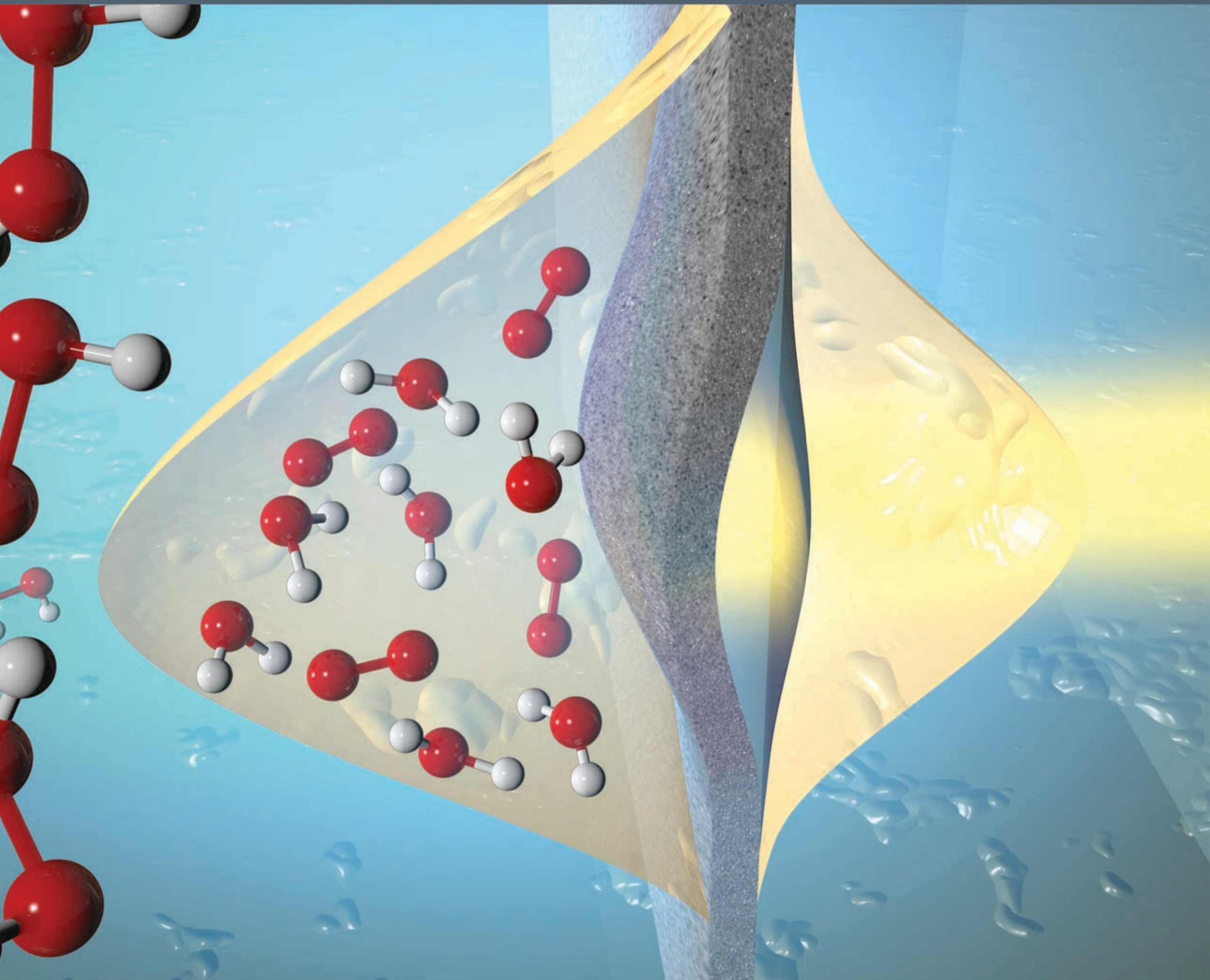
Lawrence Livermore National Laboratory



Laboratory Directed Research and Development

Annual Report FY2013

About the Cover

Theoretical studies of improvised explosives are rare, and their experimental study is expensive and technically challenging, but a necessary part of countering today's threats to national and international security. Principal investigator Sorin Bastea and his team developed important experimental data and simulations for characterizing and modeling the detonation behavior of explosives in the Laboratory Directed Research and Development project, "Detonation Performance of Improvised Explosives via Reactive Flow Simulations and Diamond Anvil Experiments" (11-ERD-067). Project results may enable development of ultrafast methods for assessing sensitivity and perhaps even performance in energetic formulations, and could additionally open new avenues for the shock synthesis of materials. Researchers determined that shock wave excitation enables the observation of chemical kinetics in a very small volume, where the rate of heating is limited only by the transit time of the shock wave through the volume. Effectively, this is in situ chemistry in a very small beaker. The research was recently featured as the cover story in the scientific journal, *Journal of Physical Chemistry A*. The simulation of ultrafast shock initiation of exothermic chemistry in hydrogen peroxide shown here was provided by Liam Krauss of the Livermore Computing visualization team, which specializes in multimedia presentation of scientific results obtained through high-performance computing.

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Acknowledgments

The Laboratory Directed Research and Development Program extends its sincere appreciation to the principal investigators of fiscal year 2013 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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Director's Statement

William H. Goldstein, Director

The Laboratory Directed Research and Development (LDRD) Program was conceived as a bold initiative, and scientific and technical risk are essential attributes of a portfolio that expands Lawrence Livermore National Laboratory's capability to serve our national security mission. 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 science and technology, maintaining our core competencies, building new capabilities, and reaching beyond the immediate challenges toward the future, the LDRD Program enables us to fulfill our national security mission in an evolving global context.

The LDRD Program is the largest single source of internal investment in our future, and for fiscal year 2013, the LDRD budget of $83.2 million supported 152 projects. 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 have impact on 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 strategic plan

- Developing programs in strategic focus areas, guided by the strategic plan, where we have chosen to build or expand our influence

- Looking beyond the immediate challenges to future opportunities

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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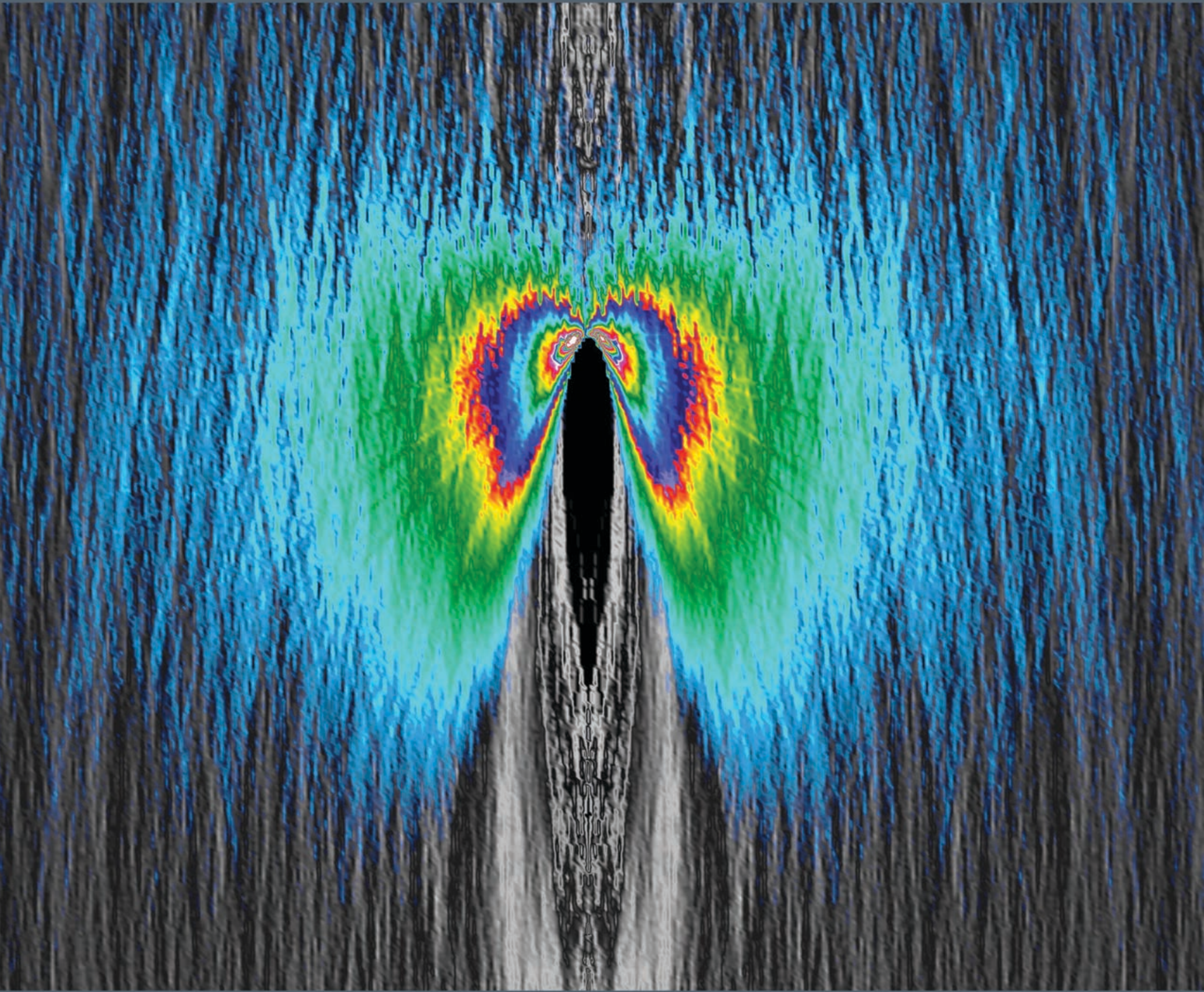


Laboratory Directed Research and Development

Annual Report FY2013

About Lawrence Livermore National Laboratory

A premier applied-science laboratory, Lawrence Livermore National Laboratory (LLNL) has earned the reputation as a leader in providing science and technology solutions to the most pressing national and global security problems.

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 missions.

About 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 excellent 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 and technology 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, and energy and environmental security, as well as cutting-edge science and technology and engineering in high-energy-density matter, high-performance computing and simulation, materials and chemistry at the extremes, information systems, measurements and experimental science, and energy manipulation.

About the *FY 2013 Laboratory Directed Research and Development Annual Report*

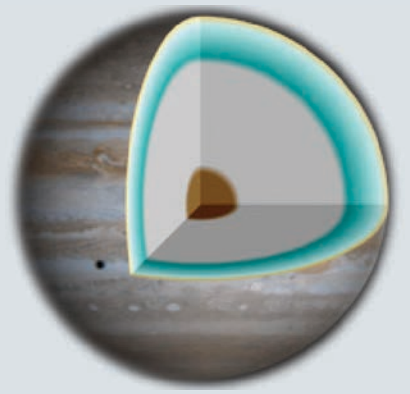
The LDRD annual report for fiscal year 2013 (FY13) provides a summary of LDRD-funded projects for the fiscal year and consists of two parts:

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 FY13, and a list of selected publications and presentations that resulted from the research.

Highlights of Accomplishments for the Fiscal Year

In FY13, 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 *Strategic Investment Roadmap*, as well as for critical national needs. Here, we provide examples of the highlights of accomplishments for various mission focus areas and foundational science, technology, and engineering.



Gas giant planets such as Jupiter (pictured) and Saturn, and possibly some recently discovered exoplanets, may contain large amounts of metallic hydrogen (depicted at the center).

Stockpile Stewardship

Hydrogen is one of the most important elements in the periodic table and a material that must be well understood to maintain the safety and security of the nation's stockpile of nuclear weapons because of its central role in nuclear fusion. However, despite decades of intense efforts from many experimental high-pressure groups, little concrete information is known about details of how hydrogen behaves at high pressures and temperatures. An alternative approach to experiments is the use of high-performance computers to simulate the activity of electrons under those conditions. Such simulations in the past have shown that under specific conditions, hydrogen becomes metallic, turning from an insulator into a conductor. A Livermore LDRD team, which is searching for metallic hydrogen from "first-principles" simulations, is performing breakthrough calculations of the fundamental electronic and optical properties of hydrogen at high pressure (13-LW-004). Their work is critical for the Laboratory, specifically for

- Developing next-generation, first-principles simulation methods of materials in nuclear weapons
- Understanding the complex physical processes that occur in weapons
- Placing the Laboratory in a leading role in the development of electronic structure methods
- Reinforcing Livermore's position as one of the leading centers for the study of materials at high pressure

The team's goal is to clarify the phase diagram of hydrogen where molecules break down—in effect, melt—under the effects of pressure. This is where hydrogen turns from a insulator into a metal. Where possible, their simulations will be compared to experimental results. In FY13, the team used a combination of numerical methods to calculate the free energy of the most important structures of both solid molecular and atomic hydrogen, at a single high temperature, allowing them to determine the correct structure of the solid as a function of pressure. An accurate understanding of hydrogen's properties is crucial not only for stockpile stewardship but also for astrophysics, planetary physics, inertial-confinement fusion, and energy production.

Nuclear Threat Reduction

The ability to detect hard x-ray or soft gamma-ray emissions from special nuclear material plays an important role in our ability to search for weapons, perform attribution in the event of an attack or interdiction of a device, and strengthen safeguards for treaty obligations. An LDRD team is developing hard x-ray mirrors to dramatically improve detection performance by increasing the photons collected from weak sources or by filtering out a background signal that obscures an important spectral signature (13-ERD-048). This work is important for the Laboratory's work in nuclear nonproliferation, specifically the development of new technologies to advance nuclear detection and diagnostics.

