intend to use ignition experiments at the National Ignition Facility to prioritize microphysics research. This effort will extend current data analysis techniques to include statistical and topological multivariable methods to test for self-consistency between microphysics models and inertial-confinement fusion data. This proposed work will lead a worldwide effort to establish a predictive control of burning plasma for fundamental science and potential future applications such as energy production.

We expect that this effort will lead to a new generation of benchmarked microphysics models, resulting in new insights for optimizing and using the burning plasma state. We

will develop new high-energy-density platforms and engage a broad scientific constituency. Specifically, this effort will benchmark equation-of-state models for target capsule ablators (used to compress the laser ignition target) and fuel and test fundamental assumptions such as the Thomas–Fermi limit relevant to quantum mechanics, which are implicit in these models. We will determine seeds for instability generated by chemistry or rapid compression rates. Key transport and kinetic quantities will be measured to benchmark simulations for optimizing burning plasma. Our results will enable more predictive simulations of inertial-confinement hohlraum target capsules, fuel assembly, stagnation pressure, and yield. In addition, this work will engage a new generation of scientists focused on high-energy-density research at LLNL and other national laboratories.

Mission Relevance

Developing an enhanced predictive capability for initiating and optimizing fusion burn is important for science-based stockpile stewardship, a core LLNL strategic mission. Our research will provide the benchmarking data and computational tools to enable more predictive manipulation of high-energy-density matter, with a specific focus of optimizing a burning plasma state at the National Ignition Facility and other high-energy lasers.

FY14 Accomplishments and Results

In FY14 we (1) performed the first measurements of the equation of state for hydrogen and its conductivity at tenfold compression, of the sound speed for hydrogen at pressures up to 3 Mbar, of the hydrogen Gruneissen coefficient (a key temperature-dependent physical characteristic responsible for thermoelastic efficiency of materials) to 3 Mbar, and of the sound speed of carbon–hydrogen ablators; (2) measured thermal conductivity for carbon–hydrogen and beryllium to several electronvolts; (3) performed the first stopping-power measurements in the warm dense-matter regime; (4) designed ablator and deuterium equation-of-state measurements for the National Ignition Facility; (5) performed the first measurements of gold emissivity in the regime relevant to fusion-ignition hohlraum target capsules; (6) performed measurement, analysis, and comparison of hohlraum L and M electron-shell spectra with theory; and (7) developed a road map for advanced nonlocal thermodynamic equilibrium theory.

Proposed Work for FY15

In FY15 we will (1) conduct a workshop on burn physics and advanced fusion concepts; (2) conduct the first equation-of-state measurements at the National Ignition Facility of ignition ablators and perhaps fuel into the many-terapascal regime; (3) extend hydrogen equation-of-state measurements to higher compression; (4) measure stopping power for warm dense matter with coupling constants greater than one and in the electron degeneration regime; (5) extend nonlocal thermodynamic equilibrium emissivity measurements to the hohlraum high-density regime; (6) benchmark and improve theory for thermal conductivity, nonlocal thermodynamic equilibrium, and equation of state; and (7) begin to identify potential ways to manipulate hot-spot conditions given emerging physics models.

Plasma Interactions with Mixed Materials and Impurity Transport

Thomas Rognlien (14-ERD-101)

Abstract

Magnetic fusion energy devices must be designed to control the interaction between the hot fusing plasma and surrounding solid- or liquid-wall materials. Exhausting plasma sputters undesirable wall material back into the confinement region, and for high-power operation, solid walls may reach melting limits, severely limiting or prohibiting operation. Predicting the behavior of such systems, especially when multiple wall materials are present, is a very challenging scientific problem. We propose to combine the expertise of Livermore's materials modeling with edge-plasma modeling and experimental diagnostics to enable validated simulation plasma and material interaction models of plasma-induced changes to tungsten and beryllium materials in magnetic fusion energy devices. A set of specific issues will be addressed for tungsten and tungsten–beryllium materials—such as those that will be used in the ITER international fusion research reactor in the south of France—including sputtering, impurity transport in plasmas, and material surface evolution. We will combine upgrades to existing plasma and material codes and validation with experimental data.

We expect to develop and apply physics-based models of turbulent and collisional impurity transport through the edge plasma to the core and compare the results with experimental data. We will assess the role of tungsten dust particles and mobilization of observed tungsten tendrils in contributing to the inward flux of impurities, compute the rate of material mixing of different materials at various locations in a tokamak magnetic fusion reactor, and the properties of this new mixed material. All these issues are central to guiding the operation of high-power tokamaks such as ITER, as well as designing new devices. The extended computation tools developed and physics insight gained will enable us to effectively address new issues that will likely arise, such as use of new materials.

Mission Relevance

Magnetic fusion energy faces the challenge of interfacing fusion plasmas with surrounding materials. Our research addresses this issue and contributes directly to the Laboratory's core competency in high-energy-density science and advanced materials and manufacturing. In addition, our project supports Livermore's initiative to grow both national and international collaborations in magnetic fusion science.

FY14 Accomplishments and Results

In FY14 we (1) added impurity species to the BOUT++ turbulence code; (2) accelerated work on the multiphysics code DUSTT/UEDGE which is used to model both dust transport (DUSTT) and edge-plasma transport (UEDGE); (3) developed the polymer-chain model for tungsten tendrils, (4) compared BOUT++ and UEDGE impurity transport simulations in two dimensions; (5) modified our original plan for 2014 installation of a laser blow-off system at

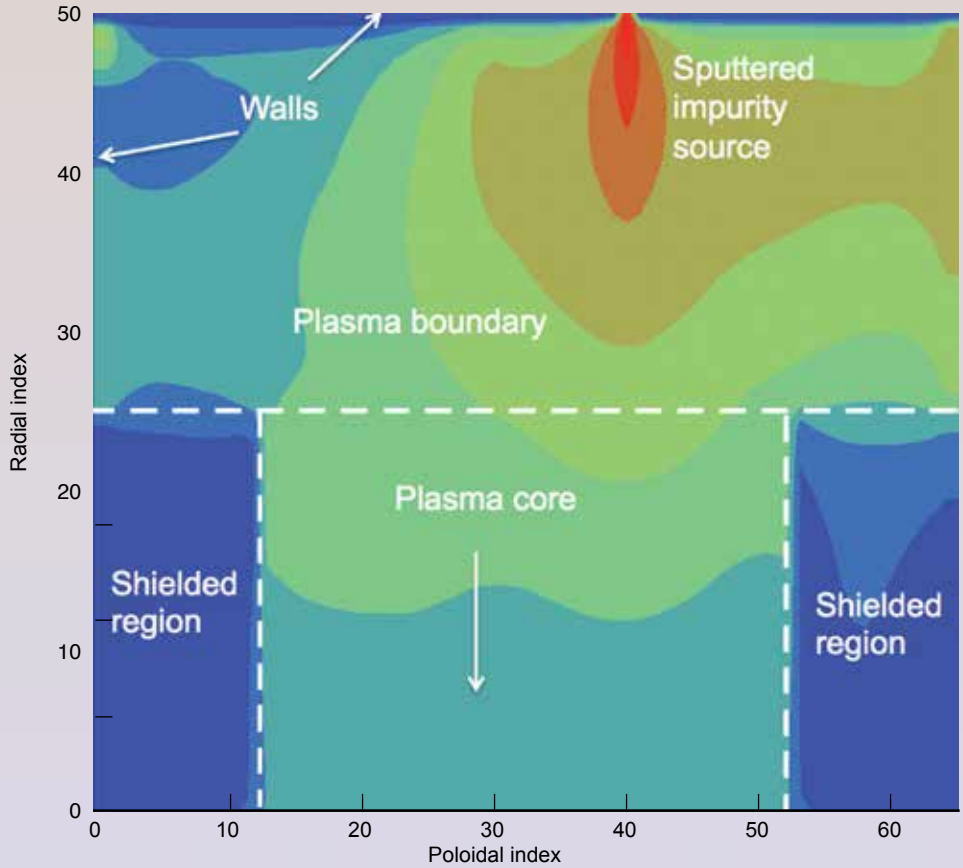
the DIII-D fusion reactor in San Diego to the NSTX-U tokamak at Princeton Plasma Physics Laboratory in 2015 because of a shift in DIII-D operations, as well as NSTX-U plans for increasing metallic wall coverage and LLNL impurity diagnostics there; and (6) accelerated modeling work by adding interatomic potentials for beryllium–tungsten systems to the LAMMPS molecular dynamics code and began simulations.

Proposed Work for FY15

In FY15 we will (1) install the laser blow-off system at NSTX-U and perform initial operation, (2) develop numerical tools to compare NSTX-U impurity emission data with UEDGE and BOUT++ simulations, (3) perform BOUT++ simulations of turbulence-driven impurity transport in NSTX-U, (4) develop a fast two-dimensional model for tungsten-ion transport to mimic three-dimensional BOUT++ results, (4) continue beryllium and tungsten–beryllium sputtering molecular dynamics simulations and begin kinetic Monte Carlo code calculations of surface mixing, and (5) compare, as data is available, tungsten experimental dust transport with DUSTT and UEDGE simulations.

Publications and Presentations

Pigarov, A., et al., 2014. "Modeling of tungsten dust transport in ITER with multi-physics code DUSTT/UEDGE." *Contrib. Plasma Phys*. **54**(4–6), 615. LLNL-JRNL-663068. *http://dx.doi.org/10.1002/ctpp.201410088*



Simulation of impurity density to quantify how wall impurities migrate into the hot plasma core region of a tokamak magnetic fusion energy device under the influence of plasma turbulence. The results are presented in a rectangular format for ease of viewing—the real geometry is that of an annulus where the radial dimension is much smaller than the poloidal circumference.

Developing a Compact, High-Power Pulsed Generator System

Robert Yamamoto (14-LW-009)

Abstract

Pulsed power sources are employed today in many arenas, where they play an essential role. In pulsed power, energy is accumulated over a relatively long period of time and then released very quickly, thus increasing instantaneous power. Examples range from their use in research employing particle accelerators and laser systems to military ordnance applications. Although condenser banks are the most commonly used pulsed-power sources, their relatively low-volumetric energy density presents a problem for applications requiring hundreds of megajoules of pulsed electrical energy. We propose to investigate a new approach to the pulsed power problem through the design, construction, and testing of a model pulsed generator. The central theme of our project is to construct and test a model based on the concept of a Halbach array of magnetic-field components. We will create the design from our computer simulations, and then compare the experimental results with those simulations. To achieve the maximum gain from the design, we will exploit flywheel technology based on rotors fabricated from a high-strength carbon–fiber composite. This new approach to pulsed energy systems will benefit greatly from the on-going development at the Laboratory of such flywheels for use in bulk energy storage.

As predicted by theory and computer simulation, we expect to achieve generator power densities that are an order of magnitude higher than the highest achieved in Compulsators, invented in the 1980s at the University of Texas and studied for several decades as one route to the solution of the power density problem. Our investigation will be aimed at experimentally confirming our theoretical predictions using a scaled-down version of the new generator that we will design and test. Our simulations predict a peak pulsed power output of 200 GW at a pulse length of 5 ms, delivered by a unit with a rotor that is 1 m in outside radius and 1.4-m long.

Mission Relevance

Pulsed power sources are used to generate and apply energetic beams and high-power energy pulses, with applications in radiation hydrodynamics and inertial-confinement fusion that support central Laboratory missions in national and energy security. In addition, pulsed power technology is central to the LLNL core competency in high-energy-density physics and measurements of material properties relevant to advanced materials and manufacturing.

FY14 Accomplishments and Results

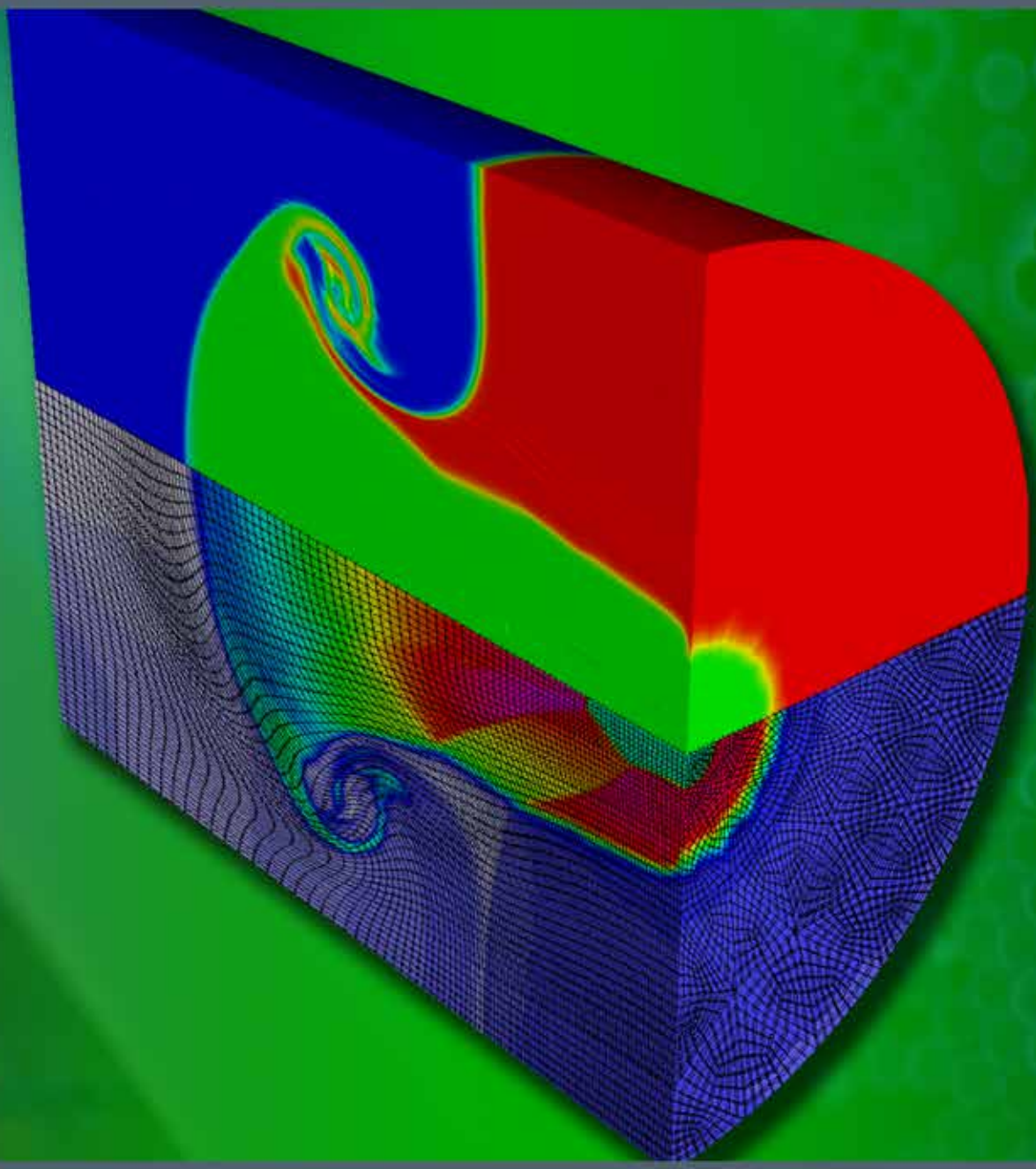
In FY14 we (1) completed the concept, preliminary, and final design for our model pulsed generator; (2) performed structural, rotational dynamics, and magnetic detailed analyses; (3) completed key fabrication drawings and contacted vendors for fabrication; (4) specified and ordered the magnetic coupler; (5) identified required sensor diagnostics as well as the vacuum vessel along with vacuum pumping

equipment; (6) established the coil design and developed coil-winding fabrication techniques; and (7) procured the two large rotors and the rotor and shaft assembly, the main bearings for the rotating system, the stator, and the litz wire (a cable used in electronics to carry alternating current) that will be used for the Halbach array coils.

Proposed Work for FY15

For FY15 we propose to (1) complete the entire fabrication drawings and procure the remaining hardware; (2) install vacuum pumping equipment and pressure-monitoring equipment on the vacuum vessel; (3) complete assembly of the high-power generator and install it in the vacuum vessel, along with all diagnostics and power feed through; (4) install the generator drive motor on the vacuum vessel and mate it to the magnetic coupler; (5) commence testing of the high-power generator; and (6) record and analyze test data and compare actual performance to modeled predictions.

High-Performance Computing



FY2014

Lawrence Livermore National Laboratory

An Open Framework to Explore Node-Level Programming Models for Exascale Architectures

Chunhua Liao (12-ERD-026)

Abstract

High-performance computing at the exascale will require node architectures that have thousands of cores, a deep memory hierarchy, and heterogeneous components. This will significantly increase the complexity of designing and adopting programming models that map applications to these architectures. Coupled with the fact that standardized node-level programming models often lag several years behind their target architectures, a significant risk exists that no model will be available for programming exascale architectures when the machines are finally deployed. Our objective is to develop an open framework to assist users, both programming-model researchers and application developers, in building node-level programming models to explore essential exascale issues. This project will evaluate and demonstrate a framework to support the construction of various programming models for heterogeneous architectures tailored to different application requirements.

Our primary deliverable is a framework that assists users in creating various node-level programming models targeting exascale architectures. The framework will be written in the C++ programming language and iteratively released under a Unix-like open-source license, providing maximum freedom for users from both research and commercial communities. Users will be able to contribute new components, thereby continually increasing the functionality provided by our framework. If successful, this project will provide important contributions to the software foundations of high-performance computing, permitting software teams to write applications and design programming models tailored to their applications.

Mission Relevance

Ensuring that applications work well with current and future high-performance computing architectures is essential for every mission at the Laboratory. As new architectures become available, programming models will need to be updated or even overhauled to better adapt applications to these new architectures. Our project will develop in-house expertise with new programming models that will help design and use high-performance computers, in support of LLNL's core competency in high-performance computing.

FY14 Accomplishments and Results

In FY14 we continued to investigate new building blocks for node-level programming models, and used these building blocks to explore the design of new programming models. In particular, we (1) added optimization building blocks for heterogeneous computing using the NVIDIA graphic processing unit, including compile-time loop collapsing for exposing more parallelism, as well as a run-time round-robin loop

scheduler; (2) demonstrated effectiveness of the building blocks by incorporating them into the OpenMP accelerator programming model of the OpenMP 4.0 specification (OpenMP is an interface for multiple-platform, shared-memory multiprocessing programming); and (3) documented the overall concept of the project and how it enables customization and hybridization using a miniature application derived from the real-world BoxLib software library (used for writing parallel, block-structured adaptive mesh-refinement applications) as a driven application.

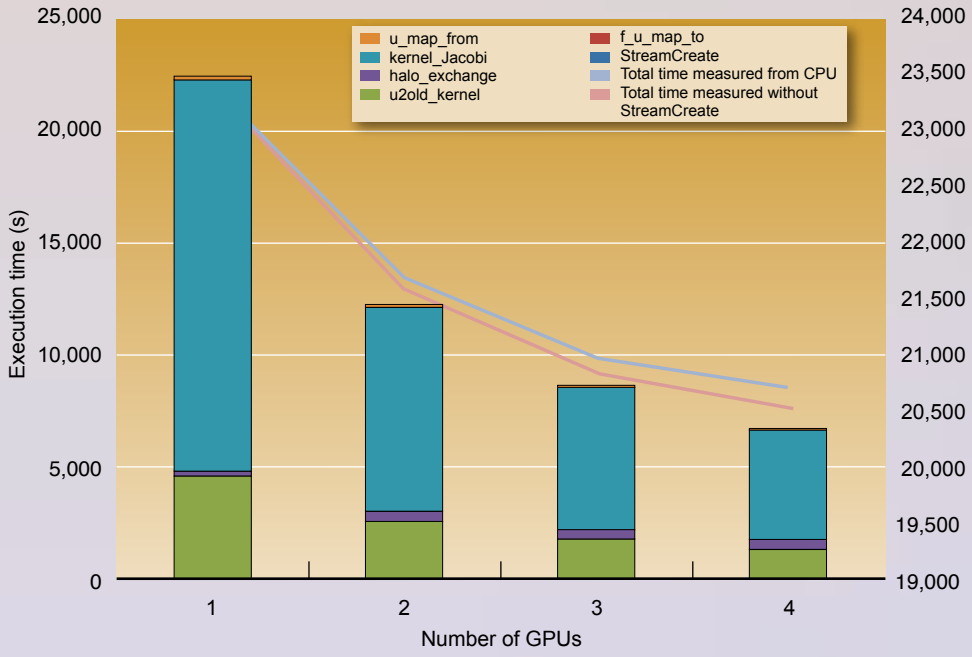
Proposed Work for FY15

In FY15 we plan to (1) document the developed building blocks and the example programming models developed by leveraging those building blocks, (2) study new building blocks that can exploit the local scratch-pad memory of computational accelerators, (3) perform further performance analysis for the example programming models—results will guide us in identifying opportunities for new optimization building blocks, and (4) progress to compliance with the OpenACC 2.0 application programming interface, adding features such as reduction operations and asynchronous computation.

Publications and Presentations

Liao, C., et al., 2013. "Early experiences with the OpenMP accelerator model," *OpenMP in the Era of Low Power Devices and Accelerators*, vol. 8122, p. 84. Springer, New York, NY. LLNL-CONF-636479.

Lidman, J., et al., 2014. *An automated performance-aware approach to reliability transformations*. Euro-Par 2014 Parallel Processing 20th Intl. Conf., Porto, Portugal, Aug. 25–29, 2014. LLNL-CONF-658972.



Using building blocks provided by our programming model framework, we built a directive-based programming model to support programming multiple graphic processing units (GPUs). This figure shows the performance obtained for Jacobi, which is a popular algorithm for solving differential equations. We obtained scalable speedup for almost all components of the Jacobi benchmark.

Lidman, J., et al., 2012. *ROSE::FTTransform—A source-to-source translation framework for exascale fault-tolerance research*. 2nd Intl. Workshop Fault-Tolerance for HPC at Extreme Scale (FTXS 2012), Boston, MA, June 25–28, 2012. LLNL-CONF-541631.

Ma, H., et al., 2013. *Symbolic analysis of concurrency errors in OpenMP programs*. 2013 Intl. Conf. Parallel Processing, The 42nd Ann. Conf., Lyon, France, Oct. 1–4, 2013. LLNL-CONF-640048.

Rahman, S., et al., 2012. *Studying the impact of application-level optimizations on the power consumption of multi-core architectures*. ACM Intl. Conf. Computing Frontiers, Cagliari, Italy, May 15–17, 2012. LLNL-CONF-599780.

A Linearly Scalable Algorithm for First-Principles Molecular Dynamics at Exascale

Jean-Luc Fattebert (12-ERD-048)

Abstract

Current molecular dynamics algorithms with $O(N^3)$ complexity—that is, requiring computational resources of calculations that increase with the cube of N, where N is the number of atoms in the system—will not be able to take full advantage of the orders-of-magnitude increase in computational power expected by the end of the decade. We will therefore develop and implement a first-principles molecular dynamics simulation technology with reduced complexity—$O(N)$ (linearly scalable) instead of $O(N^3)$—to simulate molecular systems. We will focus on making the capability truly scalable and reliable for routine use in applications involving thousands of atoms simulated with many thousands of processors. To this end, we will also develop a faster convergence solver for the sparse representation of solutions, implement the $O(N)$ algorithm needed for sparse parallel linear algebra, develop and implement an algorithm for constant-pressure simulations, and use the first-principles molecular dynamics computer code MGmol, which is based on the density functional theory used to investigate the electronic structure of many-body systems, to implement new algorithms based on real-space finite differences on a uniform mesh.

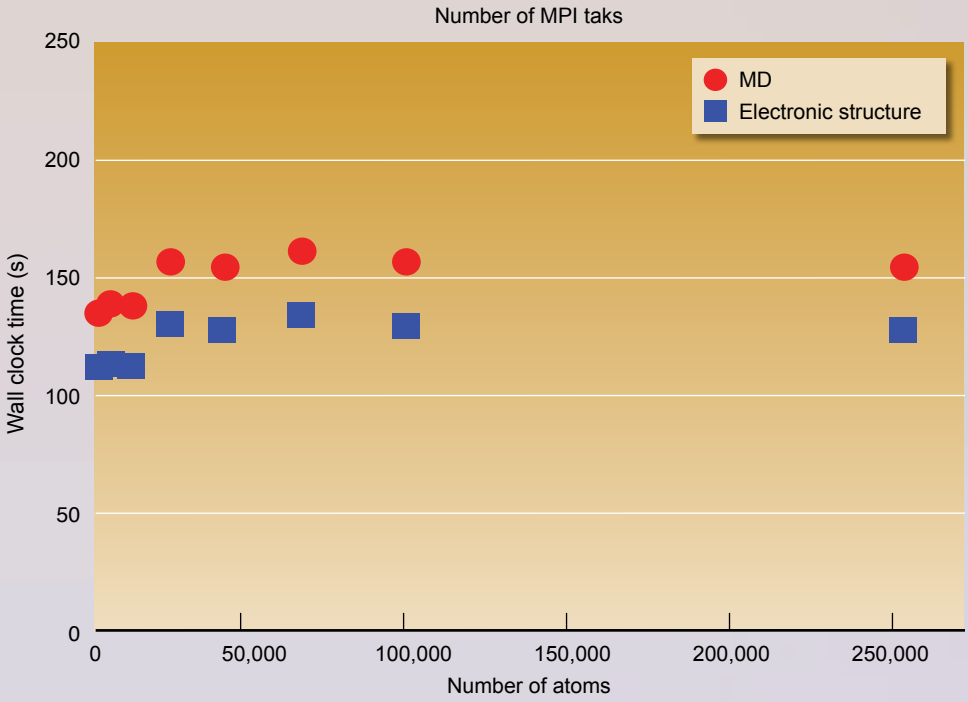
The reduced-complexity $O(N)$ algorithm we develop should be able to simulate hundreds of thousands of atoms from first principles on exascale computers. This capability will enable research with more realistic models of matter than we use today, involving, for instance, more realistic defects and more complicated molecular structures for the study of nucleation in materials or calculation of the equation of state of polymers with realistic molecular structures.

Mission Relevance

An *O(N)* complexity algorithm will enable quantum molecular dynamics calculations at an unprecedented scale and accuracy on DOEs next-generation supercomputers, providing insight at the molecular level in various fields such as materials in the extreme environment of fusion energy and the toxicity of chemical agents, in support of the Laboratory's missions in national security, energy security, and fundamental science. In addition, developing molecular dynamics simulation technology using Livermore supercomputers is directly aligned with the Laboratory's core competency in high-performance computing.

FY14 Accomplishments and Results

In FY14 we (1) completed parallelization of the atomic coordinates and related quantities and computations in our MGmol code, leading to a fully distributed code; (2) adapted various part of the code to enable scaling of input and output; (3) extended our weak-scaling studies on the Vulcan BlueGene/Q supercomputer, demonstrating excellent weak scaling up to 250,000 atoms, using 250,000 message passing interface tasks; (4) implemented, to speed up our time-to-solution, new data structures for storing data to perform linear system solutions; (5) began adding threading to the code, using OpenMP directives (an interface for multiple-platform,



Weak scaling on IBM's BlueGene/Q-class supercomputer for one first-principles molecular dynamics (MD) time step using our new *O(N)* algorithm.

shared-memory multiprocessing programming); and (6) began to apply, working with application scientists, our methodology to modeling of solutions with low concentration of charged ions.

Project Summary

We have developed a truly scalable first-principles molecular dynamic algorithm with complexity of *O(N)*, which is based on density functional theory and avoids global computational communications. The computational model uses a general nonorthogonal orbital formulation for the density functional theory energy functional, which requires knowledge of selected elements of the inverse of the associated overlap matrix. We have developed a scalable algorithm for approximately computing selected entries of that matrix, based on an approximate inverse technique. The new algorithm exploits sparsity and uses nearest-neighbor communication to provide a computational scheme capable of extreme scalability. Accuracy is controlled by the mesh spacing of the finite-difference discretization, the size of the localization regions in which the electronic orbitals are confined, and a cutoff beyond which the entries of the overlap matrix can be omitted when computing selected entries of its inverse. We have demonstrated the algorithm's excellent parallel scaling for up to 250,000 atoms on 250,000 processors, with a wall-clock time of *O(1)* minute per molecular dynamics step.

Publications and Presentations

Fattebert, J. L., 2013. *Short-range* O(N) *algorithms for first-principles molecular dynamics at extreme scales*. ACSR Exascale Math Working Group Workshop, Washington, DC, Aug. 21–22, 2013. LLNL-TR-635778.

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Osei-Kuffuor, D., and J. L. Fattebert, 2014. "Accurate and scalable *O(N)* algorithm for first-principles molecular-dynamics computations on large parallel computers." *Phys. Rev. Lett*. **112**, 046401. LLNL-JRNL-641855. *http://dx.doi.org/10.1103/PhysRevLett.112.046401*

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Whole-Heart Modeling on High-Performance Computing Systems

David Richards (13-ERD-035)

Abstract

Sudden death from cardiac arrest is the most common cause of death worldwide, accounting for close to 300,000 deaths annually in the U.S. alone. A deep and mechanistic understanding of cardiac dysfunction, with all the complex interdependencies, requires more than just conceptual models and experimental observations. Despite vast improvements in diagnostic capability, in situ measurement of a beating heart at the fidelity necessary to understand and predict its behavior is not on the horizon. Thus, computer simulation is an essential tool for improving our knowledge of cardiac dysfunction. However, cardiac simulation codes are limited in physical scope, spatial resolution, and practical duration of time simulated. We propose to take major steps toward the development of a whole-heart modeling capability that would contain electrophysiological and mechanical components coupled together in a scalable framework. We intend to use a highly scalable cardiac electrophysiological code we developed in collaboration with IBM for Lawrence Livermore's Sequoia BlueGene/Q supercomputer and leverage initial development of a mechanics code as a launching point for our whole-heart modeling effort, eventually leading to a practical whole-heart modeling capability.

We expect to develop a mechanics component for a scalable, coupled electrophysiology and mechanics code that we will use to study cardiac resynchronization therapy, such as specialized pacemakers to re-coordinate the action of the right and left ventricles. We will also develop a vision on how to extend this coupled model to include vascular fluidics. These achievements will represent major progress toward the development of a scalable whole-heart modeling capability. The increased fidelity of our simulations could enable patient-specific therapy, leading to superior devices and pharmaceuticals, with potential investment from device manufacturers, pharmaceutical companies, contract research organizations, and medical software companies.

Mission Relevance

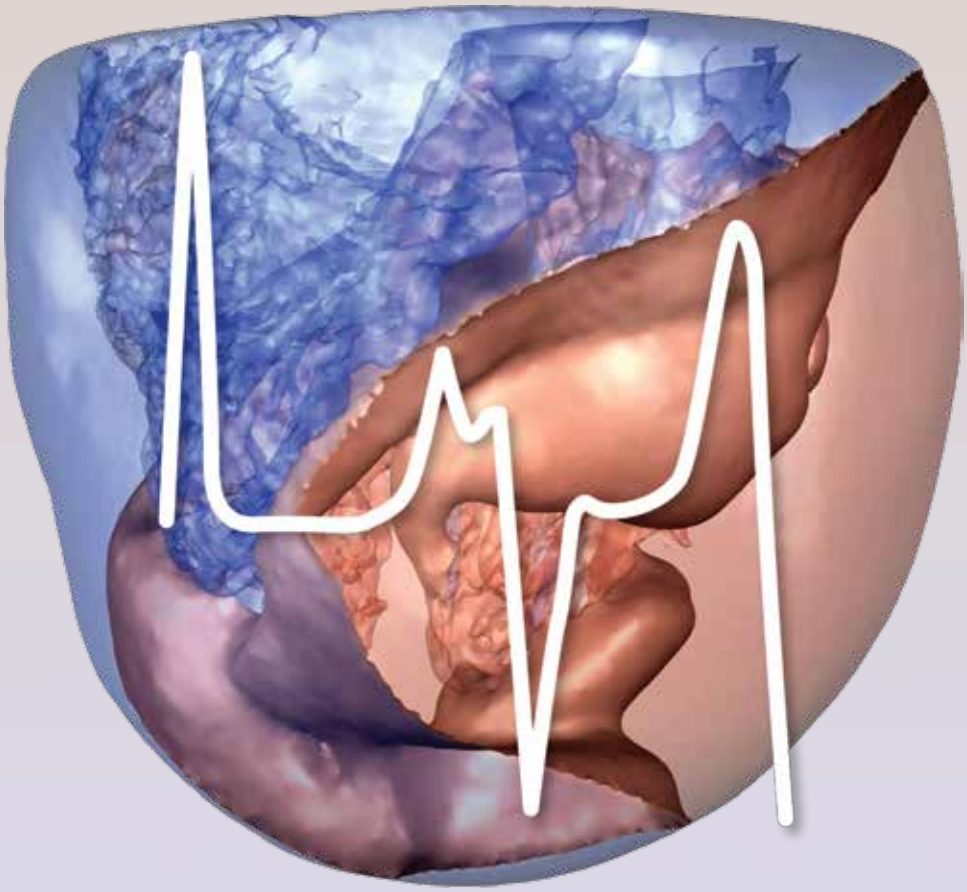
The proposed research into development of massively parallel simulation capabilities on supercomputers is relevant to the Livermore core competency in high-performance computing. Furthermore, it supports the core competency in bioscience and bioengineering to accurately simulate biological systems across scales and to quickly and efficiently manipulate these systems to improve public and environmental health. Finally, the work targets industrial investment in LLNL's computational science capability through the High Performance Computing Innovation Center, helping attract and retain an elite crop of computational scientists.

FY14 Accomplishments and Results

In FY14 we (1) performed studies of drug effects on arrhythmia (improper heart beat); (2) improved the TT06 model, which is used for modeling human ventricular single cells, to better describe action-potential decay (in muscle cells, an action potential is the first step in the chain of events leading to contraction) and improved stimulation protocols for electrophysiology simulations; (3) performed initial mechanics studies including validation against other codes and evaluation of the effect of increased mesh resolution and anatomic fidelity on results; (4) demonstrated coupling from calcium ion concentrations predicted by our electrophysiology code to the contractile model in our mechanics code; (5) improved the performance and stability of the mechanics code; and (6) completed a tool chain to go from medical image data to simulated electrocardiograms and mechanical deformation.

Project Summary

Working closely with colleagues at IBM, we have developed a whole-heart simulation capability that includes both electrophysiological and mechanical components coupled together in a scalable framework suitable for high-performance computing systems such as Sequoia. Cardiac simulations on these systems complement existing conceptual models and experimental observations to improve our knowledge of cardiac dysfunction. We used our electrophysiology capabilities to perform simulations of the effects of various drugs on



Simulation of the electrical activation of the heart. Calculated electrocardiogram traces reveal normal beats and arrhythmias.

heart function and evaluate the drugs' tendency to produce arrhythmias. For the mechanical component, we developed a new numerical approach that uses active tensions and a mixed pressure-displacement finite-element formulation to compute the mechanical deformation of a beating heart at high resolution. The result is the first iterative solver capable of handling a high-resolution mesh of the human heart with 1.7 million degrees of freedom, achieving unprecedented accuracy. Follow-on work will involve the development of a multi-scale model that can simulate drug-induced toxicity to the heart at the atomic scale, ion channel scale, and organ scale. Our ultimate goal is to develop, validate, and demonstrate an unprecedented, purely computational capability to predict drug-induced toxicity to the heart given a drug's chemical structure.

Publications and Presentations

Gurev, V., et al., 2014. "A high-resolution computational model of the deforming human heart." *Biomech. Model. Mechanobiol*. DOI 10.1007/s10237-014-0639-8. LLNL-JRNL-655988.

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Richards, D. F., et al., 2013. "Toward real-time simulation of cardiac electrophysiology in a human heart at near-cellular resolution." *Comput. Meth. Biomech. Biomed. Eng*. **16**, 802. LLNL-JRNL-607581.

Task Mapping on Complex Computer Network Topologies for Improved Performance

Abhinav Bhatele (13-ERD-055)

Abstract

As processors have become faster over the years, the cost of a prototypical computing operation, such as a floating-point addition, has rapidly grown smaller. On the other hand, the cost of communicating data has become proportionately higher. Maximizing data locality and minimizing data movement, both on and off the server node, is critical to optimizing communication and overall application performance as well as reducing energy costs. We propose to produce tools to analyze parallel applications for communication inefficiencies, investigate techniques, and develop models to understand network congestion on supercomputers, and design, implement, and evaluate algorithms for mapping tasks in a parallel application to the underlying computer network topology to improve performance. Our work will focus on Livermore codes running on parallel machines at LLNL.