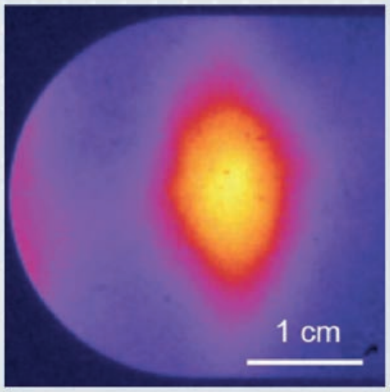
In FY13, the team built new multilayer substrates and successfully demonstrated their reflectivity at the highest-ever energies. They also developed the first predictive model of multilayer reflective properties. In the coming year, the team will extrapolate results from prototype optics into a full system for nuclear security applications. For its experiments, the team is using 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 the highest photon energies. In the project's first year, the team successfully

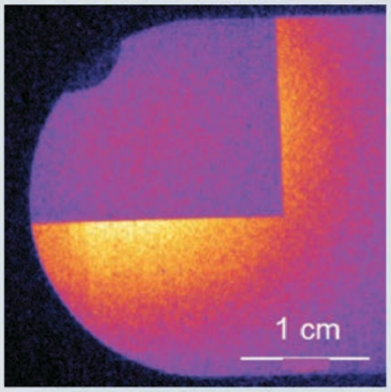
- Demonstrated the first-ever multilayer reflectivity above 500-keV
- Developed the first predictive model of multilayer reflective properties
- Identified materials for use in the optics as well as metrology techniques

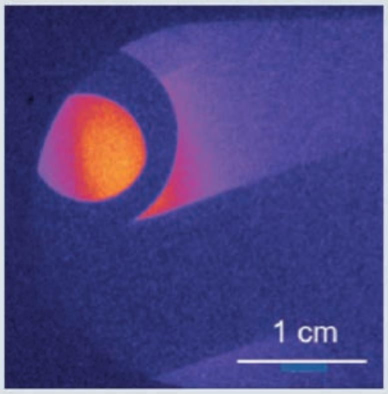
In the coming year, the team is extrapolating the results from focusing on optics into a full prototype system for nuclear security applications.

High-Energy-Density Physics

A new Livermore project seeks to create a compact, ultrafast hard x-ray source at Livermore that would allow researchers to perform multiple experiments in a single shot (13-LW-076). This source will join a growing number of advanced lasers at Livermore for materials research and nondestructive characterizations. For stockpile stewardship, this new hard x-ray source will help reduce uncertainties in plasma properties.







Betatron x-ray radiographs obtained at the Jupiter Laser Facility reveal images of a (top) beam profile, (center) silicon wafer, and (bottom) gold ignition target capsule.

The LDRD team is leveraging Livermore's unique expertise in accelerator, laser, and x-ray sciences and using both modeling and experiments of laser and plasma interaction to develop the source at the Callisto laser at the Laboratory's Jupiter Laser Facility. In FY13, the team developed the source—the size of a large tabletop—at Callisto, produced some of the highest betatron energies ever demonstrated, and performed preliminary imaging experiments at lower energies. Their new code for electron trajectories and betatron radiation agreed with experimental results, while a second code was used to analyze phase contrast images from the source. In the coming year, the team will

- Perform an experiment at Callisto to characterize the source using a longer-pulse and higher-energy laser, and probe a laser-driven shock using the resulting x rays
- Explore the possibility of future betatron x-ray source development on large facilities, including the OMEGA laser at the Laboratory for Laser Energetics in Rochester, New York, and Livermore's Advanced Radiography Capability
- Collaborate with the SLAC National Accelerator Laboratory in Menlo Park, California, to develop pump and probe extended x-ray absorption fine-structure experiments using the new x-ray source
- Collaborate with Rutherford Appleton Laboratory in the United Kingdom to perform x-ray phase-contrast imaging experiments using the source at the Livermore Gemini laser

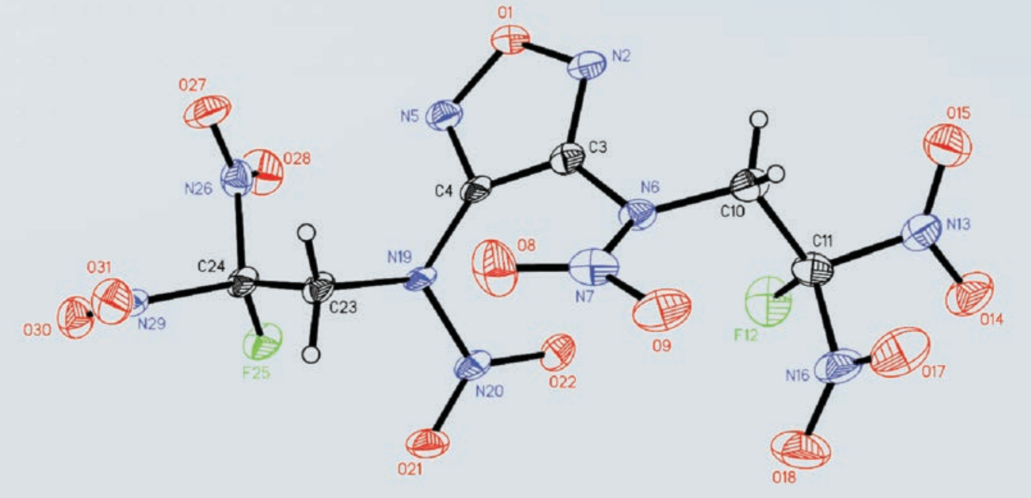
Once the source is complete, potential users will span the disciplines of ultrafast material characterization and imaging in industry, medicine, chemistry, protein crystallography, biology, and inertial fusion sciences.

Energetic Materials

Investigators at Livermore are some of the few in the U.S. who are synthesizing new energetic compounds. An LDRD team seeks to develop two such materials, both with fewer deleterious environmental and health effects than existing compounds (12-ERD-066). The two are highly oxidized energetic compounds for use as replacements for ammonium perchlorate in rocket propellants and liquid energetic plasticizers for propellant and explosive uses. The development of these new energetic materials

- Gives weapon designers new materials to achieve enhanced performance and reduced sensitivity, leading to a safer and more secure stockpile
- Leads to a better understanding of synthesis efforts in foreign countries
- Provides much needed data on the performance, synthesis, and sensitivity of improvised explosives, in support of national security

In FY13 the team synthesized several new compounds, pressed them into pellets, measured their heat of combustion, and performed safety testing. In the coming year, the team will continue to investigate new materials, and one in particular because of the discovery that it has improved thermal stability. The new compounds and intermediates may have pharmaceutical applications because they are structurally similar to known biologically active compounds.



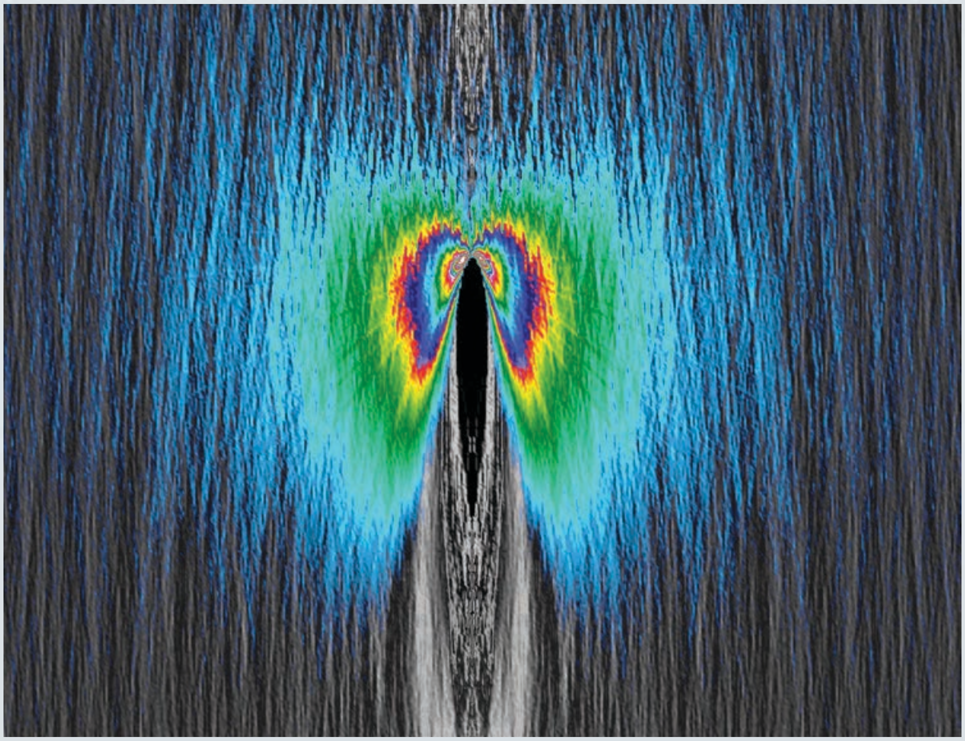
Crystal structure of LLM-209, a new highly energetic compound we have developed.

Inertial-Confinement Fusion

Enhancing fundamental understanding of complex non-ideal plasmas and validating models used in stockpile stewardship and National Ignition Facility codes is critically important to inertial-confinement fusion energy and the U.S. program of reliability testing and maintenance of its nuclear weapons without the use of nuclear testing.

The LDRD project examining transport properties of dense plasmas and a new hybrid simulation technique for matter at extreme conditions (12-SI-005) is applying a recently developed, world-class massively parallel molecular dynamics code for hot dense matter to better understand model uncertainties for plasmas related to thermal conductivity and stopping power from ion collisions with electrons. The rate at which charged particles slow down as they interact with matter is an important indicator of the underlying particle interactions, and simulations of this process help test the accuracy of models of important processes of energy and particle flow, such as thermal conductivity and diffusion.

The LDRD researchers recently reported the results of large-scale molecular dynamics simulations of charged-particle stopping in a classical electron gas. Calculations simulated approximately 10^4 to 10^6 particles, which is orders of magnitude more than have been used in previous studies. The calculations spanned a range of coupling



Wake potential of a fast charged-ion projectile in a dense plasma background.

regimes, from weak to moderately strong, allowing the team to evaluate various stopping-power models currently in use and to determine the ranges over which the models' predictions are valid. The work is also significant because it shows how large-scale molecular dynamics simulations can be used to extend theories of plasma behavior. The enhanced code will

- Simulate strongly coupled, non-ideal, and degenerate plasmas known as warm dense matter
- Address species diffusivity and equation-of-state issues in the regime of warm dense matter
- Provide insight into existing theories of complex plasmas, including mixtures, and motivate developments of new theories and experiments.

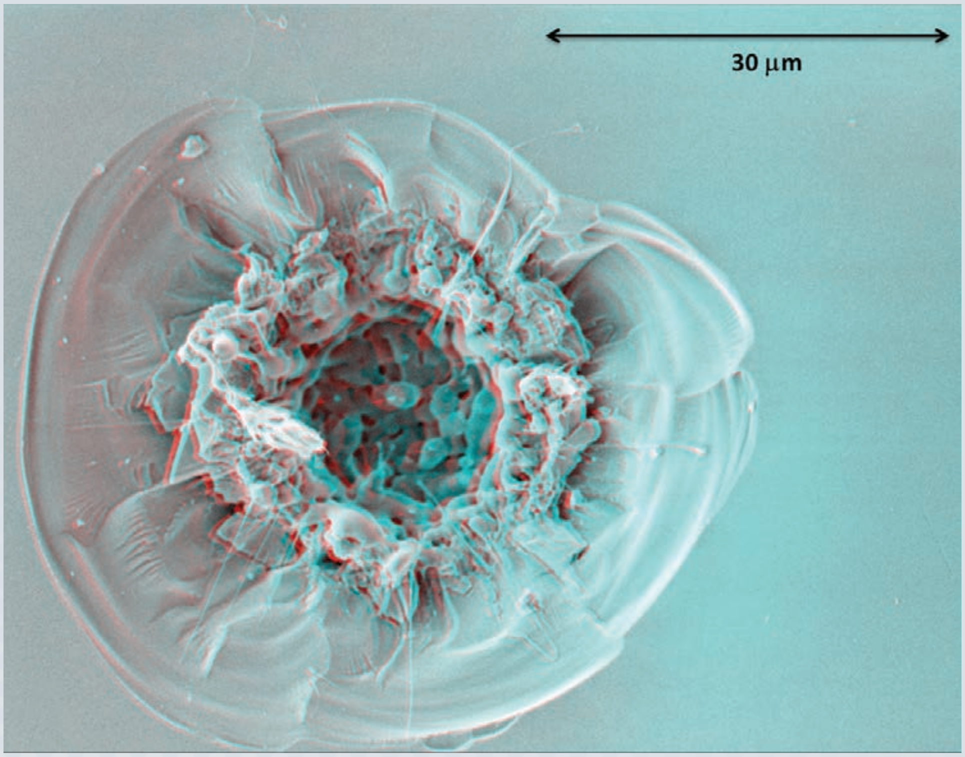
For the coming year, the team will develop practical models and extend the application of thermal and electrical conductivity simulations to inertial-confinement fusion efforts.

Lasers and Optics

Laser optic surface damage is of concern for high-power laser facilities because even damage sites initially much smaller than the diameter of a human hair in size can quickly grow large enough to destroy the optic, resulting in damage to downstream optics as well. In fact, laser-induced optics damage will remain for the foreseeable future a key constraint on the operation of inertial-confinement fusion laser facilities—which are critical for the Laboratory's missions in stockpile stewardship and advanced lasers and applications. The internal features of damage sites are still undiscovered. A recently concluded LDRD project performed a series of experiments to understand the nature of laser-induced damage to laser optics (11-ERD-030). To that end, they

- Prepared damage sites to selectively isolate candidate chemical and mechanical attributes
- Measured energy deposition to locate attributes that drive growth
- Examined atypical sites—those that exhibit growth behavior on the extremes of the distribution
- Identified the most quickly and most slowly growing site types and tested the sensitivity of various diagnostic techniques

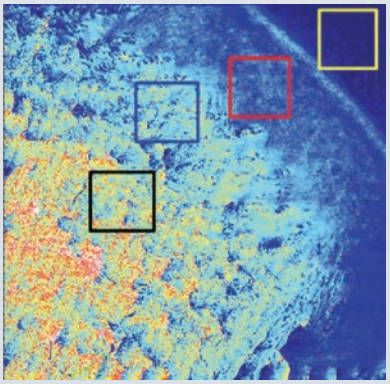
Their findings about the amount of sub-surface fracture surrounding a damage site, the presence of organic compounds, amount of microstructure, integrated photoluminescence, and bulk stress, among others, were used to produce a growth model based on simple fracture mechanics. The model proved to be in good agreement with observed growth rates as a function of laser beam energy on the exit surface of silica.



A three-dimensional image of a damage site on the exit surface of a silica optic.