The proposed tools will help code-development teams to decide whether the communication in a computer application can benefit from better mapping. The tools

will also help us in identifying phases in the application that are most severely affected by congestion. We expect this work will lead to development of tools that can measure computational resource competition (contention) during an application run. The analytical models will help us understand why certain mappings lead to higher overall congestion or more hot spots on the network. Our deliverable will be mapping algorithms that can be applied to a parallel application to optimize its communication performance. Optimizing the majority of applications that run on supercomputers can reduce the workload for Livermore computing resources, increasing their efficiency in terms of job throughput and enabling more science simulations to be performed with the same resources. Our work will also help ensure that LLNL achieves maximum utilization of its computational capabilities as we approach the exascale era of a quintillion floating point operations each second.

Mission Relevance

The proposed work is directly aligned with Livermore's score competency in high-performance computing. Our work will include fundamental computer science research on large-scale graph-embedding algorithms, and the tools developed will be used to improve the performance of highly scalable high-performance computing applications for LLNL programs.

FY14 Accomplishments and Results

In FY14 we (1) characterized the communication behavior of parallel applications such as pF3D (a laser–plasma interaction code), AMG (an algebraic multigrid method), and MILC (a lattice quantum-chromodynamics code); (2) developed a message-level, congestion-aware, iterative model for network traffic on a network based on Dragonfly topology, which enables more-efficient router connectivity in a computer network; (3) implemented a trace-driven network simulator over a massively parallel discrete-event simulation framework; (4) developed generic mapping algorithms that optimize for different metrics, adding support for various graph partitioning and mapping libraries in a generic task-mapping framework; (5) explored mapping and graph-partitioning algorithms with select Laboratory programmatic codes; and (6) studied the impact of job placement and inter-job interference on application performance on machines that deploy the Dragonfly network.

Proposed Work for FY15

In FY15 we propose to (1) tailor task-mapping algorithms for specific LLNL simulation codes, (2) evaluate the performance benefits from task mapping of various application codes, and (3) develop an accurate understanding of supercomputer networks and develop congestion models for various topologies.

Publications and Presentations

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Scalable High-Order Computational Multiphysics at Extreme Scale

Charles Still (14-SI-002)

Abstract

Simulation via high-performance computing is an essential cross-cutting capability of the Laboratory. Most multiphysics simulation tools were developed in an era when floating-point operations were the performance-limiting factor. However, data motion and memory capacity are now the limiting factors because energy considerations are driving the design of high-performance computing architectures. This effectively inverts the computing model used for the design of many current codes. We propose

to establish a new breed of multiphysics simulation capabilities based on high-order finite-element formulations, where algorithms are designed from inception to take advantage of future hardware features. This will be accomplished by developing coupled high-order finite-element numerical methods for multiphysics simulations involving arbitrary Lagrangian–Eulerian hydrodynamics (mathematical description of fluid flow in a shocked substance), multigroup radiation-diffusion, and magneto-hydrodynamics. The numerical algorithm development will be complemented with a computer science research effort to show that high-order simulation technology is scalable, well-suited to advanced extreme-scale architectures, and offers multiphysics modeling capabilities superior to current algorithms.

If successful, we expect to deliver new high-order multiphysics algorithms, together with a prototype implementation that obtains high performance on multiple disparate advanced computer architectures as a proof of principle. Once proven, the concepts, methodologies, and many of the computational techniques can be extended to apply to diverse science and engineering areas. Our ultimate goal is to establish the mathematical and computer science research foundation for a new breed of multiphysics codes, well suited to emerging architectures, which will expand the state of the art in high-fidelity modeling. This effort will continue the long history of LLNL leadership in the development of world-class simulation technology.

Mission Relevance

Developing improved multiphysics codes are central to the Laboratory's strategic focus area in stockpile stewardship. Our goal is to demonstrate a new simulation capability—a scalable high-order, coupled multiphysics simulation—through development of a performance-portable code designed to work well on emerging advanced architectures that will form the basis for the technology platforms of LLNL's Advanced Simulation and Computing. Our research will provide a strong base in building an exascale (1,000-petaflop operating system) computing initiative, and supports the core competency in high-performance computing to simulate the behavior and performance of complex systems and expanding capabilities to exascale computing and beyond.

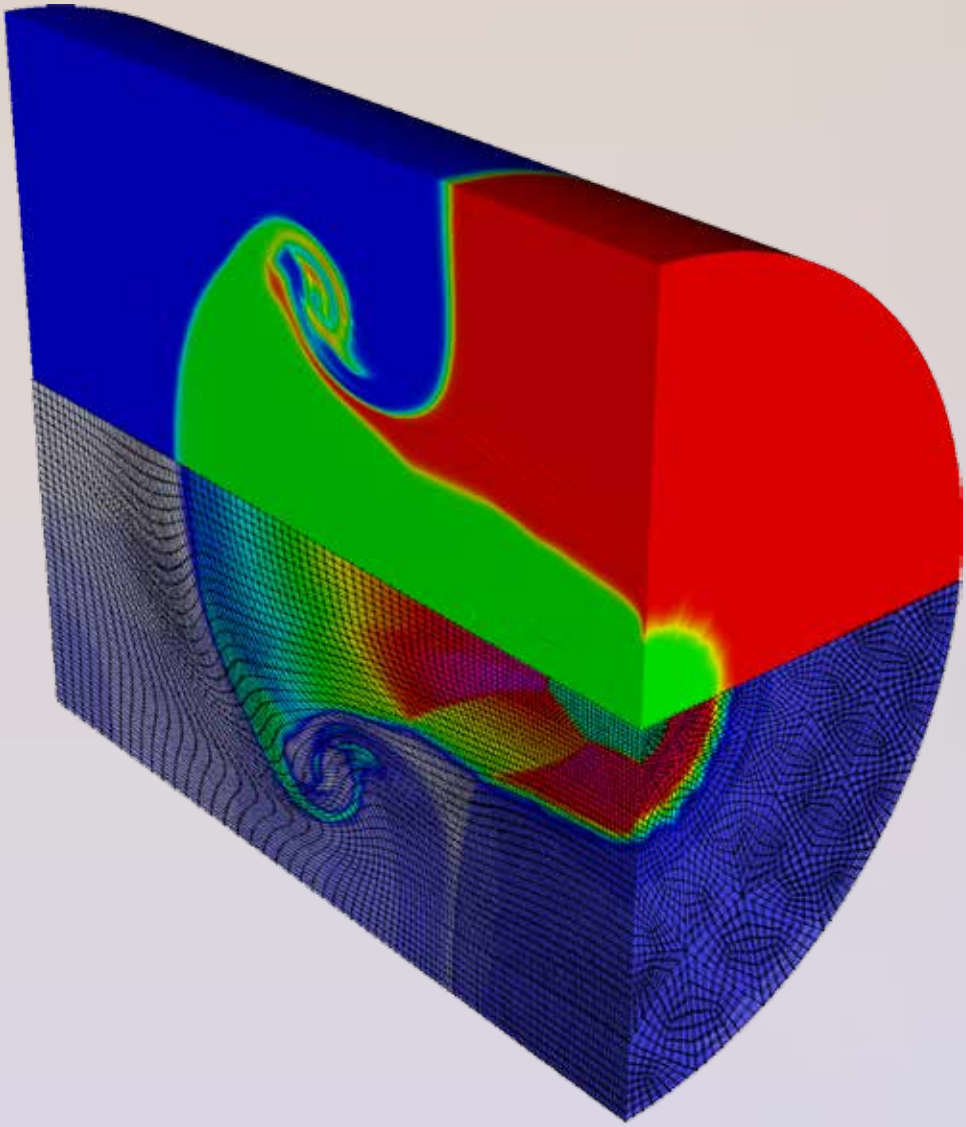
FY14 Accomplishments and Results

In FY14 we (1) completed, implemented (in the BLAST high-order finite-element hydrodynamics software), and demonstrated the scalability of the high-order, single-material curvilinear arbitrary Lagrangian–Eulerian algorithm; (2) developed and implemented the multiple-material Lagrangian algorithm and high-order generalizations of closure models; (3) completed a multiple-material arbitrary Lagrangian–Eulerian algorithm using high-order material indicator functions and applied it to two- and three-dimensional arbitrary Lagrangian–Eulerian benchmarks; (4) proposed a new high-order method for ensuring monotonic radiation-diffusion fields; (5) developed a performance model and assessment of gas dynamics in Lagrangian hydrodynamics; (6) mapped the model to future architectures; (7) identified performance gaps in the model in the current

implementation; (8) investigated code transformations and new finite-element algorithms to address the gaps, leading to a 2- to 12-fold speedup in the Lagrange phase; and (9) began study of the arbitrary Lagrangian–Eulerian performance characteristics.

Proposed Work for FY15

In FY15 we will (1) propose a high-order finite-element numerical method for multigroup diffusion, (2) explore the accuracy and robustness of the new approach on challenging two- and three-dimensional radiation-diffusion benchmarks, (3) develop algorithms for high-order implicit and explicit coupling to arbitrary Lagrangian–Eulerian hydrodynamics, (4) investigate methods for coupling radiation material properties (opacities) to multiple-



Our new high-order finite-element algorithms enable first-ever high-order multiple-material arbitrary Lagrangian–Eulerian hydrodynamic simulations on highly unstructured three-dimensional meshes. Shown are the mesh and total density (bottom) and the three materials (top) from a parallel multiple-material simulation in the BLAST shock hydrodynamics code using third-order spatial and time discretizations (transforming continuous attributes into discrete ones).

material arbitrary Lagrangian–Eulerian algorithms, (5) deliver a performance model and assessment for stress and strain in a Lagrangian hydrodynamics code and a subset of multiple-material arbitrary Lagrangian–Eulerian algorithms, (6) map the performance model to future architectures, (7) identify performance gaps to the model in the current implementation, and (8) explore mechanisms for closing those gaps in the multiple-material arbitrary Lagrangian–Eulerian subset.

Publications and Presentations

Anderson, R., et al., 2014. *A high-order finite element approach for treating multi-material zones in ALE hydrodynamics*. 11th World Congress Computational Mechanics (WCCM XI), Barcelona, Spain, July 20–25, 2014. LLNL-PRES-657195.

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Parallel Time Integration for High-Performance Computing

Jacob Schroder (14-ERD-013)

Abstract

Growth in high-performance computing will come from more central processing unit cores, not faster clock speeds. Previously, faster clock speeds decreased the compute time per time step, and thus allowed for more time steps while not increasing the overall compute time. However, clock speeds are no longer increasing, which will inevitably lead to increases in compute time. We propose to develop an algorithm to compute multiple time steps simultaneously in parallel, and thus accelerate time-stepping methods and provide a solution to the time-integration bottleneck. We will apply this algorithm iteratively and with scalability using multigrid methods, targeting parabolic and hyperbolic equations. Multigrid achieves its efficiency by applying two complementary processes, smoothing and coarse-grid correction, over successively coarser grids for a given problem. We propose a nonintrusive multilevel algorithm that uses successively coarser time scales to accelerate the solution at the original fine-time scale.

By computing multiple time steps in parallel, we expect our proposed algorithm to expose new parallelism in the time dimension and have the potential to dramatically decrease the overall time to solution for time-stepping methods and to allow for greater machine utilization on future computer architectures. Most current methods such as the parallel-in-time algorithm and its variants for solution of the general

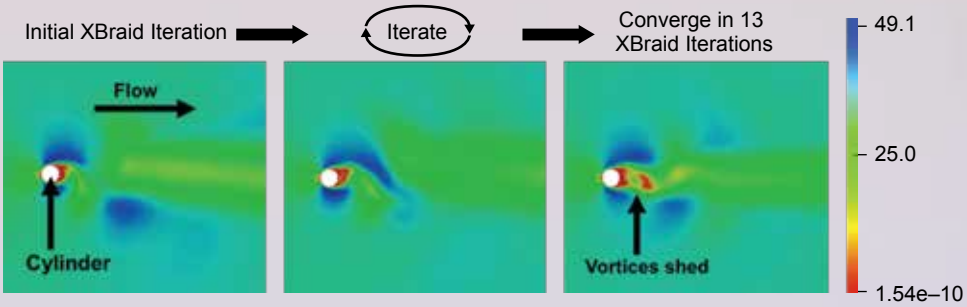
nonlinear system of ordinary differential equations are only two-level methods, and hence exhibit limited concurrency. We will develop new scalable, multigrid techniques for parallel time integration. While multilevel parallel-in-time methods may seem improbable, preliminary proof-of-concept results using the MATLAB computing language for algorithm development already shows optimal results for parabolic problems using implicit or explicit schemes.

Mission Relevance

Our proposed algorithm to expose new parallelism in the time dimension will allow Laboratory codes to run faster by taking advantage of the huge increase in concurrency coming with exascale computing, and supports high-performance computing and simulation, along with several LLNL mission areas that depend on these computing resources. Essentially, all Livermore multiphysics production codes make use of explicit time stepping, and are therefore subject to the serial time-integration bottleneck. The need for parallel time integration is imminent, and developing this technology and associated expertise helps the Laboratory maintain its leadership in high-performance computing ecosystems.

FY14 Accomplishments and Results

In FY14 we (1) validated our parallel-in-time method for model parabolic problems by showing parallel speedups of tenfold for the two-dimensional model problem and sixfold in three dimensions; (2) tested and validated the method for convection-diffusion problems and problems with a variable diffusion coefficient; (3) continued our theoretical study, which has already yielded sharp two-grid convergence bounds; (4) began our theoretical and practical study of hyperbolic problems with the linear advection equation; (5) implemented a general parallel-in-time software framework (XBraid) in the C programming language, whose design provides constructs that map efficiently to typical machine instructions and which interfaces with the MFEM (multi-layer finite-element method) discretization library—this framework undergoes



Our parallel-in-time software, XBraid, is applied nonintrusively to a compressible Navier–Stokes code for computational fluid dynamics, Strand2D. The test problem models vortex shedding over a cylinder. The maximum parallel speedup observed over sequential time stepping is 7.5 times faster than with other methods. Here, we see the solution at the final time step as XBraid iterates over the entire space–time domain. XBraid converges in only 13 iterations to the solution at the 5,120th time step.

regular regression testing and group review to ensure high quality; and (6) began our theoretical and practical study of hyperbolic problems with linear advection.

Proposed Work for FY15

In FY15 we will (1) focus on hyperbolic problems, beginning with linear hyperbolic equations in one dimension (e.g., wave and advection), moving to two- and three-dimensions, if time allows; (2) investigate the effect of shocks on coarse timescales and explore implicit and explicit time-integration schemes, which requires simultaneous coarsening in space—this is a difficult problem area for which we maintain the risk-mitigating alternative of an intrusive full space–time approach; and (3) continue work from FY14 concerning parabolic problems and extend our parallel software framework to carry out our parallel validation studies.

Publications and Presentations

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Computation Power at Scale

Barry Rountree (14-ERD-065)

Abstract

The field of high-performance computing faces a profound change as we move towards exascale computation with one quintillion floating point operations per second. For the first time, users will have to optimize codes in the presence of limited and variable electrical power. While it may be initially possible to hide these limitations by over-provisioning and under-utilizing scarce power resources, we will demonstrate how addressing these limitations can lead to full utilization of power resources and an order-of-magnitude improvement in throughput. We propose to optimize the three difficult conditions of model creation for performance prediction, job scheduling, and run-time optimization, all under specific power bounds. We will investigate a hardware over-provisioning strategy, in which more compute nodes are resident in the supercomputing center than can be powered fully. Intuitively, hardware over-provisioning allows the cluster to use maximum machine-room power, and therefore achieve maximum performance, by judicious scheduling of per-node power. To do so, we will leverage LLNL's hardware resources, our expertise in job scheduling, and continuing relationships with our academic partner, the University of Arizona.

Exascale computing presents a new performance problem of how to best get the most science out of each watt, rather than out of each node. Any code run at scale will have to address this issue, and if we do not have solutions ready for code teams when we take delivery of our first power-limited systems, the result will be unnecessarily poor performance. Our ultimate goal is to influence the design of the first several generations of exascale systems and their software ecosystems to maximize performance per watt of power. We will produce a configuration-based model, power-aware job scheduler, and run-time system. We expect to influence the design of exascale systems, having demonstrated that power-aware approaches will reliably result in significant performance improvements.

Mission Relevance

Many core missions in national and energy security at the Laboratory are dependent on the predictive simulation capability of large-scale computers, which are moving into the exascale realm. Exascale systems will be intrinsically power limited. Our research will enable optimized software in a new era of power-constrained supercomputing, in support of LLNL's core competency in high-performance computing.

FY14 Accomplishments and Results

In FY14 we (1) implemented a chained linear-programming model to determine a tight bound on theoretical best performance of arbitrary code under arbitrary power bounds, (2) partnered with the University of Arizona to add a postdoctoral researcher dedicated to this project, (3) modified the SLURM (Simple Linux Utility for Resource Management) simulator to test multiple power-bound scheduling heuristics, and (4) created a power lab that will allow power and thermal measurement and control on cutting-edge computer resources, including those from Intel, Advanced Micro Devices, and NVIDIA computer manufacturers. The software we are developing is being incorporated into the Red Hat Enterprise Linux operating system and Performance Application Programming Interface software.

Proposed Work for FY15

In FY15 we propose to focus on power balancing between computational jobs, assuming that applications inform the run-time system of their needs. Specifically, we will (1) develop a complete multiple-node model, (2) develop new power-aware scheduling algorithms, (3) add several features to the run-time system functionality and begin internode power balancing, (4) begin additional work enabled by the power lab to characterize performance over power and thermal bounds, and (5) characterize the power and performance of the InfiniBand computer network communication link, nonvolatile random access memory, accelerators, and solid-state drives.

Inertial-Confinement Fusion Science and Technology

FY2014

Lawrence Livermore National Laboratory

Hydrogen Ice Layers for Inertial-Confinement Fusion Targets

Bernard Kozioziemski (12-ERD-032)

Abstract

Targets for inertial-confinement fusion comprise layers of condensed hydrogen fuel inside spherical capsules. The layers must be easily reproducible and very smooth. Numerous experiments have shown that these requirements can only be met by using a nearly perfect single crystal of solid hydrogen. The formation of these high-quality layers depends on creating and isolating a single crystal of the solid and then slowly cooling the melt to freeze the remaining liquid. The current success rate of this process is subject to the random nature of nucleation and the resulting seed crystal used to grow these layers. This method results in a range of layer qualities, many of which do not meet target specifications. This reduced yield currently constrains the shot rate for layered targets that can be achieved at the National Ignition Facility and other laser facilities. We propose to develop a deterministic seeding process leading to reproducible high-quality target ice layers.

If successful, this project will provide the scientific understanding needed to develop a layering process and modifications to the target that will allow high-quality layers to be grown reproducibly in a predictable time. In addition to the scientific insight into heterogeneous nucleation that will be gained, this work will also improve the Laboratory's ability to conduct inertial-confinement fusion research by reducing and making more predictable the time required to perform shots.

Mission Relevance

This project advances inertial-confinement fusion research and therefore supports the Laboratory's strategic focus area in inertial-confinement fusion science and technology relevant to LLNL missions in stockpile stewardship, energy security, and high-energy-density science.

FY14 Accomplishments and Results

In FY14 we (1) measured super-cooling of hydrogen and deuterium for 28 substrates, and found that these ranged in temperature from 0.005 to 0.3 K—argon and krypton provided the smallest super-coolings; (2) performed molecular dynamics simulations and showed that hydrogen nucleates on krypton because of their overlapping potential minima—a test of electro-freezing found no nucleation effect of the electric voltage up to 600 V; (3) developed a layering simulator to predict the impact of process changes on layering outcomes, and showed that both helium-3 age and correct identification of a good layer are of fundamental importance; (4) tested a predictive control method to improve the reliability of isolating a single seed—it melted the solid in half the time of the current method; (5) found a linear mass uptake to be caused by

shell geometry and not a change in crystallographic direction; and (6) modeled radio-frequency heating using non-resonant frequencies and found that the power coupled into the deuterium vapor phase was small compared to the shell.

Project Summary

We have developed an experimental system that can readily be used to test the super-cooling of hydrogen on new template materials as they become available, as well as a framework for understanding the results. The rare-gas solids were found to promote nucleation of solid hydrogen better than other materials and were important in aiding the process of understanding super-cooling effects. Highly ordered graphite was found to promote solid nucleation nearly as well as the rare gases, and is more practical to implement. We have shown that an applied electric field does not result in nucleation of solid hydrogen from the liquid phase. A model of the thermal transport inside of a fusion shell, including the effects of evaporation and condensation, was developed to explain the observed seed location in National Ignition Facility experiments. The model explains that a cold location forms because of the position-dependent evaporation rate, and that the migration of the cold location developed because of a change in diffusion rates as helium-3 builds up in the shell. Finally, we developed a process model to test ideas for improving the rate of producing ignition-quality fuel layers. The National Ignition Facility program will test the highly ordered graphite seed attached to a fusion target shell. We will collaborate with the Laboratory for Laser Energetics at the University of Rochester and the Schafer Corporation in Livermore on identifying additional template candidates and testing these with the experimental platform created for this project.

Publications and Presentations

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Van Cleve, E., et al., 2014. "Hydrogen crystallization in low-density aerogels." J. *Phys.: Condens. Matter.* **26**. 225004. LLNL-JRNL-663197.

Zepeda-Ruiz, L., R. Dylla-Spears, and B. Kozioziemski, 2013. *Molecular dynamics simulations of nucleation and growth of D-ice layers.* LLNL-PRES-589795.

Next-Generation Process for Tritium Recovery from Fusion Power-Plant Blankets

Susana Reyes (13-ERD-056)

Abstract

Laser inertial fusion energy can achieve a self-sustained fuel cycle by breeding tritium in a lithium blanket. The leading candidate for efficient tritium extraction is a lithium halide chemistry system that uses centrifugal contactors to extract tritium into the salt phase and then recovers it via electrolysis. The objective of our project is to develop a safe and reliable process for tritium recovery that replaces volatile and corrosive halides with more benign chemical compounds and eliminates the need for rotating machinery. Key goals include demonstration of extraction feasibility; development of a design methodology for the electrolyzer unit, including development of computational fluid dynamics models; and advancing the development of a reliable non-centrifugal contactor to eliminate a key source of vibration and operational risk.

The proposed work, if successful, will enable replacing the problematic lithium halide salts with benign lithium hydroxide or perhaps lithium carbonate, thereby avoiding corrosion and volatility problems. We will develop a new contactor that will eliminate the need for high-temperature, high-speed centrifugal contactors. Finding an alternative and efficient tritium recovery process from lithium would have a major impact on the design, operation, and cost of tritium separation processes envisioned for virtually all planned fusion reactors. Records of invention and provisional patents have been prepared to protect intellectual property, thereby providing the Laboratory with a unique position in the event that the promise of the new process for tritium recovery is realized.

Mission Relevance

The development of this next-generation tritium recovery process for laser inertial fusion energy will be key for the optimization of the fuel cycle and will ultimately allow for an attractive and self-sustained solution for closing the fusion fuel cycle. A new enabling technology for tritium recovery from fusion blankets should emerge, which will provide energy security benefits beyond laser inertial fusion energy to both the DOE and NNSA.

FY14 Accomplishments and Results

In FY14 we (1) re-scoped the project, in response to a limited budget, with the goal of focusing on completing theoretical models and associated analysis; (2) performed a review of the existing model for the contactor and addressed inconsistencies; (3) developed a systems model for the blanket system to identify key parameters for system performance and minimization of tritium inventories; (4) performed additional computational fluid dynamics simulations to assess the efficiency of separation of lithium and salt; and (5) integrated the blanket system in an overall model for the fuel cycle to perform a systems assessment for a closed fusion fuel cycle based on a lithium blanket.

Proposed Work for FY15

In FY15 we will (1) complete all milestones for the conceptual design of the alternative contactor and separator system with no moving parts, (2) perform an initial demonstration of alternative chemistry that will allow the replacement of hazardous halides with a benign electrolyte, (3) perform a series of experiments to determine key aspects of the mass transfer kinetics that are crucial for determining efficiency of the process, and (4) complete and document an overall system-level model for the fuel cycle, including blanket systems, to allow for systems studies of a fusion fuel cycle using a lithium blanket.

Publications and Presentations

Reyes, S., 2013. "LIFE: A sustainable solution for developing safe, clean fusion power." *Health Phys.* **104**, 641. LLNL-TR-576952.

Reyes, S., et al., 2013. "LIFE tritium processing: A sustainable solution for closing the fusion fuel cycle." *Fusion Sci. Tech.* **64**, 187. LLNL-TR-576952.

Transient Loading Effects on Structural Materials for Laser Inertial Fusion Energy

Ryan Hunt (13-ERD-058)

Abstract

Our objective is to assess—quantitatively and in an accelerated manner—the effect of transient loads caused by thermal, pressure, and vibrational pulses on structural steels in an inertial fusion energy engine. Although the behavior of the steels is reasonably understood in steady-state conditions, the greatest threat to an inertial fusion energy engine will be a combination of thermally activated creep from high operating temperature and pulsed stresses from x rays, pressure waves, and subsequent surface cracks. No existing knowledge base applies to this regime, which is critical to building a predictive, benchmark model of materials performance to guide engine design. We will address this shortage by building and performing cyclic, creep, and thermal pulse experiments in appropriate environments and at relevant stresses.

We will produce a knowledge base of material behavior that encompasses the mechanical properties of structural steels in relevant environments, including elevated temperature fatigue, creep, and crack propagation, as well as the accelerative effects on the properties of the environment itself, specifically molten lithium with and without impurities, hot xenon, and molten lead. We will also develop an engineering model with these data as a benchmark. This will significantly expand our knowledge of the survivability of structural materials in these extreme environments and thereby enable us to better judge structural material lifetime and determine the optimum balance between risk and performance in an inertial fusion energy engine design.

Mission Relevance

This project will improve our understanding of candidate structural materials for achieving an inertial fusion energy engine as a potentially revolutionary carbon-free energy technology, in support of the Laboratory's strategic focus area in inertial-confinement fusion science and technology and core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

Because of an early decision to modify our scope in proportion to allotted budget, our accomplishments have focused exclusively on mechanical fatigue characterization rather than also examining creep and thermal pulse cycling. Specifically, in FY14 we established experimental capabilities to assess the mechanical properties of relevant structural steels in a platform that can be used to evaluate high-cycle fatigue and fatigue crack growth in environments such as high-temperature argon and molten lithium (see figure). In addition, we measured fatigue cracking properties of alloy materials with respect to temperature and environment at room and elevated temperatures.

Proposed Work for FY15

In FY15 we will (1) examine the constant-strain fatigue life of relevant structural steels in relation to temperature and environment, which is an unexplored area of knowledge important to inertial and magnetic fusion; (2) develop an initial characterization of additively manufactured materials, and develop a collaboration with the Naval Postgraduate School in Monterey, California, with the objective

Our experimental facility capable of loading samples under high-cycle fatigue at temperatures up to 600° C while submerged in a liquid lithium bath.

of studying solid-state spray deposition of oxide-dispersion strengthened steels; and (3) characterize test parts from this collaboration and from Livermore's on-site additive manufacturing processes.

High-Temperature Plasma Chemistry Kinetics Test Bed

Michael Armstrong (14-ERD-077)

Abstract

The field of high-temperature chemistry in partially ionized plasmas underpins the application of plasma-enhanced chemical vapor deposition in the semiconductor industry, and is of great importance for both magnetic fusion and inertial fusion energy. We propose to experimentally determine chemical concentrations and kinetics, in both the gas phase and at surfaces, of carbon and lead compounds in a high-temperature noble gas plasma. We will also investigate the reaction path of hydrogen (as a proxy for deuterium and tritium) for recovery and to avoid deposition on the first wall in a fusion energy chamber—the response of the chamber wall to plasma-environment chemistry is substantially unknown. We will use a plasma-torch-based flow vessel with optical diagnostics to characterize temperature and species concentrations under conditions similar to those found in laser fusion energy chambers. These data will be synthesized into a comprehensive model able to represent the gas phase and surface chemistry across the whole range of fusion energy operating conditions. In addition, the development of detailed kinetic models and quantitative experimental data will have direct relevance to plasma-environment chemistry and plasma-enhanced chemical vapor deposition.

We expect to experimentally determine chemical concentrations and kinetics, in both the gas phase and at surfaces, of carbon and lead compounds in a high-temperature noble gas plasma. These data will be synthesized into a comprehensive model able to represent the gas phase and surface chemistry across the whole range of operating conditions relevant to fusion energy. To provide a set of self-consistent conditions constrained by a system model, we will use the characteristic parameters for an inertial fusion system to define the nominal phase area of interest for our investigation. However, the methods, models, and data resulting from this project will be relevant to the broader fusion community and to the wider subjects of surface deposition and material synthesis (particularly carbon chemistry) via plasma-enhanced chemical vapor deposition.

Mission Relevance

The success of this effort will help establish technical foundations to support the LLNL strategic focus area in inertial-confinement fusion science and technology and core competency in high-energy-density science, and will support a potential future role in the increasingly important plasma-based processing industry.

FY14 Accomplishments and Results

In FY14 we (1) established a collaboration with Stanford University for development of the inductively coupled plasma system and obtained several plasma flow reactors, including a commercial instrument (currently at LLNL), a custom atmospheric pressure unit, and a custom low-pressure unit (both at Stanford); (2) performed calculations for the chemical species of interest, to guide the experimental design; (3) narrowed the possible diagnostic techniques to two-photon laser-induced fluorescence for hydrogen and oxygen, and narrowband infrared absorption for detection of small molecular species; (4) evaluated implementation of a residual gas analyzer for the detection of larger molecules, given the likely small concentration of larger molecular species (hydrocarbon–hydrocarbon chains and rings); (5) performed flow calculations for reactors to address issues such as temperature and turbulent mixing; and (6) tested the atomic hydrogen diagnostic and purchased quantum cascade lasers for narrowband infrared detection of acetylene, which is a precursor to the formation of graphene and graphene-derived species such as carbon nanotubes.

Proposed Work for FY15

For FY15 we will continue simulations to guide the selection of optical diagnostics at LLNL, in parallel with the development of the inductively coupled plasma system. Specifically, we will (1) implement nonlinear detection techniques, such as two-photon laser-induced fluorescence and degenerate four-wave mixing, that are sufficiently sensitive; (2) implement a small residual gas analyzer, if simulations indicate substantially lower concentrations of large molecules of more than a few atoms; and (3) investigate, in parallel, the mechanical properties of cryogenic xenon to determine its suitability as a material for a hohlraum target capsule for laser fusion ignition.

Information Systems and Data Science

FY2014

Lawrence Livermore National Laboratory

Adaptive Sampling Theory for Very-High-Throughput Data Streams

Ana Paula de Oliveira Sales (11-ERD-035)

Abstract

For predictive modeling techniques to be useful for processing electronic data streams of the scope and scale encountered in cyber security and intelligence applications, it is critical that statistical inference be performed continuously, in a single pass, and with an update rate at least as fast as arrival of the data. We propose research that will deliver an intelligent and strategic sampling theory to effectively close the widening gap between rates of analysis and observation. This will enable statistical inference to be conducted in real time on data streams previously addressed only by retrospective techniques.

A central concern in cyber security and intelligence is continuous surveillance, which is necessarily a matter of sequential inference. Our primary deliverable will be a body of work, both theoretical and represented in the form of statistical learning algorithms, of novel information theoretic approaches for adaptive sub-sampling of very-high-throughput data streams to effect orders-of-magnitude increases in ingestion rates for filter-based learning algorithms. This will serve as a key component of analytic surveillance systems, and will be accomplished while both minimizing the effects of uncertainty introduced by sub-sampling as well as maintaining mathematical guarantees of estimation consistency. We will produce a stand-alone optimized and extremely high-throughput data analysis system, with capabilities to turn on and off adaptive sampling.

Mission Relevance

This research will provide a suite of capabilities that will support many aspects of large-scale streaming data analysis at LLNL, and in particular supports the Laboratory's strategic cyber, space, and intelligence strategic focus. Our methodology will form a key and crucial component for a variety of analytic surveillance systems and will help the Laboratory's reputation as a leader in analysis of cyber security and intelligence data.

FY14 Accomplishments and Results

In FY14 our goal was to further improve the data processing rate of our particle learning tool, and package the code into a release version along with documentation. We have successfully accomplished both goals. Specifically, we (1) implemented a distributed version of particle learning in Storm, which allows for processing of streaming data in parallel over nodes in a compute cluster; (2) extended particle learning to perform adaptive sampling such that the data processing rates can be automatically adjusted to optimally match the data arrival rates—adaptive sampling weights the computational burden of processing each data point versus the expected improvement in the model density estimate to determine whether or not a data point is used to update the model, and as data arrival rates increase, adaptive sampling becomes more stringent, updating the model only with the most information-rich observations; (3) demonstrated the utility of adaptive sampling and parallel streaming

processing approaches to scale particle learning with the data arrival rates, and have integrated them into a single code base; and (4) prepared documentation on this streaming-data model inference tool.

Project Summary

The successful conclusion of this project has led to an application-agnostic tool for modeling streaming data. This tool can learn from data on-the-fly, without need to store data for posterior analysis. It allows for a wide range of tasks: density estimation, prediction, inference, clustering, and classification. We have shown its utility in a variety of applications. In particle physics we have applied it to distinguish gamma particles from neutrons. In text analysis we have performed spam detection. In cyber security this tool has been used to model user behaviors. The adaptive sampling and parallel streaming processing, via Storm implementation, have made this tool robust to streams of data of ever-increasing volumes. The implementation of this model in Storm is the first application of Storm-to-Bayesian statistical modeling to learn posterior probability densities via online inference at scale. Overall, it adds a valuable contribution to LLNL's machine-learning and statistical toolbox for big-data analytics, which is important to LLNL's mission. This work was leveraged to establish a collaboration with researchers at Kaiser Permanente Northern California, which has resulted in the expansion of parts of the modeling tool. We are continuing to search for further collaborations that may benefit from our streaming data analytic tool to identify new applications for it, as well as to further its development.

Publications and Presentations

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Efficient and Accurate Metagenomics Search Using a k-mer Index Stored in Persistent Memory

Jonathan Allen (12-ERD-033)

Abstract

Developing the capability to detect and diagnose engineered and emerging diseases across a global network is a national biosecurity research priority. Metagenomics

sequencing (the study of genetic material recovered from environmental samples) has emerged as a powerful genetic survey tool used in research for generating an unbiased and detailed description of a biological sample. We will develop novel, massively parallel algorithms to detect and identify pathogens in biological samples by searching pathogen genome databases indexed by their constituent k-mers (a specific amount of nucleic acid or amino acid sequences that can be used to identify regions within biomolecules such as DNA). This approach requires shorter laboratory preparation time prior to sequencing and no prior knowledge of the contents required for analysis. The ability to efficiently search emerging metagenomics databases presents a powerful new tool that could be used for pathogen detection and characterization.

We expect that the new software tools we create will enable orders-of-magnitude improvement in turnaround time from the submission of a biological sample's DNA to its taxonomic and functional characterization. Further, the analysis will be performed on commodity hardware—that is, computer systems manufactured by multiple vendors, incorporating components based on open standards—utilizing multiple and many-core processors combined with high-performance flash storage, making this analysis potentially deployable to field sites worldwide. The project results would demonstrate technical pathways to overcome challenging computational hurdles in metagenomics analysis, which are required to transition current research tools into technology that can be exploited by government agencies tasked with pathogen surveillance, diagnostics, and characterization.