Materials Behavior Under Extreme Conditions

To pioneer new directions in extreme matter physics, LDRD researchers are undertaking extreme compression science (12-SI-007) at fusion-class laser facilities such as Livermore's National Ignition Facilty; the Linac Coherent Light Source at the SLAC National Accelerator Laboratory in Menlo Park, California; and the OMEGA laser facility at the Laboratory for Laser Energetics in Rochester, New York. For matter at atomic pressures, the principal goals include learning to compress matter hundreds of times greater than experienced at the center of the Earth, along with developing diagnostic tools to map physics from the atomic to thermodynamic levels. Research results could help to



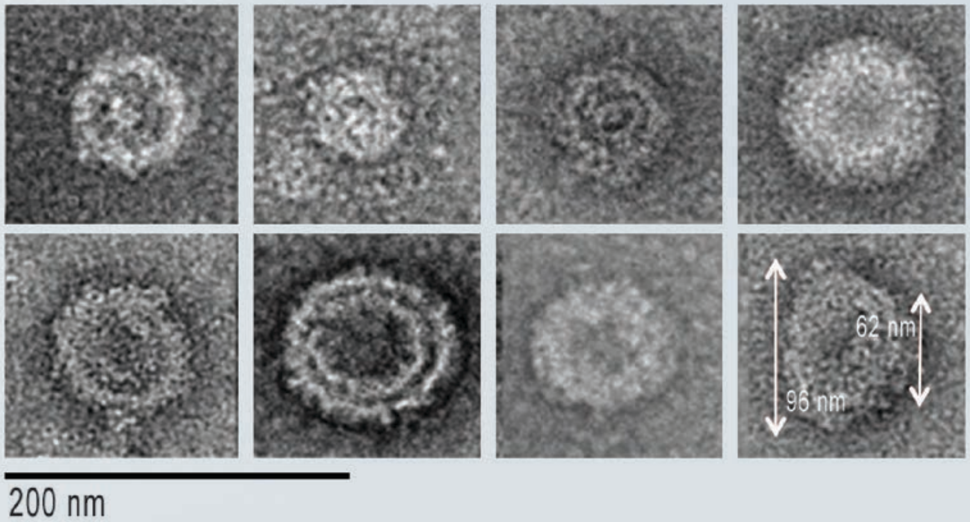
Velocity map of different states of compressed silicon highlighted by yellow, red, and blue squares. Phase transformation is highlighted by the black square.

- Facilitate identification of altogether new forms of matter on Earth
- Enable new insight to the periodic table and lead to the creation of novel materials
- Provide enhanced understanding of dynamical processes in high-density materials and the nonequilibrium of matter at extreme pressures
- Create a laboratory test bed for the wealth of new astronomical observations of planets outside the Solar System
- Determine enhanced routes to inertial fusion energy

Researchers have used a new method to investigate the micron-scale structural evolution of shock-compressed materials and measured the equation of state of compressed iron, and examined the behavior of Earth-related materials such as molybdenum, iron oxide, magnesium, and aluminum under stress. In the project's first two years, results of their materials research has resulted in 15 peer-reviewed articles in prestigious journals such as *Science*, along with a cover story for the *Journal of Applied Physics*. The project is building a large, interdisciplinary community to explore matter at near-atomic pressures, including collaborators at Stanford, Caltech, Princeton, Yale, the French Alternative Energies and Atomic Energy Commission, University of Minnesota, University of Rochester, Los Alamos National Laboratory, and the University of California, Berkeley.

Biosecurity

The nation needs fast countermeasures in case of a biological or chemical terror attack. A recently completed Livermore LDRD project focused on developing a therapeutic system that mitigates the consequences of exposure to biotoxins and chemical warfare agents (11-ERD-012). The team packaged naturally occurring cytochrome proteins into atomic-scale particles to create a drug that enhances the body's ability to metabolize certain agents, converting them into inactive compounds. This intravenously administered drug—made from a large and diverse group of enzymes that catalyze oxidation of organic substances—demonstrated a unique approach



Individual cryo-electron micrographs of nanometer-scale disks containing embedded proteins.

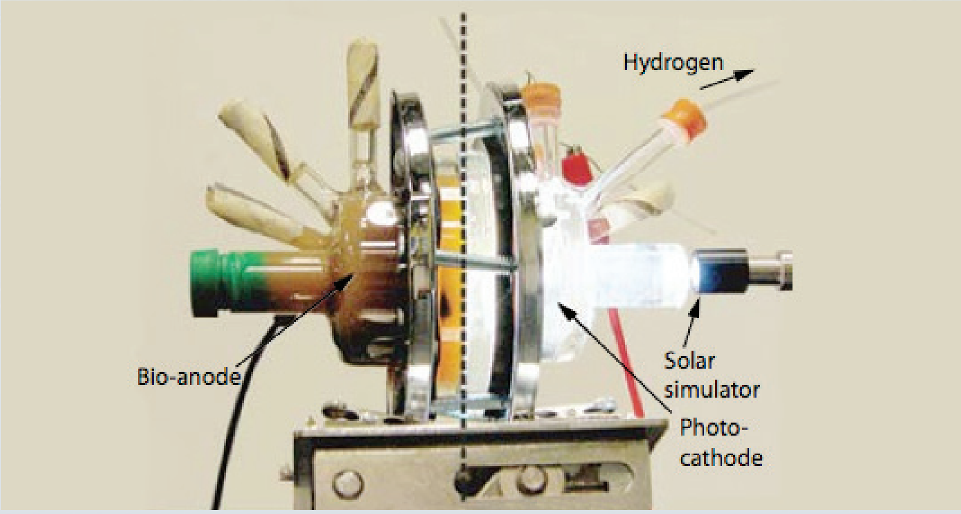
for detoxifying chemical compounds after exposure. The team developed a simple procedure for embedding proteins into fat particles, termed lipoprotein nanodisks. The team then

- Optimized reaction conditions to maintain stability and functionality of the materials in the nanodisks
- Demonstrated the nanodisks' ability to metabolize a chemical agent in rat cells
- Assessed the stability and activity of the fat and protein nanodisks in rat plasma

The nanodisks proved to be stable under physiological conditions, and maintained their functionality and stability for a full eight hours, which is sufficient time to metabolize and detoxify toxic chemicals or drugs. Because the method's effectiveness lies in bolstering metabolizing enzymes, this approach may also provide a means for responding to such scenarios as treating overdoses of prescription, over-the-counter, or illegal drugs. The team's results have generated external interest in this approach for detoxification.

Energy and Climate

Rising energy demands and the imperative to reduce carbon dioxide emissions are driving research on biofuels development. Biologically derived hydrogen is one of the most promising of these fuels and is seen as a future energy source. Its development will help to secure the nation's energy through enhanced domestic sources. The goal of an LDRD project to develop an economically viable biological hydrogen-production system (11-LW-019) was to to implement a simple and relatively straightforward strategy for hydrogen production by photosynthetic microorganisms using sunlight, sulfur- or iron-based inorganic substrates, and carbon dioxide as the feedstock. Carefully selected microorganisms with bioengineered beneficial traits would act as the



Experimental setup for hydrogen production by photosynthetic microorganisms using sunlight, sulfur- or iron-based organic substrates, and carbon dioxide as the feedstock.

biocatalysts for the process and were designed to both enhance the system efficiency of carbon dioxide removal and the net hydrogen production rate. The project team successfully demonstrated for the first time that hydrogen could be produced from a photosynthetic microbe, a method that may serve as a possible alternative to biomass-based hydrogen production. During FY13, the team

- Tested the relative importance of the three enzymes that use atmospheric nitrogen for hydrogen production in the photosynthetic bacterium *Rhodopseudomonas palustris*
- Developed a genome-scale model of metabolism of the microbe that accounts for differences in cellular biomass under various environmental conditions
- Used this model to assess the robustness of the microbe's metabolism against various genetic and environmental changes

The bacterium grew using sunlight, an inorganic substrate as a source of electrons, and carbon dioxide as the sole carbon source to produce hydrogen. Using carbon dioxide in this manner serves to "fix" it, or remove it from the atmosphere.

Cyber, Space, and Intelligence

Currently, the U.S. does not have the operational capability to provide worldwide, on-demand, medium- to high-resolution space-based imaging to the military and intelligence community. Tiny milk-box-sized nanosatellites, when placed in orbit, can act as space-based sensors for intelligence, surveillance, and reconnaissance.

The research team for a Livermore LDRD project developed a space surveillance system called STARE (space-based telescope for the actionable refinement of ephemeris), whose imaging devices have been integrated into Pathfinder satellites (13-ERD-059). After passing pre-flight qualifications, one was launched in late

2013 and another is scheduled for 2014. The team developed two new imaging technologies for three imaging payloads. Each one improved on the previous design.

- The first uses an improved Cassegrain reflector design, a type typically used in optical telescopes, as an imager
- The second incorporates temperature stabilization with the imaging system
- The third uses an altogether newly designed and patented monolithic optic design for imaging that significantly improves both optical quality and robustness

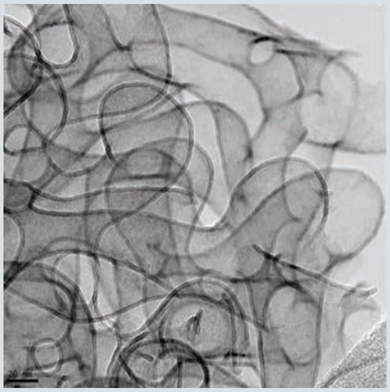
Miniature Pathfinder satellite with imaging payload.

After alignment and testing, each payload was delivered to the Pathfinder payload integrator. The team used the Cassegrain-design imager on the ground to examine and image satellites in orbit and refine their orbits. Scheduling and processing infrastructure were used to capture orbiting objects visible from a test site at Lawrence Livermore. The team validated the refinement approach successfully from the ground, with an on-orbit validation performed after launch. The design of the new monolithic optic design has already been modified and has been transferred to industry partners.

Engineered Materials

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 needed for targets to be used in the Laboratory's condensed-matter, high-energy-density physics program.

An LDRD team is using atomic-layer deposition for rapid, on-demand development of nanometer-scale (a sheet of paper is about 100,000 nanometers thick) porous bulk materials with properties tailored to specific needs (13-LW-031). Atomic-layer deposition is the technique of choice because of a recent discovery at Livermore that nanometer-thick surface coatings created in this manner drastically improve the mechanical and thermal stability as well as the catalytic activity of nanoporous gold.



Nanometer-scale tubular freestanding titanium oxide obtained by coating a gold template using atomic-layer deposition and then removing the gold core by a wet etch process.

Work to date has focused on the fabrication, characterization, and testing of various materials, including a titania-coated nanoporous gold as a lithium-ion battery electrode material. In the coming year, the team will

- Continue fabrication and characterization of atomic-layer-deposition functional bulk nanoporous gold
- Test materials for catalytic, solar energy harvesting (water splitting), as well as energy storage applications

- Continue exploring the fabrication of ultralow-density nanoscale tubular bulk materials

- Improve the etch parameters for polystyrene beads, block copolymers, and engineered micro-trusses that could serve as alternatives to nanoporous gold as a template

The goal of this LDRD project is to establish atomic-layer deposition—whether on nanoporous gold, polystyrene beads, or other template—as a general tool for the creation of engineered materials.

High-Performance Computing

Recent trends in the architecture of computer central processing units indicate that future processors will have a greatly reduced amount of memory available to each unit (or core) relative to today's architectures. The drastic reduction in memory per core is because of the high cost of dynamic random-access memory in both power and in dollars. The looming problem of memory bandwidth and capacity will affect high-performance computer applications on exascale supercomputers, which will be capable of a quintillion floating point operations per second. Data-centric applications—that is, the "big data" projects so prevalent today—are affected much more by memory latency, bandwidth, and capacity limitations than traditional high-performance computing applications. A Livermore team is designing, developing a prototype, and evaluating a data-centric node architecture that seamlessly combines dynamic and nonvolatile random-access memory (13-ERD-025). It also includes an active storage controller designed to run data-intensive kernels accessing nonvolatile random-access memory. In FY13 the team

- Engaged with vendors concerning the integration of logic and compute nodes with three-dimensional memory stacks

- Began the design of a data-intensive memory architecture, which achieved a 25% decrease in required memory bandwidth for a mesh simulation benchmark

- Demonstrated performance improvement of almost an order of magnitude for a storage application in an emulated storage controller

- Designed a new algorithm to optimize a high-performance data-intensive memory map device driver

- Implemented a two-level index for a bioinformatics database, which is a prime example of the kinds of big-data efforts this project is addressing.

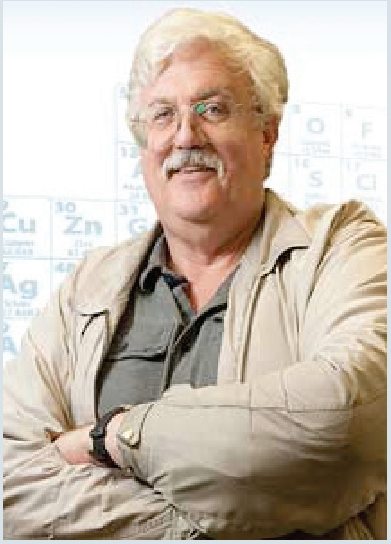
This research addresses a critical mission need for data-centric computing and benefits data science applications for both informatics and simulation data analysis.

Awards and Recognition

A primary goal of LDRD 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 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.

American Association for the Advancement of Science Fellow

Radiochemist and LDRD researcher Ken Moody has been awarded the distinction of fellow of the American Association for the Advancement of Science. Election as a fellow is an honor bestowed upon members by their peers to recognize distinguished efforts to advance science or its applications. He was one of the 40 fellows elected in the chemistry category.



Ken Moody

Moody, a 27-year Laboratory employee, has been a critical member of the team that discovered six new elements—113 through 118—among many other achievements. In 2009, he earned The American Chemical Society Division of Nuclear Chemistry and Technology's Glenn T. Seaborg Award for his work in heavy elements and nuclear forensics. In addition, he was awarded the first prize of the Joint Institute for Nuclear Research, Dubna; an R&D 100 award for "The Gamma Watermark" technology; *Popular Science's* Best of What's New Award for element 114; the Glenn T. Seaborg Award for Nuclear Chemistry of the American Chemical Society for discovering 5 elements and more than 30 isotopes and developing nuclear forensics; and the 2010 Gordon Battelle Prize for Scientific Discovery for the discovery of element 117 along with collaborators from Oak Ridge National Laboratory.

In 2012, Moody was named a Lawrence Livermore Distinguished Member of Technical Staff for his extraordinary scientific and technical contributions to the Laboratory and its missions as acknowledged by his professional peers and the larger community.

Moody is one of the creators of the discipline of nuclear forensics and applications of radiochemistry to national security and law-enforcement problems. He has served as a principal and co-investigator for a number of LDRD projects dating back to 1989 with a project to search for double-beta decay in the actinides (89-DE-013), and is currently a member of a team performing radiochemical measurements of nuclear reactions at the National Ignition Facility (13-ERD-036).

The 2013 American Physical Society Fellows from Lawrence Livermore. Charles Cerjan, Ian Thompson, Eric Schwegler, and Marilyn Schneider are shown left to right. John Moody and Pravesh Patel are shown below.