Mission Relevance

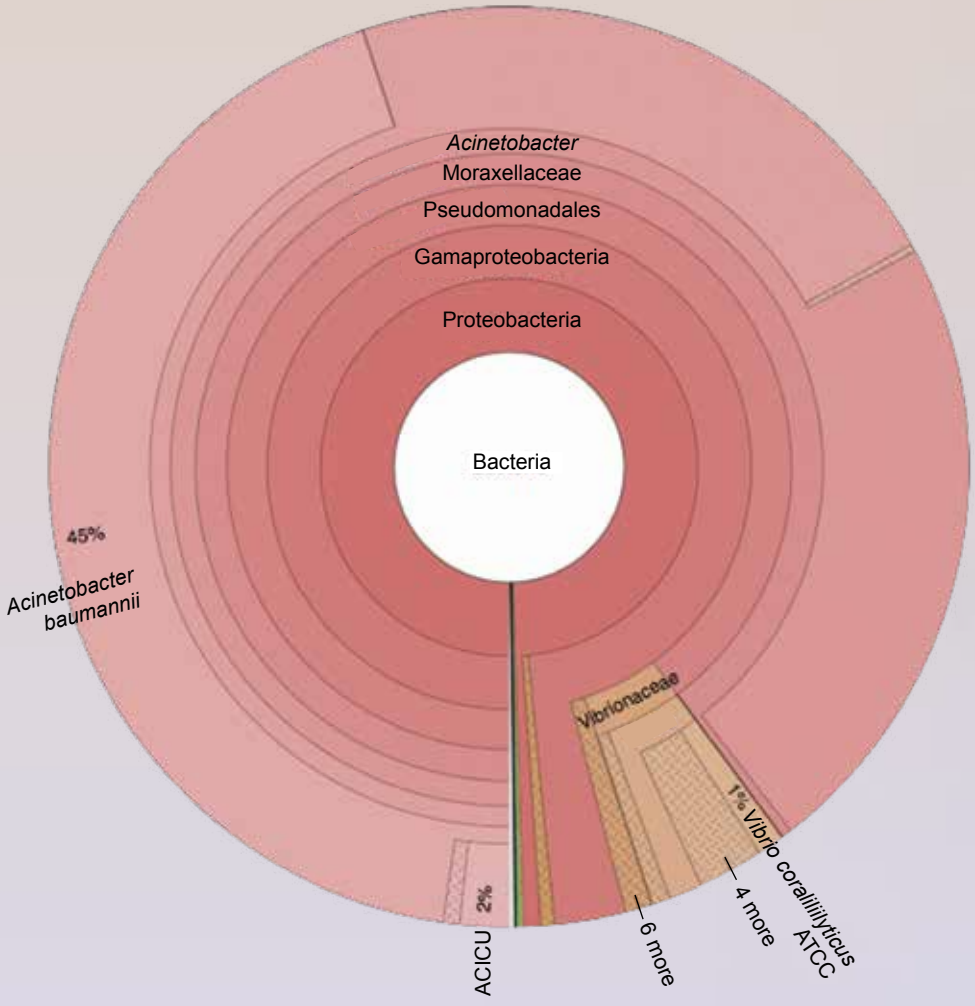
Our approach could advance metagenomic analysis for the biodefense needs of the nation, positioning us to partner with others to tackle analyses of large-scale clinical, environmental, and forensic metagenomic samples. This effort therefore supports LLNL's strategic priority in biosecurity research as well as bolsters the Laboratory's core competency in information systems and data science by using low-cost, multicore compute nodes augmented by persistent flash computer memory.

FY14 Accomplishments and Results

In FY14 we (1) developed new methods to expand the number of human host genomic variants that could be indexed and searched in a metagenomic sample, (2) indexed and searched the largest collection of searchable human genetic variants ever attempted using the complete human microbiome project data set, (3) identified a large collection of new identified microbial communities and found novel human genetic variants, (4) used content summary data to develop novel scoring schemes to determine the presence or absence of an individual organism with a dramatic reduction in false positive calls, and (5) developed a pruning scheme that allowed the index to be stored on low-cost solid-state drives while maintaining fast analysis times, thereby improving speed performance in persistent memory architectures while maintaining classification accuracy.

Project Summary

The successful conclusion of this project resulted in the design and development of a metagenomic classification software tool able to conduct microbial strain identification and gene annotation, as shown in the figure. We constructed the largest searchable genomic database available for metagenomic searches, which includes all publicly available sequenced viruses, archaea, bacteria, protozoa, fungi, human, and mitochondrial genomes from eukaryotic organisms. The software was applied to the Human Microbiome Project sample collection (characterizing microbial communities found at multiple human body sites) to demonstrate improved scaling and accuracy of the developed software. The results showed the ability to identify new microbial and host genetic sequences while maintaining run time speeds that supported analysis of large collections of metagenomic data. In summary, our project has generated a new capability that increases the number of genomic sequences that can be identified within metagenomic samples that will improve sequenced-based pathogen detection in these samples. The approach was designed to be scalable to support the analysis of a large



Bacterial metagenomic reads taken from a wound sample characterized using software created from our project showing the wound's bacterial community consisting primarily of an *Acinetobacter baumannii* strain. (Image created with the open-source software tool Krona).

numbers of samples. This new capability has attracted interest from agencies in the Department of Defense, who will provide support for integration and testing of the software into operational sequenced-based diagnostic systems for use by their customers.

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Ames, S., et al., 2014. *Design and optimization of a metagenomics analysis workflow for NVRAM.* IPDPS 14, IEEE 28th Intl. Parallel and Distributed Processing Symp., Phoenix, AZ, May 19–23, 2014. LLNL-CONF-635882.

Ames, S., et al., 2013. *Efficient K-mer indexing for metagenomics search in NVRAM.* XLDB-2013—7th Extremely Large Databases Conf., Stanford, CA, Sept. 10–11, 2013. LLNL-PRES-643608.

Ames, S., et al., 2013. "Scalable metagenomic taxonomy classification using a reference genome database." *Bioinformatics* **29**(18), 2253. LLNL-JRNL-611432. *http://bioinformatics.oxfordjournals.org/content/29/18/2253.long*

Coupled Segmentation of Industrial Computed Tomographic Images

Peer-Timo Bremer (13-ERD-002)

Abstract

Industrial volumetric inspection with computed tomographic imaging is used in applications ranging from scans of weapons to airport security to images of National

Ignition Facility target capsules. This imaging technique employs individual x-ray shots that are assembled computationally into a cross-sectional image. While each inspection application may use different modalities (such as x rays or microwaves) and offers unique challenges, the overarching problem is to find objects, materials, or other features in noisy and cluttered images corrupted by artifacts and with limited resolution. This project will advance the state of the art of industrial computed tomography segmentation. Using integrated segmentation algorithms coupled directly to reconstruction and new ensemble-based detection algorithms, we will develop a new framework significantly more capable of distinguishing objects and materials in these cluttered environments.

We will create an end-to-end pipeline of data-processing elements to advance the state of the art of computed tomographic segmentation. We expect to develop a tightly coupled solution to the three software stages of the process pipeline. In particular, we propose two primary feedback loops connected through a novel graph-based representation of an ensemble of simulations. Initially, we plan to concentrate on the airport security challenge, which has received significant attention from the Transportation Safety Administration and the Department of Homeland Security. We will develop new capabilities that can form the basis for future research, evaluation of existing technologies, and new tools. The general techniques will be broadly applicable to additional areas such as biomedical scans, geologic data, or detection of new types of homemade explosives.

Mission Relevance

Nondestructive evaluation is closely aligned with the Laboratory's national security mission and strategic focus area of stockpile stewardship. This project will significantly advance the LLNL's capabilities in x-ray imaging for nondestructive evaluation in such areas as biology and experimental physics, and employs methodologies relevant to the core competency in information systems and data science.

FY14 Accomplishments and Results

In FY14 we (1) developed a new iterative beam-hardening correction technique, (2) developed a new segmentation algorithm based on creating multiple randomized hierarchies, (3) incorporated a new machine-learning approach for the unsupervised classification of objects, and (4) integrated this technology into the engineering software package NDDAV (*N*-dimensional data analysis and visualization).

Proposed Work for FY15

In FY15 we will (1) combine multiple-hierarchy segmentation with unsupervised object detection, (2) develop semantic object classification, (3) investigate super-resolution computed tomographic reconstruction, (4) provide benchmark data to the Awareness and Localization of Explosives-Related Threats Center (a Department of Homeland Security Center of Excellence), and (5) provide an integrated software pipeline for reconstruction, segmentation, and object classification.

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Champley, K. M., and T. Bremer, 2014. *Efficient and accurate correction of beam hardening artifacts.* 3rd Intl. Conf. Image Formation in X-Ray Computed Tomography, Salt Lake City, UT, June 22–25, 2014. LLNL-CONF-649613.

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Thiagarajan, J. J., K. N. Ramamurthy, and P. T. Bremer, 2014. *Multiple kernel interpolation for inverting non-linear dimensionality reduction and dimension estimation.* LLNL-POST-653814.

Thiagarajan, J. J., et al., 2014. *Automatic image annotation using inverse maps from semantic embeddings.* IEEE Intl. Conf. Image Processing, Paris, France, Oct. 27–30, 2014. LLNL-CONF-650594.

Data-Centric Computing Architecture

Maya Gokhale (13-ERD-025)

Abstract

Recent trends in the architecture of computer central processing units indicate that future processors will have many cores integrated on a single die, with a greatly reduced amount of memory available to each core relative to today's architectures. The drastic reduction in memory per core is related to the high cost of dynamic random-access memory in both power and dollars. The looming problem of memory bandwidth and capacity will affect high-performance computer applications on exascale supercomputers. Data-intensive computing is characterized by both very large application working sets and increasingly unstructured and irregular data access patterns. Data-centric applications are affected much more by memory latency, bandwidth, and capacity limitations than traditional high-performance computing applications. Without research into new system architectures and software, the present architectural trends will severely impact LLNL's data science applications. We propose to design, prototype, and evaluate a data-centric node architecture consisting of a many-core central processing unit, a large memory that seamlessly combines dynamic and nonvolatile random-access memory, and an active storage controller based on a field-programmable gate array that can run data-intensive kernels accessing nonvolatile random-access memory. In addition, we intend

to develop massively parallel, throughput-oriented algorithms, parallel programming frameworks, and design patterns.

We expect to design data-centric node architectures, system software, and applications optimized for Livermore data science and exascale mission needs. This accomplishment will give us the opportunity to influence designs of data-intensive computing architectures, particularly in terms of memory and local storage systems, enabling cost-effective, energy-efficient large hybrid-memory architectures that can deliver high performance on demanding data-analysis workloads. In addition, we will develop caching strategies that reduce memory-access latency, improving performance of throughput-driven applications. Our active storage off-load approach would drastically reduce the bandwidth required between storage and the central processing unit, while simultaneously increasing performance and energy efficiency.

Mission Relevance

Big data—requiring the continuous processing and analysis of sensor, experimental, and simulation data—is a dominant Laboratory challenge requiring scalable, flexible architectures to match a wide range of applications and budgets. Our research addresses a critical mission need for data-centric computing and benefits data science applications for both informatics and simulation data analysis, in support of the Laboratory's core competency in information systems and data science.

FY14 Accomplishments and Results

In FY14 we (1) developed a functional simulator and emulator based on a field-programmable gate array of active memory hardware architectures to quantify memory bandwidth and energy efficiency of placing data reorganization logic in the memory, (2) implemented two system software interfaces in the simulator and emulator, (3) employed the data-intensive memory-map run-time driver on a classification algorithm for environmental genetic material and optimized the write-back algorithm, (4) implemented and optimized a new data structure to create and traverse dynamic graphs with synthetic data, and (5) implemented and evaluated two versions of streamline tracing—a memory-mapped algorithm and a user-level cache. In most instances, the memory-mapped code with system cache was faster than user-level cache.

Proposed Work for FY15

In FY15 we will (1) quantify bandwidth, energy, and performance of several active memory data reorganization and processing architectures in the field-programmable gate array and in software; (2) continue collaborations with researchers examining external processing in memory; (3) improve stability and performance of the data-intensive memory-map run-time driver over a wide range of data science and scientific simulation workloads while continuing to support operational deployment; (4) study operating system optimizations for heterogeneous persistent memories with differing latencies and bandwidths; (5) implement dynamic graph traversals and community detection algorithms in dynamic graphs stored in persistent memory; and (6) extend

the streamline tracing implementation from thread parallel programming to run-distributed-memory parallel programming.

Publications and Presentations

Ames, S., et al., 2014. *Design and optimization of a metagenomics analysis workflow for NVRAM.* IEEE Intl. Parallel Distributed Processing Symp. (IPDPS), Phoenix, AZ, May 19–23, 2014. LLNL-CONF-635882.

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Wong, D., G. S. Lloyd, and M. B. Gokhale, 2013. *A memory-mapped approach to checkpointing.* LLNL-TR-635611.

A Hybrid Content- and Concept-Based Approach to Large-Scale Video Analytics

Douglas Poland (13-ERD-046)

Abstract

The intelligence community requires tools to index and query ever-larger collections of disparate video and determine the most relevant videos with minimal missed detections (false negatives). In contrast, market-driven video indexing and retrieval tools emphasize popularity and minimizing false positives. We propose to develop a hybrid content- and concept-based approach with the expressive power of concept-based approaches (videos indexed via labels) while retaining the generality and novelty of content-based approaches (retrieving videos that share similar features typically based on image content, motion descriptors, or audio). We will develop novel data structures and algorithmic techniques to annotate, index, and query large

collections of video to support pressing needs in video analytics for intelligence applications. To this end, we will develop three key components: (1) a compact representation of video with the augmented space–time cube; (2) a graph-based knowledge-retrieval engine based on a bipartite graph of annotations, content-based features, and video clips; and (3) visual-relevance feedback, combining visual summaries of video clips and low-rank representations of video searches.

We expect to demonstrate new scalable video indexing and querying capabilities that will support pressing video analytics needs and provide a strong foundation for future work on deeper analytics. We will demonstrate these capabilities with a proof-of-concept system comprised of the proposed data structures and algorithms that will be used to evaluate the system's learning, query, and feedback performance. Our video indexing, annotation, and retrieval engine will be integrated with current search capabilities such as CAPS (Counterproliferation Analysis and Planning System), BKC (Bio-Knowledge Center), and DocEx (Document Exploitation) to create a new type of data for multiple applications.

Mission Relevance

This project supports the Laboratory's national security mission and the core competency in information systems and data science by developing the foundations of a new capability for indexing and querying video in ways that provide human analysts with the most relevant results and minimal missed detections.

FY14 Accomplishments and Results

In FY14 we (1) established the largest public research multimedia data set ever assembled—the YLI (Yahoo, Livermore, and International Computer Science Institute) corpus; (2) implemented a full set of state-of-the-art feature extractors on Livermore computing systems, including audio, visual, and motion features; (3) extracted those features on the YLI corpus; (4) continued research on hierarchical feature sets using state-of-the-art audio, visual, and motion features as starting points—in particular we are focusing on establishing a deep-learning approach to motion and on fusing audio and visual modalities in a deep-learning framework leveraging the large-memory Catalyst supercomputer at Lawrence Livermore; and (5) demonstrated proof-of-concept content-based video retrieval using "late-fusion" multimodal percepts and concepts.

Proposed Work for FY15

In FY15 we propose to continue multimodal feature research on high-performance computers. Specifically, we will (1) establish a robust graph framework that represents video activity as scenes composed of high-level multimodal concepts and activities, and combine this with sparse metadata to enable both keyword inference and propagation and keyword-based query; (2) develop relevance-feedback algorithms that leverage hierarchical concepts and percepts to expand or redirect the results as desired—this will require graphical user interface development work that enables presenting and interacting with our high-dimensional feature capability; and (3) develop, in collaboration with the International Computer Science Institute, requirements for a new type of community multimedia benchmark that requires

multimodal percepts for MediaEval15, a benchmarking initiative dedicated to evaluating new algorithms for multimedia access and retrieval.

Publications and Presentations

Choi, J., et al., in press. "The placing task: A large-scale geo-estimation challenge for social-media videos and images." *Proc. ACM Multimedia 2014 Workshop on Geotagging and Its Applications in Multimedia (GeoMM '14)*. LLNL-TR-663455.

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Planetary-Scale Agent Simulations

Peter Barnes (14-ERD-062)

Abstract

The nation faces more and more problems that are global in scale, especially in the field of national security, yet we lack the capability of simulating those problems at a worldwide scale to help develop the best possible countermeasures. We intend to develop global-scale models to predict outcomes, evaluate courses of action, and develop new analysis techniques for addressing global problems. To demonstrate the capabilities of our global modeling approach, we will develop a demonstration application based on discrete-event simulation, which models the operation of a system as a discrete sequence of events in time, with each event occurring at a particular instant and marking a change of state in the system. The discrete-event simulation we will address for this project is a pandemic biological event that simulates the spread of an evolving virus. We will tackle three fundamental research challenges that are common to discrete-event simulation: (1) the automatic reverse engineering of source code; (2) dynamic load balancing, which is a technique that distributes processing equally among processors throughout the job's life; and (3) global synchronization using only one-sided communication data.

We expect to develop and implement—on Sequoia-class supercomputers—planetary-scale models, such as a real-time model including every person on earth executing five events per second per person; a model of the entire Internet, transmitting 10 packets per second per node; and a worldwide pandemic model simulating viral evolution during replication and transmission. We will aim for planetary scale and in so doing learn a tremendous amount about the underlying technical challenges.

Mission Relevance

This project supports the Laboratory's core competency in information systems and data science by developing a new, scalable discrete-event simulation system that will support the predictive modeling of complex systems critical to national security. This

project also supports LLNL's bioscience and bioengineering core competency by advancing computational biology tools for outcome prediction, especially using genomic sequences, related protein sequences, and protein structure models to help predict how viruses might evolve.

FY14 Accomplishments and Results

In FY14 we (1) developed a new approach to implementing reverse methods for optimistic discrete-event simulation using Backstroke, an open-source reverse-code generation framework—this new method is much more powerful in that it supports the entire C and C++ languages, for any application with available source code; (2) developed an initial implementation of Backstroke that supports every language feature with the exception of allocating arrays; (3) designed and implemented coupling between ROSS (Rensselaer's Optimistic Simulation System) with the Charm++ run-time system in collaboration with Rensselaer Poytechnic University in New York and the University of Illinois at Urbana-Champaign; and (4) designed and employed a new implementation of our viral evolution model. The new Backstroke approach has greatly simplified our interface between the optimistic parallel discrete-event simulation and the ns-3 simulator, an open-source discrete-event network simulator for Internet systems.

Proposed Work for FY15

In FY15 we will (1) complete the second version of the Backstroke implementation, (2) complete coupling of the ROSS and Charm++ systems, (3) develop performance metrics for the ROSS–Charm++ system, (4) develop approaches for dynamic load measurement, (5) complete implementation of the viral evolution model, (6) design coupling of the viral evolution model with the disease epidemic model EpiSimdemics, (7) evaluate the feasibility of supporting the ns-3 simulator in an optimistic parallel discrete-event simulation engine, and (8) begin implementation of the interface between the optimistic parallel discrete-event simulation engine and the ns-3 simulator.

Publications and Presentations

Barnes, P. D., et al., 2014. *Extreme scale optimistic parallel discrete event simulation with dynamic load balancing*. 2014 Winter Simulation Conf., Savannah, GA, Dec. 7–10, 2014. LLNL-PRES-663468.

The Livermore Brain: Massive Deep-Learning Networks Enabled by High-Performance Computing

Barry Chen (14-ERD-100)

Abstract

The ability to automatically detect patterns in massive sets of unlabeled data is becoming increasingly important as new, advanced types of sensors come into use. However, the biggest obstacle to applying machine learning to this and other areas

of national security data science is the sheer volume of unlabeled data that must be analyzed. Deep learning is a computational approach for identifying patterns in such unlabeled data, and has already been shown to often outperform traditional systems built on hand-engineered features. The goal of this project is to develop the learning algorithms that would form the basis of massive deep-learning neural networks for effectively capturing complex spatiotemporal patterns in massive data sets, thereby helping to address nationally important problems. Our approach consists of three parts: (1) scale up existing deep-learning algorithms to run on high-performance computing platforms, (2) develop new deep-learning algorithms to model high-dimensional time-varying signals, and (3) validate the new deep-learning algorithms by applying them to three complex data problems—classifying audio and video content, detecting anomalies in wide-area aerial video, and modeling network behavior.

If successful, we will develop the world's largest deep-learning network, including next-generation, deep-learning architectures and algorithms that effectively identify complex spatiotemporal patterns in massive, unlabeled data sets, thus establishing LLNL as a leader in machine learning on high-performance computing platforms. We will also demonstrate statistically significant improvements in classification rates on three mission-relevant problems and develop the expertise for effectively using deep-learning methods in a wide range of data-science applications.

Mission Relevance

In addition to advancing the specific applications of classifying audio and video content, detecting anomalies in wide-area surveillance video, and modeling computer network behavior relevant to the Laboratory's strategic focus area in cyber security, space, and intelligence, this project also supports the Laboratory's national security mission by developing technologies that are broadly applicable to other mission-relevant applications, including general threat detection, National Ignition Facility system monitoring and prediction, and the validation of advanced manufacturing parts. Overall, the project is well aligned with LLNL's core competency in information systems and data science.

FY14 Accomplishments and Results

In FY14 we (1) adapted Stanford's deep-learning codes for graphic processing units to LLNL's high-performance computer cluster and repeated the Google artificial brain experiment, training a 1-billion parameter net on images from video; (2) determined that our data-intensive memory-mapping libraries would optimize data throughput more effectively than our communication and routing libraries, and began integration of these libraries into our code—we will complete throughput benchmarking in FY15; (3) demonstrated an impressive 150-fold increase in net size, creating and training (using 100 nodes of Livermore's Edge supercomputer) a massive 15-billion parameter net on 99.3 million images from the newly released Yahoo! Flickr Creative Commons data set, as shown in the figure; (4) showed that our massive net automatically learns complex patterns in images such as fireworks, airplanes, towers, and text; (5) created baseline classification systems for image and video action

classification applications; and (6) developed new learning algorithms for space–time receptive fields and validated our approach on both synthetic and real video.

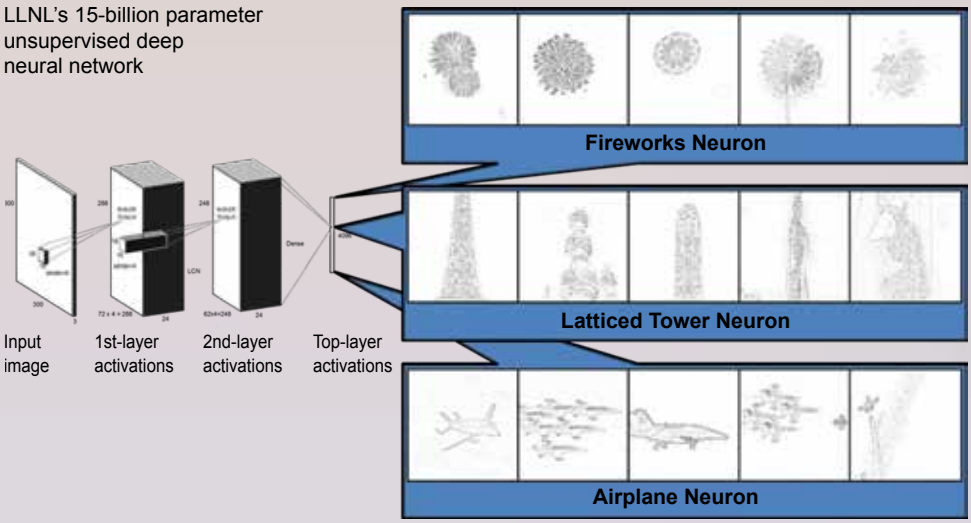
Proposed Work for FY15

For FY15 we will (1) research dynamically changing space–time receptive fields and show that our new approach improves video action classification, (2) investigate new artificial neutral networks used for learning efficient coding ("autoencoders") that learn to predict future observations to better model temporal patterns and improve prediction performance, (3) research new algorithms to combine predictive autoencoder networks with conventional ones, and (4) develop and enhance the Fastlab neural net software to fully utilize graphic processing unit nodes on Edge and large central processing unit clusters (e.g., LLNL Open Computing Facility's Cab and Sierra systems), optimizing speed performance on both architectures.

Publications and Presentations

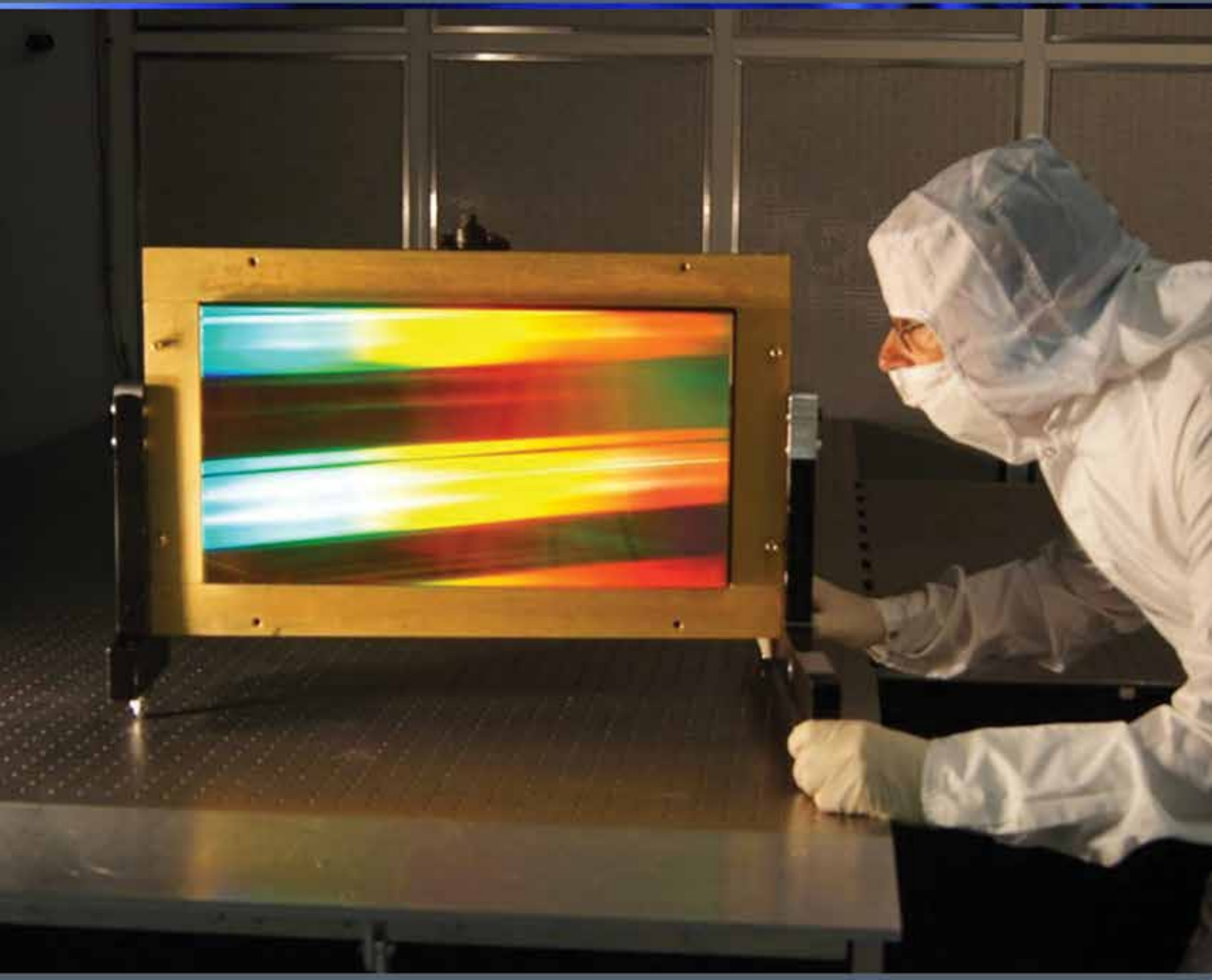
Boakye, K., et al., 2014. *HPC enabled massive deep learning networks for big data.* LLNL-ABS-662780.

Ni, K., et al., 2014. *Large-scale deep learning on the YFCC100M dataset.* LLNL-CONF-661841.



Using graphic processing units in high-performance computer clusters, we trained massive unsupervised neural networks to automatically learn semantically meaningful patterns (e.g., fireworks, towers, and airplanes) in 99.3 million unlabeled images from the newly released Yahoo! Flickr Creative Commons data set.

Lasers and Optical Materials Science and Technology



FY2014

Lawrence Livermore National Laboratory

Probing Atomic-Scale Transient Phenomena Using High-Intensity X Rays

Stefan Hau-Riege (12-ERD-021)

Abstract

The newly built Linac Coherent Light Source (LCLS) at the SLAC National Accelerator Laboratory at Stanford enables atomic-resolution imaging with femtosecond time resolution. This capability can enable advances in functional biology as well as enabling the development of storage technologies to harness renewable energy sources and obtain energy independence. These diverse, important issues can be addressed only through symbiotic fundamental research in physics, biology, chemistry, engineering, and mathematics, which is possible with well-designed experiments at the LCLS. However, samples exposed to the LCLS are severely damaged by radiation in an unchartered regime of x-ray and matter interaction. Without understanding the damage mechanism, the diffraction data cannot be analyzed correctly. Therefore, we propose to undertake fundamental research in the emerging field of x-ray-induced transient nanometer-scale phenomena. We will provide a validated predictive capability for high-intensity x-ray and matter interactions at the atomic level. At the same time, we will validate and improve x-ray plasma diagnostics methods for taking femtosecond snapshots of the dynamics of equilibration processes taking place at the nanometer-length scale.

We expect to greatly enhance our understanding of atomic-scale intense x-ray and matter interaction as well as establish the foundational knowledge needed to enable LCLS imaging. We will develop experimentally validated realistic models for high-intensity x-ray and matter interaction, obtain femtosecond snapshots of the dynamics of equilibration processes in materials excited by x rays, and develop an understanding of x-ray and matter interaction processes in nanostructures. This interplay of modeling and experiments forms the backbone of our proposal.

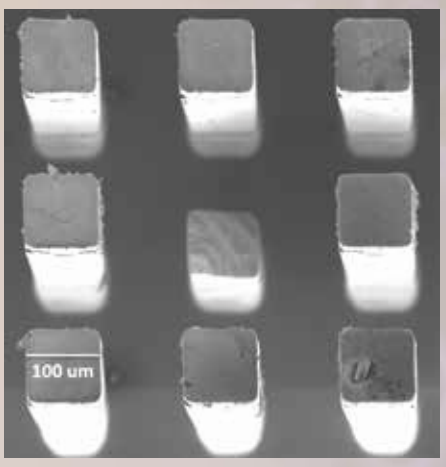
Mission Relevance

Our proposed research program will sustain and further develop LLNL's international leadership in x-ray plasma probing and solid-damage physics through meaningful, timely research. Our project is directly relevant to many Livermore strategic mission thrusts, including stockpile stewardship science and securing energy independence for the nation, as well as supporting the core competency in lasers and optical materials science and technology. In addition, this high-profile project will attract top postdoctoral researchers and allow us to engage in international collaborations.

FY14 Accomplishments and Results

In FY14 we (1) completed the post-exposure analysis of our samples irradiated by the LCLS beam, (2) were awarded beam time to perform ultrafast imaging on carbon nanometer-scale tube arrays, (3) performed imaging of carbon nanotube arrays,

and (4) continued the development of our x-ray interaction model, applying it to study the electron dynamics in x-ray irradiated samples.



Tall silicon mirrors (aspect ratio of 1:7) for back-reflecting high-intensity x-ray pulses. The center mirror had been exposed to an x-ray free-electron laser pulse and was obliterated, while the surrounding pillars were not affected.

Project Summary

The successful conclusion of this project resulted in a greatly enhanced understanding of atomic-scale intense x-ray and matter interaction. In particular, we studied the feasibility of using silicon single crystals as back-reflectors for high-intensity, hard x-ray pulses. Our enhanced understanding of the radiation-induced processes enabled us to develop novel x-ray back-reflectors that can withstand focused x-ray free-electron laser beams. In another x-ray free-electron laser experiment, we demonstrated imaging of carbon nanotube forests. In addition, we developed modeling capabilities to study the effect of high-intensity x-ray radiation on Bragg diffraction from silicon and diamond, the nonequilibrium electron dynamics in materials driven by high-intensity x-ray pulses, and the photoelectron dynamics in diffractive imaging of biological samples using x-ray free-electron laser beams. We also supported experiments to study the effects of high-atomic-number elements on local atomic displacements in solid-density materials. We are discussing, with the DOE Office of Fusion Energy Sciences, the possibility of forming an x-ray free-electron laser modeling program based on the results obtained with this project.

Publications and Presentations

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High-Fluence, Multipulse Laser Surface Damage: Absorbers, Mechanisms, and Mitigation

Jeffrey Bude (12-ERD-023)

Abstract

The lifetime and performance of optical systems designed to guide high-photon fluxes are limited by degradation and damage to key optical components at high-photon fluences (radiant energy). Even high-quality optical surfaces without flaws can degrade as a result of extensive multipulse optical stress and can suffer damage from absorption by damage precursors. The mechanisms of this degradation and the nature of these precursors are unknown. We will employ a suite of integrated tasks that closely link processing, characterization, and modeling to develop a scientific understanding of the mechanisms that govern high-fluence optical damage and degradation, and develop techniques to improve the high-fluence lifetime for optical glasses and other related optical materials.

We expect to determine the physical mechanisms of high-fluence damage initiation in optical materials, including links between absorption and damage and the nature of damage-precursor absorption. We will also identify the physical origin of high-fluence surface damage precursors on optical glasses and the processes that introduce them onto surfaces during fabrication. We will develop processes to reduce whole-optic fluence damage by reducing precursors and modulation from etched flaws. We will also develop accelerated multipulse optical-stress protocols to characterize degradation from billions of pulses and to clarify degradation mechanisms. This work will advance understanding of laser–matter interactions and extend operational lifetime and performance of high-fluence laser systems.

Mission Relevance

This work directly addresses ignition and stockpile stewardship challenges by optimizing utilization of large inertial-confinement fusion laser systems, and also supports inertial fusion energy science and technology strategic focus area. Reduction of high-fluence damage on optical glasses will allow fusion-class lasers to operate at higher fluences and with reduced sensitivity to contrast, in support of the Laboratory's core competency in lasers and optical materials science and technology.
Understanding how optical surfaces degrade under extreme multipulse stress will guide design and use of optics for fusion energy systems, and understanding how absorption leads to damage can clarify the mechanisms of laser damage from contamination, damage in coatings, and the role of radiation-induced defects on damage in optics. More broadly, understanding laser–matter interactions is a frontier problem in condensed matter physics.

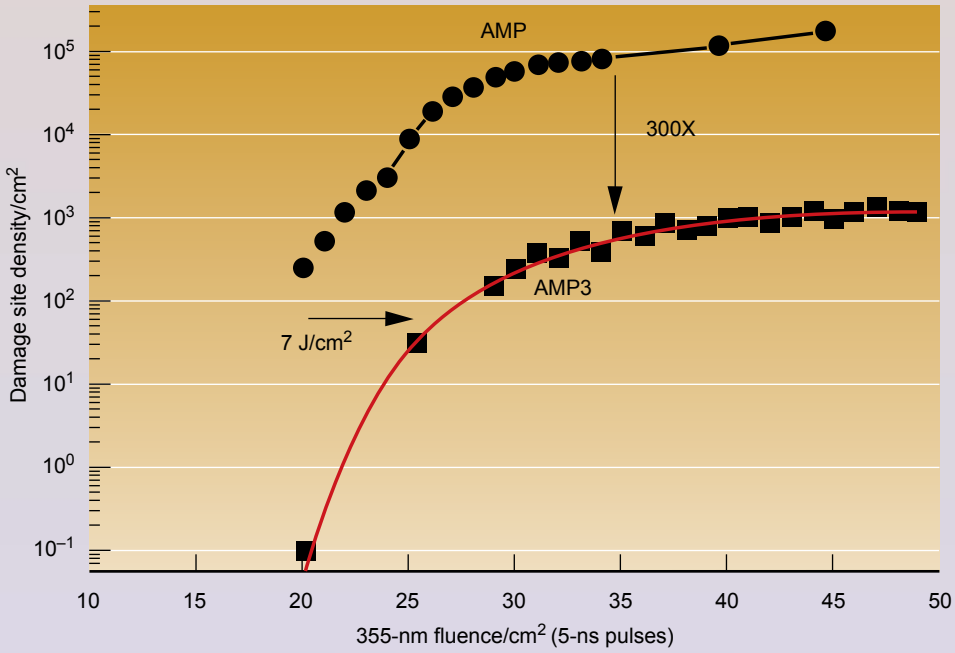
FY14 Accomplishments and Results

In FY14 we (1) studied the physical characteristics of high-fluence precursors, finding that they must absorb enough light to reach 3,000 K and must be strongly bonded to

an optics surface; (2) assessed the importance of trace impurities in processing liquids, finding that parts-per-billion levels were sufficient to induce damage, even for nominally transparent species and especially for impurities introduced in the last stages of the process; (3) achieved a reduction of over 2,000 in damage density and an increase of almost a factor of 2 in damage thresholds for silica; and (4) completed a study of multipulse optical degradation for silica out to 1 billion pulses, determining that the primary mode of degradation was transmission loss because of photo-generated surface defects.

Project Summary

The successful conclusion of this study provided a new understanding of optical damage and degradation for silica at high pulse fluence and longtime multipulse exposure, and demonstrated a means to control or mitigate these effects. We found that the dominant laser damage precursors at high fluence are submicron precipitates of trace ionic and organic impurities in processing chemicals. Defects in these precipitates absorb enough laser energy to reach temperatures that can initiate micron-sized damage sites. Processes were developed to reduce the probability of precipitation during wet chemical processing and drying—we achieved a 2,000-fold reduction in damage density that extends useful operation fluences by almost a factor of 2. We also studied degradation of silica surfaces exposed to billions of laser pulses as a function of laser parameters at 355 nm. Defects generated photochemically within 1 nm of the surface lead to transmission loss of up to 4% per surface. Defect generation is most severe at low pulse intensities and is offset at high intensities by photochemical processes that destroy these defects. Transmission loss can also be suppressed by atmospheric oxygen. Laboratory programmatic support will enable us to continue work to fully transfer the optics processes developed in this research



A comparison of damage density between the Advanced Mitigation Process (AMP) and the more complex optics treatment process AMP3, which was optimized to reduce the probability of precipitation from wet chemical processes we developed. We achieved a 300-fold reduction in overall damage density and a 7J/cm² shift to higher fluences.

project to full-scale optics production and use on the National Ignition Facility at Livermore. Extension of the work on giga-pulse optical degradation to new materials for use in high-average-power laser systems will also be supported.

Publications and Presentations

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Novel Multiple-Gigahertz Electron Beams for Advanced X-Ray and Gamma-Ray Light Sources

David Gibson (12-ERD-040)

Abstract

Our objective is to design, model, assemble, and demonstrate a high-brightness electron-beam source that is capable of generating electron bunches in each wakefield period, or bucket, of the drive radio frequency. Using X-band radio frequency (11.424 GHz) maximizes the number of bunches that can be generated, up to 1,000 bunches per pulse, at a 120-Hz

repetition rate. Such an electron source would significantly increase the flux and/or brightness of electron-based light sources (such as Compton-scattering sources and free-electron lasers). The research objective will be accomplished by building a new multiple-gigahertz-compatible photo-injector and a gigahertz-compatible cathode illumination laser, which will be integrated with existing radio-frequency power and electron accelerator hardware to make beam measurements.

If the project is successful, we will have demonstrated a multiple-gigahertz electron beam suitable for use in a Compton-scattering-based gamma-ray (MEGa-Ray) source. This would allow the gamma-ray flux and overall source brightness to be significantly increased, while simultaneously simplifying the design of the associated photo-injector drive laser, accelerator, dark-current mitigation hardware, interaction laser, and interaction region. The time structure of the gamma-ray source opens the possibility of time-resolved or stroboscopic measurements with picosecond-scale resolution. In the process of conducting this research, we will also learn what changes, if any, are needed to the accelerator hardware to accelerate the high-repetition-rate bunch train to a few hundred megaelectronvolts.

Mission Relevance

This effort supports the Laboratory's core competency in lasers and optical materials science and technology to address national nuclear missions. Generation of a multiple-gigahertz electron bunch train with small-per-bunch charge can allow a higher flux and higher brightness MEGa-Ray source than the current single-bunch system provides, improving the quality of desired fundamental nuclear physics measurements, as well as expanding the envelope of feasible nuclear photonics applications for national and international security.

FY14 Accomplishments and Results

In FY14 we (1) brought the new X-band accelerator system online (see figure), which included several weeks of conditioning the radio-frequency structures and measurement of fields, and completed the drive laser-beam transport to the cathode; (2) conducted beam performance studies and improvements during the commissioning phase of the accelerator, where we generated a 28-MeV,100-pC electron beam with measured emittance below 1-mm mrad, and compared the measured beam performance with PARMELA accelerator simulation program models to verify the system was operating as expected; (3) demonstrated the production of two electron beams, spaced 87.5-ps apart in time, and verified we were able to accelerate both bunches concurrently to the same final energy; and (4) designed and built an interaction region, including a beam-diversion chicane, focusing quadrupole magnets, and bending spectrometer, along with an interaction laser and focusing, insertion, and extraction optics that will allow for x-ray generation at 30 keV. Because of a reduction in funding, we did not perform a frequency conversion demonstration of the gigahertz laser system developed in the first two years of the project.

Project Summary

The successful completion of this project has resulted in the demonstration of a novel, 11-GHz repetition-rate fiber-based laser that is precisely synchronized with accelerator drive radio-frequency, and is therefore able to generate over 500 bunches of electrons with 87.5-ps spacing. Coupled with a high-quantum-efficiency photocathode, and the X-band accelerator system we commissioned, this laser technology can significantly increase the average beam current available from radio-requency accelerators, and can consequently increase the flux available from a variety of beam-based light sources, including systems based on Compton-scattering and free-electron laser sources such as the LInac Coherent Light Source at Stanford. We have made initial steps toward this goal by demonstrating the successful acceleration of two electron bunches through a system without significant inter-bunch beam effects that cause beam degradation. We are pursuing sponsors from a variety of areas for support to integrate the laser and radio-frequency technologies, as well as to operate the x-ray source as an experimental station.

Publications and Presentations

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Finalizing installation of our new X-band accelerator at Lawrence Livermore, which can significantly increase the average beam current available from radio-requency accelerators, and consequently increase the flux available from a variety of beam-based light sources.

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Marsh, R. A., et al., 2013. *LLNL X-band test station status.* North American Particle Accelerator Conf. (NA-PAC13), Pasadena, CA, Sept. 29–Oct. 4, 2013. LLNL-CONF-636365.

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Marsh, R. A., et al., 2012. *Ultracompact accelerator technology for a next-generation gamma-ray source.* 2012 IEEE Particle Accelerator Conf. (IPAC), New Orleans, LA, May 20–25, 2012. LLNL-PROC-557854.

Ionic Dopant Pairs for High-Fluence Filters

Kathleen Schaffers (12-ERD-041)

Abstract

Our objective is to explore and extend the current methods and fundamental understanding of how to achieve precise control of the oxidation state of colored dopant ions (particularly copper and iron ion pairs) in a glass host. With this understanding, we will develop an optic with the appropriate absorption characteristics for a suitable red blocker (high transmission at 351 nm, low transmission at 1,053 nm) for high-peak-

power lasers. This will be accomplished through experimental glass melting, doping with metallic ions and ion pairs, material and spectroscopic characterization, and developing a predictive model for choosing laser optical glasses.

The scientific understanding developed from this study will provide the tools to formulate a model for predicting the reduced-to-oxidized ratio of a dopant ion in specific glass hosts as well as the spectral positioning and width of absorption bands within a glass. We will also develop a capability to fabricate robust optical filters with a controllable quantity of the desired oxidation state of copper and iron dopant ions and the appropriate spectral characteristics. In addition, we will determine a suitable glass that also meets the solarization, damage resistance, and manufacturability requirements for high-power laser applications.

Mission Relevance

The basic knowledge gained from this study can improve upon LLNL's leadership in producing optics with extreme requirements for specific optical applications, in support of the Laboratory's core competency in lasers and optical materials science and technology. In particular, the development of a red blocker optic for use in high-peak-power laser systems such as the National Ignition Facility will be critical to improving efficiency.

FY14 Accomplishments and Results

In FY14 we (1) gained a fundamental understanding of ligand field effects and how this affects the absorption peak and position; (2) developed a model to simulate optical properties of the dopant ions and host materials, which successfully predicted the absorption position and coordination environment of the dopant ion for certain systems; (3) studied the high-fluence behavior of both glasses and solutions with specific emphasis on the solarization and spectroscopic properties of materials; and (4) identified the desirable copper and iron dopants and iron–tin redox dopant pairs to produce a red blocker optic.

Project Summary

At the conclusion of this project, we achieved a fundamental understanding of the chemical processes to tailor the optical properties of dopants in various glass hosts for use in high-fluence systems as optical filters and other absorbing optics. We have an understanding of ligand field theory and how it relates to solution and glass chemistry. We also determined that ligand field strength, oxidation state, coordination environment, and redox pairs greatly affect the peak position and height of the absorption peak. Using this knowledge, we identified dopant ions that yield the correct spectroscopy in copper and iron–tin glass hosts. These dopants were incorporated into a significant number of glasses, and high-fluence laser experiments were performed on many of the glasses yielding the correct spectroscopy. The technology and understanding we gained have been transferred to two vendors for

continued development and scaling to a usable optical filter. The future focus will be to determine the correct host (providing the required spectroscopy and solarization resistance) and scale it to the appropriate sizes for large laser systems.

Publications and Presentations

Demos, S. G., et al., 2013. "Change of self-focusing behavior of phosphate glass resulting from exposure to ultraviolet nanosecond laser pulses." *Optic. Express* **21**(4), 4854. LLNL-JRNL-598992.

Laser Lethality Experimentation, Modeling, and Simulation Capability

W. Howard Lowdermilk (12-ERD-050)

Abstract

Existing experimental data and models are inadequate for developing systems to counter the threat of modern ballistic missiles. The need therefore exists for an experimentally validated, predictive modeling and simulation capability to optimize the design and performance of anti-ballistic-missile laser weapon systems and to reduce the need for costly full-scale testing. Our goal is an experimentally validated model for the laser-induced fracture and fragmentation of thin metal plates and pressure vessels in single- and multiple-layer configurations. We will conduct laser interaction experiments and measure thermal and physical properties using Laboratory equipment and facilities to enable and guide the development of a new ALE3D (arbitrary Lagrangian–Eulerian three-dimensional) code and to validate the resulting laser–target interaction models.

We will produce measurements of the thermal and physical properties of selected materials in relevant regimes of temperature and stress loading, characterizations of the laser-induced fracture and fragmentation of thin metal plates and pressure vessels in single- and multiple-layer arrangement, and ALE3D capabilities for modeling laser–target interaction, culminating in an experimentally validated model. We will also demonstrate our new model's capability and practicality for countering laser lethality problems. This capability will enable the timely and cost-effective design and optimization of anti-missile laser weapon systems needed to defend against modern ballistic missiles.

Mission Relevance

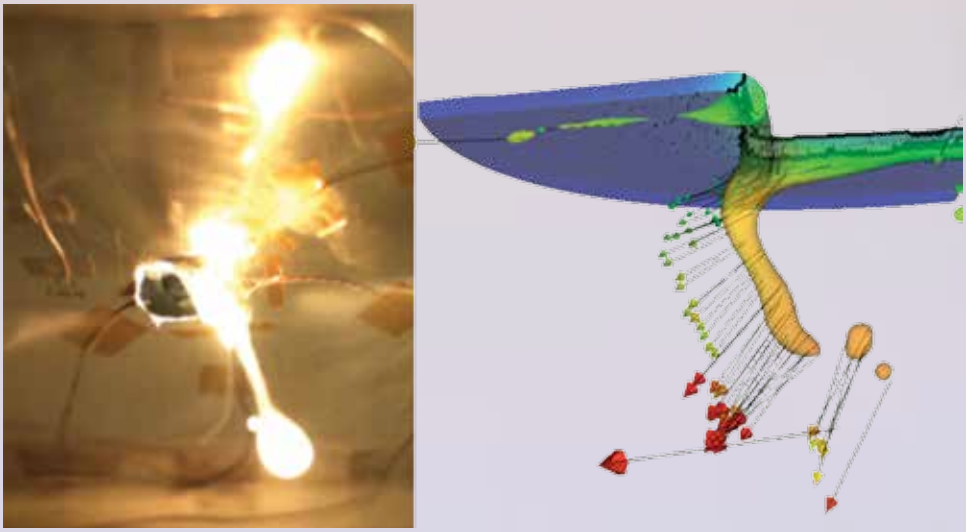
This project directly supports LLNL's national security mission by meeting the currently unfilled need for a validated, predictive modeling capability to evaluate laser lethality and missile vulnerability for laser-based anti-missile systems, as well as supports the core competency in lasers and optical materials science and technology. In addition, the capability to be developed will also be applicable to similar fracture and fragmentation problems in support of stockpile stewardship and the Laboratory's energy security mission.

FY14 Accomplishments and Results

In FY14 we (1) modified the ALE3D code to simulate the effects of material melting under laser irradiation conditions, which included the effects of temperature-dependent surface tension, evaporative cooling, recoil momentum, residual thermal stress, and gravity; (2) completed development and validation of our unique tool for a broad range of laser–material interaction and laser lethality problems; (3) developed a simple physical model explaining the formation of metallic droplets, which, in the absence of gravity, will delay laser penetration for extended periods of time; (4) demonstrated experimentally that melt under the effect of surface tension evolves in droplets with a cross section much smaller then the melted area; and (5) completed experiments and modeling of laser-beam penetration through an aluminum sheet and the subsequent interaction with a pressurized vessel.

Project Summary

During the course of our project, we developed an experimental configuration to measure the temperature-dependent absorptivity measurement for diode-pumped alkali lasers at 795-nm wavelengths, and the measured values were used in our subsequent modeling efforts. We made the first known measurements of a loaded material under intense laser heating, in which a compact laser diode array was used as a laser simulator. We demonstrated that calculations with a modified Johnson–Cook strength and damage model describe well the temperature-dependent strains up to the failure point. We also modified LLNL's ALE3D code to perform continuous modeling of the heating process and melt evolution, along with effects of temperature-dependent surface tension, evaporation cooling, and recoil momentum. As a result, we developed and validated a unique tool for a broad range of laser-lethality applications. Finally, we demonstrated that a blob of hot metal resulting from laser penetration through a metal shell is able to damage sensitive elements within a target, as shown in the figure. The conditions of our laser-penetration experiments are similar to the process of additive manufacturing. In additive manufacturing, the powder and substrate must be melted, but heating must remain below plasma formation and



A melt blob formation for a 1-mm aluminum sheet irradiated at 200 W/cm^2, in which the blob itself can damage sensitive elements in a target. On the right, the modeling results for similar conditions. Arrows indicates the melt velocity.

extensive evaporation levels. As a result, the version of ALE3D we developed and validated in the penetration experiments is now widely used in Livermore's additive manufacturing activities. In addition, our method to measure temperature-dependent absorptivity has been modified to measure powder absorptivity, and is now incorporated in many additive manufacturing applications both within and outside Lawrence Livermore.

Publications and Presentations

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Lowdermilk, H., et al., 2013. *Toward predictive laser lethality*. LLNL-PRES-606012.

Rubenchik, A. M., et al., 2013. *Calibration of infrared thermal imaging camera*. LLNL-PRES-642735.

Rubenchik, A. M., et al., 2012. *Laser–material interaction experiments using a diode array to simulate a DE weapon-class source*. LLNL-PRES-512874.

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Wu S. S., et al., 2014. *Laser heating and penetration of aluminum alloys at 0.8-micron wavelength*. Directed Energy Systems Symp., Monterey, CA, Aug. 25–28, 2014. LLNL-CONF-659418

Wu, S. S., et al., 2013. *Near-IR laser interaction with composite materials*. LLNL- PRES-606013.

Multilayer Thin-Film Science for Core Missions

Regina Soufli (12-ERD-055)

Abstract

Unique scientific facilities and strategic missions are emerging that require beyond-state-of-the-art multilayer thin-film coatings to produce efficient optical systems for laser fusion systems. Such advanced multilayer coatings do not exist today. We propose new research on fundamental multilayer thin-film science topics including roughness and microstructure manipulation, corrosion mitigation, defect reduction, and smoothing. We will develop high-performance, ultrashort-period, corrosion-resistant x-ray multilayer coatings and defect-free dielectric multilayer coatings for applications to inertial-confinement fusion and inertial fusion energy programs.

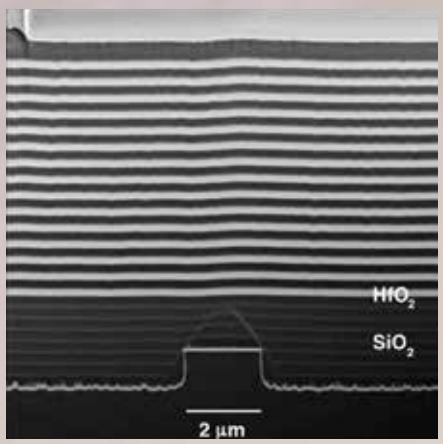
We expect our advanced ultrashort-period and corrosion-resistant multilayer coatings will enable x-ray imaging at the Linac Coherent Light Source free-electron laser at the SLAC National Accelerator Laboratory at Stanford, and significant improvements in National Ignition Facility diagnostics at Livermore. The new coatings will also provide enhancements to nuclear radiation-detection systems, as well as increase scientific capabilities for optics relevant to next-generation solar physics and astrophysics missions. Our defect-free dielectric multilayer coatings will be resistant to laser damage and would be an enabling technology for successful operation of fusion energy programs at Lawrence Livermore.

Mission Relevance

The proposed research is well aligned with Livermore's strategic mission thrust area of advanced laser optical systems and applications, through development of multilayer coatings for wavelengths near and below 6.8 nm—a key technical need for x-ray astronomy, radiation detection, photolithography, microscopy, National Ignition Facility diagnostics, and free-electron laser experiments. The project also supports the Laboratory's core competency in advanced materials and manufacturing.

FY14 Accomplishments and Results

In FY14 we (1) investigated the physics of spontaneously intermixed, partially amorphous aluminum–magnesium layers used as barriers against atmospheric corrosion in magnesium–silicon carbide multilayers, using techniques such as large-



Defect planarization increases multilayer mirror laser resistance to exceed 100 J/cm^2 at a wavelength of 1,064 nm and 10-ns pulse length.

angle x-ray diffraction and transmission electron microscopy; (2) completed the study of lifetime stability properties of magnesium–silicon carbide multilayers, with and without corrosion barriers, on samples aged for up to three years; and (3) successfully planarized (smoothed) both substrate and coating defects on subscale optics for high-fluence laser applications over multiple shots.

Project Summary

Magnesium–silicon carbide multilayer mirrors are essential in applications such as solar physics telescopes and in laser sources in the 25- to 80-nm wavelength region. By the conclusion of this project, we successfully (1) elucidated the physics of atmospheric corrosion in magnesium–silicon carbide multilayers; (2) demonstrated efficient aluminum–magnesium corrosion-barrier layers for magnesium–silicon carbide multilayers; (3) discovered and elucidated the physics of spontaneous intermixing and amorphization of nanometer-scale aluminum and magnesium layers; and (4) demonstrated corrosion-resistant magnesium–silicon carbide coatings with high reflectance in up to three narrow bands at 25 to 80 nm. In our planarized mirrors effort, we demonstrated greater than 125-J/cm^2 laser resistance (1,064-nm wavelength and 10-ns pulse length) for a single shot (see figure) and 50 J/cm^2 for 600 shots. A laser resistance of 33.5 J/cm^2 (scaled from 3- to 10-ns pulse length) has been demonstrated over a 1-cm^2 area. We are in discussions with sponsors who are interested in implementing the corrosion-resistant magnesium–silicon carbide coatings developed as part of this project in the aforementioned applications. Research Electro-Optics, Inc. in Boulder, Colorado and CSIRO, who manufactures precision optics for U.S. observatories, have both expressed an interest in our defect planarization technology for commercial applications.

Publications and Presentations

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The Next Generation of Gamma-Ray Sources: Dual-Isotope Notch Observation

Christopher Ebbers (12-ERD-060)

Abstract

We propose to develop a dual-isotope notch observation (DINO) system, a revolutionary detector arrangement that, when coupled with a mono-energetic gama-ray (MEGa-ray) source, will enable the unambiguous detection of special nuclear materials, isotope-specific imaging, and nuclear assay. For a DINO system, if a resonant isotope of interest is present in an examined object, then the resonant photons at the peak of a MEGa-ray interrogation beam will be heavily absorbed in the object, creating a "notch" in the transmitted beam. The MEGa-rays are a new class of light source with extraordinary qualities created by Compton scattering of short-duration laser pulses interacting with relativistic electrons. Our objective is to develop a nuclear detection system capable of efficiently using the next generation of laser-based gamma-ray, or MEGa-ray, sources being developed at LLNL and elsewhere. We will create the computational tools necessary to model such detectors, including enabling Lawrence Livermore's radiation transport code Mercury, or equivalent, to handle nuclear photonics. Using those tools, we will model DINO detector activities specific to the MEGa-ray source. In addition, we will build and test a DINO detector for source characterization.

Upon successful completion, we will have developed the physics code base and modeled, built, and demonstrated a DINO detector configuration for characterizing MEGa-ray sources, with an unprecedented level of isotopic-specificity detection that is two to three orders of magnitude faster than current technology. The suite of computational tools for modeling and optimizing DINO-type detectors will enhance the physics modeling capability of the Mercury code for nuclear resonance fluorescence.

Mission Relevance

By developing technology critical to the success of MEGa-ray sources, this effort supports the Laboratory's core competency of advanced laser optical systems

for establishing nuclear photo-science as a new scientific discipline to address nuclear security missions. In addition to their potential as basic research tools in nuclear photonics, narrowband-width photon sources such as MEGa-ray are expected to have a number of applications in key Livermore mission areas such as detecting highly enriched uranium for counterterrorism, precision assays of nuclear fuels and nuclear waste for counter-proliferation, and surveillance for stockpile stewardship.

FY14 Accomplishments and Results

In FY14 we (1) performed, in collaboration with the Nuclear Engineering Department at the University of California, Berkeley, analysis of experimental data from the High-Intensity Gamma-Ray Source at Duke University; (2) determined, in collaboration with Nuclear Engineering and the Nuclear Security Science and Policy Institute at Texas A&M University, the use and necessary modifications to LLNL-based codes for nuclear resonance fluorescence detection scenarios; (3) began preliminary characterization for the use of a novel narrowband spectrometer as a source for DINO-type scenarios; and (4) demonstrated the isotopic-specific detection of shielded lithium compounds and components at the Institut Laue-Langevin in Grenoble, France.

Project Summary

The project was successfully concluded with preliminary data analysis by the University of California, Berkeley Department of Nuclear Engineering. A conceptual development of a DINO-type detector was developed, along with research into modeling, design, use, and detection of narrowband radiation in the presence of large background-radiation terms. We continued the development of and added to the number of trained users for the LLNL high-fidelity multiple-particle transport code COG, which is now the sole code that contains correct physical modeling for the accurate prediction of nuclear resonance fluorescence detection scenarios. Finally, the experiment at Institut Laue-Langevin has led to several new innovative and potentially patentable methods regarding the use of narrowband radiation sources for the detection of isotopic-specific components. A conceptual detector design and a code base suitable for narrowband nuclear resonance fluorescence detection have been developed. Continuation of this research will require development of an actual narrowband source of tunable gamma-rays, presumably generated via Compton-based scattering of electrons from an energetic laser source. Currently a 30-MeV capable source has been demonstrated, with the potential for more energetic sources in the future.

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Giga-Shot Optical Laser Demonstrator

Robert Deri (13-SI-001)

Abstract

Enhanced understanding of laser-induced optical damage phenomena has historically been a key enabler for development of high-energy laser systems because their output energy is ultimately limited by the ability of their component materials to withstand high-pulse laser energy. Currently, almost no information on laser damage above 10 Hz over billions of pulses (giga-shots) is available. For example, small flaws that do not grow over exposures to a relatively small number of pulses might grow over a larger number of shots and thus impose unforeseen limits on operating conditions. This type of information is critical for designing optimized laser systems. Our objective is to develop the capability to explore optical materials damage under pulsed exposure over extreme timescales. Questions to be addressed include how long will mitigated damage sites in fused silica last and how do damage thresholds scale with pulse separation for closely spaced pulses.

We expect to develop a novel high-energy, high-repetition-rate laser capable of exploring optical materials behavior and optical damage testing for extremely long exposures. A successful project will enable the first optical damage data on extreme timescales of one billion pulses for key laser materials. It will quantify the robustness to long exposures for a variety of materials such as silica with mitigated or small

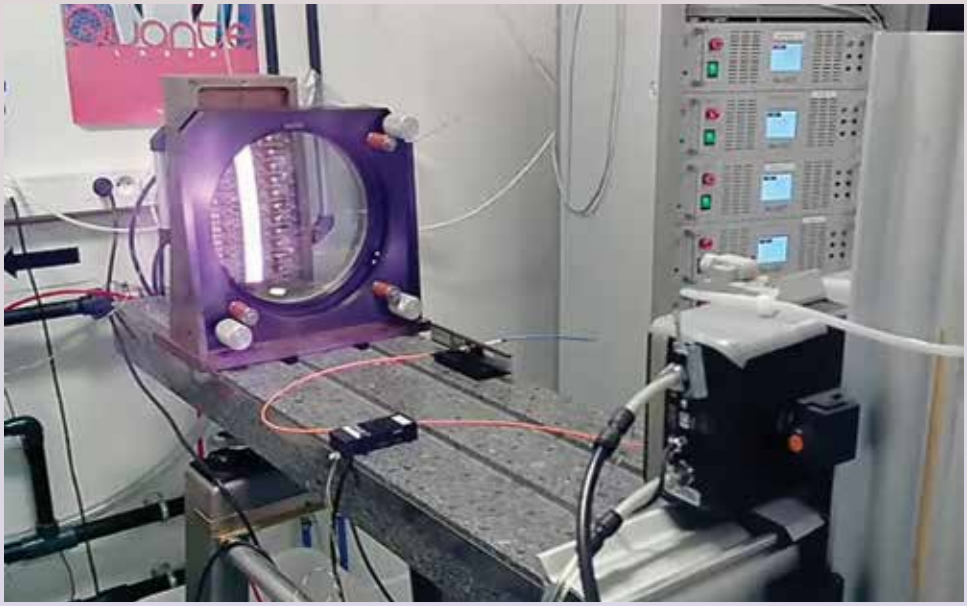
damage sites that show no damage growth for moderate pulse counts. We will explore the dependence of long-term damage on exposure conditions, providing the data necessary to devise damage mitigation and preconditioning strategies. These results will provide the insights needed to design the next generation of reliable, high-energy pulsed lasers for scientific exploration and industrial processing requiring improved energy, efficiency, reliability, and cost.

Mission Relevance

Our research will enable the design of reliable lasers with higher-energy pulses for next-generation systems important to the Laboratory and in support of the core competency in lasers and optical materials science and technology. These include the use of high-energy pulsed lasers for scientific exploration in high-energy-density science, instrumentation for gamma-ray sources and accelerators, defense and security applications such as space debris clearing, and for inertial fusion energy drivers. Such laser systems are also of interest for industrial materials processing such as laser peening and high-velocity laser-assisted deposition.

FY14 Accomplishments and Results

In FY14 we (1) completed the optical design of the entire laser system and began its implementation; (2) demonstrated that the front end provided sufficient output energy; (3) fabricated the laser amplifier gain slabs and began production of other thermal and mechanical components; (4) completed a detailed pump diode redesign to mitigate material availability issues, resulting in some delay of these components; (5) completed the fabrication of most pump delivery optics; (6) completed fabrication of the pump diode arrays (shown in figure), demonstrated their operation in the laboratory, and showed that they fully met their design specifications; and (7) secured



A diode array for our gigashot optical laser demonstrator operating with a power meter at 120-Hz repetition rate.

most of the cavity optics for high-energy operation of the overall beam line. This work has positioned us to proceed with research on our novel amplifier head design in FY15.

Proposed Work for FY15

In FY15 we will (1) complete a demonstration of the laser's capabilities, including maximizing the output pulse energy and repetition-rate performance; (2) modify the design to improve thermo-optical performance, based on what was learned from experimental work performed in FY14—in particular, we will explore methods for mitigating thermal wavefront and birefringence distortions, such as employing alternative beam-line architectures and amplifier head mechanical designs; and (3) complete the project with an extended (giga-shot) period of laser operation, which will demonstrate the system's capability of performing experiments over such extreme timescales and its utility for use in characterizing the behavior of optical materials over extreme exposure counts.

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A Compact, Femtosecond Hard X-Ray Source for Materials Characterization and High-Energy-Density Science

Felicie Albert (13-LW-076)

Abstract

New x-ray techniques are an important tool for investigating materials related to energy conversion, high-energy-density science, and manufacturing. With a new generation of ultrafast x-ray sources, coupled with novel pump and probe tools and capabilities, we can gain insight into materials behavior under extremely high temperatures and pressures. Our objective is to develop a novel compact femtosecond x-ray source using the 15-J, 15-fs Callisto laser at Livermore's Jupiter Laser Facility. We will develop the

source using modeling and experiments of laser and plasma interaction. Two additional experimental campaigns—time-resolved x-ray absorption spectroscopy and x-ray Thomson scattering—will demonstrate the source's potential as a new x-ray capability.

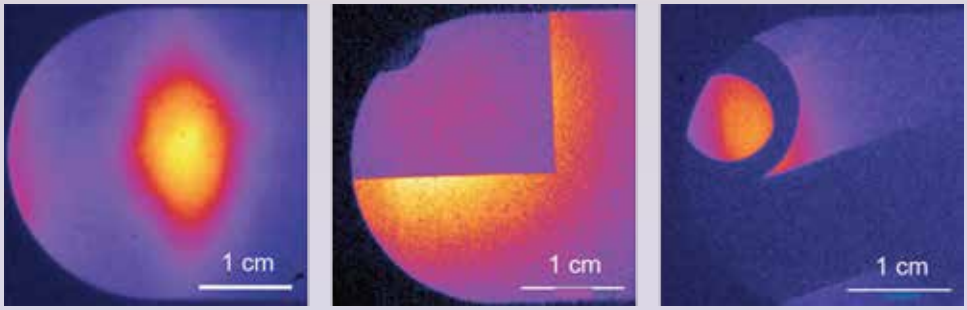
We expect to create a new x-ray source capability that is ultrafast (femtosecond), broadband, and collimated. We intend to achieve x-ray source properties of up to 100-keV x-ray energy, a pulse duration of less than 60 fs, x-ray flux greater than 10^8 photons per shot, and source size less than 1 m. We will demonstrate this new x-ray source as a viable capability for materials and high-energy-density sciences, which will lead to numerous follow-on experiments. In addition, we will obtain results with this source from time-resolved x-ray spectroscopy and dynamic warm-dense plasma studies obtained through femtosecond pump and probe experiments. Potential users of this source span the disciplines of ultrafast material characterization and imaging in industry, medicine, chemistry, protein crystallography, biology, and inertial fusion sciences.

Mission Relevance

This project is closely aligned with the Laboratory's core competency in lasers and optical materials science and technology. It leverages Livermore's expertise in accelerator, laser, and x-ray sciences and will strengthen leadership in developing novel, ultrafast x-ray light sources. The project will provide a path to better understanding of material properties and phase transitions, which is important for stockpile stewardship and in situ material characterization during manufacturing. Our research will also help reduce uncertainties in plasma properties, another important goal for stockpile stewardship science.

FY14 Accomplishments and Results

In FY14 we (1) performed source characterization at the Callisto laser; (2) observed and simulated new electron beam dynamics effects in laser wakefield accelerators; (3) secured beam time at Livermore's Titan facility to develop the betatron x-ray source in the picosecond regime; (4) secured beam time at the Astra Gemini laser in the United Kingdom for an experiment using betatron radiation to perform time-resolved x-ray phase-contrast imaging and absorption spectroscopy on a shocked iron sample, and obtained x-ray phase-contrast images of shocks in iron and silicon targets; and (5) studied potential applications of our x-ray source for high-energy-density science experiments.



Betatron x-ray radiographs obtained at the Jupiter Laser Facility in Livermore reveal images of a laser beam profile (left), silicon wafer (center), and gold ignition target capsule (right).

Proposed Work for FY15

In FY15 we will (1) implement and use betatron x-rays for time-resolved x-ray absorption studies at the 25-TW Matter in Extreme Conditions station at SLAC's Linac Coherent Light Source at Stanford; (2) characterize the source (spectrum, collimation, duration, and size) and use it to measure high-energy-density properties of a laser-heated aluminum sample; (3) measure x-ray spectrum transmitted through the sample over a broad frequency range (opacity) as well as near the aluminum K-edge (sudden attenuation increase) at 1.56 keV using extended x-ray absorption fine-structure spectroscopy and x-ray absorption near-edge structure spectroscopy to obtain time-resolved information on the aluminum plasma evolution (density, electron, and ion temperatures) under well-controlled high-energy-density conditions; and (4) complete subsequent data analysis and simulations.

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Enhancing Laser-Driven Ion Beams by Self-Guiding of Intense and Ultrashort Laser Pulses in Plasma

Derrek Drachenberg (13-FS-006)

Abstract

Generating high-energy, high-quality proton and ion beams is important to Livermore's missions in high-energy-density science, fusion, and nonproliferation, as well as for future medical applications. Laser-driven proton sources are compact and low cost compared to typical radio-frequency accelerators. We propose to test the feasibility of a new method to enhance the yield of laser-driven acceleration of ions and protons. Energetic ions can be produced via intense laser interactions with a thin solid target. However, the efficiency of acceleration can be greatly reduced if the laser pulse has low temporal contrast, and enhancing contrast requires expensive and complex modifications in the laser system. Here we plan to use self-guiding and self-focusing of intense and ultrashort laser pulses in plasma to suppress the pre-pulse intensity before reaching the target, and demonstrate enhanced ion acceleration.

We propose to test the feasibility of a new method to enhance the yield of laser-driven acceleration of ions and protons by employing self-guiding intense and ultrashort laser pulses in a gas cell prior to impacting the solid target foil where the ions are generated. This proposed method shifts the complex technology of pulse cleaning and intensity enhancement from the laser to a simple modification of the target: pre-pulses are not guided in the gas cell, therefore the foil stays intact before the main laser pulse arrives, which is guided in the gas cell with its intensity increased. The prospect of an increased yield will improve the signal-to-noise ratio in radiographs with a simpler laser system. Likewise, the production of higher-energy ions paves the path towards a compact ion

source for various applications. Upon successful demonstration of enhanced ion acceleration, this contrast enhancement scheme can be generalized to any experiment demanding high contrast and high intensity, as a novel and inexpensive approach to mitigating inherent limitations from the laser system. It can be leveraged towards development of highest-energy proton and ion sources on future large-scale laser facilities.

Mission Relevance

This proton acceleration methodology has never been demonstrated before. This work will impact laser-based particle acceleration technologies and high-energy-density physics, which support multiple Laboratory mission focus areas as well as the science, technology, and engineering core competencies in lasers and optical materials and the study of properties of matter under extreme conditions of temperature and pressure in high-energy-density science.

FY14 Accomplishments and Results

In FY14 we determined that the method of pre-pulse cleaning by self-guiding the laser pulse through a low-density plasma is feasible. We observed that the maximum proton beam energy does not depend on foil thickness, which indicates a successful reduction of the on-target pre-pulse intensity.

Project Summary

We developed and fabricated a modified target design integrating the laser wakefield acceleration gas-cell design with an angled foil holder for proton beam generation via target normal sheath acceleration, in which relativistic electrons accelerated by intense laser light propagate through a target and build up a strong electric field at the rear surface. Because of the strong field strength, the atoms at the target surface are ionized and accelerate in the target's direction. We conducted experiments at Livermore's Jupiter Laser Facility using the Callisto laser. Diagnostics for measuring target normal sheath acceleration proton-beam characteristics included radio-chromic film stacks, a Thomson parabola ion spectrometer, and imaging plates. The self-guiding length in the gas cell for optimized contrast enhancement was estimated to be about 4 mm and compared to experimental results, and we compared proton beam acceleration with and without self-guiding. Further analysis of the current data should be extended to understand the scaling of proton energy to foil thickness for the entire proton spectrum, and additional experiments will be required to determine the cause of the reduction in proton cutoff energy with self-guiding. Finally, demonstration of ion beam generation with high repetition-rate short-pulse laser systems is desired.