American Physical Society Fellows

All six fellows of the American Physical Society named from LLNL in 2013 were LDRD-supported researchers and represent the largest number from any DOE laboratory this year. Election is limited to no more than half of one percent of the association's membership for a given year.

John Moody

Pravesh Patel

- **Charles Cerjan** was cited was cited for seminal contributions to time-dependent Schrodinger equation propagation algorithms and their applications, the development of laser-produced plasma sources for advanced lithography, and the investigation of the basic mechanism of magnetic multilayer material response and its application to magnetic storage devices. Cerjan has been a principal or co-investigator for a number of LDRD projects dating back to 1985, including a theoretical investigation of intense field interactions with atoms (85-LW-018), target fabrication science and technology (05-SI-005), nuclear astrophysics at the National Ignition Facility for studying reactions of stars on Earth (08-ERD-066), and currently, advanced inertial fusion target designs and experiments for transformative energy applications (11-SI-002).

- **John Moody** was cited in the plasma physics category for pioneering experiments contributing to understanding propagation, scattering, transmission and redirection of high-intensity laser beams in large-scale plasmas for inertial-confinement fusion. He is currently a co-investigator for research into a compact, femtosecond hard x-ray source for materials characterization and high-energy-density science (13-LW-076).

- **Pravesh Patel** was cited in the plasma physics category for pioneering contributions in the science of ultra-intense laser interaction with matter, and particle acceleration and applications to creating and probing high-energy-density plasma states, as well as for his leadership in advancing the fast-ignition concept for inertial-confinement fusion. He has served as both principal and co-investigator on numerous high-energy-density science projects including reaching isochoric states of matter by ultrashort-pulse proton heating (02-ERD-

006), creation of a neutron star atmosphere (04-ERD-028), opacity of the solar interior (05-ERD-045), and currently as a co-investigator for advanced inertial fusion target designs and experiments for transformative energy applications (11-SI-002).

- **Eric Schwegler** was named by the Division of Computational Physics for his important contributions to the development of linear-scaling electronic structure theory and the use of first-principles methods to examine the properties of aqueous solutions, nanomaterials, and matter under extreme conditions. He has been a co-investigator and principal researcher for numerous LDRD projects dating back to 2001, including applied biological simulations (01-SI-012), nonequilibrium phase transitions (04-ERD-108), laser-induced surface damage in optical materials (06-ERD-035), the properties of confined water and fluid flow at the nanometer-scale (06-ERD-039), the physics and chemistry of the interiors of large planets (09-SI-005), and mix at the atomic scale (10-ERD-004). He is currently a co-investigator for a project investigating the equation of state of polymers under extreme conditions with quantum accuracy (12-ERD-052).

- **Marilyn Schneider** was recommended for fellowship by the American Physical Society Topical Group on Instrumentation and Measurement Science for her outstanding contributions to x-ray measurements from laser-produced plasmas. She has served as a principal investigator for an LDRD project examining the development of tunable radiation sources for material science studies and simulation of radiation transport in dense astrophysical plasmas (05-ERD-068), as well as high-temperature thermal X-radiation sources at short-pulse lasers (08-ERD-024).

- **Ian Thompson** was selected by the Division of Nuclear Physics for his development and application of all-order treatments of nuclear-cluster dynamics in peripheral reactions, leading to a new understanding of halo nuclei within a few-body framework. He is currently a co-investigator for research into nuclear plasma physics (11-ERD-069).

Presidential Early Career Award for Science and Engineering

Miguel Morales-Silva has been named for a 2013 Presidential Early Career Award for Science and Engineering for his leading-edge research in condensed matter physics. Using advanced computational techniques such as density functional theory and quantum Monte Carlo, Morales-Silva studies materials at extreme pressure and temperature on some of the world's most powerful supercomputers. His work is important to stockpile stewardship to ensure the safety, security, and reliability of the nation's nuclear deterrent without underground testing. In addition, this research also provides scientists with a

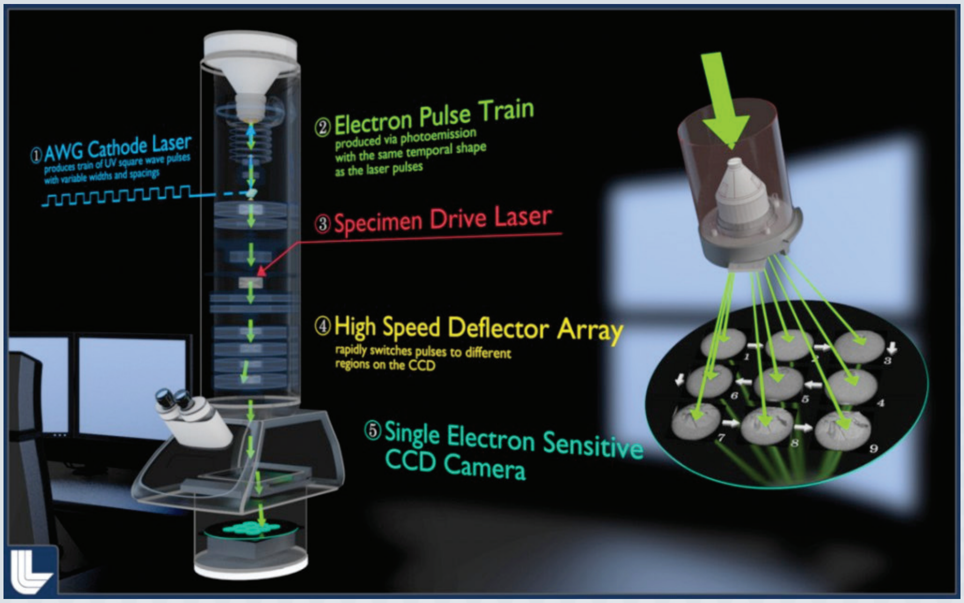
Miguel Morales-Silva

better understanding of planet formation. The early career presidential awards are the highest honor bestowed by the United States government on science and engineering professionals in the early stages of their independent research careers. Established by President Clinton in 1996, the awards are coordinated by the Office of Science and Technology Policy within the Executive Office of the President. Award winners are selected for their pursuit of innovative science and technology and their commitment to community service as demonstrated through scientific leadership, public education, or community outreach. Morales-Silva is currently the lead investigator for an LDRD project on the search for metallic hydrogen (13-LW-004), as well as a co-investigator for an effort to benchmark calculations of the fundamental electronic properties of selected materials across the periodic table using quantum Monte Carlo calculations, which rely on repeated random sampling to compute results (13-ERD-067).

R&D 100 Awards

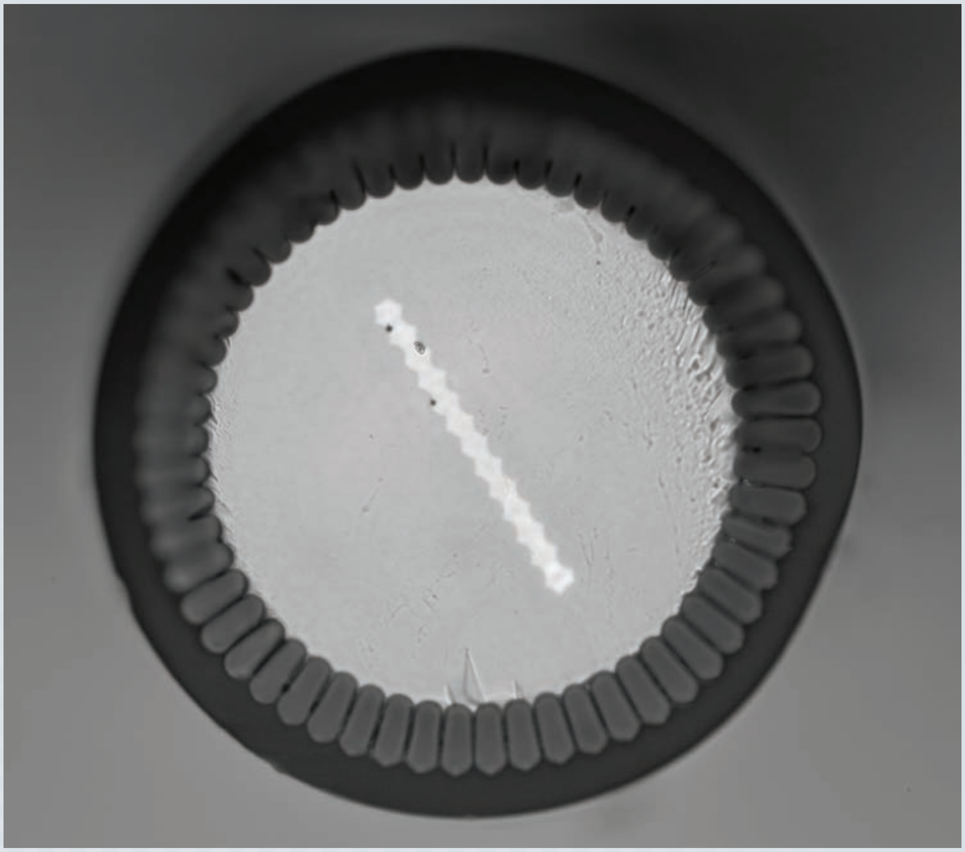
In 2013, LDRD-supported technologies received two of five awards presented to the Laboratory in the R&D 100 Awards competition, which selects the world's top 100 innovations with commercial potential.

- The **Movie Mode Dynamic Transmission Electron Microscope** captures billionth-of-a-meter-scale images at frame rates more than 100,000 times faster than those of conventional techniques. This revolutionary imaging technique—an extension of its R&D 100 Award-winning predecessor—enables the capture of super-fast processes as they occur, including microstructural changes, phase transformations, and chemical reactions in nanometer-scale structured materials and, potentially, biological organisms. The microscope could enable



Movie Mode Dynamic Transmission Electron Microscope direct observation of molecular interactions, such as protein-to-protein binding and host interactions with pathogens. Industrial partner Integrated Dynamic Electron Solutions expects to begin taking orders for technology in the next several months. As shown in the simplified schematic, the device has a waveform generation laser system that delivers shaped laser pulses and a low-aberration, high-speed deflector array that is precisely synchronized with the laser to deflect each pulse to a different area on the camera. Development of this instrument was initially supported by an LDRD project on time-resolved transitions via dynamic transmission electron microscopy (06-ERD-007).

- A technology named **Efficient Mode Converters for High-Power Fiber Amplifiers** overcomes a key limitation of high-power fiber lasers by maintaining beam quality as power is increased. The innovators behind this technology replaced the circular core used in conventional fiber lasers with a rectangular design that accommodates high-energy-output fiber amplifiers by spreading the laser power across the rectangular core's larger area mode, so the modes can be amplified without damaging the fiber. This technology will increase the power of high-quality fiber lasers from several to potentially 100 kW, for applications in defense and even solid-state lasers for machining, among others. An LDRD project investigating the science and technology of unconventional fiber waveguides for emerging laser missions (10-SI-006) provided support that enabled this award-winning mode converter.



Cross section of an ytterbium-doped optical ribbon fiber, less than a hundredth of an inch wide, fabricated at the Livermore fiber-draw tower. The lighted rectangular ribbon core is seen in the center. This fiber successfully demonstrated greater than two-hundredfold amplification of a single higher-order laser mode without distortion.

Federal Laboratory Consortium's Far West Region Competition

Lawrence Livermore garnered four awards in the Far West Regional competition in the latest Federal Laboratory Consortium awards for excellence in technology transfer in the fall of 2013. One Outstanding Commercialization Success Award went to "Digital Polymerase Chain Reaction (PCR)," a rapid and highly sensitive method for detecting genetic material in individual droplets. The award was presented to LLNL and the startup QuantaLife (and its successor company) from Pleasanton, California, for commercializing an LLNL digital polymerase chain reaction technology as the Droplet Digital Polymerase Chain Reaction System, known as the most accurate genetic analysis platform currently available. The technology had origins in an LDRD project under principal investigator Chris Bailey, the viral discovery platform (08-SI-002), in which existing and new laboratory developments were integrated into a comprehensive approach for the rapid identification and characterization of viruses in clinical samples.

LLNL was also awarded an Outstanding Partnership Award for "Rapid Viability Polymerase Chain Reaction," a rapid method for detecting viable anthrax-causing spores and developed in collaboration with the U.S. Environmental Protection Agency. The technology had origins in an LDRD project under principal investigator Staci Kane on viability-based detection methods for pathogens in complex environmental samples (08-ERD-025).

Edward Teller Award

Jim Hammer

On August 20, 2013, the American Nuclear Society announced the recognition of Livermore's Jim Hammer as one of two recipients of the 2013 Edward Teller Award. Hammer, who is also one of the Laboratory's Distinguished Member of Technical Staff, is co-inventor of the fast-ignition approach to nuclear fusion energy. The Edward Teller Award recognizes pioneering research and leadership in the use of laser and ion-particle beams to produce unique, high-temperature and high-density matter for scientific research and for controlled thermonuclear fusion. Through its honors and awards program, the American Nuclear Society recognizes the exceptional accomplishments of nuclear science and technology professionals. Hammer has served as principal and co-investigator as well as a reviewer for numerous LDRD projects. His latest role is as a co-investigator for work to generate and characterize matter at extreme gigabar pressures at Livermore's National Ignition Facility (13-ERD-073).

New Mineral Named in Honor of LDRD Researcher

A new garnet mineral ($Ca_3Ti_2SiAl_2O_{12}$), recently discovered in a refractory inclusion in the Allende meteorite, has been named "hutcheonite" in honor of LLNL's Ian Hutcheon, who has made numerous contributions to the study of meteorites and what they can tell us about the evolution of the early solar system. The name was

Cosmochemist Ian Hutcheon holds a piece of the meteorite Allende, which contains some of the oldest objects in the solar system. A new mineral, hutcheonite, is named in honor of Hutcheon.

chosen by the mineral's discoverers, Sasha Krot (University of Hawaii) and Chi Ma (Caltech). Both the structure and name of the mineral were recently officially approved by the International Mineralogical Association. Hutcheon was the principal LDRD investigator for a 1996 project on subduction and mantle recycling (96-ERI-005), and has subsequently served as principal and co-investigator for projects such as the study of Martian carbonates (98-ERD-042), absolute chronometers and correlated isotopic anomalies in meteorites (01-ERI-004), cosmochemical forensics (07-ERI-005), probing the organization of the cell membrane (08-LW-015), a nuclear forensics integrated approach for rapid response (10-SI-016), and currently, accelerator and secondary-ion mass spectrometry for analysis of coastal carbon flux (11-ERD-066).

NATO Munitions Safety Award for Technical Achievement

Phil Pagoria received the 2013 NATO Munitions Safety Award for Technical Achievement in recognition of his discovery and development of the energetic molecule LLM-105, which has potential applications in enhancing the safety of nuclear and conventional weapons. He was nominated for the award by the U.S. Army and the Office of the Secretary of Defense for Acquisition, Technology, and Logistics. He has worked in the energetic materials synthesis area at LLNL for the past 27 years and currently serves as its Explosives Safety Committee chairperson. Pagoria's work over the years was supported by the LDRD Program through multiple projects dating back to 1991. Currently, he is the principal investigator for a project examining new energetic materials (12-ERD-066). The award is given annually to an individual or team to acknowledge significant advances in munitions safety technology and is supported by NATO's 13 member states.

Phil Pagoria (right) receives the 2013 NATO Munition Safety Award for Technical Achievement from Technical Specialist Officer Manfred Becker.

Susana Reyes

American Nuclear Society's Mary Jane's Oestmann Professional Women's Achievement Award

This American Nuclear Society award recognizes outstanding personal dedication and technical achievement by a woman in the fields of nuclear science, engineering, research, or education and is presented at the association's winter meeting. In 2012, LDRD researcher Susana Reyes, was named the 20th award winner, recognizing her leadership in developing detailed hazard and safety analyses for both inertial and magnetic fusion facilities, including present and future power reactors. Reyes is currently the principal investigator for a next-generation process for tritium recovery from fusion power plant blankets (13-ERD-056).

Best HPC Collaboration Between Government and Industry

An LDRD project on applying high-performance computing and simulation to future energy challenges (12-ERD-074) was part of an *HPCwire* 2013 editor's choice award to LLNL for best government and industry collaboration, which honored Livermore and IBM on the Vulcan supercomputer's energy incubator program. The program helps to illustrate the benefits of supercomputing to private industry through external application of supercomputer technologies and expertise to energy applications.

Director's S&T Award

The Director's S&T Award honors Lawrence Livermore researchers whose scientific or engineering advances have had a significant impact on the Laboratory's mission and its sponsors. Two of the four award-winning projects had LDRD support.

- **"Development of Revolutionary New Plastic Scintillators"** is an outgrowth of an LDRD project headed by Natalia Zaitseva on salicylic acid derivatives as a new class of scintillators for high-energy neutron detection (07-ERD-045). The team produced novel scintillator materials, published their experimental efforts, and licensed the technology to a firm that is now selling the materials for national security applications. This technology makes it possible to produce very large and inexpensive upgrades to detection systems such as those used by the U.S. Customs and Border Protection and other governmental agencies, and has opened a new research era for radiation detection.

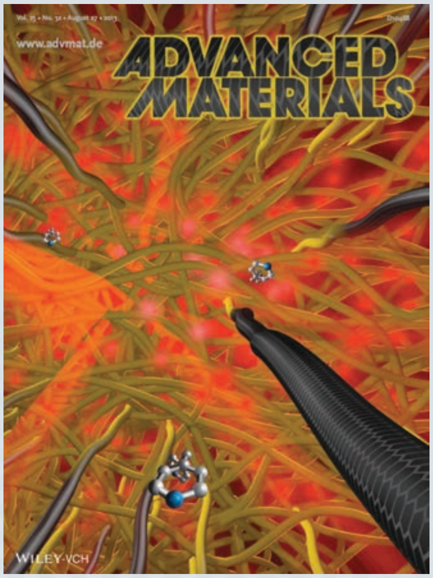
- The award for **"Development of High-Density Carbon Capsules"** had origins in an LDRD project led by Peter Amendt on target fabrication science and technology (05-SI-005). The award recognized successfully developed targets with a diamond outer layer that have recently led to much progress in inertial-confinement fusion, including a record yield of 1.8 quadrillion neutrons, for an experiment in June 2013. The team developed the material science necessary to fabricate diamond targets with controlled grain-sized, texture, density, and doping.

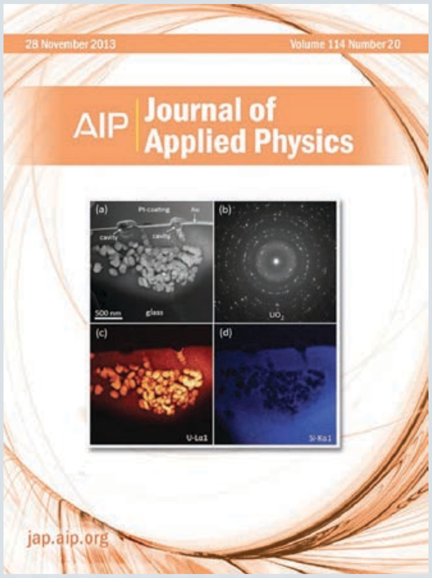
Journal Covers

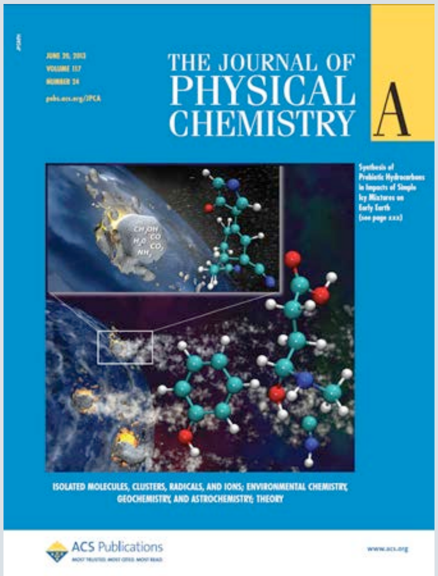
In a paper featured on the cover of *Advanced Materials*, LDRD researcher Tiziana Bond and colleagues at Switzerland's Institute of Energy Technology describe an innovative sensor based on surface-enhanced Raman spectroscopy with which the team was able to detect an organic species in a concentration of a few hundred femtomoles per liter. Raman spectroscopy works by illuminating molecules with a single frequency of light—usually from a laser—then using the frequency pattern of the resulting scattered light as a fingerprint to identify the subject substance. Work was supported by an LDRD project for a sub-wavelegth plasmon laser (12-ERD-065).

A November 2013 cover of the *Journal of Applied Physics* featured work developed in an LDRD project headed by Ian Hutcheon on an integrated approach for rapid response using nuclear forensics (10-SI-016). The team reported on their study of materials subjected to a laboratory-generated plasma that simulates the conditions that produce nuclear fallout. This material represents crucial evidence in post-detonation nuclear forensics to determine the type of device detonated and the source of its nuclear material, for instance.

A cover article in the *Journal of Physical Chemistry A* on the synthesis of prebiotic hydrocarbons in impacts of simple icy mixtures on early Earth has origins in an LDRD project on the equation of state of polymers under extreme conditions with quantum accuracy (12-ERD-052), with Nir Goldman as principal investigator. The possibility exists that both prebiotic raw materials and energy may have been simultaneously delivered to the Earth by a cometary impact. Cometary ices are predominantly water, but contain other constituents important to prebiotic aqueous chemistry, such as ammonia and methanol, and an impact can provide an abundant supply of energy to drive chemical reactions to create the organic compounds. Complete knowledge of the chemical properties of prebiotic mixtures under extreme thermodynamic conditions is needed to understand the role of impact events in the formation of life-building compounds both on early Earth and on other planets that may have played a role in the origin of life.







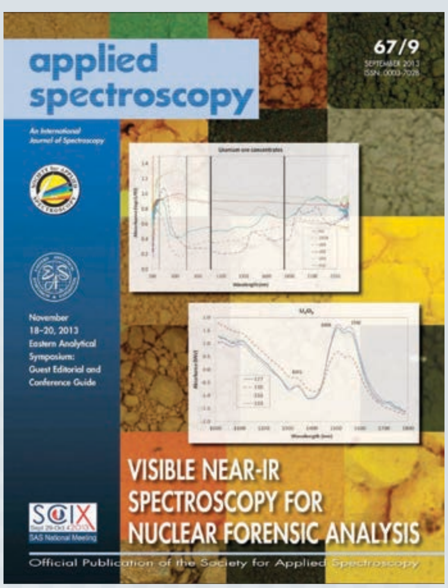
In a cover article for the September 2013 issue of *Applied Spectroscopy,* the application of visible and near-infrared reflectance spectroscopy to nuclear forensic analysis and attribution involving uranium ore concentrates is reported. The application of near-infrared spectroscopy as a noncontact, nondestructive method to rapidly analyze concentrates for species or process information is part of an LDRD research effort on an integrated approach for rapid response with nuclear forensics (10-SI-016) led by Ian Hutcheon.

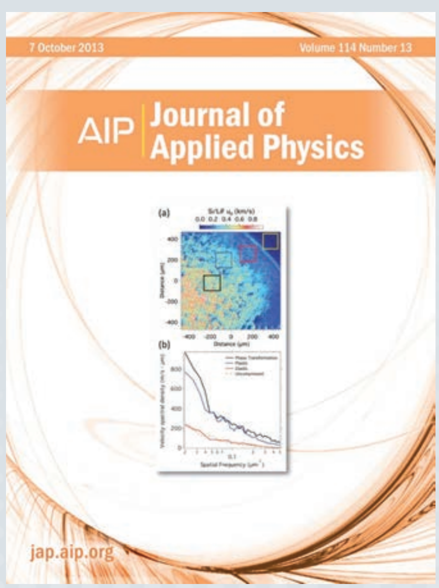
The cover from the October 2013 issue of the *Journal of Applied Physics* featured an article based on an LDRD project headed by Jon Eggert in extreme compression science (12-SI-007), in which researchers used a new method to investigate micrometer-scale structural evolution of shock-compressed materials. Understanding the nature and dynamics of heterogeneous flow in materials subjected to shock loading is important for many fields, ranging from high-speed collisions to inertial-confinement fusion.

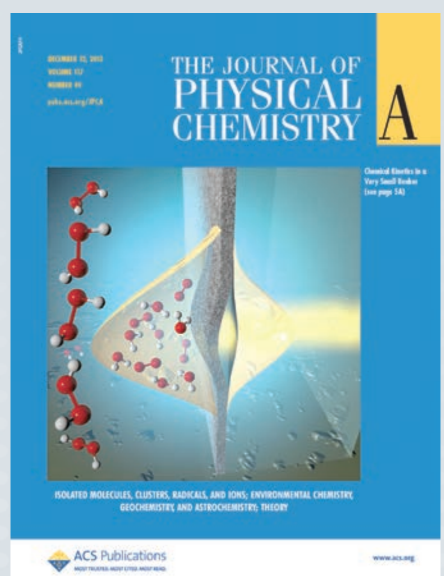
Work from an LDRD project on the detonation performance of improvised explosives via reactive flow simulations and diamond anvil experiments (11-ERD-067) headed by Sorin Bastea was featured on the cover of the December 2013 issue of the *Journal of Physical Chemistry A*. Project results may enable development of ultrafast methods for assessing sensitivity and perhaps even performance in energetic formulations, and could additionally open new avenues for the shock synthesis of materials.

Gordon Bell Prize Finalist

The Gordon Bell Prize is awarded each year to recognize outstanding achievement in high-performance computing. Principal investigator Pavlos Vranas and his project team performing research into illuminating the dark Universe with the Sequoia supercomputer (13-ERD-023), entered the Gordon Bell prize competition and were







selected as one of the six finalists from around the world. Their entry, "The Origin of Mass," describes how work was done using algorithms and code implementation on Livermore's BlueGene/Q high-performance computing platform to simulate theories that can explain the nature of dark matter and lead to experiments that will detect it. The purpose of the award is to track the progress over time of parallel computing, with particular emphasis on rewarding innovation in applying high-performance computing to applications in science, engineering, and large-scale data analytics.

Best Paper Awards

At the IEEE International Parallel and Distributed Processing Symposium held in Boston in May 2013, LDRD research into advanced algorithm technology for exascale multiphysics simulation (11-ERD-017) resulted in a best-paper award ("Exploring Traditional and Emerging Parallel Programming Models using a Proxy Application") for team members Charles Still, Ian Karlin, Jonathon Cohen, Jeffrey Keasler, and Daniel Laney.

Christos Faloutsos and Tina Eliassi-Rad received a best interdisciplinary paper award from the Association for Computing Machinery at CIKM 2012, their 21st International Conference on Information and Knowledge Management, based on work for an LDRD project on continuous network cartography (13-SI-004), headed by principal investigator Celeste Matarazzo.

The *International Journal of Energetic Materials and Chemical Propulsion* awarded Kyle Sullivan a best-paper award based on work for an LDRD project on scalable high-volume micro-manufacturing techniques for three-dimensional mesoscale components (11-SI-005) headed by researcher Christopher Spadaccini.

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 2013 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 is indispensable to the Department because it enables the laboratories to position themselves to advance our national security mission and respond to our Nation's future research needs."[3]

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[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 14, 2013).

[2] *Managing for High-Quality Science and Engineering at the NNSA National Security Laboratories* <http://www.nap.edu/catalog.php?record_id=13367> (retrieved March 14, 2013).

[3] *Report on Laboratory Directed Research and Development (LDRD) at the DOE National Laboratories, Report to Congress, June 2013* <https://ldrdrpt.doe.gov/PUBLICdocument/congress.pdf> (retrieved March 14, 2013).

At LLNL in 2013, Laboratory Director Parney Albright (through October 2013) and Deputy Director for Science and Technology William Goldstein (who assumes role of Laboratory director in 2014) were responsible for the LDRD Program. Execution of the program was delegated to the director of the LDRD Program Office, William Craig (through August 2013) and Ken Jackson, interim director (through November 2013). Rokaya Al Ayat assumes leadership of the LDRD Program in 2014. The LDRD Program at LLNL is in compliance with DOE Order 413.2B and other relevant DOE orders and guidelines.

Program Structure

Project Categories

The LDRD Program at LLNL consists of three major project categories: Strategic Initiative (SI), Exploratory Research (ER), and Laboratory-Wide (LW) competition. During the year, the LDRD Program also funds a few projects in a fourth category, Feasibility Study/Project Definition (FS).

Strategic Initiative

The SI 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 Lawrence Livermore's *Strategic Investment Roadmap*, 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 capabilities.