Publications and Presentations

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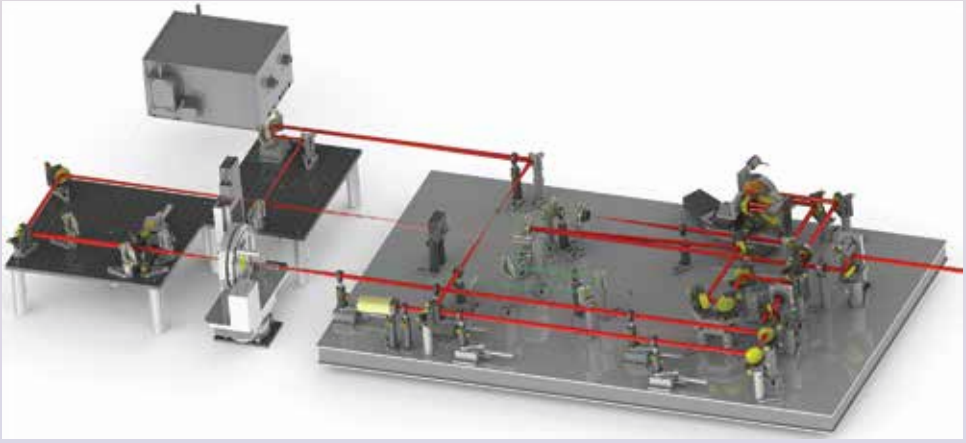
Picosecond Laser Interactions with Materials: Mechanisms, Material Lifetime, and Performance Optimization

Ted Laurence (14-ERD-014)

Abstract

High-peak-power laser pulses, when focused to relativistic intensities, can drive intense secondary sources such as energetic electron beams, proton beams, and kiloelectronvolt to megaelectronvolt x-ray sources that are highly relevant for important DOE and defense missions such as time-resolved backlighting of dense materials or future laser-driven compact accelerators and their broad range of applications. Laser-induced damage of optical materials is a particularly interesting example of laser and material interactions in which the material itself is destroyed. We propose to understand the key mechanisms of picosecond laser-induced material modification in materials used for reflective and diffractive optics and for nonlinear materials and lenses, and understand the limits of these materials as a function of laser parameters and processing and environmental conditions. We will develop techniques to measure the key material parameters believed to be important in this regime, including multiple-photon absorption, free-carrier absorption, and avalanche breakdown in both intrinsic and defective materials, and we will use these measurements to develop models of optical lifetime. Finally, based on these results, we will develop processes or materials that improve the lifetime and performance of optics for short-pulse systems.

We expect to determine intrinsic, fundamental damage thresholds and extrinsic surface defect effects for dielectric materials used for coating short-pulse optical surfaces. In addition, we will characterize chemical and defect modification processes in short-pulse laser material caused by high-average-power conditions with short pulses. We expect to perform measurements of elementary excitation processes and the channels by which energy stored in the electronic system is transferred to the material in the solid state, and simulate material response in this intensity range. Identification of dominant failure mechanisms will enable material and process solutions to improve the reliability of optics for picosecond-class systems. We intend to explore conditioning by slowly ramping laser intensities as a mitigation strategy.



We have developed a new laser-based pump and probe experimental setup that will be used to understand how optical surface defects affect the processes that lead to laser-induced damage with picosecond lasers.

Mission Relevance

Optics for advanced high-energy laser systems are required to withstand high peak power at pulse lengths from under 1 to 100 ps—some under high-repetition-rate, high-average-power conditions. By establishing test parameters and clarifying fundamental mechanisms, we will help guide the design and operation of optics under these extreme conditions. This work is in support of the Laboratory's core competency in lasers and optical materials science and technology that contributes to the design of future high-energy and high-peak-power laser systems.

FY14 Accomplishments and Results

In FY14 we (1) obtained preliminary evidence that extrinsic surface factors affect damage performance of fused silica surfaces, in contrast to previous claims that damage for 1-ps pulses and below are solely because of intrinsic, bulk material properties; (2) completed testing the damage behavior of fused silica from 1 to 30 ps for both fused silica and deposited silica coatings; (3) completed our small beam-damage test station and pump and probe setup, shown in the figure, and expect commissioning in early FY15; (4) developed new software control systems for picosecond laser damage testing and analysis; and (5) developed a new collaboration with the Laboratory for Laser Energetics at the University of Rochester to produce single-layer and multiple-layer coatings for our experiments. This new collaboration will allow us to study effects of deposition processes on laser damage performance in the picosecond regime, especially to determine dominant failure mechanisms and explore mitigation strategies.

Proposed Work for FY15

In FY15 we will (1) extend the work on fused silica surfaces to include samples with single- and multiple-layer coatings of silica, alumina, and hafnia and include laser damage performance studies and measurements of multiple-photon absorption rates and free-carrier absorption rates; (2) begin studies of multiple-pulse effects for understanding damage growth and surface degradation; and (3) use our collaboration with the Laboratory for Laser Energetics to explore more promising mitigation efforts related to deposition processes such as electron-beam deposition, rather than laser-based mitigation and conditioning processes.

Thermal Management of High-Heat-Flux Laser Diodes Using Liquid-to-Vapor Phase Change

Jack Kotovsky (14-ERD-040)

Abstract

Semiconductor laser diodes are the preferred light source to pump laser gain media because of their higher brightness and narrower emission spectra, which facilitate more powerful and more efficient laser systems for a wide variety of LLNL mission-relevant applications, including fusion energy, laser-assisted manufacturing, and directed-energy

systems. However, the fundamental limitation in state-of-the-art laser diode systems remains the removal of heat from the diodes to the cooling fluid. Improved cooling would extend both the lifetime and the use of these laser systems. We will develop a laser diode package that uses an ultrathin heat pipe or open-loop, two-phase flow system to conduct heat away from diode surfaces during continuous operation. We will utilize the Laboratory's expertise in micro-electromechanical systems packaging, material joining, heat transfer, and laser diodes to develop a solution that removes very large (greater than 1 kW/cm^2) heat fluxes while maintaining a uniform diode temperature. By increasing power output per diode chip, this improvement will redefine the cost–performance relationship and expand the potential use of laser diodes for Laboratory missions.

We will investigate the use of a cooling-fluid phase change to enhance heat removal and thereby enable new applications. Specifically, we will deliver (1) a liquid–vapor phase-change model for heat pipe operation, (2) a working prototype for heat fluxes and related performance data, (3) an integrated laser diode and cooling-system package design, (4) a combined thermal and structural model of laser diodes and cooling systems, and (5) a fabricated one-bar laser diode and cooling-system package and related performance data.

Mission Relevance

This project supports Laboratory missions in national and energy security and strategic focus area in inertial-confinement fusion science and technology by expanding the lifetime and increasing the potential uses of semiconductor laser diodes, which are critical for applications such as laser inertial fusion energy, materials processing such as laser peening and laser-assisted deposition and cladding, and high-average-power lasers for directed energy, generation of mono-energetic gamma rays, and other scientific instruments. Fundamental research in expanding the potential for laser diodes is also aligned with the Laboratory's core competency in lasers and optical materials science and technology.

FY14 Accomplishments and Results

In FY14 we (1) produced results from thermal models of laser diode cooling systems under consideration—open (flow through) and closed (heat pipe) designs both continue to hold promise of excellent thermal performance and are being pursued in parallel; (2) explored, based on flow models, varied wicking structures for the closed system and decided on a rectangular cross-sectioned architecture verses a porous metallic structure; (3) chose silicon for the coolant structure because it can be shaped to deliver relevant, micro-scale geometries and has excellent thermal properties; (4) quantified the prototype wicking channel's performance with a video system and selected geometries for the closed-system design in process; (5) designed and built an open-geometry prototype system, which is ready for testing in FY15, ahead of schedule; (6) constructed manifold assemblies to join the open prototypes to a fluidic test platform, which was designed and completed for device characterization in FY15; and (7) tested deposited layers to match coolant structure to diode bars with regard to expansion.

Proposed Work for FY15

In FY15 we will (1) continue hardware development to produce prototypes of both the closed and open systems, (2) conduct testing and quantification of their performance throughout the year and evolve the designs based on these results and ongoing use of computer-based models, (3) fabricate repeating-unit designs for both systems unless there is a clear motivation to select a single system, and (4) develop micro-electromechanical system layout, etching, bonding, metallization, and dicing and cutting processes.

Understanding the Creation and Reduction of Surface Microscale Roughness During Processing of Glass Optics

Tayyab Suratwala (14-ERD-042)

Abstract

Optics with low-scatter and high-damage-threshold surfaces are important for high-peak-power and high-average-power laser systems. In addition to enabling the higher fluence and higher power operation of these systems, optics with such improved surfaces could also provide greater flexibility in designing future lasers. Techniques recently developed to improve the damage thresholds of glass optics have done so at the expense of surface smoothness, which is significantly degraded as a result. The need therefore exists for processes to produce low-roughness optics, with reduced light scatter, reduced laser contrast, and improved laser-damage resistance. In this project, we will develop a scientific understanding of the microscopic and molecular interactions that occur during polishing and post-processing and how they influence the roughness of glass surfaces. This investigation will include a thorough study of the microscopic interactions occurring during polishing and post-chemical treatments. We will then use our new knowledge to develop cost-effective methods to achieve very low roughness—that is, near-atomic-level smoothness—on optical glass surfaces both before and after post-processing.

Our major deliverables are a science-based understanding of the microscopic and molecular-level interactions occurring during polishing and post-processing—with an emphasis on the creation, control, and prediction of roughness—and novel, low-cost processes to achieve low-roughness surfaces during polishing and after post-processing. Our processes will be compatible with other novel finishing processes, such as convergent polishing.

Mission Relevance

By aiding in developing improved finishing processes for advanced laser optics—delivering lower surface roughness, reduced optical scatter, and potentially greater resistance to high-fluence laser damage—this project supports Lawrence Livermore's mission in stockpile stewardship and its core competency in lasers and optical materials science and technology.

FY14 Accomplishments and Results

In FY14 we (1) completed the facility setup of polishers and workpiece-cleaning tooling tailored for controlling surface microroughness, (2) performed the first set of polishing experiments and chemical and mechanical characterization of the nature and mechanism of impurity (potassium and cerium) penetration into the outermost work-hardened polishing layer (Bielby layer), (3) developed a new nanometer-scale scratching technique based on atomic force microscopy to measure single-particle removal function during polishing, (4) performed polishing experiments relating pad roughness and hardness to workpiece roughness and material removal rate, and (5) developed a quartz-crystal microbalance flow-cell system to study the effect of workpiece roughness on redeposition.

Proposed Work for FY15

In FY15 we will (1) conduct additional polishing experiments at various slurry compositions and pH to further describe the Bielby polishing layer; (2) formulate a chemical and structural model for creation of the Bielby layer and its influence on surface roughness; (3) complete single-particle removal measurements as a function of particle material, size, velocity, and load; (4) investigate the role of pH, slurry viscosity, and slurry composition on workpiece roughness during polishing; (5) conduct experiments on post-polishing treatments on workpiece roughness; and (6) measure polishing reaction-product adsorption and precipitation kinetics using a quartz-crystal microbalance flow-cell system.

Publications and Presentations

Suratwala, T., 2014. "Microscopic removal function and the relationship between slurry particle size distribution and workpiece roughness during pad polishing." *J. Am. Ceram. Soc.* **97**, 81. LLNL-JRNL-635673. *http://dx.doi.org/10.1111/jace.12631*

Multichannel Air-Guiding Fibers to Transport Extreme Laser Beams and Enable High-Flux Particle Accelerators

Michael Messerly (14-ERD-070)

Abstract

Optical fibers have become increasingly important not only for telecommunications but also for high-power lasers. Fiber lasers are efficient, compact, and robust and have applications in missile defense, medicine, and guide-star lasers, as well as high-flux laser-based particle accelerators. However, nonlinear propagation artifacts limit the powers and pulse energies of fiber lasers. We recently suggested that a tradeoff between beam self-focusing and stimulated Raman scattering will limit conventional fiber lasers to average powers of 37 kW, well below the 100 kW to 1 MW needed for missile defense. We propose to develop a new waveguide class of air-guided multichannel fibers.

Compared to existing air-guided fibers, our fibers will show that an array of resonant holes resolves stability problems. One version would be able to transport photons for the Laboratory's envisioned missile-defense lasers. The other, tuned to different wavelengths, would be able to transport pulses having megawatt to gigawatt peak powers.

We believe that to achieve and transport the average powers needed for defense-class lasers and the peak powers needed for DOE-class particle accelerators, we will need new types of optical fibers. We expect to demonstrate, through modeling, fabrication, and testing, that air-guided multichannel optical fibers can transport missile-defense class lasers of 100-kW average power and laser pulses suitable for surgical and micro-machining applications in the gigawatt peak-power range. We also expect to demonstrate the first air-guided multichannel fibers that propagate modes appropriate for laser-based particle acceleration. Furthermore, we expect to establish baseline competencies for fabricating all types of hollow-core photonic crystal fibers, enabling custom fibers for aerospace and medical applications. Specifically, we intend to fabricate and test hollow-channel fibers with a flattened mode on a hexagonal grid. The fibers will have relatively thick webs to dissipate and tolerate laser-generated heat and to permit transport of high-power laser light while maintaining good beam quality. The fibers will be computer modeled and then fabricated on the LLNL fiber draw-tower facility and tested both at Livermore and at collaborator facilities.

Mission Relevance

Our research supports the national security mission by providing a safe, vibration-insensitive conduit of extreme light from a centrally located laser to a target or beam director. The work also supports the Laboratory's core competency in lasers and optical materials science and technology by proving paths to new sensors and lasers, such as

Fabricating photonic crystal optical fibers as part of our research into hollow-core fibers that will dissipate and tolerate laser-generated heat and permit transport of high-power laser light while maintaining good beam quality.

long-length gas sensors and compact gas lasers. Long-term, the project supports DOE's particle acceleration mission by providing a path to high-flux laser-based particle accelerators, allowing a thousandfold increase in energy or reduction in size over accelerators based on radio waves.

FY14 Accomplishments and Results

In FY14 we developed models and software to analyze air-guided and patterned-mode optical fibers. The air-guided fibers included those in which negative curvature regions bind the light, which promise extremely low losses. The patterned-mode fibers included both half- and quarter-wave tiles. The latter represent a new concept and may improve bending performance significantly. We also developed processes for fabricating air-guided fibers. This work began by studying and adopting the methods of the University of Bath, an acknowledged leader in this field. In addition, over the final months of FY14, we developed an alternative technique that is more reproducible and that seems able to achieve much thinner structures than those obtained with the Bath technique. We have not yet tested prototype fibers with pulses having 100-MW peak powers. The fine glass structures in air-core fibers must be very thin, on the order of one-sixth the wavelength of the guided light. While our prototypes may guide infrared light, they cannot yet guide near-infrared or visible light, our target wavelength ranges. We believe that our new fabrication method will allow us to achieve this in FY15.

Proposed Work for FY15

In FY15 we plan to (1) design, fabricate, and test two new fibers—a multichannel fiber, selectively doping the channels' cores with a rare earth to stabilize the preferred flattened mode, and a fiber that supports a multichannel vortex mode, which would be the first of its kind; (2) model the effects of twist on multichannel guides, with which we expect to demonstrate that it is possible to selectively radiate unwanted modes; and (3) develop experimental techniques for filling and sealing hollow fibers with gases in preparation for future laser development.

Short-Wavelength, High-Power Fiber-Laser Sources

Paul Pax (14-ERD-078)

Abstract

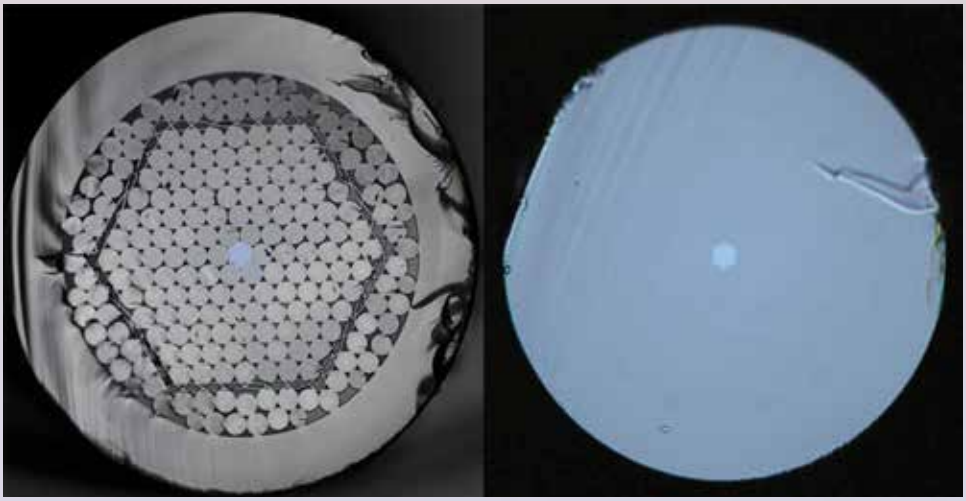
High-power lasers are at the core of defense applications such as directed-energy systems for missile or artillery defense, secure laser communications, and remote sensing. Fiber-based laser sources are natural candidates for these applications, offering unmatched beam quality, efficiency, thermal management, and reliability. However, the overwhelming majority of work on fiber lasers has been in the near- or mid-infrared region of the spectrum. Operation at shorter wavelengths has been overlooked, despite advantages such as beam control. Shorter wavelengths (i.e., visible versus infrared light)

would reduce the size and the weight of a directed-energy device by a factor of four, or for fixed aperture, increase the power density on target by a factor of four. We propose to evaluate and demonstrate the prospects of novel short-wavelength fiber lasers. We will develop new materials, fabrication methods, and fiber designs; use new blue diode pumps; and push frequency conversion to high power. We will explore several approaches to extending high-power fiber-laser technology to short-wavelength operation. An important feature of our research will be the development of doped glass fabrication by the sol-gel method for producing solid materials from small molecules, which is not yet widely used in this application.

Developing approaches to extend high-power, fiber-laser technology to short-wavelength operation will improve or enable multiple defense applications, such as directed-energy systems and submarine communications. In addition, we will develop modeling and design expertise, develop new glass and fiber fabrication techniques applicable to the LLNL fiber optic draw tower, and gain expertise in high-power harmonic generation, all of which will position us for a lead role in kilowatt power scaling. We intend to evaluate three approaches: (1) novel active species lasing in the visible region, pumped with blue diodes; (2) neodymium-doped lasers at 900 nm; and (3) high power and efficiency frequency conversion. We will fabricate new fibers, test new lasers, and evaluate power scaling, building on existing LLNL resources and expertise.

Mission Relevance

High-power, short-wavelength fiber lasers will benefit defense technologies in support of a central Laboratory strategic mission in defense and a core competency in lasers and optical materials science and technology, specifically in the area of directed-energy systems and secure and covert communications. Success would also benefit nondefense applications, such as guide stars for astronomy and sources for high-brightness color displays.



The laser fiber on the left shows an intermediate stage in the process of drawing the preform starting materials to the finished fiber. At the center is the neodymium-doped core, which is the lasing medium. The outer hexagon allows an air gap into the fiber cross section, forming a confining pump-light cladding. The image on the right shows the finished fiber, in this case without cladding.

FY14 Accomplishments and Results

In FY14 we (1) designed a W-shaped profile fiber for short wavelength lasing on the neodymium 3-level transition around 930 nm, to suppress the 4-level gain around 1,060 nm; (2) procured a first glass sample—unfortunately, delivery was delayed, and worse, the glass suffered high losses and concentration quenching, making it unusable for our purposes; (3) engaged another vendor who uses a different fabrication process, and procured a glass sample on a best-effort basis; (4) fabricated and tested a simple step-index fiber from the second vendor's glass, which shows much better loss and quenching properties; (5) quantified the lasing properties, and engaged in discussion with the vendor regarding another iteration; and (6) constructed a simple fiber-laser model to benchmark to our experimental results.

Proposed Work for FY15

In FY15 we will (1) extend our current work on novel wavelength fiber lasers by developing a fiber source operating directly in the visible wavelengths, enabled by recent and ongoing commercial development of blue nitride diode lasers for applications in data storage and lighting; (2) take advantage of these new blue nitride diodes to fabricate a samarium-doped active fiber, and build and test a fiber laser operating at 651 nm; (3) engage with potential vendors for the starting glass material; and (4) pursue an alternative approach based on Livermore sol-gel technology.

High-Average-Power Diffraction Pulse Compression Gratings Enabling Next-Generation Ultrafast Laser Systems

Leon Haefner (14-ERD-084)

Abstract

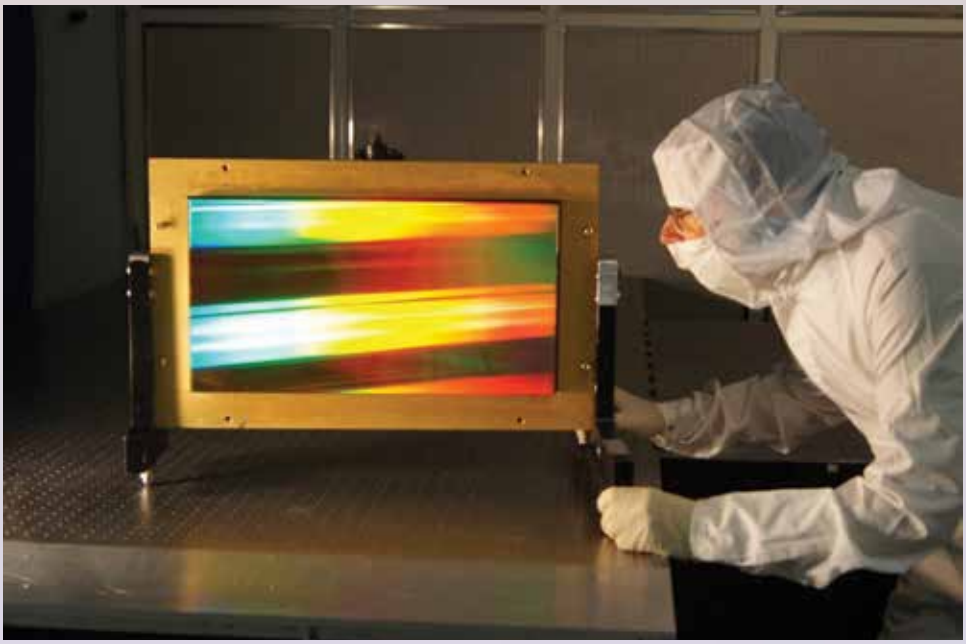
For more than 15 years, the Laboratory has been a leader in the design, development, and fabrication of meter-scale diffraction gratings used to stretch or compress pulses to modify laser energy for femtosecond laser systems. The gratings are key to achieve high laser irradiance, and are used in short-pulse compressors in a scheme referred to as chirped pulse amplification. High-average-power ultrafast lasers are required for machining or driving high-flux secondary sources such as coherent or incoherent x-ray generation and particle accelerators. However, new laser capabilities require new gratings. The main challenge in operating at high average power is thermal management in the laser amplifiers and control and mitigation of thermally induced distortions in the diffractive pulse-compression gratings. We propose to develop new pulse-compression gratings to enable high-average-power (>100 W) and enhance existing systems by up to 25%. We intend to (1) develop and engineer the process for fabricating broadband, metallic gratings capable of operating at 300-W average power; (2) understand the impact on short-pulse laser performance of deposited heat in large-area gratings and implement active heat removal; (3) explore and develop a nonplanar laser pulse-compressor design; (4) design

and develop high-average-power, high-efficiency, nonplanar, multiple-layer dielectric diffraction gratings supporting 30-fs pulses or better; and (5) construct a test laser.

If successful, we will design, develop, and demonstrate actively cooled, metallic gratings capable of the shortest pulse compression of femtoseconds at high average power greater than 300 W (a tenfold increase in average power capability), as well as high-efficiency multilayer dielectric gratings capable of short-pulse compression greater than 30 fs with very high efficiency. We will develop an understanding of the impact on short-pulse laser performance of deposited heat in large-area gratings and implement active heat removal. Our new designs will enable energetic high-average-power ultrafast lasers and improve the compressor efficiency up to 25%. Success will also enhance the Laboratory's grating manufacturing capabilities and provide numerous opportunities for collaboration with external high-energy petawatt laser projects. It will further pave the path to the next generation of ultrahigh-repetition-rate ultrafast laser systems employing diode-pumped solid-state lasers.

Mission Relevance

High-average-power, ultrafast lasers are highly relevant to defense, homeland security, fundamental science, biological, and medical applications because of their compactness, high brilliance, and potentially lower cost than conventional sources. Ultrafast laser drivers require a new generation of diffractive optics that can withstand high average power and actively manage residual heat. Our work addresses these challenges and will enable high-average-power, short-pulse laser operations never before achieved, in support of the Laboratory's core competency in lasers and optical materials science and technology.



A diffraction grating suitable for a petawatt-class laser pulse compressor. Hafnium oxide is deposited onto an ultralow expansion glass substrate. A 1,480-line/mm groove structure in the hafnium oxide is fabricated using holographic lithography and then coated with a gold layer.

FY14 Accomplishments and Results

In FY14 we (1) developed the process to deposit gold on hafnium oxide; (2) performed successful fabrication and re-processing of a 1,480-line/mm grating patterned and etched into a hafnium oxide layer on a 4-in.-diameter ultralow expansion substrate; (3) demonstrated a 2-in.-diameter complete gold-overcoated grating deposited on hafnium oxide; (4) designed the test laser system and established the front end that provides the initial, seed laser beam; (5) developed the conceptual design for the grating test stand; (6) designed and initiated the production of optics for the laser pulse stretcher; (7) fabricated a full-scale petawatt diffraction grating that met the above specifications (shown in figure); and (8) commissioned the test laser system. Because of a reduced budget, we delayed the development and testing of the multilayer dielectric gratings and compressor geometries until FY15.

Proposed Work for FY15

In FY15 we plan to concentrate our efforts on (1) modeling, understanding, and optimizing the electric field distribution and suppression of guiding mode resonances in the underlying multilayer dielectric stack in the diffraction gratings; (2) develop broadband multilayer dielectric grating designs and fabricate test samples; (3) design and demonstrate a proof-of-concept for out-of-plane short-pulse compressor geometries using the test laser system; (4) characterize and perform damage testing of test grating samples; and (5) model, design, and test an active cooling concept for average-power gratings.

Publications and Presentations

Britten, J. A., D. A. Alessi, and L. C. Haefner, 2014. *A new class of metal diffraction gratings tuned for maximum efficiency and bandwidth for the next generation high intensity petawatt lasers.* Intl. Conf. Ultrahigh Intensity Lasers, 2014, Goa, India, Oct. 12–17, 2014. LLNL-ABS-654675.

Laser–Matter Coupling Mechanisms Under Varying Chemical and Particulate Surface Configurations

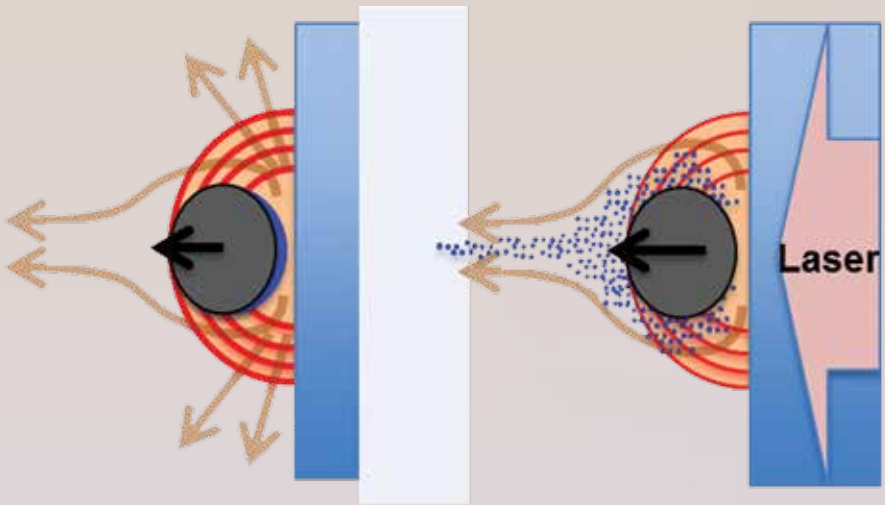
Manyalibo Matthews (14-ERD-098)

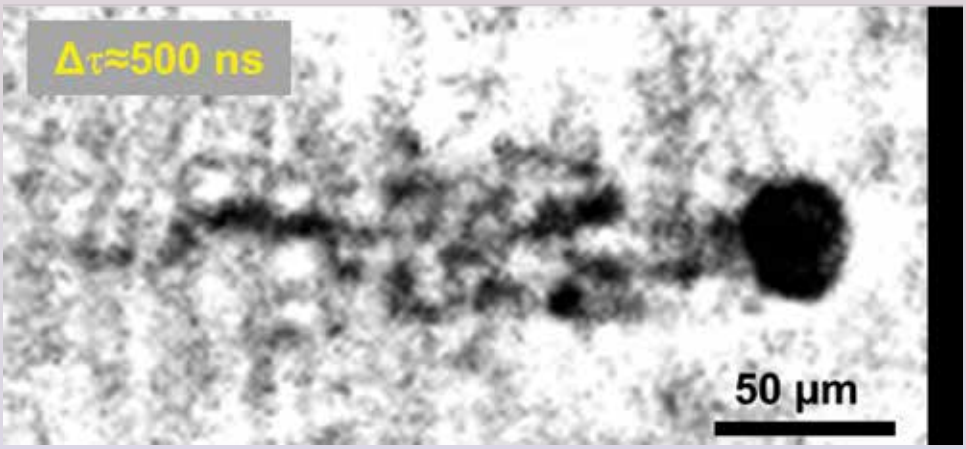
Abstract

Particulate contamination negatively impacts a wide range of high-performance materials and devices, including microelectronics, space optics, lasers, and nuclear reactors. Such contamination is especially problematic in high-photon-flux lasers. This problem calls for improved optical designs with contamination-mitigation strategies for high-power laser systems. We will study the morphological and chemical evolution of surface-bound microscopic-scale particles in the presence of high-power laser light and those particles'

interaction with an underlying optical surface to better understand laser–particle material interactions over temporal scales of pulsed-to-continuous waves. To this end, we will (1) develop a suitable test chamber and diagnostic suite; (2) conduct experiments with different particle–surface pairs, chemical environments, and laser conditions; (3) simulate laser absorption, material response, and subsequent effects on laser beam propagation; and (4) explore mitigation strategies, including pre-pulse laser cleaning, chemical passivation, and coatings. Our primary focus will be on mirrors and windows in diode-pumped alkali lasers and high-power laser systems related to ignition fusion research.

We expect to develop a functioning test bench that can be used to examine particle-driven damage events in situ and under a wide variety of conditions. We will also establish a computational capability that allows accurate simulation of laser and particle interactions at high laser powers. In addition, we will generate findings addressing the propensity for damage from specific particulate contamination in





Laser ablation of idealized spherical metal contaminants from fused silica optics, shown schematically and captured using nanosecond pump–probe imaging. Significant material spall is observed, which can degrade the optic.

optics used in both high-power, continuous-wavelength lasers and high-energy, pulsed-power lasers. Finally, we will deliver an effective scheme for using coatings or vapor chemistry treatments to limit both local and nonlocal damage (that is, damage generated by phase objects).

Mission Relevance

Our research on thermal transport in laser-heated metal on surfaces can advance laser-based additive manufacturing technologies for stockpile stewardship applications. This project also enhances the performance of high-power laser systems in support of the Laboratory's core competency in lasers and optical materials science and technology and can extend our understanding of the effects of contamination on light propagation in the large, high-performance optics used in cyber security, space, and intelligence applications.

FY14 Accomplishments and Results

FY14 experiments consisted of large-aperture laser tests where a large number of particles bound to optic surfaces could be tested at once. These experiments also enable small-beam dynamic testing where the time-resolved response of individual particles can be probed. Simulation and modeling efforts focused on applying laser ablation theory to particle–optic interfaces, and simulating the effect of laser–particle ablation pits on final optics performance (degradation) using the Fourier optics theory, which addresses the wave properties of light. Specifically, we (1) correlated haze-scattering sites with aluminum and borosilicate particle-ablation events, revealing a large asymmetry in damage behavior between material types; (2) successfully executed pump–probe measurements of laser-ejected tungsten, aluminum, and iron particles on fused silica exit surfaces, clarifying momentum-transfer mechanisms through comparison with theory; (3) executed experiments on dielectric mirror coatings contaminated with titanium particles; (4) measured time-resolved plasma heating associated with laser–particle ablation; and (5) developed analytic solutions to far-field scattering patterns created by laser–particle ablation events.

Proposed Work for FY15

In FY15 we will (1) develop a model for ablation-driven diffraction from phase objects; (2) commission an environmentally controlled test chamber for pump–probe experiments to explore pressure and humidity effects; (3) conduct a review of diagnostics and alternatives, and repeat FY14 experiments as needed; (4) develop an electromagnetic code for a particle–light coupling model; and (5) extend our study to include the recently discovered tightly bound debris category of contamination on optics.

Publications and Presentations

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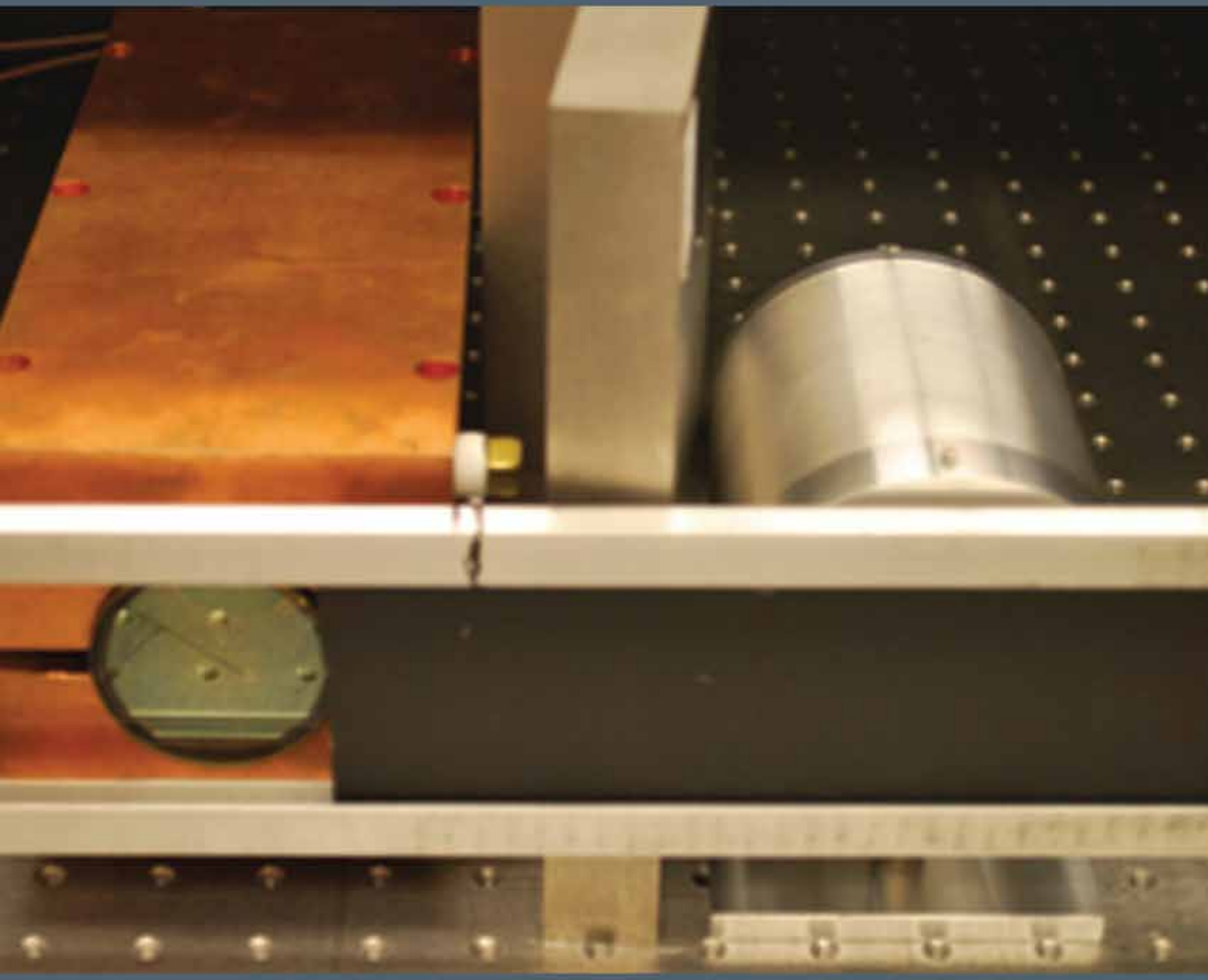
Nuclear Science and Technology



FY2014

Lawrence Livermore National Laboratory

Ultrahigh-Burn-Up Nuclear Fuels

Patrice Erne Turchi (12-SI-008)

Abstract

One of the key questions for the U.S. as it seeks to incorporate nuclear energy in its clean-energy strategy is how to more completely burn nuclear fuel in its power plants. We propose to make significant advances in the basic science for the development and qualification of advanced, ultrahigh-burn-up nuclear fuel. To achieve this goal, we will couple modern computational materials modeling, fabrication, and characterization capabilities and targeted performance-testing experiments using ion-beam facilities. This project will establish the scientific foundation for selecting the optimum fuel type for advanced reactor concepts.

Our work combines a robust experimental program with validated modeling. We will experimentally quantify the stability and kinetics of phase transformations, inter-diffusion, microstructural evolution, micromechanical properties, and the influence of severe radiation environments on fuel performance. Ultimately, we will have a validated model for advanced nuclear energy materials under extreme conditions of radiation, temperature, and evolving chemistry. We will also have a science-based path forward to an optimized inert matrix fuel, while contributing to the development of a validated nuclear-fuel database.

Mission Relevance

Our approach to developing the science of advanced nuclear energy fuels aligns well with the Laboratory's energy and national security missions. Development of both advanced fuel cycles and hybrid fusion–fission concepts face the same scientific challenges. This research will extend LLNL capabilities and further enable actinide science for high-energy-density science, energy manipulation, and materials on demand, and is relevant to the core competency in nuclear science and technology.

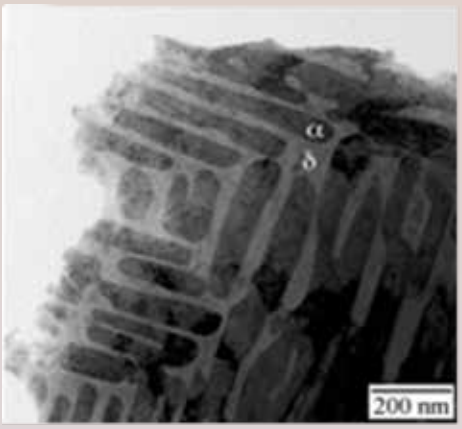
FY14 Accomplishments and Results

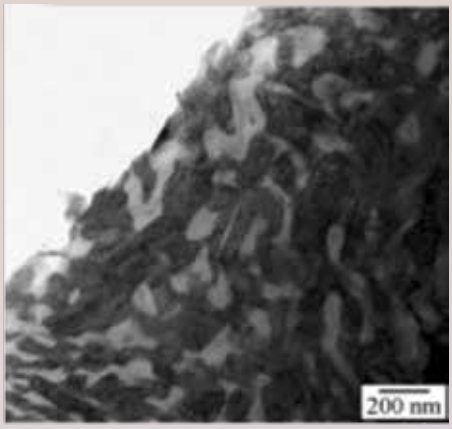
In FY14 we characterized samples irradiated in FY13. Specifically, we (1) examined the role of daughter products on the stability of gadolinium–plutonium and aluminum–plutonium alloys—by recasting the ab initio energetics for the body-centered cubic phase in the CALPHAD framework (computer coupling of phase diagrams and thermochemistry), the plutonium–uranium phase diagram has been successfully thermodynamically re-assessed; (2) combined the plutonium–uranium thermodynamic functions with the plutonium–gadolinium and uranium–gadolinium functions to predict the ternary plutonium–uranium–gadolinium system, which has never been assessed or reported in the literature; (3) showed, through our predictions, that a small amount of uranium affects the plutonium–gadolinium phase stability by precipitating the other complex phases, and hence may impact swelling and mechanical integrity during aging; (4) determined that both plutonium–uranium and plutonium–uranium–gadolinium phase diagrams and property diagrams can be used to design experiments that are

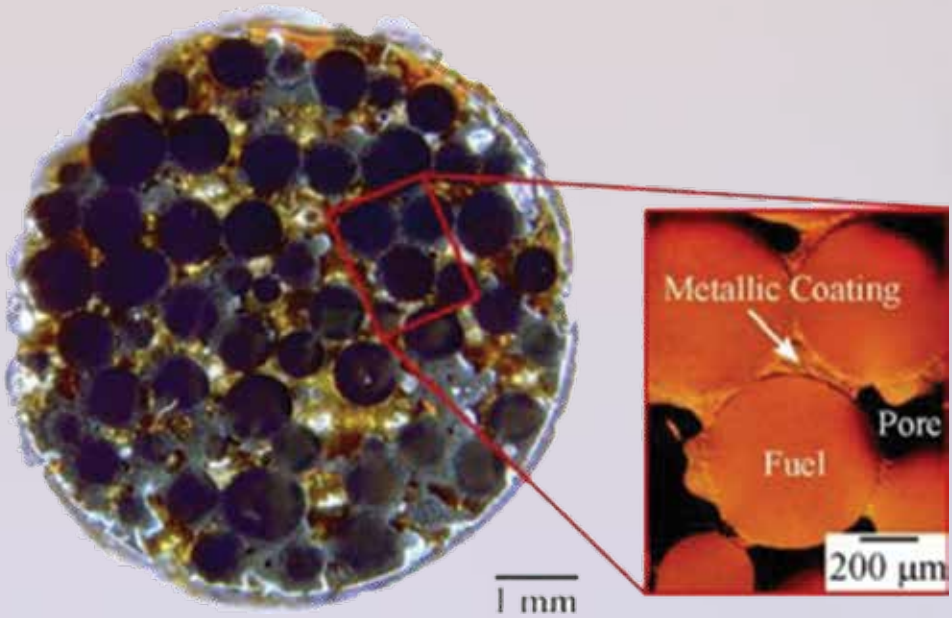
usually costly, challenging, and time-consuming—indeed, these preliminary diagrams can be used to identify compositions and temperature ranges where maximum information can be obtained to further improve or validate the proposed thermodynamic database; (5) predicted the thermodynamic properties for the binary americium–-uranium and ternary americium–plutionium–uranium systems, and evaluated the role of americium on the stability of plutonium–uranium; and (6) characterized samples of molybdenum–uranium experimentally with x rays and transmission electron microscopy, which revealed the existence of a new phase for this alloy, and compared non-irradiated and irradiated samples of uranium–zirconium, which showed changes in microstructure morphology with consequences on fuel performance.

Project Summary

We have investigated, using an ab initio CALPHAD approach, the thermodynamic properties of a series of actinide-based alloys, including a number of binary alloy systems,







As-cast uranium–zirconium alloy shows the coexistence of alpha-phase uranium (dark contrast) and delta-phase uranium–zirconium (light contrast), seen top left. The material is composed of alternating lamellar, or plate-like, microstructures of the two phases. Loss of aligned lamellar microstructure for uranium–zirconium alloy after irradiation at Livermore's Center for Accelerator Mass Spectrometry is shown top right. A micrograph (bottom left) provides a cross-sectional view of a mock inert matrix fuel fabricated for demonstration purposes. In the mock fuel, zirconium spheres serve as a fuel surrogate. The metallic coating is made of a zirconium-based alloy, and a stainless-steel cylinder provides the cladding. A higher-magnification image (bottom right) shows the alloy coating the fuel spheres, which is important for high thermal conductivity, and also shows porous regions, which accommodate fission gas during the reactor's operation.

several ternary systems, the quaternary aluminum–molybdenum–silicon–uranium system, and a promising inert copper-based alloy metal coating. The LLNL phase-field modeling code has been updated to account for the CALPHAD data both for the thermodynamic driving force and kinetic data to study microstructure evolution as a function of quenching rate in gold–nickel and uranium–zirconium alloys. On the experimental side, we determined that transmission electron microscopy is critical in characterizing nuclear fuels by revealing for the first time the co-existence of several distinct phases of uranium–zirconium-2, and the existence of a new phase in molybdenum–uranium alloys in the uranium-rich region, where only the body-centered cubic phase is supposed to exist. Finally, the creation of material defects at the LLNL Center for Accelerator Mass Spectrometry by implanting iron ions whose energy was similar to that of a fission fragment emitted by a nuclear power plant in samples of uranium–zirconium alloys, has shown that a loss of the material's well-aligned plate-like microstructure was indicative of a loss of dimensional stability that could impact fuel's performance (and in particular swelling). Our research has clearly demonstrated the relevance of our unique capabilities in terms of modeling and experiments, especially our irradiation facility for actinide alloys. This work has contributed to a project for the Nuclear Energy Agency in the Organisation for Economic Co-operation and Development headquartered in France, for development of an international database for the thermodynamics of advanced fuel. We are also exploring a collaboration with the Korea Atomic Energy Research Institute in South Korea, which expressed an interest in our approach to the basic science of ultrahigh-burn-up advanced nuclear fuels, especially the inert-matrix fuel concept originally proposed by the Bochvar Institute in Russia, for next-generation nuclear reactors.

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Forward Path to Discovery at the Large Hadron Collider

Douglas Wright (12-ERD-051)

Abstract

After nearly 20 years of construction, the Large Hadron Collider (LHC) near Geneva, Switzerland, is poised to explore the high-energy frontier of particle physics with the world's largest and highest-energy particle accelerator. The possibility exists, however, that the lightest new particles in the framework of today's proposed physics models will be too heavy to be discovered at LHC. Lawrence Livermore is currently part of a detector upgrade project that would substantially enhance the physics reach of the Compact Muon Solenoid experiment at LHC to eliminate physics blind spots. The upgrade will detect the anomalous production of W boson pairs (subatomic particles with a positive and negative electric charge that are each other's antiparticle) through their decay to muons (unstable subatomic particles with negative charge). This provides a model-independent means of observing the presence of new subatomic particles. We propose to adapt a feedback-stabilized, ultraprecise timing system, originally developed for the Advanced Light Source at Lawrence Berkeley National Laboratory, to synchronize LHC forward-proton detectors required for the diagnostic upgrade.

We expect to enable the search for new fundamental subatomic particles through development of an advanced timing system that will be integral to a diagnostic upgrade to LHC. The key goal of the upgrade is to detect collisions in which beam protons remain intact and new physics signals are produced by collision of photons produced from the proton beams. Detecting the intact, outgoing protons requires small tracking detectors placed inside the LHC beam pipes located a few hundred meters from the interaction point. Our staged plan starts with a basic feasibility demonstration and proceeds in two steps to a fully capable LHC system proven with actual timing detectors in a test beam at the Fermi National Accelerator Laboratory near Chicago.

Mission Relevance

This project applies LLNL science and engineering capabilities to address the highest-priority mission of the particle physics program in the DOE Office of Science. A highly visible and unique role in both hardware and physics analysis at LHC will allow Lawrence Livermore to continue to attract and retain outstanding scientists, who ultimately make substantial contributions to national security programs. This frontier research project also supports Livermore's core competency in nuclear science and technology by developing diagnostics able to view complex, energetic nuclear dynamic processes.

FY14 Accomplishments and Results

In FY14 we led the analysis of a full data set of exclusive production of W boson pairs using muons. In addition, we completed the design of our reference clock system incorporating features of LHC beam bunch signals. The clock system was approved by the Compact Muon Solenoid collaboration for experimental use on LHC, and a Livermore researcher was appointed to the project steering committee for timing and trigger systems.

Project Summary

This LDRD project established LLNL leadership in high-energy physics on three fronts. First, it enabled LLNL physicists to develop a leading role in the analysis of a unique physics process, the central exclusive production of W boson pairs. The Laboratory joined an initial analysis of the Compact Muon Solenoid experiment data and then significantly expanded the analysis to include additional detection modes and developed a new technique for reducing systematic error. This analysis approach provides a unique means of accessing new physics signatures in a energy regime that is inaccessible to most other analyses. Second, in collaboration with the SLAC National Accelerator Laboratory at Stanford, LLNL developed and tested an innovative picosecond fast-timing technology that was adopted as a key component of a detector upgrade proposal to further extend the physics reach of the Compact Muon Solenoid experiment. Third, LLNL scientific leadership in the success of the proposed detector upgrade was demonstrably recognized by the physics community. Our accomplishments led to an invitation to present a proposal for support from DOE to continue participation in the data analysis and detector upgrade for the Compact Muon Solenoid at LHC.

Publications and Presentations

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Physics Beyond Feynman

Peter Beiersdorfer (12-LW-026)

Abstract

We intend to determine whether the theory of quantum electrodynamics (QED) is complete, its incompleteness having been suggested for the first time in an experiment measuring differences in energy levels in hydrogen published in *Nature* in the latter half of 2010. Of profound importance to all of physics and to our very understanding of the universe, QED is the quantum theory of the interactions of charged elementary particles with an electromagnetic field. The reported experiment, which measured the charge radius of the proton, raises the question of whether QED theory is incomplete at the level of two-loop Feynman diagrams, which are representations of the mathematical expressions governing the behavior of subatomic particles. To answer this important question, we will determine the QED two-loop term with an accuracy that is ten times greater than what has been achieved previously. These measurements will be carried out at the Livermore SuperEBIT electron-beam ion trap facility, which will produce the

necessary lead ions. Very-high-resolution grating spectrometers will be installed at this facility for observational and calibration purposes.

With the success of this project, we will demonstrate the first-ever test of two-loop QED in a bound atomic system not encumbered by the finite size of the nucleus. In the case of atomic hydrogen, this will be the proton. If we determine that QED predictions differ from measurements on the two-loop level of Feynman diagrams, our results will mean that QED theory will need to be revised. Our results could also imply that interpretation of recent measurements of the proton radius is flawed, likely requiring changes in the Standard Model of particle physics concerning nuclear interactions.

Mission Relevance

This project may profoundly alter our understanding of the fundamental forces of physics, relevant to the Laboratory's core competency in nuclear science and technology. It may thus affect the underpinnings of the physics of many Livermore missions and scientific thrusts, including atomic physics in stockpile stewardship, high-energy chemical compounds with national security relevance, energy manipulation, and materials on demand.

FY14 Accomplishments and Results

As proposed, we operated the SuperEBIT device for about three months in FY14 to collect statistically sufficient data on the element lead, and test for systematic errors. Problems with the device, which has been aging without new infrastructure investments, forced a delay of operations until the end of FY14, and the experimental portion of the project terminated on September 30. This meant that data analysis could not be completed before the end of the fiscal year.

Project Summary

The project successfully utilized the Livermore SuperEBIT facility, which was modified specifically for this project, to produce copious amounts of lead-89+, and the resulting radiation was successfully recorded with two very-high-resolution grating spectrometers and the EBIT calorimeter spectrometer. Extensive data collection periods were necessary to create a sufficient accumulation of the relevant photons for obtaining a statistically significant result. Initial analysis of the data revealed a contamination of the sought-after lead-89+ spectral line by lines from lower charge states of lead that were produced during the calibration phase of the experiment. A change in the calibration procedure ensured that subsequent measurements were free of such contamination. A set of statistically meaningful data is now available for analysis and eventual publication. We intend to submit a proposal to the DOE Office of Basic Energy Sciences to obtain support for additional research efforts to test QED and the Standard Model of particle physics.

Publications and Presentations

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Neutron Star Science with the Nuclear Spectroscopic Telescope Array

Julia Vogel (13-ERD-033)

Abstract

The Nuclear Spectroscopic Telescope Array (NuSTAR), launched in June 2012, will help scientists obtain for the first time a sensitive high-energy x-ray map of the sky with extraordinary resolution. This pioneering telescope will aid in the understanding of how stars explode, producing elements like calcium that end up in bone and teeth. Lawrence Livermore is a founding member of the NuSTAR project, with key personnel on its optics team. We propose that LLNL also assume a leadership role in the science performed with NuSTAR with analysis of observations of the different neutron star classes identified in the last decade that are still a mystery to astrophysicists. These studies will not only help understand newly discovered astrophysical phenomena or emission processes for members of the neutron star family, but will also expand the utility of neutron star observations for addressing broader questions in astrophysics and physics. For example, neutron stars provide an excellent laboratory to study exotic and extreme physical phenomena, such as the equation of state of the densest matter known, the behavior of matter in extreme magnetic fields, and the effects of general relativity. At the same time, knowing their accurate populations has profound implications for understanding the life cycle of massive stars, star collapse, and overall galactic evolution.

We expect to identify new behavior of neutron stars, proving or refuting model predictions and advancing our understanding of these complex objects. We will provide proper interpretation of NuSTAR observations, which requires appropriate models of telescope and detector performance and knowledge of how NuSTAR instruments modulate the intrinsic x-ray emission. Our research will be of great interest to the astrophysics community. The successful execution of this project will provide LLNL with a leading presence in x-ray astronomy and astrophysics, facilitate Laboratory participation in next-generation experiments, help develop and retain a workforce of highly qualified scientists and engineers, and position LLNL for future astrophysics and x-ray optics projects.

Mission Relevance

Participation in space science is an important element in LLNL's cyber and space security and intelligence strategic focus area. Providing next-generation capabilities for security of space requires a trained workforce with demonstrated excellence in

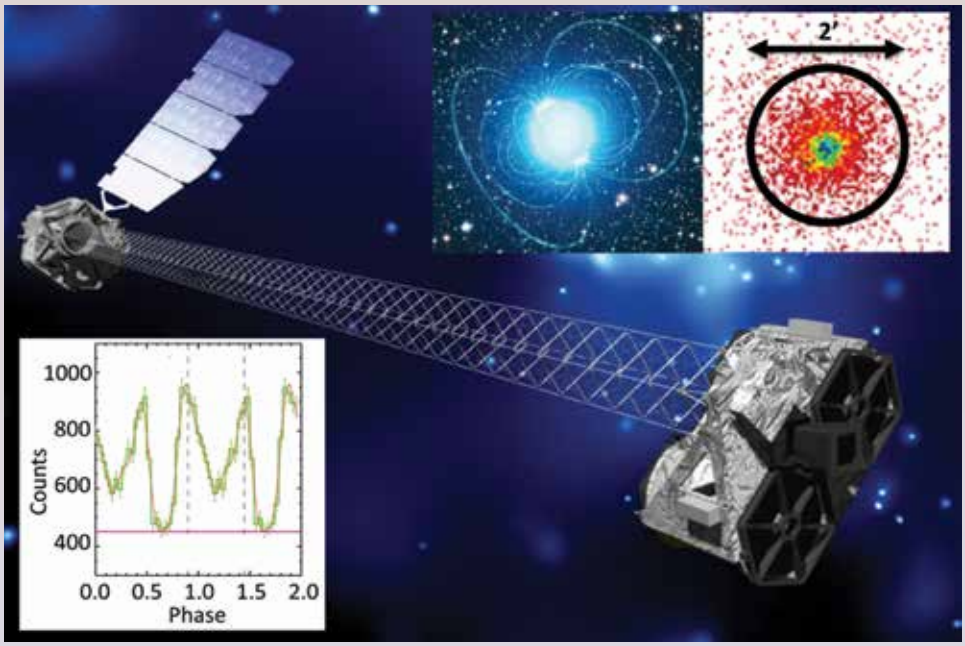
conceiving, fabricating, and performing science with advanced instrumentation. Our analysis of NuSTAR observations of neutron stars will strengthen the Laboratory's preeminence in x-ray astronomy as well as Livermore's core competency in high-energy-density science and nuclear science and technology.

FY14 Accomplishments and Results

In FY14 we (1) refined the NuSTAR optics models and focused on analysis and scientific interpretation of x-ray data for neutron stars in light of current and emerging models, (2) developed and refined algorithms for neutron star data analysis and applied those algorithms to NuSTAR data from magnetar observations (neutron stars with very high magnetic fields), and (3) significantly contributed to or led the data analysis and publication of results for magnetars 1E 2259+586 (see figure) and 1E 1841-045, the recently detected magnetar SGR J1745-2900 near the Galactic center, and the Geminga neutron star.

Proposed Work for FY15

For FY15 we propose to (1) complete the ongoing data analysis and interpretation of our results for the different classes of neutron stars currently in progress; (2) compare current and new neutron star models against our observations, including a proposed coronal electron–positron outflow model; (3) prepare for upcoming neutron star observations with NuSTAR by continuing to extend and refine our existing algorithms, and (4) analyze data once observations have taken place.



The NuSTAR observatory consists of two hard-x-ray telescopes separated from the focal plane detectors by a 10-m-long mast. Artist depiction of a magnetar indicating the magnetic field lines of the rapidly spinning neutron star and NuSTAR data acquired for magnetar 1E 2259+586 in the high-energy x-ray range of 4 to 79 keV is shown at top. Pulsations observed from the same magnetar using NuSTAR data is shown at bottom.

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Radiochemical Measurements of Nuclear Reactions at the National Ignition Facility

Dawn Shaughnessy (13-ERD-036)

Abstract

Our objective is to develop a capability for measuring neutron activation cross sections in a plasma environment. Recent results using radiochemistry of solid debris have provided the first strong evidence that an adjustable quantity of kiloelectronvolt neutrons are consistently produced at Livermore's National Ignition Facility in high-compression deuterium–tritium capsules. They have been observed through radioactive gold isotopes produced during neutron interactions in the hohlraum cylinder that houses the target capsule. Kiloelectronvolt-neutron reactions have been modeled with uncertainties up to 300%, and experiments at the National Ignition Facilty offer a unique opportunity to measure these quantities. This project builds on the existing solid-debris radiochemistry diagnostic at the facility. Metal atoms will be added to the hohlraum or incorporated into the ablator, which provides thermal protection to the capsule surface. The material will undergo neutron capture and activation, and the reaction products will be collected for analysis using our solid-debris radiochemistry diagnostic. Cross sections will be determined using gamma spectroscopy of the final samples.

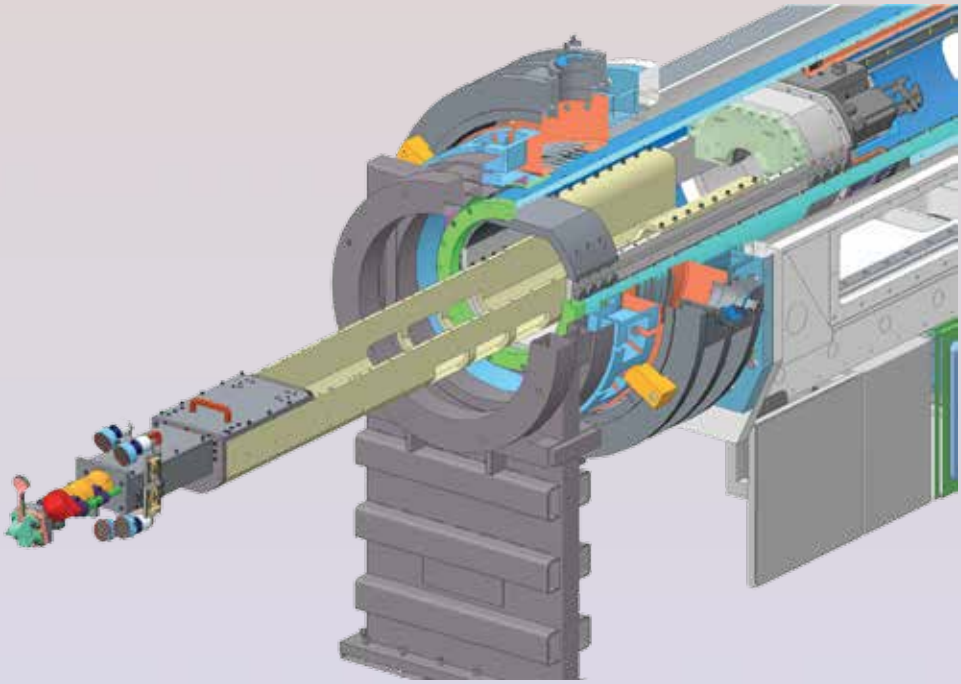
We have three primary objectives: (1) optimize the solid radiochemistry collectors, (2) incorporate selected material into the target capsule assembly and hohlraum, and (3) measure neutron capture cross sections for isotopes of yttrium, uranium, and thulium. At the conclusion of this project, we expect to have an improved solid-debris radiochemistry diagnostic that can be used to measure mission-relevant neutron cross sections and nuclear data. We also expect to be able to incorporate the selected material into both the hohlraum and target capsule for future measurements of excited-state cross sections on radioactive materials.

Mission Relevance

This project is closely aligned with Laboratory missions in stockpile stewardship and post-detonation nuclear forensics for threat reduction. In addition, research into materials to enhance the yield of laser fusion ignition capsules aligns well with the strategic focus area in inertial-confinement fusion science and technology. Finally, this work helps to promote growth in several Laboratory core competencies, most notably nuclear science and technology.

FY14 Accomplishments and Results

Accomplishments for FY14 included (1) completing the design of new solid radiochemistry collectors for the diagnostic instrument manipulator DIM 90-315 (shown in figure), providing a second line of sight in the chamber to measure spatial distribution of debris from National Ignition Facility experiments; (2) fielded 4 solid radiochemistry collectors on DIM 90-315, bringing the total up to 12; (3) tested these collectors on several shots, with data indicating that collection is higher on this diagnostic instrument manipulator than with the other two manipulators; (4) completed a design



Our newly designed solid radiochemistry collector is positioned at the front of the nose cone of the diagnostic instrument manipulator at the National Ignition Facility. Two collectors are fielded on the top and two collectors are at the bottom of the nose cone. The collectors will provide a capability for measuring neutron activation cross sections and nuclear data in a plasma environment with radiochemistry of solid debris.

for adding materials to the outside of the hohlraum in 0.5- or 1-mm-thick pieces; (5) constructed a hohlraum that has 3 rare earth materials on the surface of the hohlraum for an upcoming shot—the first shot will have 0.5-mm foils of neodymium and thulium on the outside of the hohlraum and a second target request has been made for 0.9-mm curved foils of neodymium, gadolinium, and thulium; and (6) completed the initial design for a large-area collector for the diagnostic instrument manipulator, which will be available for a March 2015 shot.

Proposed Work for FY15

In FY15 we will (1) conduct a shot with an indirect-drive exploding pusher with thulium, neodymium, and gadolinium on the outside, which will allow us to measure neutron capture cross sections at 14 MeV; (2) complete at least one additional shot with materials added to the hohlraum to understand how debris is distributed in the chamber as a function of angle; (3) complete design of the new, larger collector; and (4) add uranium-238 to the inside of the shell of a plastic symmetry capsule (i.e., a surrogate fusion target). This capsule will be used in a deuterium–tritium test shot to determine if adding materials inside the shell affects the yield performance of the capsule.

Publications and Presentations

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Complex Electronic Structure of Rare Earth Activators in Scintillators

Per Daniel Aberg (13-ERD-038)

Abstract

Interest in scintillator materials that fluoresce because of interaction with a charged particle or photon has surged recently thanks to large-scale applications in nuclear and radiological surveillance, high-energy physics, and medical imaging. One of the

current goals is to develop materials with improved energy resolution to detect fissile materials at ports, borders, and airports with a low probability of error. We propose to study the electronic energy levels of rare earth activators in these materials using theoretical modeling and experimental validation to provide information on key microscopic events such as the hole and electron-capture cross section by activators, for which there is currently no reliable method. We will implement a novel algorithm for computation of the rare earth electronic structure, in relation to crystal host bands, via atomic physics and dynamical mean field theory, and will validate the results using x-ray absorption spectroscopy and x-ray emission spectroscopy measurements.

We expect to predict the energetic position of the 4f and 5d electron orbital levels of rare earth dopants in relation to the crystal host valence and conduction bands. Thus, we can model and predict the hole and electron-capture cross-sections, which are crucial elements in understanding the entire scintillation event from the initial cascade to final emission of a detectable photon. In particular, the capture cross-sections determine how far out, from the initially dense track of electron–hole pairs, the electrons can be pushed before trapping occurs. The x-ray absorption spectroscopy and x-ray emission spectroscopy measurements will serve as direct validation of the energy levels for our theoretical effort by providing experimental data currently not available in the literature.

Mission Relevance

The ultimate goal of this project is to develop unique computational and experimental capabilities to address limiting factors in scintillator radiation detector functionality. As such, it is well aligned with the Laboratory's mission in nonproliferation. Furthermore, it supports the DOE's newly established Energy Innovation Hub and the search for rare earth element substitutes. We expect that the framework developed here will also enable incorporation of strong electron correlation effects in the equation-of-state models for actinides, which is highly relevant to stockpile stewardship science and LLNL's core competencies in nuclear science and technology and computational science and engineering.

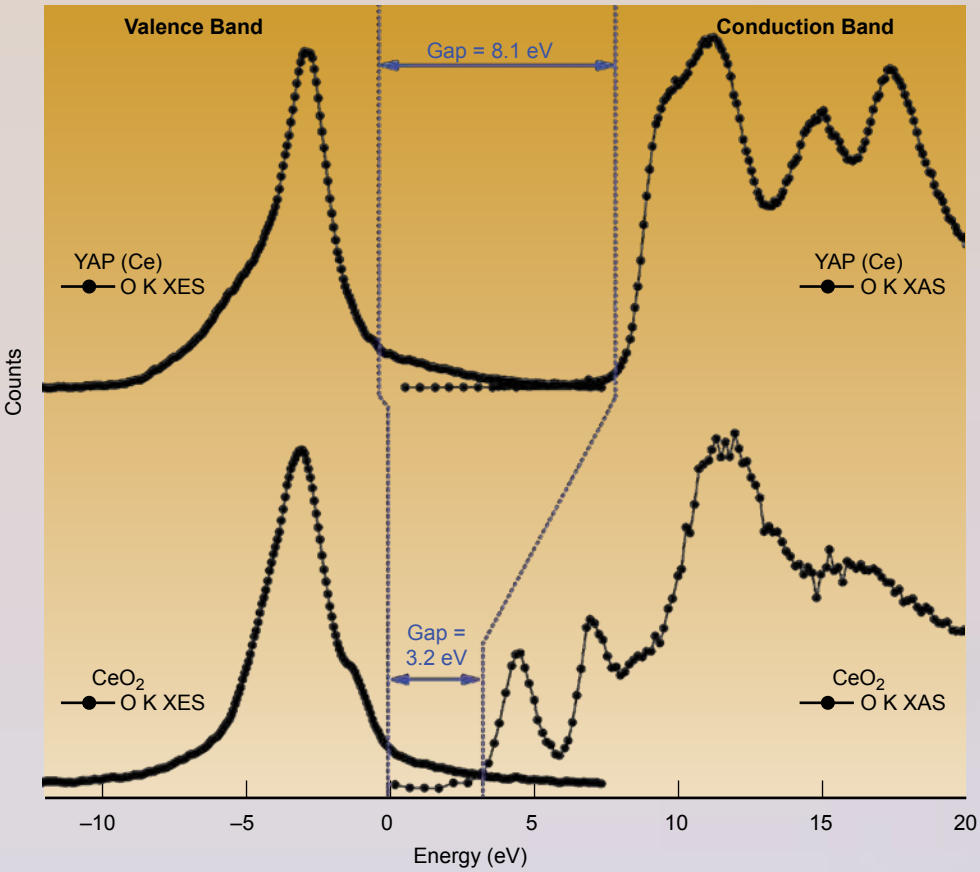
FY14 Accomplishments and Results

In FY14 we (1) completed our x-ray absorption and x-ray emission spectroscopic study of the f-orbital-levels of cesium in cesium-doped yttrium aluminum oxide; (2) began characterization of cesium-doped lanthanum bromide and strontium iodine; (3) began working on a new approach for the construction of a random-phase-approximation dielectric function—the projector augmented-wave method used in the Vienna Ab Initio Simulation Package is not sufficiently accurate, and we have therefore switched to a mixed-basis approach with initial tests showing great promise, but memory intensive for realistic systems; (4) completed re-parallelization of the GW code to calculate the energy in a many-body system of electrons, using the Parallel Basic Linear Algebra Subprograms and Scalable Linear Algebra Package libraries; and (5) devised a scheme to calculate

excited 4f and 5d electron states in a modification of the density functional theory for investigating the electronic structure of many-electron systems by introducing an f-electron chemical potential.

Proposed Work for FY15

In FY15 we will (1) complete the initial x-ray absorption and x-ray emission spectroscopic studies on rhenium-doped lanthanum bromide and strontium iodine for varying dopant concentrations, (2) finish the work on effective Hamiltonian mechanics of dynamic systems derived from random phase approximation to be able to calculate 4f and 5d energy levels with respect to the host band edges, and (3) investigate the possibility to directly calculate the screened Coulomb electronic potential matrix elements via linear response theory, and thus avoid constructing the dielectric matrix using random phase approximation.



For the first time, the band gap of a cesium-doped yttrium aluminum perovskite scintillator, YAP(Ce), has been determined by means of oxygen K-edge x-ray absorption and emission spectra from both yttrium aluminum perovskite and a reference sample. Because the soft x-ray spectroscopic method does not depend on the surface charging of the scintillator material, it is a very powerful tool to investigate the complex electronic structures of scintillator materials.

Hard X-Ray Mirrors for Nuclear Security

Marie-Anne Descalle (13-ERD-048)

Abstract

The nation continues to search for technologies and techniques to address long-standing problems and anticipate emerging challenges in nonproliferation, including the ability to search for special nuclear materials and weapons, perform attribution in the event of an attack or interdiction of a device, and strengthen the safeguards required for treaty obligations. The ability to detect hard x-ray or soft gamma-ray emissions (photons ranging to several hundred kiloelectronvolts) from special nuclear materials plays an important role in a number of these national security applications. The use of hard x-ray optics offers one path to dramatically improve performance by increasing photons collected from weak sources or by filtering out a background signal that obscures an important spectral signature. We propose to study, for the first time, photon and multilayer interactions in the hard x-ray band. We also will produce robust, inexpensive, and high-accuracy mirror substrates for hard x-ray multilayer mirrors and optics. Our efforts will include the design, execution, and analysis of an experiment at the European Synchrotron Radiation Facility in Grenoble, France, the only x-ray light source in the world where a sufficiently bright, low-divergence beam is available for basic investigations at photon energies as high as 400 keV.

We expect to develop novel hard x-ray mirror technologies and instrument concepts to detect and measure hard x-ray and soft gamma-ray emissions (photon energies between 100 and 500 keV). We will improve our understanding of photon and multilayer interactions, develop robust substrates for the optics, and develop instrument designs suitable for national security missions. A successful conclusion to this project will enable us to deploy instruments for a variety of applications of interest to various national security missions.

Mission Relevance

This project is closely aligned with the Laboratory's mission in nuclear nonproliferation. The design, development, and characterization of novel instrumentation that uses reflective hard x-ray mirrors will expand Lawrence Livermore's capabilities to contribute to national security through detection of special nuclear materials, which also supports the core competency in nuclear science and engineering.

FY14 Accomplishments and Results

During FY14 we (1) completed analysis of the 508- and 644-keV data obtained at the European Synchrotron Radiation Facility, (2) completed the move of the complex optics assembly machine on loan from Columbia University to LLNL, (3) benchmarked our code to predict the performance of optimized coating for various applications, and (4) participated in an experiment at the Livermore Jupiter Facility that resulted in the first successful demonstration of the technique in the field of high-energy-density plasmas, further expanding the applications for hard x-ray optics.

Proposed Work for FY15

In FY15 we will (1) complete the substrate research with a focus on glass slumping with electrostatic forces, and investigate the range of applicability of the process with precise metrology; (2) expand our investigation of applications spanning basic sciences to nuclear security in new energy ranges and; (3) design, build, and test at least one nested optics, taking advantage of the NuSTAR (nuclear spectroscopic telescope array) optics assembly machine at Caltech in southern California.

Publications and Presentations

Brejnholt, N. F., et al., 2013. *Challenges in characterizing multilayer coatings for use in soft gamma-ray detection and filtering*. 2013 IEEE NSS/MIC/RTSD—Nuclear Science Symp. and Medical Imaging Conf. and Room-Temperature Semiconductor X-Ray and Gamma-Ray Detectors Workshop, Seoul, Korea, Oct. 27–Nov. 2, 2013. LLNL- ABS-637594.