Exploratory Research

The ER 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 roadmap. 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 roadmap 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 one of ten research categories established by DOE that is relevant to NNSA and Laboratory missions. The ten categories are:

- Advanced Sensors and Instrumentation
- Biological Sciences
- Chemistry
- Earth and Space Sciences
- Energy Supply and Use
- Engineering and Manufacturing Processes
- Materials Science and Technology
- Mathematics and Computing Sciences
- Nuclear Science and Engineering
- Physics

Strategic Context for the FY13 Portfolio

The FY13 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 strategic roadmap 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 *Strategic Investment Roadmap* was developed by multidisciplinary teams under the guidance of the deputy director for science and technology. This document set the strategic context for the FY13 LDRD competition. As a living document, it is updated periodically to respond to our evolving mission needs. Further strategic context is provided by the *U.S. Department of Energy Strategic Plan, May 2011*[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

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[4] *U.S. Department of Energy Strategic Plan, May 2011* <http://energy.gov/sites/prod/files/DOE_2011-Strategic-Plan_Medium-Resolution_Print-Quality.pdf> (retrieved March 14, 2013).

[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 14, 2013).

America's future. In FY13, the Laboratory's LDRD Program strongly supported DOE strategic themes:

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 Laboratory's *Strategic Investment Roadmap* guides the LDRD portfolio planning process. This strategic plan describes institutional strategic goals and science and technology needs in selected mission focus areas and in critical science, technology, and engineering foundations:

Mission Focus Areas

- Stockpile Stewardship Science
- Nuclear Threat Reduction
- Cyber, Space, and Intelligence
- Biological, Chemical, and Explosives Security
- Climate and Energy Security
- Inertial Fusion Energy
- Advanced Laser Optical Systems and Applications

Science, Technology, and Engineering Foundations

- High-Energy-Density Science
- High-Performance Computing and Simulation
- Materials on Demand
- Measurement Science and Technology
- Nuclear Physics and Radiochemistry
- Radiation Detection
- Energy Manipulation
- Information and Network Systems
- Earth and Environmental Sciences
- Bioscience and Biotechnology
- Optical Materials and Targets

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.

Structure of the FY13 Portfolio

The FY13 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 FY13 the LDRD Program funded 152 projects with a total budget of $83.2M. The distribution of funding among the LDRD project categories is shown in the following pie chart.

Distribution of funding among the LDRD project categories. Total funding for FY13 was $83.2M.

Strategic Initiative

In FY13, the LDRD Program funded 12 SI projects. Although the SI category represented just about 8% of the total number of LDRD projects for FY13, it accounted for over 23% of the budget. The SI projects ranged in funding from $646K to $3M.

Exploratory Research

The LDRD Program funded 115 ER projects for FY13. The largest project category, ERs accounted for over 75% of the number of LDRD projects and over 70% of the budget for the fiscal year. Projects in this year's ER category ranged in budget from $63K to $1.9M.

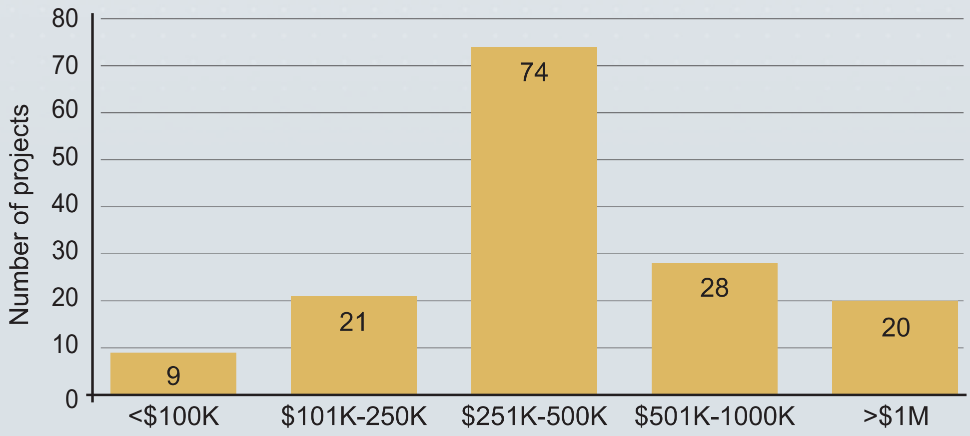
Laboratory-Wide Competition

In FY13, 18 LW projects were funded, which represent almost 12% of the LDRD projects for the year and approximately 5% of the budget. The LW projects for FY13 ranged in funding from $70 to $283K.

Feasibility Study

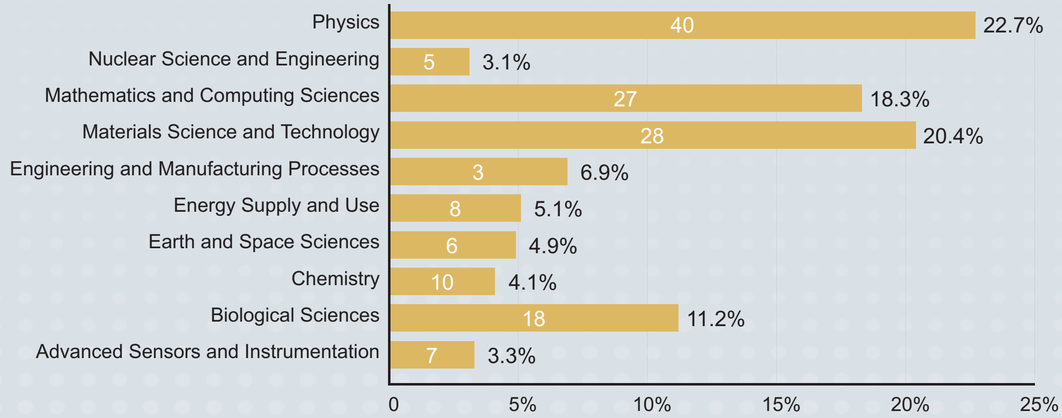
The LDRD Program funded 7 FS projects in FY13, which represent almost 5% of the LDRD projects for the year but less than 1% of the budget. The FS projects for FY13 ranged in funding from $52 to $124K.

The following bar chart shows the funding distribution by dollar amount for the 152 FY13 projects—over 62% of the projects were in the $101 to $500K range, with about 6% falling below $100K. Projects in the $501K to $1M funding range accounted for over 18% of the total, and about 13% of the projects received more than $1M. The average funding level for the 152 projects was about $547K.



Number of projects and levels of funding. The average funding level for an LDRD project in FY13 was about $547K.

Percentage of LDRD funding and number of projects for each research category for FY13 are shown in the following chart. Just over 61% of the total number of LDRD projects fall under the physics, materials science and technology, and mathematics and computing sciences categories, with costs for the three research categories at about 60% of the total LDRD budget.



Percentage of LDRD funding and number of projects in each research category in FY13.

Organization of FY13 Project Summaries

Project summaries for the LDRD FY13 annual report are organized in sections by research category (in alphabetical order). Within each research category, projects appear for the various groups including SI, ER, LW, and 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, 13-ERD-100 means the project began in FY13 and falls in the ER project category. The three-digit number (100) represents the serial number for this proposal.

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 FY13, LDRD costs at LLNL were $83.2M, which is 5.4% 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 FY09 and the percentage of total patents that were derived from LDRD 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 FY13, LDRD projects generated 52% of Livermore's total patents, even though the LDRD program was 5.4% 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	FY09	FY10	FY11	FY12	FY13
All LLNL Patents	46	54	60	78	84
LDRD Patents	20	27	32	35	44
LDRD Patents as % of total	43%	50%	53%	45%	52%

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 FY09 to FY12 account for 42% of the 787 total. In FY13, there were 156 records submitted at Livermore, with 68 (44%) 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.

Records of Invention	FY09	FY10	FY11	FY12	FY13
All LLNL Records	145	160	164	162	156
LDRD Records	56	66	59	79	68
LDRD Records as % of total	39%	41%	36%	49%	44%

Finally, LDRD plays a role in producing Laboratory copyrighted material. From FY09 to FY13, LDRD-supported projects accounted for over 27% of the 322 Livermore copyrights. In FY13, 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 FY13 there were 1,155 such articles, of which at least 293 (25%) resulted from LDRD projects. Over the last several years, the percentage of LDRD-supported articles has remained relatively consistent, with a five-year average of 22% of total Laboratory publications. The following table shows the number of journal articles per fiscal year resulting from LDRD-funded research since FY09, and the percentage of total articles that were derived from LDRD research and development.

Journal Articles	FY09	FY10	FY11	FY12	FY13
All LLNL Articles	1,311	966	994	1,016	1,155
LDRD Articles	225	227	207	230	293
LDRD Articles as % of total	17%	23%	21%	23%	25%

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

Collaborations

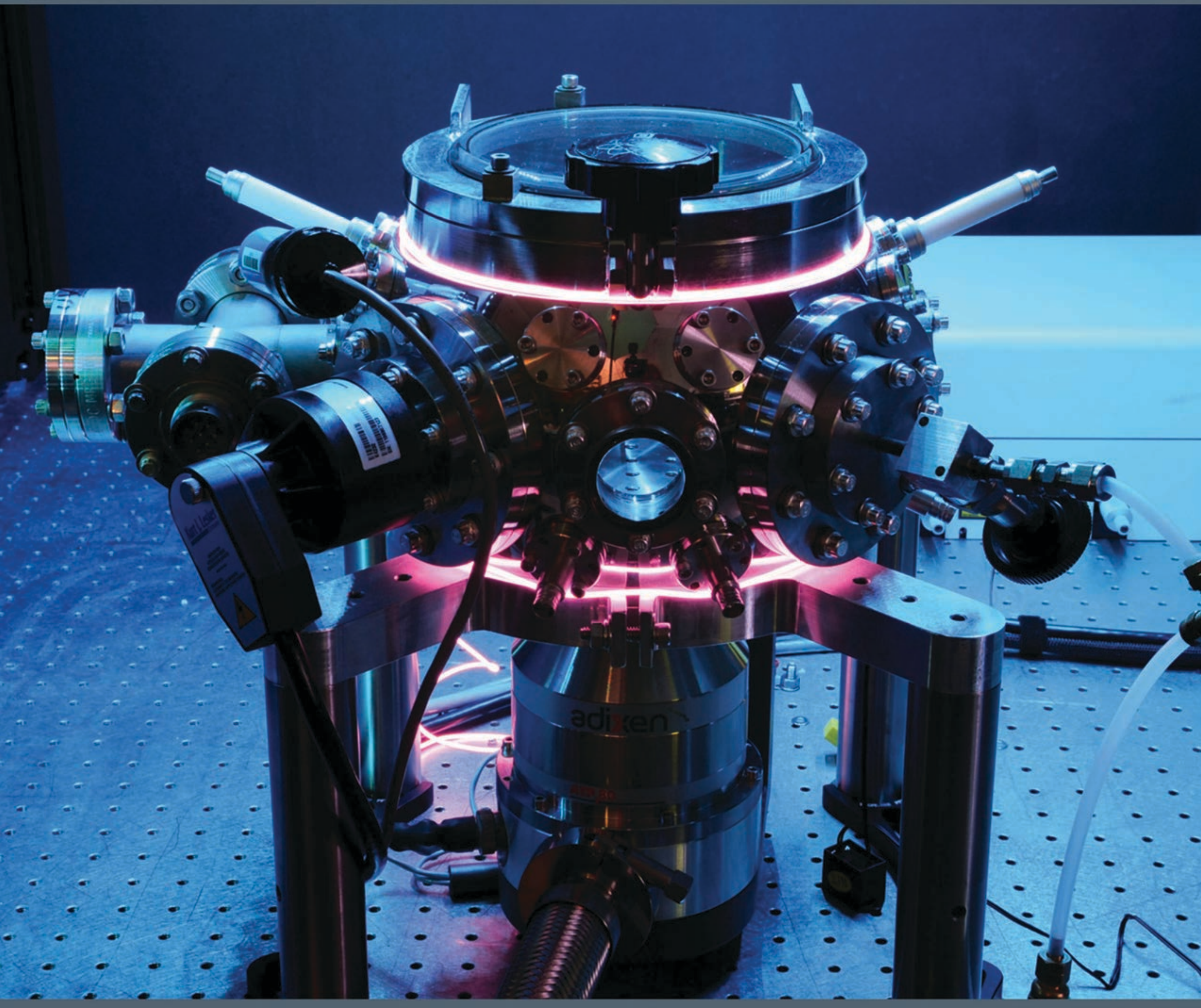
External collaborations are essential to the conduct of research and development in LDRD. By collaborating formally and informally with other national laboratories, academia, and industry, LDRD investigators are able to access world-leading facilities and knowledge—both in the U.S. and abroad—and serve 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 FY13 portfolio included 81 formal LDRD-funded collaborations involving 45 LDRD projects (30% of the total projects funded). Collaborating institutions included the University of California (10% of total collaborators), other academic institutions (65%), and other collaborators such as government agencies and industry (25%). 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 FY13, the LDRD Program supported 51% of the Laboratory postdoctoral researchers—there were a total of 243 postdoctoral researchers at LLNL in FY13, of which 123 were supported in some way by LDRD projects. The Laboratory continues significant recruitment efforts to maintain the total number of postdoctoral researchers.

Advanced Sensors and Instrumentation



Laboratory Directed Research and Development

Annual Report FY'2013

Quantification of Carbon-14 by Optical Spectrometry

Ted Ognibene (11-ERD-044)

Abstract

Although accelerator mass spectrometry has the proven sensitivity to measure carbon-14 dioxide ($^{14}CO_2$) at ultralow levels, the technology's costs, complexity, and throughput limit its usefulness in developing large-scale programs to monitor fossil fuel emissions. We propose to develop and demonstrate the use of tabletop-sized laser-based spectroscopic methods that use coupled rotational–vibrational excitation lines to quantify carbon isotope ratios from atmospheric CO_2 at concentrations as low as one carbon-14 atom per 10^{13} atoms of carbon. We will use a prototype cavity ring-down spectroscopy system to measure $^{14}CO_2$ to $^{12}CO_2$ ratios. The limits of sensitivity, selectivity, precision, measurement throughput, sample-to-sample carryover, dynamic range, and other performance metrics will be defined. Laser-based measurements of carbon-14 content will be directly benchmarked to accelerator mass spectrometry analysis from splits of the same sample.

If successful, we will develop a technique that will supersede all current carbon-14 measurement methods and maintain LLNL as the leader in the biomedical and environmental uses of radiocarbon by enabling a new, transformative carbon-14 measurement science that is applicable to environmental and biomedical research. This technology will be less expensive and simpler to operate and maintain than accelerator mass spectrometry and will be similar in size to current optical spectrometers, allowing for routine radiocarbon-based studies in any laboratory and in unattended use in remote field locations.