Brejnholt, N. F., et al., 2014. "Demonstration of multilayer reflective optics at photon energies above 0.6 MeV." *Optic. Express* **22**, 15364. LLNL-JRNL-652505. *http://dx.doi.org/10.1364/OE.22.015364*

Brejnholt, N. F., et al., 2014. *Next generation hard x-ray/soft gamma-ray optic design and implementation*. SPIE Astronomical Telescopes and Instrumentation, Montreal, Canada, June 22–27, 2014. LLNL-PROC-655710.

Descalle, M. A., et al., 2013. *Gamma-ray multilayer mirrors for homeland security applications*. 2013 IEEE NSS/MIC/RTSD—Nuclear Science Symp. and Medical Imaging Conf. and Room-Temperature Semiconductor X-Ray and Gamma-Ray Detectors Workshop, Seoul, Korea, Oct. 27–Nov. 2, 2013. LLNL-ABS-643484.

Pivovaroff, M. J., 2013. *High reflectivity multilayers for hard x-ray and soft gamma-ray applications*. LLNL-PRES-643015.

Why Is Nuclear Matter So Red?

Darren Bleuel (13-LW-003)

Abstract

We propose to investigate a dramatic enhancement in the ability of nuclei to absorb and emit photons in the low-energy red part of the spectrum, which is not predicted by nuclear theory. This enhancement was first seen in two Livermore-led experiments in 2000 and unambiguously established a decade later in another LLNL experiment with a model-independent method. We intend to lead an international effort to understand this new property of nuclear matter and provide insight into one of its most fundamental properties—the ability to emit and absorb photons. Our measurements of the systematic behavior of low-energy enhancement in the radiative strength function will

provide the data needed to develop a theoretical model for this enhancement and continue Lawrence Livermore's leading role in understanding this new phenomenon.

A low-energy enhancement of the radiative strength of nuclei has significant impact on nuclear reaction rates in high-energy-density plasma environments, including astrophysical phenomena, inertial-confined plasmas, and the interiors of nuclear devices, because of potential changes in the decay probabilities of an excited nucleus in a nuclear reaction. This, in turn, could cause significant changes in the production of nuclei used to develop stellar structure models, the interpretation of archival stockpile radiochemical data, and the evaluation of nuclear forensics data. We expect the successful conclusion of this project will enable determination of low-energy enhancement in the radiative strength function such that theorists can determine its microscopic origin and role in stellar nucleosynthesis scenarios. This information will help to inform potential neutron-capture experiments in photon-rich plasma environments, including the National Ignition Facility at Livermore, which may offer a more realistic understanding of the astrophysical setting, such as stellar interiors and supernovae.

Mission Relevance

Determining the nature of low-energy enhancement in the radiative strength function will significantly improve our understanding of the properties of nuclear matter and nucleon–nucleon interactions. Important applications include weapon aging, inertial-confinement fusion, and neutron-capture experiments in support of Laboratory missions in stockpile stewardship science and energy security, as well as the core competency in nuclear science and technology and high-energy-density matter.

FY14 Accomplishments and Results

In FY14 we (1) fabricated plutonium-239 and plutonium-242 targets, which were used for experiments at the University of Oslo to measure level densities and radiative strength functions in plutonium-240 and plutonium-243; (2) performed an osmium-192 experiment at the University of Oslo to determine the level density and radiative strength function of osmium-193; (3) began fabrication of plutonium-240, curium-248, protactinium-231, neptunium-236, and plutonium-244 targets to measure level densities and radiative strength functions over a large number of actinides; and (4) recruited a graduate student who analyzed the lanthanum-139 data set from the University of Oslo to determine level density and radiative strength function of lanthanum-140.

Proposed Work for FY15

In FY15 we propose to (1) begin a series of experiments at University of Oslo using the actinide targets of plutonium, curium, protactinium, and neptunium we fabricated to determine level densities and radiative strength over a large number of actinides; (2) complete analysis of the lanthanum-139 data set; and (3) perform and analyze the first experiment for mapping enhancements in the low-energy part of the radiative strength function of nuclear matter at the iThemba Laboratory for Accelerator Based Sciences in South Africa, using equipment we provided, with data analysis provided by local university students.

Publications and Presentations

Guttormsen, M., et al., 2013. "Constant-temperature level densities in the quasicontinuum of Th and U isotopes." *Phys. Rev. C* **88**, 024307. LLNL-JRNL-645275.

Guttormsen, M., et al., 2013. "Observation of large orbital scissors strength in actinides." *Acta Phys. Pol. B* **44**, 567. LLNL-JRNL-645274.

Guttormsen, M., et al., 2013. "Observation of large scissors resonance strength in actinides." *Phys. Rev. Lett.* **109**, 162503. LLNL-JRNL-582232.

Guttormsen, M., et al., 2014. "Scissors resonance in the quasicontinuum of Th, Pa, and U isotopes." *Phys. Rev. C* **89**, 014302. LLNL-JRNL-664178.

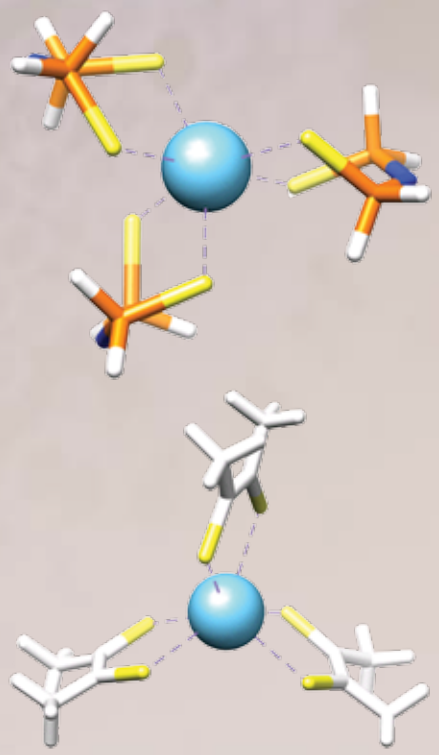
Search for Lanthanide Covalency for Enhanced Rare Earth Separations

Edmond Lau (13-LW-048)

Abstract

Rare earth elements are used in electronic displays, high-efficiency lighting, and high-performance magnets for hybrid vehicles, wind turbines, and hard drives. They are key enablers for a clean-energy economy. China now produces at least 95% of rare earth elements, but at a huge environmental cost. There is a pressing need to develop new ways to efficiently separate rare earths to minimize waste products and the environmental impact of rare earth production. We propose to develop new concepts in lanthanide bonding that will ultimately lead to novel, species-specific ligands for efficient separations. Our project is a combined experimental and theoretical effort to look for covalent interactions of lanthanides with main-group elements. We have identified a set of candidate complexes involving lanthanides of cerium, gadolinium, and lutetium with ligands containing nitrogen, sulfur, selenium, tellurium, and phosphorus. From this set, ab initio electronic structure methods will be employed to identify specific lanthanide complexes with a high degree of covalency for further experimental synthesis and characterization studies.

We expect to unambiguously establish the presence of covalent interactions between lanthanides and ligands involving main-group elements. Covalency effects are a promising new property to target in future development of chemical techniques for efficient intra-lanthanide separations, which will ultimately mitigate the environmental damage associated with rare earth production. We will compare the character of the lanthanide–ligand covalent bond in the large, medium, and small lathanides cerium, gadolinium, and lutetium, respectively. In addition, we will look for trends that can ultimately be exploited in development of species-specific ligands for separations.



Coordination geometries of lanthanum to imidodiphosphinochalcogenide (top) and dithiolene (bottom).

Mission Relevance

This project is closely aligned with the Laboratory's mission in energy security and sustainable environmental quality. Currently, separation and processing of rare earth elements carries a high environmental cost, which has limited U.S. mining efforts and led to Chinese dominance of rare earth resources. We aim to directly tackle this critical national priority by informing efforts to improve the safety and reliability of rare earth separation. In addition, our combined experimental and theory strategy for lanthanide chemistry is directly transferable to actinide efforts and the nuclear science and technology core competency at Lawrence Livermore.

FY14 Accomplishments and Results

We endeavored to synthesize ligands that have been previously reported that were sterically crowded enough to support only monometallic lanthanide complexes so that metal–ligand multiple bonds may be possible. Unfortunately, lanthanide salts are very insoluble, and we were not able to isolate the lanthanide complexes to conduct further reactivity. Density functional calculations of lanthanides coordinated by imidodiphosphinochalcogenide and dithiolene and diselenene were completed. These calculations provided some evidence that dithiolene and diselenene ligands may enable some covalency when coordinated to some of the lighter lanthanides.

Project Summary

At the conclusion of the project, creation of the sterically crowded ligands for lanthanide complexes proved to be difficult. The lanthanide salts are very insoluble and we were not able to isolate the lanthanide complexes to conduct further reactivity. Although creation of lanthanide complexes did not work out, the synthetic group was able to produce metal–ligand double bonds between uranium and nitrogen as well as bonds between uranium and the softer metal group 16 elements such as sulfur and selenium, both of which were components of the original proposal. Additionally, computational studies of imidodiphosphinochalcogenide and dithiolene and diselenene were completed. Charges and spin densities indicate that dithiolene and diselenene donate electron density to the metal ions. Follow-on support of the project might be obtained from the DOE Office of Basic Energy Science for the discovery of novel ligands for separating rare earth metals.

Publications and Presentations

Lau, E. Y., 2014. *Labwide LDRD—Final report*. LLNL-TR-663578.

Electromagnetic Manipulation of Nuclear Decay

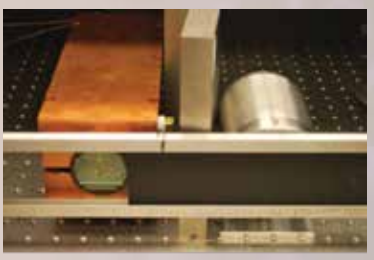
Robert Casperson (13-LW-065)

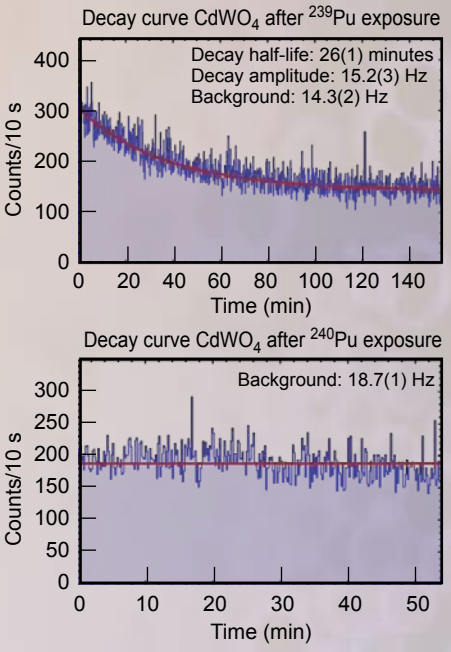
Abstract

Significant alteration of nuclear decay properties would have important consequences, ranging from novel approaches to nuclear batteries and gamma-ray lasers, to physically

interesting experiments. Quantum systems that decay by photon emission couple to the electromagnetic modes of the local environment, and by modifying these modes, one can manipulate the rate of spontaneous emission. Our main goal with this project is to observe a significant modification of the decay rate of the 26-minute, 77-eV, uranium-235 isomer by modifying the electromagnetic modes available to the isomer. To generate the uranium-235, we will use a hot atom technique to deposit nuclear recoils from plutonium-239 into a catcher material such as molybdenum or silicon. We plan to conduct two experiments. The first will use a bulk dielectric material to scale the electromagnetic modes near the nucleus and alter the decay rate. This will be accomplished with two extremely flat surfaces placed together to simulate a solid material, which allows the creation of bulk dielectrics and multilayer dielectric micro-cavities with embedded uranium-235, without disrupting the physical environment of the decaying isomer. The second experiment will use nanometer-scale multilayer mirrors to create a dielectric micro-cavity structure, which can be geometrically tuned to a range of decay rates. Both of these experiments require state-of-the-art thin-film coatings and will leverage LLNL capabilities in multilayer science.

We expect to modify the decay rate of the uranium-235 isomer by up to 60%, extending the 26-minute half-life of the isomer to over an hour. This will represent a modification over ten times larger than previous experiments attempting to alter nuclear decay rates. A successful measurement will open the door to further experiments including overall decay rate modification, as well as angular-dependent adjustments. The overall decay rate modification will be relevant to nuclear battery applications and involve suppression of the decay of long-lived isomers with low-energy transitions. The angular-dependent decay rate is applicable to advancements in the energy of gamma-ray lasers.





A photo of the vacuum system used to perform direct measurements by counting photons (top). The reflective circle on the left of the chamber is the front face of a photomultiplier tube, which was surrounded by a large, chilled, copper block. The two vertical plates are tungsten, and the aluminum cylinder on the right contains the plutonium-239 source. The long flat object in the foreground is a manual translation arm. Light production in the cadmium tungstate ($CdWO_4$) crystal after uranium recoil implantation from plutonium-239 decay shows a decay half-life of 26 minutes for the embedded isomer, which matches the vacuum decay half-life (middle). Light production in the cadmium tungstate crystal after uranium recoil implantation from plutonium-240 decay shows a flat background (bottom).

Mission Relevance

Basic research in the physics of nuclear decay provides new understandings in the fundamentals of this important process, which has impacts on many Laboratory missions, from high-energy lasers to potential energy sources, and is supportive of the core competency in nuclear science and technology.

FY14 Accomplishments and Results

In FY14 we (1) constructed molybdenum and silicon multilayer mirrors tuned to the specific energy of the uranium-235 isomer, (2) observed no significant half-life modification after pressing the mirrors together and measuring the uranium-235 isomer decay rate, (3) observed no effect after making additional measurements of bulk effects using condensed water and electron detection, (4) measured and identified scintillation light resulting from the uranium-235 isomer after embedding the isomer in various scintillators, and (5) demonstrated that the uranium-235 isomer in cadmium tungstate has a 26-minute half-life, even when embedded deep within the dense material.

Project Summary

The successful conclusion of this project showed that the uranium-235 isomer half-life is not substantially modified by embedding the isomer in a dense material, indicating

that the theory of modified spontaneous emission for atomic systems does not directly apply to the internal conversion of the isomer. Two vacuum systems were constructed for performing measurements of the isomer: the first used a microchannel plate for directly detecting internal conversion electrons, and the second utilized a chilled photomultiplier tube for detecting scintillation light from electron interactions in scintillator crystals (see figure). The electron detection measurements showed that sealing and unsealing the uranium-235 isomer in a bulk material results in apparent isomer population gains and losses, which are unrelated to an altered decay half-life and call into question past measurements where the uranium-235 isomer was embedded in silver. The photon detection measurements using cadmium tungstate resulted in the first measurement and identification of scintillation light from the uranium-235 isomer decay. The decay half-life of this isomer in cadmium tungstate was shown to be 26 minutes, which matches the vacuum decay half-life. The project demonstrated that the uranium-235 isomer half-life is not substantially modified by embedding the isomer in a dense material, which answers the primary motivating question for this project. As such, additional research will not be pursued on this topic.

Publications and Presentations

Casperson, R. J., et al., 2014. *Progress in electromagnetic alteration of nuclear decay properties*. LLNL-PRES-652506.

Casperson, R. J., et al., 2014. *The search for electromagnetic alteration of U-235m decay*. LLNL-TR-663152.

Hughes, R. O., et al., 2014. *Electromagnetic alteration of nuclear decay properties*. LLNL-POST-656355.

Nuclear Fission in a Plasma

Walid Younes (14-ERD-034)

Abstract

Fission plays a critical role in the nucleosynthesis of heavy elements—however, fission-fragment properties have never been calculated for astrophysical plasmas in 75 years of fission physics. Some of the same physics will also play a role in fission at the National Ignition Facility (NIF). We propose to calculate how fission-fragment properties are modified by the tremendous plasma densities inside the crusts of neutron stars, as well as the high neutron fluxes in neutron stars and at NIF. Our theory effort focuses on plutonium-240 fission in those environments, with similar effects expected for other actinides. We will calculate the effect of both neutron-star and NIF plasmas on fission by building on existing Laboratory codes and techniques for the microscopic description of fission, and incorporating nucleus and plasma coupling.

Nuclear fission can be significantly altered inside a plasma. We do not currently know precisely what species of fragments are produced by fission inside neutron-star and supernovae plasmas, or what their energies and other properties are. We expect to provide the first-ever predictions of these quantities, shedding light on the origin of the elements heavier than iron in nature. We will also provide the interpretation needed by proposed fission experiments at NIF. If successful, the project also opens the door to further research in nucleosynthesis, atomic nuclear coupling effects at NIF, and muonic atoms, which share similar physics with the proposed work and are relevant to both fundamental science and applications to energy production in nuclear reactors.

Mission Relevance

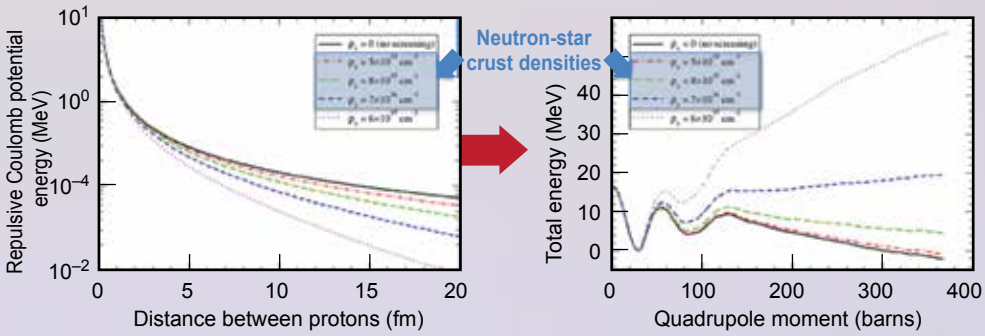
This project is of direct relevance in the Laboratory's nuclear science and technology core competency by calculating nuclear properties in plasma environments, addressing fundamental questions in the nucleosynthesis of elements heavier than iron, and shedding light on the formation of the heaviest elements in nature by studying fission—the process that limits the maximum size of those elements in the astrophysical environments where they are born. Furthermore, this project develops a critical competency for studies at the interface between nuclear and high-energy-density science that will directly benefit the analysis and interpretation of nuclear diagnostics at NIF, and more generally advance stockpile stewardship science.

FY14 Accomplishments and Results

In FY14 we (1) implemented, in our fission code, electron screening that affects the Coulomb force from interactions between electrons inside the plasma; (2) performed benchmark calculations that show significant effects from screening for fission in plasmas at neutron-star densities; (3) performed preliminary calculations showing effects from the initial-state population that could be important as we approach NIF plasma conditions; and (4) began work on optimizing the nucleon interaction and implementing the number-projection formalism, which will improve the accuracy of calculated fission-fragment properties.

Proposed Work for FY15

In FY15 we will focus on the study of the initial-state population and its effect on fragment properties for nuclei fissioning in a plasma. Specifically, we will calculate the initial states



Microscopic calculations of fission showing the weakening of the Coulomb repulsive force between protons (left plot) with increasing electron density, and the resulting barrier against fission (right plot). Electron densities relevant to neutron-star crusts are highlighted.

and their properties. In addition, we will model, for the first time, the population of those states and calculate their subsequent fission as a function of plasma conditions.

Publications and Presentations

Younes, W., 2014. *Fission theory: From the Earth to neutron stars*. Nuclear Symmetries and Stewardship Science, Berkeley, CA, May 1–2, 2014. LLNL-PRES-654254.

Younes, W., 2014. *Nuclear fission inside astrophysical plasmas*. 248th ACS National Meeting and Exposition, San Francisco, CA, Aug. 10–14, 2014. LLNL-ABS-652231.

The World's Lowest Nuclear State in Thorium-299m

Stephan Friedrich (14-LW-073)

Abstract

Thorium-229 is known to have the lowest-energy isomer of any isotope, with the first excited state of thorium-229m only 7.6 ± 0.5 eV above the ground state. Despite intensive work for 35 years and repeated false claims of discovery, no direct emission from thorium-229m has ever been observed. We propose to use our superconducting high-resolution soft x-ray detector to unambiguously detect the emission from this isotope and measure its energy and lifetime. This will allow construction of lasers at this energy to study photo–nuclear interactions and build ultraprecise nuclear clocks. Nuclear clocks have wide applications in metrology, fundamental science, and security applications, enabling measurement of postulated changes in the fine-structure constant over time to characterize the strength of electromagnetic interaction between elementary charged particles. Our superconducting x-ray detectors have a detection threshold of about 1.5 eV and an energy resolution of around 2 eV in the energy range of interest. We have also shown that background levels are below an event per minute, and thus are sufficiently low to detect emission from thorium-229m.

We intend to directly measure the decay of the thorium-229 isomer for the first time since its discovery 40 years ago. We will use a state-of-the-art superconducting tunnel junction to detect the decay of the isomer independent of its decay mode. We also expect to determine its energy with an accuracy of ±0.05 eV, which improves the current value by an order of magnitude, and to determine its lifetime (and thus its line width). Because nuclear states are much less affected by external perturbations, an accurate measurement of the thorium-229m energy and lifetime will enable the construction of ultraprecise nuclear clocks based on lasers tuned to the transition energy. These nuclear clocks, in turn, will improve measurements in quantum computing and fundamental physics.

Mission Relevance

Nuclear clocks will enable measurements of the constancy of the fine-structure constant, or more precise quantum bits for quantum computing and associated security

applications. In addition, characterizing the decay of thorium-229 experimentally will lead to a better understanding of low-energy nuclear states and their calculated lifetimes, in support of the Laboratory's core competency in nuclear science and technology, both key capabilities relevant to LLNL's national security mission.

FY14 Accomplishments and Results

In FY14 we (1) installed an optical fiber to calibrate our superconducting detector in the energy range of the expected thorium-229m signal of approximately 7.5 eV; (2) exposed our detectors to a pulsed 355-nm laser, and observed distinct peaks from the absorption of up to 10 photons per pulse (~3.5 eV per photon); (3) measured an energy resolution between 0.76 and 2 eV and demonstrated a detection threshold of about 1.5 eV and a calibration accuracy of ±0.01 eV, more than sufficient for the proposed experiments; and (4) installed a uranium-233 source and observed a peak at an energy of 6.43 eV. However, we have not been able to reproduce this peak under nominally identical conditions. Occasionally, we have also observed peaks at approximately 20, 22, and 40 eV, but we currently do not understand the origin of these peaks.

Proposed Work for FY15

We suspect that the appearance and disappearance of peaks at different energies (some tantalizingly close to the sought-after thorium-229m signal) are due to residual gases freezing out on the detector surface at 0.1 K. The resulting ice can either block the thorium-229m recoil nuclei or cause fluorescence when excited by alphas from the uranium-233 source. Therefore, in FY15 we will (1) purge the detector cryostat with clean nitrogen gas, and will subsequently test purging with argon gas; (2) replace the uranium-233 source with uranium-234 and plutonium-239 sources to understand which peaks are specific to uranium-233 and which ones are from fluorescence that can be excited by any alpha decay; and (3) install a getter pump near the detector to preferentially absorb these gases, in the event that residual gases are responsible for the irreproducible results.

Solving the Reactor Antineutrino Anomaly

Stephen Padgett (14-LW-087)

Abstract

Nuclear reactors provide the highest intensity source of man-made antineutrinos, which are produced by the beta decay of the neutron-rich fission products created in the reactor. As a result, these antineutrinos are of great interest for fundamental neutrino physics, nonproliferation monitoring, and reactor physics. However, the antineutrino signal that is observed in reactor experiments is only about 94% of the expected signal, which has attracted much attention and is referred to as the reactor antineutrino anomaly. This anomaly may indicate the existence of a fourth, sterile neutrino that could be a form of hot, dark matter. We propose to explore an explanation to the reactor antineutrino anomaly and provide the first measurement of the beta-energy spectrum for a short-lived fission

product by studying yttrium-96 beta decay. The results will experimentally test the assumptions currently being made about how the energy is shared between the beta particle and antineutrino in fission-product beta decay. This work will improve the precision of reactor flux and spectrum predictions, which is important for fundamental neutrino physics and nonproliferation monitoring of nuclear reactors.

We will develop a novel experimental approach to precisely measure the shape of the beta-energy spectra for fission products and apply it to measure the spectrum of yttrium-96, the most important isotope for the reactor antineutrino signal. The spectral shape factor will be obtained from comparison to the allowed beta-decay spectrum in lithium-8 decay. This measurement will, for the first time, test the assumed beta-energy spectrum shape commonly used in flux predictions. These results are needed to confirm or refute the existence of a fourth sterile neutrino.

Mission Relevance

This experimental work strengthens the LLNL core competencies of nuclear science and technology, specifically on the nature of neutrinos and dark matter. The possible existence of sterile neutrinos and the reactor antineutrino anomaly are of great interest to the physics community—high-profile fundamental science research such as this helps to attract top nuclear scientists to Livermore. A more reliable prediction of the antineutrino signal is also needed to augment the work on nonproliferation monitoring, which supports the Laboratory's mission in nuclear threat reduction.

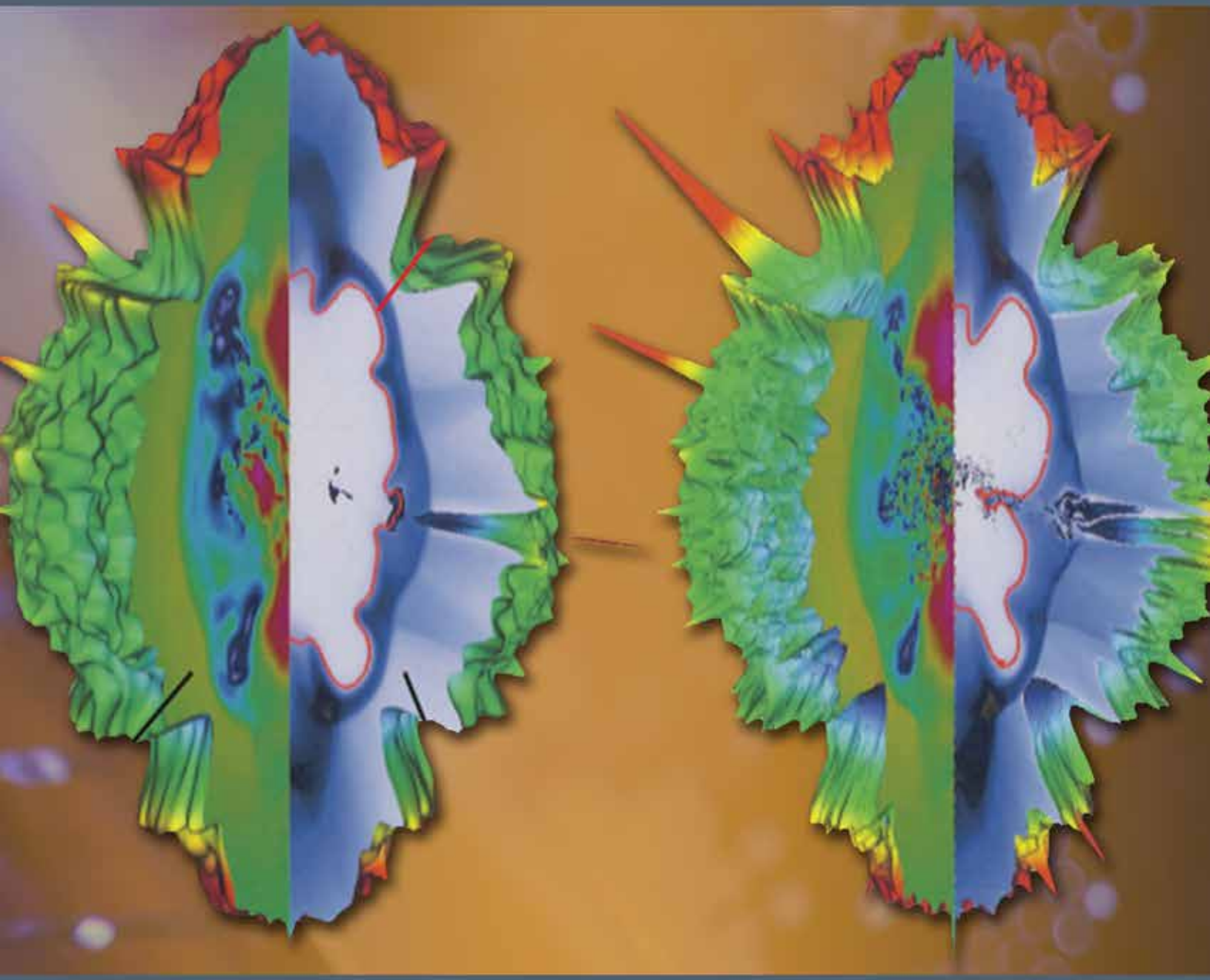
FY14 Accomplishments and Results

In FY14 we (1) designed and built the optimized plastic scintillator detectors needed for the beta spectral-shape measurements, (2) obtained laser beam time for measurements on lithium-8 decay for calibration and determined detector response characterization for beta spectral-shape measurements, and (3) analyzed data taken on antimony-134 decay with current detectors, which has similar physics to yttrium-96 and rubidium-92 decay, to establish the data analysis tools needed for future beta spectroscopy with our experimental setups.

Proposed Work for FY15

For FY15 we plan to (1) complete the lithium-8 calibration measurement and analyze the resulting data, (2) complete the detector response characterization needed for final measurement of yttrium-96 beta decay, and (3) conduct measurements on yttrium-96 decay and analyze data to determine the impact on future reactor antineutrino anomaly predictions.

Stockpile Stewardship



FY2014

Lawrence Livermore National Laboratory

The Role of Plasma Electromagnetic Fields in Anomalous Mass Diffusion: Applications to High-Energy-Density Science

Peter Amendt (11-ERD-075)

Abstract

Our objective is to conduct a comprehensive study of anomalous diffusive effects in plasmas relevant to LLNL's core missions in stockpile stewardship and inertial-confinement fusion. This work takes advantage of the Laboratory's expertise in high-energy-density science to explore a number of existing anomalies in the growing inertial-confinement fusion and high-energy-density science database that are not explained by conventional methods such as hydrodynamic mix. We propose to develop tools for including ion diffusive phenomena in our physical and computational descriptions of various laboratory phenomena. Analytical methods will be developed and particle-in-cell simulations performed to arrive at a detailed understanding of barodiffusion (diffusion of species brought about by pressure gradients) and thermal diffusion, with eventual incorporation into LLNL's suite of radiation-hydrodynamics production codes.

Most of the physical phenomena underlying the Laboratory's core missions of stockpile stewardship and inertial-confinement fusion revolve around the nature of hydrodynamic (collisional) shocks. However, the underlying medium is often a plasma with self-generated fields. Understanding the morphology of shocks in plasmas, especially low-Mach-number shocks, is a key deliverable of our proposed research. The understanding gained in this investigation will be used in tandem with the theoretical framework of barodiffusion and thermal diffusion to arrive at a description of shock-based anomalous diffusion. Adoption and eventual implementation of the resulting models in the Laboratory's weapons program codes is anticipated.

Mission Relevance

The proposed research, which we will perform in collaboration with researchers at the Massachusetts Institute of Technology, will explore the physics of low-Mach-number, collisional plasma shocks and their role in anomalous, nonclassical mass diffusion. A comprehensive understanding of non-fluid (plasma) shock behavior is central to several core missions of the Laboratory, including stockpile stewardship and the pursuit of fusion at Livermore's National Ignition Facility.

FY14 Accomplishments and Results

In FY14 we discovered a physical phenomenon in multiple-fluid simulations using the three-dimensional, electromagnetic LSP particle-in-cell code for large-scale plasma simulations. This phenomenon consists of an ion-bunching effect following shock transit of a classical interface undergoing diffusive relaxation. As a shock is launched across the interface region, rearward ions are injected ahead of the shock front and are collected into a co-moving bunch before collisions with the background fluid

eventually degrade them. Such a phenomenon, if validated with further simulations and experiments, may have the effect of flooding the hot-spot fuel with more mass near the "shock flash" than conventional simulations predict.

Project Summary

Our research has helped provide worldwide exposure to the potential implications of plasma kinetic and multiple-fluid effects in inertial-confinement fusion implosions. We developed a plasma version of the familiar adiabatic (no loss or gain of heat) lapse-rate phenomenon from atmospheric physics and applied it to capsule implosions and dynamics of hohlraum cylinders housing the target capsule. We also studied the behavior of multiple-species plasma shocks with self-generated electric fields using the LSP hybrid particle-in-cell code, successfully benchmarking this behavior against analytic theory. A new phenomenon of ion bunching from shock traversal of an interface is predicted to potentially affect early formation of the central hot spot by flooding the gaseous fuel with solid fuel at the time of shock flash.

Publications and Presentations

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Transport Properties of Dense Plasmas and a New Hybrid Simulation Technique for Matter at Extreme Conditions

Frank Graziani (12-SI-005)

Abstract

We propose to apply a recently developed, massively parallel molecular dynamics code for hot dense matter—the ddcMD (domain decomposition molecular-dynamics) code—to better understand model uncertainties for plasmas related to thermal conductivity and stopping power from ion collisions with electrons. These issues are critically important to stockpile stewardship and laser fusion energy. In addition, the project will significantly extend the ddcMD code's current capability to the regime of warm dense matter, where strong coupling exists and the electrons have significant degeneracy. The code will simulate strongly coupled, nonideal, and degenerate plasmas known as warm dense matter, and will address species diffusivity and equation-of-state issues. It will provide insight into existing theories of complex

plasmas, including mixtures, and motivate developments of new theories and experiments.

The proposed project will enable us to determine regime diffusivity for warm dense matter and equation of state for low- and high-atomic-number plasma mixtures. We will study their behavior as a function of atomic number and high-atomic-number impurity concentration. These results will be compared to current model implementations in burn codes. In the regime of hot dense matter, the proposed project will produce stopping power and thermal conductivities for low- and high-atomic-number mixtures. We will also implement a new treatment of dynamic electrons in the regime of hot dense matter and study the implicit limit of electron kinetic equations and the utility of this approach for thermonuclear burn.

Mission Relevance

This project will improve predictive capability of Livermore's Advanced Simulation and Computing codes and will facilitate the design of ignition experiments at the National Ignition Facility in support of stockpile stewardship science and energy security for the nation. These goals will be accomplished by enhancing our fundamental understanding of complex nonideal plasmas and by validating models used in stockpile stewardship and National Ignition Facility codes.

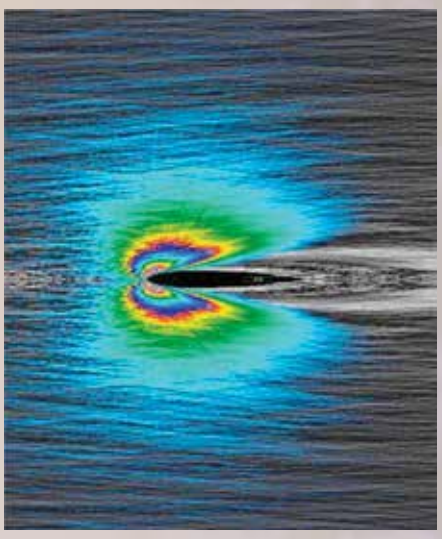
FY14 Accomplishments and Results

In FY14 we (1) performed large-scale molecular dynamics simulations of classical, charged particle stopping on the Sequoia supercomputer at Livermore—simulations spanned the weak to strongly coupled regime; (2) made comparisons with disparate kinetic theories to establish regimes of validity; (3) used molecular dynamics to investigate the micro-field distribution in plasmas, providing insights into the nature of continuum lowering; (4) used our molecular dynamics capability to investigate underlying assumptions in opacity codes; (5) used our molecular dynamics capability along with theory to investigate electrical and thermal conductivity, making comparisons with theory and other codes; (6) wrote a three-dimensional quantum hydrodynamics code for solving the stopping of fast ions in warm dense matter, performing simulations and comparing these to data taken at Livermore's Jupiter Laser Facility; (7) studied species diffusivity in warm dense matter with molecular dynamics and theory, focusing on the effect of asymmetric plasmas where one species is low atomic number and the other species is high atomic number; (8) developed new theoretical capabilities with kinetic-theory molecular dynamics and quantum hydrodynamics; and (9) developed a new numerical method for solving the quantum Lenard–Balescu equation for interaction in a plasma, demonstrating the method on simple test problems.

Project Summary

The successful conclusion of this project has provided LLNL with a new computational and theoretical capability for studying warm dense and hot dense matter, including

out-of-equilibrium systems. The backbone of this multi-institutional effort—the Cimarron Project—is the massively parallel molecular dynamics code ddcMD, developed at Lawrence Livermore National Laboratory. The project's focus is material conditions such as those in inertial-confinement fusion experiments, and in many stellar interiors: high temperatures, high densities, significant electromagnetic fields, mixtures of high- and low-atomic-number elements, and non-Maxwellian particle distributions. Of particular importance is our ability to incorporate key atomic, radiative, and nuclear processes into this classical molecular dynamics code, so that their interacting effects under nonideal plasma conditions can be investigated. This molecular dynamics capability has been applied to a variety of microphysics issues including plasma diffusivity, conductivity, stopping power, and continuum lowering. In addition, we have developed a new set of theoretical tools for investigating the microphysics of warm and hot dense matter based on quantum kinetic theory and quantum hydrodynamics. Finally, we have developed the theoretical basis for a hybrid method that ties together quantum kinetic theory and classical molecular dynamics. The National Boost Initiative, along with support from Los Alamos National Laboratory, is enabling continuation of the Cimarron effort.



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Asteroid Deflection

Paul Miller (12-ERD-005)

Abstract

We propose to address a challenge expressed in a presidential science adviser call for significantly more analysis of asteroid deflection methods that could be used to protect Earth in the event of an impending collision. The Nuclear Regulatory Commission reported to Congress that nuclear explosives are currently the only option to defend Earth against large asteroids, or, for smaller asteroids, when the time before a collision is short. We intend to explore approaches employing nuclear devices to deflect near-Earth objects on course to collide with Earth. Effects of such collisions range from localized disasters to massive global devastation. We will develop scenarios with a range of threat compositions, sizes, dynamics, and times to impact, and optimize parameters such as height of burst and yield for a nuclear deflection response. We will collaborate with experts in academia and the National Aeronautics and Space Administration. Much of our work will be unclassified, using generic sources for the studies.

We expect to develop threat scenarios and evaluate and optimize a variety of nuclear approaches, develop an asteroid breakup modeling capability, and increase the expertise in and understanding of threats from near-Earth objects. We will assess U.S. devices for their applicability against a range of potential threats. This project will substantially improve our understanding of options available to disrupt or divert objects on a collision course with Earth.

Mission Relevance

Our proposed research draws directly on LLNL's nuclear-design capability for a mission of national interest in ensuring international and domestic security. In addition, the work expands upon our traditional role of stockpile stewardship because nuclear explosives represent one of the major options for asteroid deflection.

FY14 Accomplishments and Results

In FY14 we (1) refined and expanded our scenarios and simulations with our suite of codes, including developing a scenario based on the asteroid Bennu and several standardized test problems; (2) used test problems to validate our code results, including tests of energy deposition using different codes; (3) completed a preliminary assessment of U.S. capabilities and their effectiveness for a range of object sizes; and (4) further developed our collaborative interactions, including joint exercises with the National Aeronautics and Space Administration, Federal Emergency Management Agency, and Defense Advanced Research Projects Agency, as well as ongoing collaborations with the National Aeronautics and Space Administration's Goddard Spaceflight and Ames Research centers, the University of Colorado, Arizona State University, and a National Nuclear Security Administration tri-laboratory collaboration with Los Alamos and Sandia national laboratories.

Project Summary

Over the course of the project, we established a suite of modeling and assessment capabilities for use in planetary defense from asteroid impacts. These include (1) estimation of deflection uncertainties; (2) the ability to model solid, fractured, or rubble asteroids, with realistic shapes (see figure); (3) a detailed understanding of the energy deposition from a nuclear blast; (4) a capability to model kinetic impacts; (5) development of simulation tools for modeling both deflection and disruption; (6) definition of scenarios for exploration and comparison; (7) development of test problems for validation and verification; (8) a capability to model orbital dispersal; (9) the beginning of improved equations-of-state, fracture and failure, porosity, and strength models; and (10) a preliminary assessment of existing U.S. deflection capabilities. All of this served to stimulate the formation of a follow-on NNSA tri-laboratory project on the topic of planetary defense from asteroid impacts in collaboration with Los Alamos and Sandia national laboratories.

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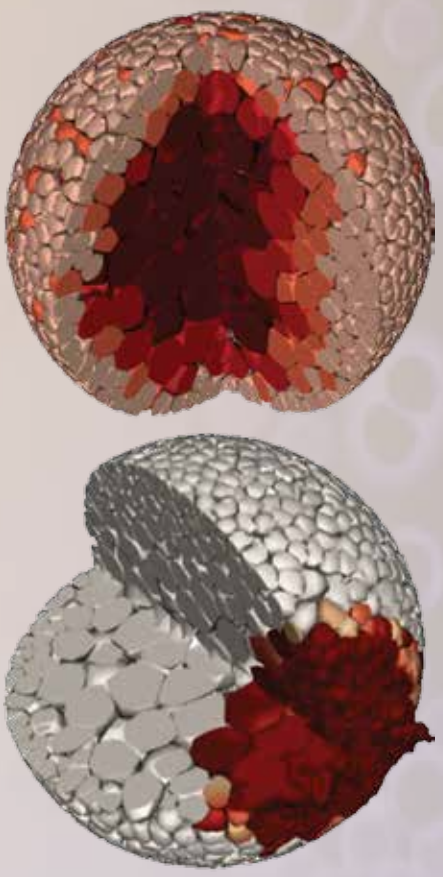

Initial configuration of a hypothetical asteroid with a section removed to view the interior structure (top). Colors designate different size ranges of boulders. Snapshot of early-time response to energy deposition from a nuclear blast on the front surface, with colors representing velocity magnitude (bottom).

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Predicting Weapon Headspace Gas Atmosphere for Modeling Component Compatibility and Aging

Elizabeth Glascoe (12-ERD-046)

Abstract

As the nuclear stockpile ages and undergoes life-extension measures, concerns about material compatibility and aging arise. In a warhead, material incompatibilities and degradation can result in a loss of functionality. Moreover, these problems may appear after decades of apparent compatibility or after a change made as part of a life-extension program. We currently lack a science-based understanding of what constitutes the gas-phase signatures of such undesirable material changes that are detectable in systems surveillance. Our goal is to develop a capability for assessing material compatibilities with age, specifically, a model that simulates the reactive transport of volatile species through materials. The model will be based on fundamental physical and chemical properties of the materials and will be versatile enough to apply to different geometries, sizes, and arrangements. Our approach of using a reactive transport code to predict material compatibilities could have wide-ranging applications in military, aerospace, medicine, and other fields.

We will develop a new methodology and capability to predict material compatibilities. Our computational model will simulate the transport and chemical reactions of volatile species and thereby predict the resulting constituents of headspace gas (in the unfilled space in a container). To this end, we will characterize and quantify the fundamental physics of transport, sorption, and chemical-reaction kinetics and mechanisms, creating a method for assessing the long-term compatibility of warhead materials.

Mission Relevance

This project supports the Laboratory's stockpile stewardship mission by providing a capability to predict the chemical compatibility of materials over the long term. It

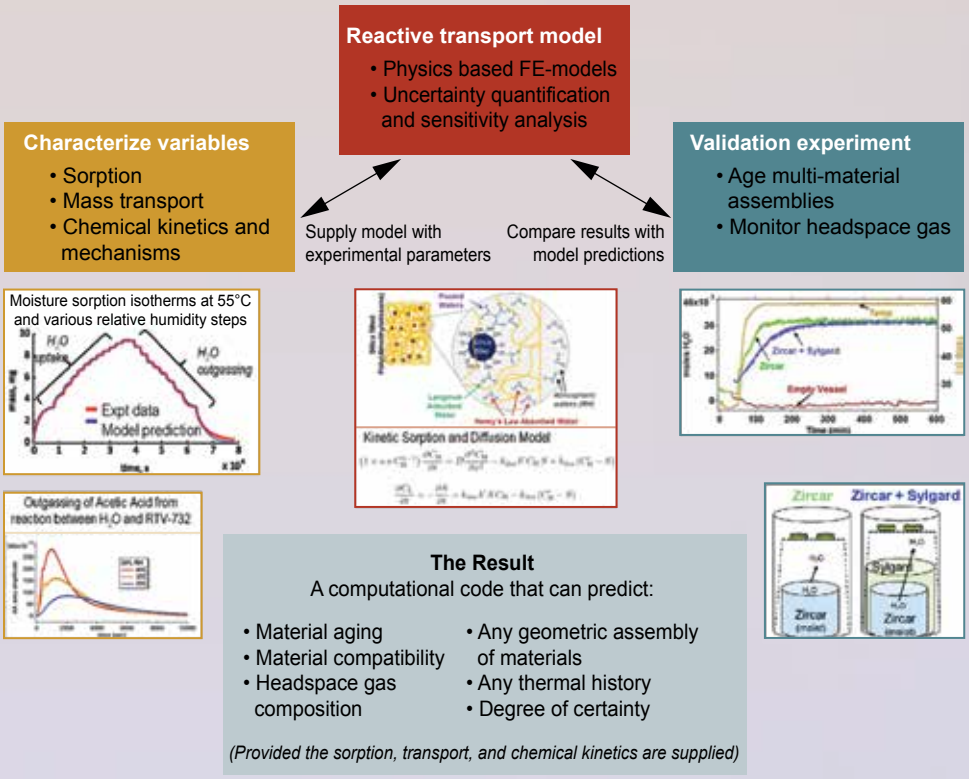
enables greater predictive foresight in selecting replacement materials for stockpile life-extension programs.

FY14 Accomplishments and Results

In FY14 we (1) tested and validated our dynamic sorption and diffusion model against one-dimensional experiments of moisture uptake and outgassing, using numerous materials for validation; (2) identified multiple short-comings in the model that were corrected and improved; (3) developed and tested uncertainty quantification and sensitivity analysis modeling methodologies; (4) performed sorption and diffusion experiments on materials using acetic acid rather than water vapor; (5) introduced and tested chemical reaction modeling to the model; and (6) completed kinetic validation experiments using RTV-732 silicone sealant and a one-dimensional wafer over multiple temperatures and humidities.

Project Summary

This project was successful in establishing an unique capability for measuring and modeling material compatibility, as shown in the figure. It can serve as a model for the rest of the weapons complex and be deployed to multiple applications. Our primary experimental developments included establishment of (1) a new technique for measuring vapor uptake and outgassing, (2) multiple-material aging methods, and (3) moisture-based chemical reaction quantification methods based on quadruple-mass-spectrometry and heat-flow calorimetry. Our primary modeling accomplishment included development of a



We established a unique capability for predicting material compatibility by measuring and modeling sorption, diffusion, and chemical reaction kinetics of vapors in materials.

dynamic sorption and diffusion model based on absorption, adsorption, and pooling. Uncertainty quantification and sensitivity analysis methods using Livermore's PSUADE code (problem-solving environment for uncertainty analysis and design exploration) were applied to the model, and we introduced chemical reactions modeling as well. The combination of these mechanisms to model compatibility and aging in a system context has not been used previously, and represents a new state of the art in this area. The DOE weapons program will provide support for further development and utilization of our capability, and additional funding opportunities are being explored relevant to munitions technology development, shale-gas production, and countering chemical warfare agents.

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A Model-Reduction Approach to Line-By-Line Calculations for Opacity Codes

Carlos Iglesias (12-ERD-047)

Abstract

The physical properties of stars depend upon the transport of energy from their nuclear cores to their surface. Although energy can be transferred out from the center by conduction and convection, radiation transport is the most important mechanism. In turn, the transport of photons depends on transparency of the intervening matter, termed the radiative opacity. Consequently, opacity plays a key role in determining the evolution,

luminosity, and instabilities of stars and even the eventual fate of the universe. We propose to improve opacity calculations by developing an accurate, efficient model-reduction approach to calculating bound–bound radiative transitions in many-electron ions. Opacity calculations for many-electron systems presently contain uncertainties because of the Gaussian approximation used to describe transition-array spectra. This research will bridge the extremes between the fast but potentially inaccurate Gaussian approximation presently in use and the exact but computationally expensive line-by-line calculation, with an expression that preserves higher moments of the transition array. We will develop an alternative method for computing moments of the transition array and incorporate the algorithms into LLNL opacity codes.

Our main goal with this project is to develop a model-reduction approach to many-electron ions that is accurate and effective. If successful, this project will develop a new algorithm that will allow an accurate and efficient optimization of the number of effective spectral lines required to generate a more realistic representation of bound–bound spectra. This result will significantly reduce computational time and allow opacity codes to handle larger problems without increasing computational effort.

Mission Relevance

Stellar models and Laboratory applications, such as stockpile stewardship experiments at LLNL's laser facilities, rely on accurate descriptions of energy transport. By reducing the computational time required to calculate bound–bound spectra, this research furthers understanding of energy transport for the Laboratory's stockpile stewardship strategic focus area and supports the national security mission.

FY14 Accomplishments and Results

In FY14 we extended the partially resolved transition-array concept to the super-configuration approach and implemented it in Livermore's VISTA, a relativistic, local thermodynamic-equilibrium opacity code. This extension describes limited configuration-interaction effects important when other explanations of angular momentum coupling for light or heavy atoms fail, without relying on previous approximations.

Project Summary

Our partially resolved transition-array concept was developed in the first two years and have been implemented into Livermore's TOPAZ, a line-by-line opacity code. The method increases the computational speed of opacity codes using line-by-line approaches by two orders of magnitude. In addition, the partially resolved transition-array concept was also extended to the super-configuration approach and implemented into Livermore's VISTA, a relativistic, local thermodynamic-equilibrium opacity code. The latter extension not only describes limited configuration interaction effects important when other explanations of angular momentum coupling fail, without relying on the previous approximations, but more importantly it shows a clear view of how future opacity codes will be structured. The recent development of the partially resolved transition-array concept is essential for the design of next-generation opacity codes.

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Early-Phase Hydrodynamic Instability Development in National Ignition Facility Capsules

Daniel Clark (12-ERD-058)

Abstract

Recently, the importance of the effect of early-phase Richtmyer–Meshkov instability growth on later Rayleigh–Taylor hydrodynamic instability has been appreciated in determining the pulse shape needed to achieve fusion ignition in National Ignition Facility (NIF) experiments. Richtmyer–Meshkov instability occurs when two fluids of different density are accelerated, and the Rayleigh–Taylor instability is the result of a lighter fluid pushing a heavier one at the interface between the two fluids. We will develop a quantitative understanding of the role of early-phase Richtmyer–Meshkov growth in determining the phase and amplitude of later Rayleigh–Taylor growth in NIF fusion implosions. To this end, we will combine simulations using the radiation-hydrodynamics code HYDRA with published and original analytic work. Although the immediate scope of the project is limited to indirectly driven NIF implosions and to theoretical and simulation-based investigations, our results will be relevant to other applications in stockpile stewardship science, NIF uses of ignition, and target design for inertial fusion energy.

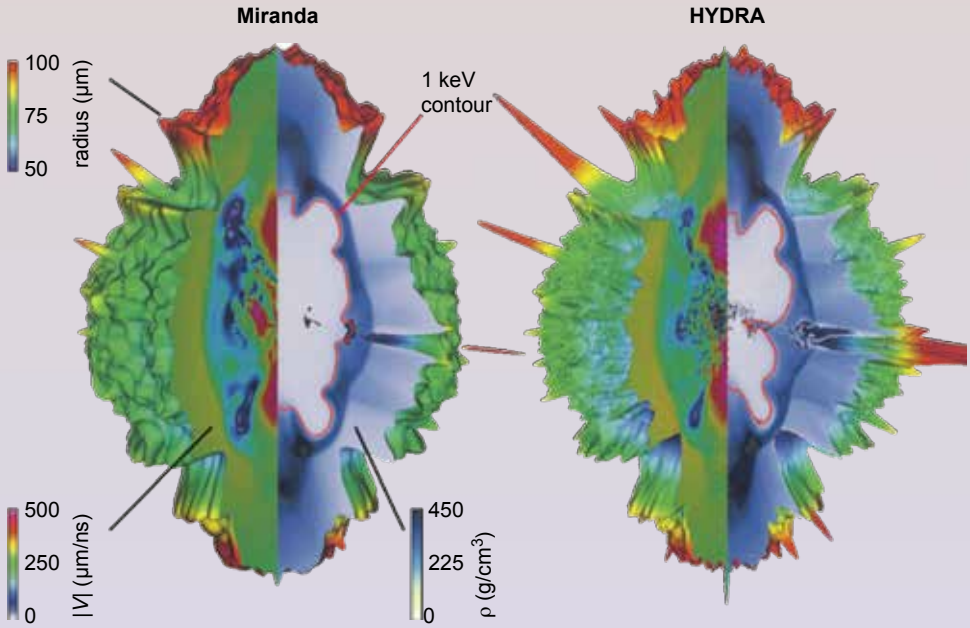
Our three main deliverables will be (1) a detailed and quantitative understanding of the connection of Richtmyer–Meshkov phase growth to subsequent Rayleigh–Taylor growth in NIF implosions; (2) an understanding of the influence of equation of state, opacity, and other physics uncertainties in how Richtmyer–Meshkov instability growth affects Rayleigh–Taylor growth; and (3) a comparison of our results with mixed-model results previously obtained at LLNL. A possible additional result will be experimental designs to test our theoretical results on NIF.

Mission Relevance

By helping achieve a more complete understanding of hydrodynamic instabilities and mix, this project will reduce uncertainties in stockpile stewardship science applications, may uncover new physics relevant to stockpile stewardship, and will further LLNL's missions in nuclear and energy security, including NIF ignition, in which hydrodynamic instabilities play a central role. In addition, by understanding details of instability development more thoroughly and by precisely determining acceptable stability boundaries, this project will also help improve ignition-capsule hydrodynamic stability and therefore enable the high-gain target designs required for inertial fusion energy.

FY14 Accomplishments and Results

In FY14 we (1) finalized a fairly accurate simplified analytical model of the combined effects of Richtmyer–Meshkov and Rayleigh–Taylor in determining ablation-front growth-factor spectra; (2) performed an assessment of growth-factor sensitivity to opacity and M-band (2- to 5-keV radiation) variations—an assessment of equation-of-state sensitivity remains incomplete at this time, and this research avenue was de-emphasized given the apparent agreement between simulations and recent NIF experimental results, suggesting that either these sensitivities are small or the current models are sufficiently accurate that these sensitivities are not a concern; (3) shifted our emphasis to exploiting our validated simulation methodology to develop improved implosion designs, and the resulting two improved implosion designs were tested experimentally at NIF; and (4) completed several three-dimensional simulation comparisons between Miranda (a hydrodynamics code for simulating Rayleigh–Taylor and Richtmyer–Meshkov instability growth) and HYDRA, showing good agreement (see figure) and highlighting the importance of viscosity in suppressing turbulence in three-dimensional NIF hot spots.



Comparison of three-dimensional Miranda and HYDRA radiation-hydrodynamics codes simulations of a National Ignition Facility shot at Livermore. The fuel–ablator interface is shown at the exterior of each figure with a scale in the upper left. Velocity magnitude is shown on the left interior for each figure (scale is shown on the lower left), and density on the right interior (scale is shown on the lower right). In addition, a contour showing the 1-keV hot-spot boundary is also shown on the right interior side in red. The large amount of distortion because of instability growth and other sources of asymmetry is apparent. Despite the very different numerics of Miranda and HYDRA, the high level of agreement between the two simulations is evident.

Project Summary

Our research has substantially deepened and extended our understanding of hydrodynamic instabilities relevant to NIF. The importance of Richtmyer–Meshkov and Rayleigh–Taylor instabilities in implosions at NIF has long been appreciated and extensively studied, but a detailed understanding of the cooperation of these two instabilities to produce the stability characteristics typical of the implosions had remained incomplete. With our improved understanding of instability growth, the alternate implosion designs developed as part of this project have also substantially impacted experimental plans for NIF and hopefully will result in progress towards the final goal of ignition. Our three-dimensional simulations performed with Miranda have altered our appreciation of the importance of viscosity in NIF hot-spot evolution and overturned nearly three decades of common wisdom that viscosity is unimportant. Finally, two postdoctoral researchers have been trained as part of this work, and both have been hired as permanent staff members. The understanding gained and concepts developed during this project have been adopted by the NIF mix campaign and will continue to be explored experimentally.

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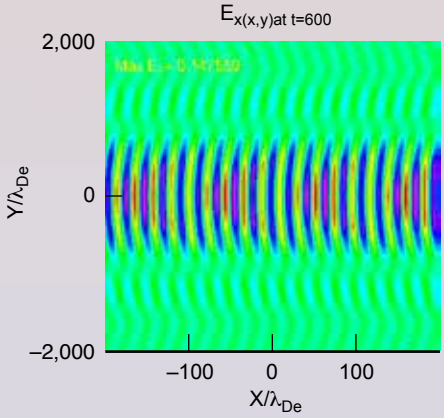
Theory and Simulation of Large-Amplitude Electron Plasma and Ion Acoustic Waves with an Innovative Vlasov Code

Richard Berger (12-ERD-061)

Abstract

The goal of the National Ignition Campaign is to implode capsules and ignite the deuterium–tritium fuel with x rays converted from laser light in a high-atomic-number plasma. If successful, this is a potential path to fusion energy, a safe and carbon-free source of energy. Experiments at Livermore's National Ignition Facility to realize this goal depend on predictable propagation of the 192 laser beams through hot dense plasma. We propose to examine processes that affect laser light propagation directly and the effects on the distribution of electrons from plasma waves excited by laser light. Specifically, we intend to improve understanding of the kinetic processes, described by the Vlasov equation, that determine the nonlinear state of large-amplitude electron plasma waves and ion acoustic waves and the self-consistent distribution of electrons and ions associated with these waves in two dimensions. We will use Livermore-developed 2D + 2V Vlasov codes (two dimensions in space and velocity), to study nonlinear ion acoustic and electron plasma waves in hot dense plasmas.

We expect to establish the dependence of transverse and longitudinal modulation instability of an electron plasma wave-on-wave amplitude and wavelength over the Debye length (part of the thermodynamic description of large systems of mobile charges). We will also study the nonlinear evolution of two-species ion acoustic waves, specifically the slow mode of carbon–hydrogen plasmas. We will create a multiple-ion species Vlasov code by generalizing the choice of boundary conditions in the laser–plasma interaction code VALHALLA (Vlasov adaptive limited high-order algorithms for laser applications), as well as input options, and consider collisions in VALHALLA for inclusion, if feasible. Nonlinear ion acoustic waves will be studied for single- and multiple-ion species with VALHALLA using the same methods developed to study electron plasma waves. The results will be applicable not only to the understanding of stimulated Raman and Brillouin light-scattering processes, but also to other effects such as two-plasmon decay and ion acoustic waves driven by an inter-penetrating plasma.



A traveling electron plasma wave undergoing simultaneous self-focusing and longitudinal modulational instability.

Mission Relevance

Our research on laser light propagation and the effects of plasma waves on electron distribution and hard x-ray generation relevant to nuclear fusion supports the Laboratory's stockpile stewardship strategic focus area as well as the inertial fusion science and technology strategic focus area.

FY14 Accomplishments and Results

In FY14 we (1) added to LOKI, our two-dimensional Vlasov code, a new pitch-angle-scattering algorithm that reproduced known physical results for plasma wave damping to high accuracy, (2) wrote new post-processing tools for the refactored LOKI, (3) conducted two-dimensional ion acoustic-wave simulations, and (4) showed that the threshold for the Langmuir wave-decay instability requires wave amplitudes too large to be present in our Vlasov simulations.

Project Summary

Our research provided the Laboratory with a highly-scalable, parallelized, accurate 2D + 2V Vlasov simulation code, built with very modern architecture that will allow continued improvements without restructuring. To our knowledge, this code has no Vlasov competitors. We provided an assessment of the nonlinear evolution of large-amplitude plasma waves that differs markedly from that provided by the standard method for solving the Vlasov–Maxwell system, the particle-in-cell method. This alternative view demands deeper understanding of the role statistical noise plays in particle-in-cell simulations. To that end, new collaborations have been established with Los Alamos National Laboratory and the French Atomic Energy Commission to compare the particle-in-cell method with Vlasov. The physical processes we have studied will be the basis for developing new models of nonlinear interactions that will be added to LLNL's primary laser–plasma interaction simulation code, pF3D. Those modifications may allow modeling beyond the current empirically established laser–plasma-interaction "playbooks." We have developed collaborations to explore experimental tests of our results and will use this unique capability to attract new staff.

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Banks, J. W., et al., 2013. *Steady electron plasma waves in 2D + 2V*. 43rd Anomalous Absorption Conf., Stevenson, WA, July 7–12, 2013. LLNL-POST-640095.

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New Energetic Materials

Philip Pagoria (12-ERD-066)

Abstract

With few investigators in the U.S. developing new energetic compounds, we propose to identify methods that will provide two such compounds. Together, they would improve current and future weapon systems, while at the same time having fewer deleterious environmental and health effects. The two are highly oxidized energetic compounds for use as replacements for ammonium perchlorate in rocket propellants and liquid energetic plasticizers for both propellant and explosives applications. We envision discovering new reaction pathways that will be important for the scientific community because of the structural similarities to biologically active compounds.

If successful, we will provide new energetic compounds of utility to current and future weapon designers, along with increased understanding and expertise in energetic materials synthesis. Our new compounds will be fully characterized with respect to sensitivity, equation of state, and thermal stability—important parameters for weapon designers and modelers. The new compounds and intermediates may have pharmaceutical applications because they are structurally similar to known biologically active compounds. This research will help to fill the gap in basic research in energetic materials synthesis in the U.S.

Mission Relevance

This project overlaps with Laboratory missions in biochemistry and stockpile stewardship. The development of these new compounds will give weapon designers new materials to achieve enhanced performance and reduced sensitivity, leading to a safer and more secure stockpile. This effort also will lead to a better understanding of synthesis efforts in foreign countries and of the performance, synthesis, and sensitivity of homemade explosives, in support of national security.

FY14 Accomplishments and Results

In FY14 we (1) structurally characterized several new energetic compounds by spectroscopic techniques and by single-crystal x-ray diffraction, as well as having their small-scale safety-test responses measured; (2) synthesized several precursor compounds and converted them to target energetic compounds; (3) prepared two new energetic compounds from the fluorodinitroethyl group to improve thermal properties; (4) scaled the synthesis of LLM-204 (shown in figure), LLM-208, and LLM-209 up to 25-g amounts; (5) formulated the three compounds with the synthetic rubber polyisobutylene to create energetic plasticizers and submitted them to a disc acceleration experiment to determine detonation performance; and (6) synthesized a new energetic plasticizer, LLM-222, as well as an energetic plasticizer precursor at the 1-g scale.

Project Summary

Eight new energetic compounds containing polynitroethylamino- and fluorodinitroethylamino-groups and nitrogen–oxygen heterocycles were created. These materials were synthesized at different scales, some in excess of 25 g, and we determined material structures and handling safety. The heats of formation were either estimated or calculated from the measured heat of combustion data, and results were used along with measured material densities to predict detonation performance and specific impulse of the compounds using the LLNL-developed Cheetah code for predicting explosive detonation performance based on thermochemistry and detonation physics. The compounds LLM-201, LLM-204, LLM-208, and LLM-209 were all scaled-up to the 25-g level, formulated, and submitted for disc acceleration performance testing. Essentially, these new materials are on the shelf for designers to consider in current and future weapon systems. Our suite of compounds has been submitted to various Department of Defense agencies for consideration for further development. Of particular interest is LLM-201, which has attractive sensitivity and thermal properties as a melt-pour ingredient. The compound LLM-208 is a dense, thermally stable ingredient that meets the metrics for a propellant oxidizer material. The approach taken in this project to couple stable nitrogen–oxygen heterocycles with what are typically thermally unstable polynitroalkyl groups to generate new compounds with improved thermal stability could be extended to other energetic systems in the future. The development of the new energetic plasticizer LLM-222 has been reported to various defense agencies and it (along with other plasticizer precursors) will be scaled-up under the DOE and Department of Defense Joint Munitions Program.

Publications and Presentations

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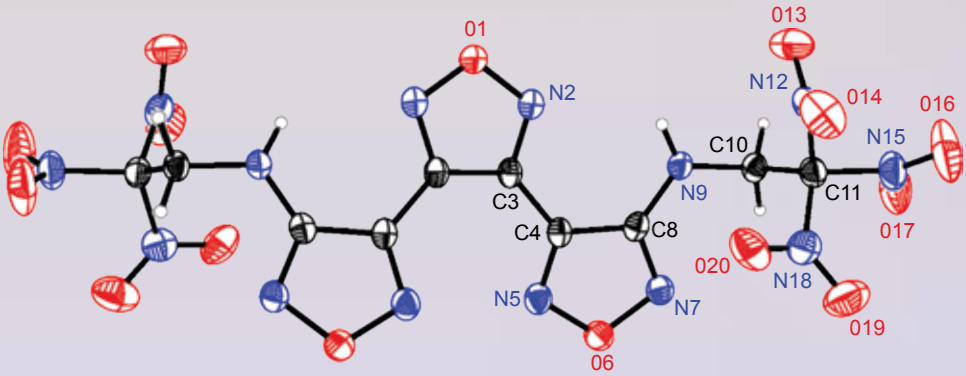

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Application of Imposed Magnetic Fields to Ignition and Thermonuclear Burn at the National Ignition Facility

L. John Perkins (14-ERD-028)

Abstract

Achieving ignition and thermonuclear burn at Livermore's National Ignition Facility (NIF) would provide data relevant to stockpile stewardship in the nuclear regime unattainable outside of an underground nuclear test. Although NIF has returned fusion data of unprecedented quality, fusion yields remain a factor of ten too low for bootstrap heating, which is the self-heating process whereby alpha particles from the deuterium–tritium reaction in the target capsule deposit their energy back into the fuel and raise the temperature, resulting in a significant increase in the thermonuclear reaction rate. Applying a seed magnetic field that would compress ignition targets to greater than 10,000 T under implosion could provide a path to ignition by relaxing conditions required for ignition and propagating burn. Using a magnetized NIF ignition target, we will test the ability to achieve ignition in NIF capsules that would otherwise fail because of adverse hydrodynamic instabilities. We will accomplish this goal with a hohlraum hollow cylinder that has a cavity coil driven by a co-located pulsed-power supply and a deuterium–tritium gas ignition test capsule, demonstrating detectable alpha-particle heating through magnetized volumetric burn. If successful, this project will lead to the fielding of a cryogenic-layered, solid deuterium–tritium ignition capsule in NIF ignition research.

We will show that imposed magnetic fields compressed to high values under NIF implosions can significantly relax the conditions for achieving ignition. To this end, we will develop a magnetized hohlraum and an ignition gas capsule to assess fusion burn physics under high magnetic fields. Specifically, we will (1) produce an integrated design of the proposed magnetized hohlraum, the deuterium–tritium gas ignition test target, and the follow-on cryogenic-layered deuterium–tritium target; (2) design, construct, and test a pulsed-power supply and transmission line capable of supporting a 40-T axial hohlraum field; and (3) conduct proof-of-principle tests on NIF of the magnetized ignition platform to demonstrate detectable fusion alpha heating in compressed magnetic fields.

Mission Relevance

This project supports the Laboratory's mission in national and energy security by advancing NIF ignition platforms and ignition probability, improving models for predictive capability, contributing to weapons physics data, and supporting stockpile stewardship and high-energy-density burning plasma science, as well as the strategic focus area in inertial-confinement fusion science and technology.

FY14 Accomplishments and Results

In FY14 we (1) produced designs of cryogenic-layered ignition targets and warm deuterium–tritium volumetric-burn gas targets with imposed magnetic fields; (2) performed two-dimensional simulations with our LASNEX and HYDRA radiation-hydrodynamics codes that demonstrate expected increases in ignition margins and suggest that present sub-marginal NIF designs may reach the ignition regime under imposed magnetic fields; (3) performed initial assessment of hohlraum plasma conditions under a magnetic field via two-dimensional integrated simulations that indicate there is potential for suppression of hot-electron preheat; (4) designed the NIF power supply, transmission lines, and coil system to provide hohlraum fields of approximately 70 T, and placed orders for a custom-fit capacitor for integration in the NIF diagnostic instrument manipulator; (5) performed modeling of the coupled hohlraum and coil magneto-hydrodynamic issues with the three-dimensional arbitrary Lagrangian–Eulerian code, which indicated that uranium (but not gold) hohlraums should permit adequate field penetration; and (6) scoped NIF integration issues and fielding options, and produced the initial three-dimensional computer-aided design drawings for power supply integration.

Proposed Work for FY15

In FY15 we will (1) produce final integrated designs for cryogenic-layered and warm deuterium–tritium gas targets with an imposed magnetic field for fielding in FY16; (2) assess ignition margin and stability for the targets; (3) integrate two-dimensional hohlraum physics simulations with the coupled HYDRA radiation-hydrodynamic code and ZUMA hybrid particle-in-cell code, addressing magnetic-field-dependent suppression of hot-electron transport and beneficial effects on capsule preheat; (4) fabricate four hohlraum–coil targets; (5) construct and test power supply, transmission lines, and hohlraum and coil on a stand-alone test stand, and characterize the magnetic field during the test; (6) develop final designs for NIF siting and integration for fielding in FY16, and select fielding options; and (7) produce final three-dimensional computer-aided-design drawings and a NIF system-integration map.

Publications and Presentations

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Advanced Double-Shell Target Designs for Inertial Fusion Energy

Peter Amendt (14-ERD-031)

Abstract

We propose to develop innovative approaches to achieving double-shell ignition for targets on advanced laser systems, providing both a platform for studying high-atomic number and fuel mix and a starting point for developing high-gain designs for potential inertial fusion energy applications. The motivation for this project is to identify and further develop a complementary alternative to central-hot-spot ignition. Our plan is to modernize and adapt a former design—cognizant of advances since then in materials science, simulations, and target material and laser constraints—and implement innovative approaches to potentially simplify target fabrication while maximizing performance margin. The techniques to be applied will consist of analysis and one- and two-dimensional radiation-hydrodynamic simulations.

If successful, we expect an effort by the Laboratory towards pursuing a double-shell approach to demonstrate fusion ignition. Such an effort would provide a complementary approach to realizing the challenge of laboratory-scale ignition. A successful demonstration of double-shell ignition would inspire interest in developing high-gain versions of the target for potential inertial fusion energy applications. Our specific goals are to (1) modernize the original target design to accommodate the latest design techniques and available materials; (2) explore innovative designs for testing that could facilitate and simplify the target fabrication process, improve performance margins, and lessen exposure to debilitating mix; (3) propose opportunity shots at Livermore's National Ignition Facility that could test key performance aspects of the revised and improved designs; and (4) propose and develop advanced designs that can accommodate higher gains for providing a potential path to inertial fusion energy.

Mission Relevance

The proposed research sustains the core competency of nuclear science and technology through development of a high-atomic-number target ignition platform and meets the imperative of long-range, mission-directed research in the areas of inertial-confinement fusion energy and stockpile stewardship. This project will also

be effective for training early- and mid-career scientists in the methods of double-shell target design and stockpile stewardship applications.

FY14 Accomplishments and Results

In FY14 we developed a new ignition double-shell design that has a virtually interface-free inner shell. An often-expressed concern with past designs has been the vulnerability to performance-degrading mix between the inner shell with a high atomic number and the low-atomic-number deuterium–tritium fuel. The density mismatch across this problematic material interface (or Atwood number) is what drives the high potential for mix. Our FY14 breakthrough in the double-shell design is the projected use of additive manufacturing techniques for creating graded density foams and lead- and silica-based glasses to remove all classical interfaces from the inner shell. In this sense, the former issues with fuel and inner-shell mix exposure can be dealt with using three-dimensional nanometer-scale lithographic printing techniques.

Proposed Work for FY15

Our proposed research for FY15 consists of (1) simplifying the interface-free inner-shell design with regard to target fabrication and manufacturing, (2) performing one-dimensional simulations with turbulence mix models, (3) executing multiple-mode simulation studies of the design to assess and further minimize the role of destructive hydrodynamic instabilities, and (4) perform optimization studies of hohlraum ignition target capsules to maximize performance margins at the laser-fusion scale and to facilitate future laser operations in testing double-shell performance.

Publications and Presentations

Amendt, P., J. Milovich, and G. Zimmerman, 2014. *Interface-free ignition double-shell designs for high margin to inner-shell mix*. LLNL-PRES-655306.

LDRD

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