Mission Relevance

The proposed technology will enable the regional monitoring of fossil fuel combustion to verify carbon emissions and ensure adherence to emission limits, in support of the Laboratory's mission in enhancing the nation's energy and environmental security.

FY13 Accomplishments and Results

In FY13 we demonstrated the feasibility of our approach to measure $^{14}CO_2$ using cavity ring-down spectroscopy. Specifically, we (1) constructed, because of difficulties in achieving reliable operation of the laser at the preferred wavelength, a prototype system around a laser operating at a nonoptimal wavelength; (2) prepared samples containing 1-mg carbon with carbon-14 to carbon-12 ratio concentrations ranging from 1 to 100 times contemporary atmospheric concentrations; (3) converted samples to CO_2 and injected them into our cavity ring-down spectroscopy system, recorded the spectrum, and determined a detection limit of our prototype system of 20 times the contemporary atmospheric concentration of carbon-14 to carbon-12; (4) measured sample splits using accelerator mass spectrometry for comparison; and (5) determined the required operational parameters for a system that will be capable of measuring $^{14}CO_2$ at levels below contemporary atmospheric levels.

Project Summary

The successful completion of this project demonstrated the proof-of-principle approach in measuring the $^{14}CO_2$ to $^{12}CO_2$ ratio using a laser-based technique called cavity ring-down spectroscopy. We constructed a prototype system and made measurements of samples containing carbon-14 ranging from 1 to 100 times the contemporary atmospheric concentration of carbon-14 to carbon-12 and determined a detection limit of 20 times the contemporary atmospheric concentration. We also performed measurements using stable gas isotopic variations of CO_2 and modeled the cavity ring-down spectroscopy spectrum. From these measurements, we defined the operational parameters necessary for a system capable of measuring $^{14}CO_2$ to $^{12}CO_2$ ratios at concentrations below contemporary atmospheric levels. The National Institutes of Health, specifically the National Resource for Biomedical Accelerator Mass Spectrometry, as well as the LLNL Lawrence Scholar Program, will provide support for our ongoing efforts to construct the next-generation instrument.

Publications and Presentations

Ognibene, T. J., and A. D. McCartt, 2013. Q*uantification of ^{14}C by optical spectrometry*. LLNL-TR-645844.

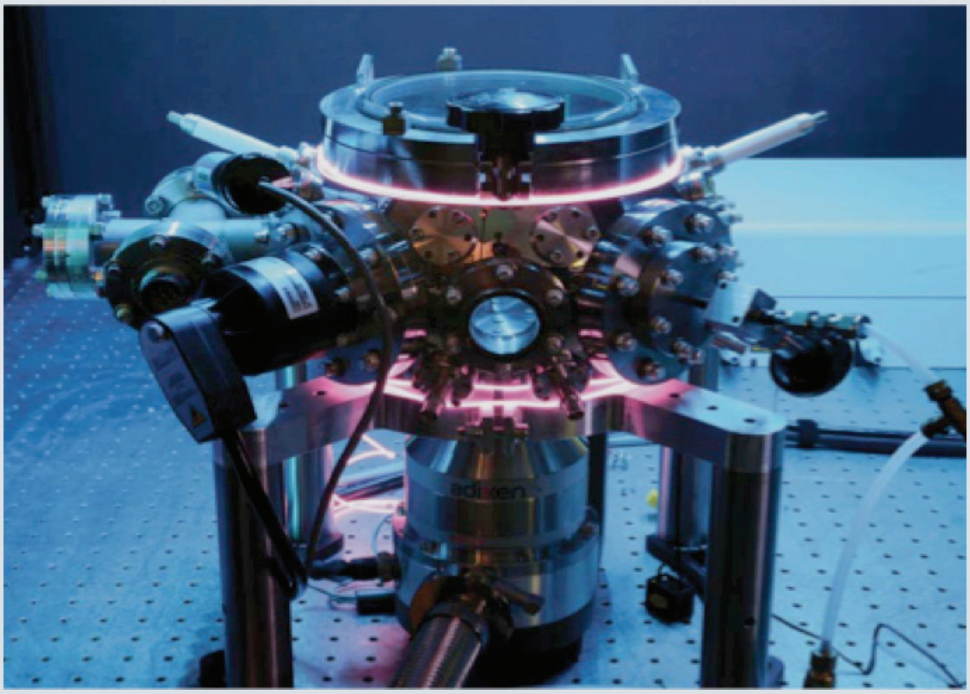
Ultrafast, Sensitive Gamma and Neutron Detectors

Corey Bennett (11-ERD-052)

Abstract

The National Ignition Facility is enabling the weapons physics and high-energy-density scientific communities to investigate previously inaccessible plasma physics regimes, particularly phenomena associated with the deuterium and tritium fusion burn as a function of plasma conditions. Measuring the signatures of gamma rays, neutrons, and charged particles emitted by these burning plasma experiments is exceedingly difficult because of the very short lifetimes and relatively low signal levels. Any credible understanding of the dynamics of nuclear burn will require measurements at a temporal resolution of approximately 1 ps. To meet the challenge of measuring these relatively weak gamma, neutron, proton, and x-ray (in some cases) signals from fusion-class experiments, we propose to develop a very-high-speed detection capability with extremely good particle-measurement quantum efficiencies. This will be done by building on existing LLNL successes in a new class of radiation sensors (“radoptic” sensors), which utilize optical interferometry, and associated high-speed recording technologies.

To create sensitive, high-speed, current-mode gamma and neutron detectors, we will (1) develop techniques for converting gamma and neutron signals to charged-particle signals (usually electrons), (2) develop electron optics techniques for concentrating



Electron optics test bed for development of an ultrafast and sensitive detector of sub-atomic particles and photons emitted in burning plasma experiments.

electron fluences from the converters and directing them to a slightly modified x-ray radoptic detector, and (3) explore alternative sensor materials using a combination of modeling and laboratory experiments to improve radiation sensitivity, temporal response, radoptic efficiency, and optical transparency in candidate sensor materials.

Mission Relevance

This project supports the Laboratory's mission in stockpile stewardship science by developing innovative diagnostics able to view complex, highly energetic dynamic processes in three dimensions and with sub-picosecond temporal resolution.

FY13 Accomplishment and Results

In FY13 we (1) modeled radiation-initiated electron cascades in radoptic detectors both analytically and with Monte Carlo random sampling computational methods; (2) determined that the electron cascade is on the order of 100 fs and is not expected to effect the time response of the detector—the characteristic size of the electron cloud is typically less than 2 µm, enabling high spatial-resolution imaging; and (3) demonstrated an integrated system in which a pulse of 20-keV electrons was detected with a radoptic detector and recorded with less than picosecond resolution utilizing a time-lens recording system.

Project Summary

We have investigated a new class of ultrafast neutron and gamma-ray detection systems based on radoptics in which incident radiation produces a change in the index of refraction of a semiconductor optical cavity and results in the modulation of a reflected optical probe beam. We modeled systems for stopping and converting the incident radiation to an electron cloud in the semiconductor and experimentally demonstrated an integrated radoptic detector and time-lens recorder system in which a pulse of 20-keV electrons incident on one of these devices modulated by an optical probe reflected off the device and was recorded as a single shot with less than picosecond resolution. It's believed that this type of ultrafast neutron and gamma-ray detection will be critical to future stockpile science experiments planned at the National Ignition Facility, specifically those involving fusion burn dynamics.

Publications and Presentations

London, R. A., et al., in press. "Electron cascades in sensors for optical detection of ionizing radiation." *J. Appl. Phys.* LLNL-JRNL-638710.

Vernon, S. P., et al., 2012. "X-ray bang-time and fusion reaction history at picosecond resolution using radoptic detection." *Rev. Sci. Instrum.* **83**(10), 10D307. LLNL-CONF-555292.

Resonantly Detected Photo-Acoustic Raman Spectroscopy as a New Analytical Method and Micro-Volume Probe

Jerry Carter (11-ERD-061)

Abstract

There is a recognized need for non-optical methods to detect gas- and solid-phase materials for multiple national security and scientific applications. We propose to develop a new analytical measurement technique by combining quartz tuning-fork technology and acoustic resonators with optical fiber-based photo-acoustic Raman spectroscopy (PARS) for in situ gas- and solid-phase sample analysis. A piezoelectric crystal quartz in the form of a tuning fork will be utilized as the narrowband microphone for detecting acoustic signals generated by the stimulated Raman of a gas sample in an acoustic cell. Advantages of our system include reduced size, a rugged device for operation in a wide range of environments, high sensitivity with low sample volume, and immunity to environmental noise.

We expect that our research will significantly improve the sensitivity of PARS and provide a viable alternative to traditional optical-signal-based measurement techniques that suffer from a number of limitations for multiple applications. For many uses, we expect our tuning fork and acoustic resonator PARS will have significant benefits in terms of sensitivity, selectivity, noise suppression, and form factor. We also anticipate that this technology combined with optical fibers will enable remote sensing for national security applications such as weapon material lifetime diagnostics for enhanced surveillance or optical lifetime predictions for advanced fusion-class laser systems.

Mission Relevance

This project supports LLNL's national security mission by developing the basis for a new analytical diagnostic technique with applications in stockpile stewardship. Furthermore, this technology has potential applications in combustion and environmental research, homeland security, and biomedicine.

FY13 Accomplishments and Results

In FY13 we (1) modeled the acoustic pulse for 266-nm pulsed excitation in a binary gas (hydrogen with nitrogen) with and without dissipation, finding that in the dissipation case, the acoustic pulse shape was significantly broadened compared to the non-dissipation case; (2) conducted non-resonant, high-temporal-resolution experiments using 266-nm excitation in the same binary gas mixture to study the noise in our PARS measurements; (3) identified radio-frequency noise, noise from gas cell windows, and most importantly, an unknown acoustic signal generated at the laser focus that was clearly not a PARS signal; (4) conducted laser power studies that revealed the acoustic power varied with the laser energy to the fourth power, suggesting a four-photon process; (5) concluded that multiphoton ionization of nitrogen is the major source of noise—laser energy absorbed by the gas will eventually thermalize, generate heat, and produce a change in pressure and thus an acoustic signal; and (6) determined that this background signal—which originated at the laser focus and obscured any underlying PARS signal—was sufficiently large and variable and expected to have overlapping frequency content with our PARS signal and could not be filtered or suppressed.

Project Summary

We have developed the first dynamic Raman scattering and heat deposition model that accounts for acoustic absorption. The system model employed the Raman process to predict the temperature change in the active region, a thermoacoustic radiated pressure field, and the receiver response for an acoustic microphone. We developed a multipass Raman converter for ultraviolet excitation that was successfully operated with hydrogen, producing efficient first-Stokes beam conversion. We were the first to demonstrate high-repetition-rate ultraviolet Raman conversion of hydrogen using a multipass Raman converter and performed the first ultraviolet experiments of hydrogen for resonant and non-resonant PARS. We were unable to validate the PARS model because of a non-PARS acoustic signal generated simultaneously at the laser focus. However, we conducted laser power studies using high-temporal-resolution acoustic measurements and determined the acoustic power varied with the laser energy to the fourth power, suggesting a four-photon process. We have tentatively concluded that multiphoton ionization of nitrogen is a major source of noise, preventing a detectable PARS signal of hydrogen with ultraviolet excitation. Beginning in FY14, LLNL program funding has been awarded for the development of remote Raman gas sensors using optical detection methods. The new project is partly based on ultraviolet excitation, and this LDRD project laid important groundwork by bringing together a multidisciplinary group of scientists with different areas of expertise, including Raman, acoustics, and stimulated Raman, which has significantly increased the breadth of understanding for this technology.

Publications and Presentations

Carter, J. C., et al., 2013. *UV Excited Photoacoustic Raman*. LLNL-TR-646859.

Chambers, D. H., and J. C. Carter, 2012. "Modeling of photoacoustic Raman spectroscopy." *J. Acoust. Soc. Am.* **128**(4), 2412. LLNL-PRES-484444.

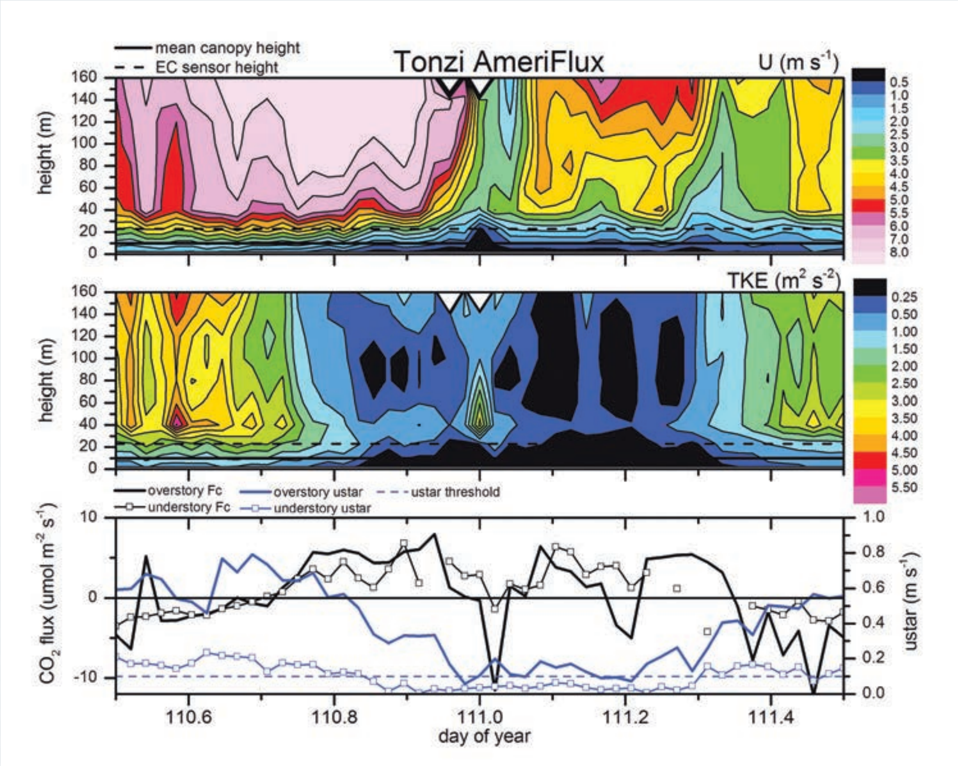
Chambers, D. H., and J. C. Carter, 2011. "Modeling of photoacoustic Raman spectroscopy with dissipation." *J. Acoust. Soc. Am.* **132**(3), 1994. LLNL-PRES-592954.

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



We are using a balloon-borne laser radar system to measure overstory and understory tree canopy wind flows to better understand the significance of cold air drainage and turbulence on the accuracy of eddy covariance flux measurements. Data were collected during spring at the Tonzi AmeriFlux tower in the Sierra foothills in California. These measurements were performed simultaneously with carbon fluxes to identify the atmospheric drivers of anomalous flux measurements. Contour plots of 30-min. mean wind speed and turbulence kinetic energy (TKE) are shown in the top panels, with 30-min. mean carbon dioxide (CO_2) fluxes and friction velocity (ustar) in the bottom panel.

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 to 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 tools 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 roadmap, 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.

FY13 Accomplishments and Results

In FY13 we (1) performed a second set of intensive field campaigns at Tonzi Ranch and the Wind River Field Station, and performed intensive field campaigns at Site 300; (2) continued input forcing in ACASA for Tonzi and Wind River using balloon-borne instrument platform data at the planetary boundary layer and began validating selected output variables (e.g., turbulence kinetic energy, wind speed, and soil CO_2 flux) using soil respiration and boundary-layer observations; (3) began calculating the errors associated with ACASA versus other methods to assess model performance, and began filling gaps in flux data in the FLUXNET network of micrometeorological tower data sets using ACASA; (4) received official AmeriFlux status for the flux tower at Site 300, now called the Diablo flux tower; (5) completed extensive modeling improvements to ACASA for the Tonzi site to include seasonality, understory, and overstory flux partitioning for the leaf area index, and added temporally variable photosynthetic equations; and (6) added the Southern Great Plains Atmospheric

Radiation Measurement site for flux data collection using light detection and ranging remote sensing technology to strengthen collaborations with AmeriFlux, Lawrence Berkeley National Laboratory, and Department of Energy Office of Science.

Proposed Work for FY14

In FY14 we will (1) complete the ACASA simulations, including refining them for Tonzi (its unique water and ecosystem interactions requires additional model adjustments to further improve simulation of CO_2 fluxes), finish validating modeled CO_2 fluxes for Wind River, and run ACASA for Site 300; (2) further analyze unique planetary boundary layer data sets with collaborators at the University of California, Berkeley, and the University of Washington that were originally collected to complement ACASA but are revealing unexpected and significant large-scale flow effects on the flux tower data; (3) submit our 2013 Site 300 flux data to AmeriFlux; and (4) actively collaborate with AmeriFlux and the NEON ecosystem flux network to communicate our results in reducing CO_2 uncertainty.

Publications and Presentations

Osuna, J. L., et al., 2013. *Preliminary results of modeling turbulence kinetic energy at AmeriFlux sites for assessing flux quality.* American Geophysical Union Fall Mtg., San Francisco, CA, Dec. 3, 2012, and North American Carbon Program/AmeriFlux Ann. Mtg., Albuquerque, NM, Feb. 25–28, 2013. LLNL-POST-607033.

Osuna, J. L., S. Wharton, M. Falk, 2013. *A new approach for reducing uncertainty in biospheric CO_2 flux.* North American Carbon Program/AmeriFlux Ann. Mtg., Albuquerque, NM, Feb 25–28, 2013. LLNL-PRES-582996.

Wharton, S., 2013. *LIDAR applications for the carbon flux and wind energy communities.* Invited talk at the University of Oklahoma, Norman, OK, Mar. 11, 2013. LLNL-PRES-627420.

Wharton, S., 2013. *Long-term carbon sink in an old-growth forest: Lessons learned from 15 years of CO_2 fluxes at the Wind River Field Station*. Invited talk at Oregon State University, Corvallis, OR, May 20, 2013. LLNL-PRES-637433.

Wharton, S., and M. Alai, 2012. *LLNL LIDAR Research*. LLNL-PRES-558140.

Wharton, S., et al., 2013. *LLNL hi-resolution evapotranspiration modeling for accurate irrigation management and scheduling tools.* Fresno Agricultural Technology Showcase, Fresno, CA, Aug. 13, 2013. LLNL-PRES-615401.

Wharton, S., et al., 2013. *Atmospheric LIDAR provides insight into land surface-atmosphere exchange at AmeriFlux towers.* American Geophysical Union Fall Mtg., San Francisco, CA, Dec. 3, 2012, and North American Carbon Program/AmeriFlux Ann. Mtg., Albuquerque, NM, Feb. 25–28, 2013. LLNL-POST-605972.

Sub-Wavelength Plasmon Laser

Tiziana Bond (12-ERD-065)

Abstract

We propose to develop a nanometer-scale plasmon laser (a plasmon is a quasi-particle resulting from the quantization of plasma oscillations) that overcomes the diffraction limits of current laser diodes. Sub-wavelength size is achievable because of the strong field confinement of surface plasmon modes in metallic nanowires. The proposed nanoscale vertical laser structure is amenable to high-density, large-area fabrication techniques and requires simpler processing, providing a cost-effective alternative to existing diode lasers. The plasmon waveguide configuration can potentially deliver a two-order-of-magnitude reduction in device size compared to current state-of-the-art vertical-cavity surface-emitting lasers. In addition, there would be a proportional increase in wafer device density, as well as a reduction in laser-array dimension beyond the current state of the art. Generating intense, nanoscale localized optical-frequency fields presents many possibilities for prospective applications in nanoscience and nanotechnology. Example applications include near-field nonlinear-optical probing, high-speed imaging, intense heat harvesting and transfer, ultradense information storage, multichannel parallel-data communication, multiplexed miniature spectroscopy, enhanced metrology, and nanoscale modifications useful in nanolithography techniques.

We expect to develop a nanoscale laser that is significantly smaller than the wavelength of its emitted light, which is possible because of strong field confinement of plasmon modes in metallic nanostructures. We intend to design a plasmon resonant cavity and characterize it with and without gain media. We will also conduct spectroscopic characterization of dyes and/or quantum dots (semiconductors whose electronic characteristics are closely related to the size and shape of the individual crystal). The output versus input power will be measured to determine lasing threshold and efficiency that, along with spontaneous versus stimulated recombination rates, will give a complete picture of device performance.

Mission Relevance

By developing an optical meta-material coating for sub-wavelength optical lasers, we support the Laboratory's strategic mission thrusts in advanced laser optical systems and applications with high-density light sources, and in biosecurity with integrated spectroscopic sources for laboratory-on-a-chip applications. Our proposed nanoscale laser also has possible applications relevant to many other LLNL missions, including solar cells for enhancing energy security, forensic attribution for countering nuclear threats, and stockpile stewardship science with high-explosive multiplexed detection.

FY13 Accomplishments and Results

In FY13 we (1) evaluated the photoluminescent enhancement of quantum dots because they are more stable, suffer less quenching than laser dyes, and offer a sharper separation between ultraviolet absorption and visible emission bands;

(2) designed metallic nanopillar arrays to align (or not) with emission or absorption to separately investigate each effect; (3) optimized pillar fabrication by using electron beam evaporation followed by cryoetching for a straighter pillar with higher aspect ratio; (4) tuned the excitation wavelength from 430 to 530 nm using a super-continuum source filtered with a monochromator, with the structure showing a reflectance resonance around 490 nm—the photoluminescent enhancement showed a strong dependence on the excitation wavelength around the resonant wavelength and the control measurement showed no photoluminescent enhancement; (5) used near-field microscopy to investigate the near-field distribution; and (6) performed near-field microscopy measurements, after designing and fabricating a comb structure to allow the probe to access the field between the pillars, which showed a strong field confinement between the nanopillars.

Project Summary

We proposed novel, deep sub-wavelength nanometer-scale lasers based on metallic plasmonic-assisted nanometer-scale resonator arrays, enabling reach beyond the optical diffraction limit. With such technology we could attain nanolaser arrays on a large scale by leveraging our tunable plasmonic nanocavity platform currently produced on 4-in. wafers. For this project we verified the feasibility of optically pumped nanolaser arrays by demonstrating the enhancement of spontaneous emission of gain material of both dyes and quantum dots embedded in the substrate. Given the ability to define multiple resonances in our metallic nanopillar structures, one main goal has been to align them with both the absorption and emission spectrum of the gain media, to enhance the dipole cross section and decay rates. We have measured both an increase in fast-decay rates of twentyfold (on par with the literature) and fast-decay amplitudes of up to 45%, as recorded at the same wavelengths, indicating that the faster decay is mainly associated with radiative rather than nonradiative channels. The wavelength dependence of the two parameters also clearly indicates the influence of plasmonic resonances. Creating the emission conditions by appropriate loss and gain balance—that is, for radiative decay channels to overcome the nonradiative (lossy) channels—is part of the critical path for demonstrating full lasing.

Publications and Presentations

Bond, T., et al., 2012. *Black plasmon nanoresonators*. 6th Intl. Conf. Quantum, Nano, and Micro Technologies (ICQNM12), Rome, Italy, Aug. 19–24, 2012. LLNL-PRES-557605.

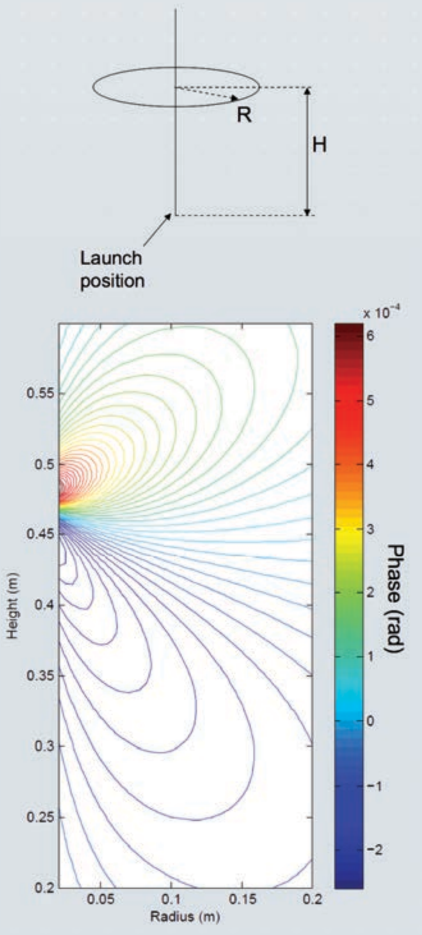
Bond, T., et al., 2012. *"Tunable plasmonic nanogap resonator."* Proc. 20th IEEE Intl. Conf. Network Protocols (ICNP12), Austin, TX, Oct. 30–Nov. 2, 2012. LLNL-PROC-562156.

Bora, M. et al., 2012. *"Photoluminescence enhancement in plasmon resonant cavities."* Proc. 2012 MRS Fall Mtg. and Exhibit, Boston, MA, Nov. 25–30, 2012. LLNL-POST-548131.

Bora, M., et al. 2013. "Plasmon black metals in resonant cavities." *Appl. Phys. Lett.* **102**, 251105. LLNL-JRNL-425128.

High-Precision Test of the Gravitational Inverse-Square Law with an Atom Interferometer

Stephen Libby (12-LW-009)



The modeled phase difference in a cold-atom gradiometer (top) from test ring loops having an equivalent cross section of 0.5 x 0.5 cm and differing radius and vertical placement (bottom). The density is that of mercury, and the atom trajectory apex is at 47 cm.

Abstract

The Newtonian gravitational inverse-square law has been assumed to be valid from the quantum gravity "Planck scale" to the cosmological scale. However, new developments in physics suggest that gravity might be modified at short length scales. This would manifest itself as a Yukawa term—a modification to the usual Newtonian gravitational potential. In addition, the Newtonian coupling gravitational constant is by far the least accurately known fundamental constant in nature. We propose to apply an existing gravity gradiometer based on atom interferometry to the study of possible modifications of Newtonian gravity at high accuracy at the centimeter-to-meter scale and to measure the gravitational constant at new levels of precision.

We expect, if successful, to make a significant contribution to fundamental physics by constraining grand unification models and the possible interplay of dark energy and particle physics by improving bounds on non-Newtonian mass-coupled forces ranging in scale from centimeters to meters. Additionally, this work will directly improve our general ability to extract small gravitational anomaly signals at close range, greatly aiding our ability to apply variants of the gravity gradiometer device to emergency response and nuclear threat monitoring.

Mission Relevance

Our efforts to apply gravity gradiometers to set new, highly precise bounds on Newtonian gravity at the centimeter-to-meter scale is well aligned with several of the Laboratory's strategic missions, as well as with the broader science, technology, and engineering foundations. Specifically, our research supports nuclear threat reduction through the application of a gravity gradiometer to emergency response and treaty verification, advanced optical systems with the application of precision optical metrology to source-mass quantification, and fundamental physics research with exploration of the fundamental physics of gravity.

FY13 Accomplishments and Results

In FY13 we (1) successfully retrofitted the AOSense gravity gradiometer system to include a narrow line-width master laser and increased atom flux; (2) completed the analytic and computational signal response (including quantum corrections) and sensitivity analyses of the gradiometer, proof mass, and translator system required for high-precision gravity, including a detailed system error budget and discovery of novel, optimal test configurations; and (3) designed and developed the high-precision, hollow-cylinder, 150-kg nonmagnetic heavy-metal proof-mass translation system, accommodating the computational model error budget motivated by the fundamental physics test geometry.

Project Summary

Our purpose was to apply the existing AOSense, Inc. gravity gradiometer to extend the bounds on possible modifications of Newtonian gravity (new mass coupled forces) at the centimeter scale, and to improve the accuracy of the overall coupling gravitational constant to 10^{-4}. To do this, we successfully retrofitted the AOSense gravity gradiometer system (adding a narrow line-width master laser and increased atom flux) for high signal-to-noise measurements. Using Wigner functions, we completed a full analytic and computational signal response (including quantum corrections) for the atom interferometer to accuracies required for designing and interpreting high-precision, near-field interferometer experiments. We computed a detailed system error budget and discovered several novel, optimal test configurations. Following our computational model error budget, we designed and built a high-precision proof-mass translation system and procured split, hollow-cylinder 150-kg heavy-metal proof masses. After final machining and metrology of the proof masses, we will be ready to begin our gravity experiments. We expect, if successful, to make a significant contribution to fundamental physics by constraining grand unification models and the possible interplay of dark energy and particle physics by improving bounds on non-Newtonian mass-coupled forces over the centimeter-length scale. Additionally, the signal analysis and cold atom interferometer modeling needed has already directly improved our ability to extract small gravitational anomaly signals at close range with cold atom-based sensors, greatly aiding the range of application of our gravity gradiometer devices currently under parallel development for emergency response and nuclear threat monitoring.

Publications and Presentations

Fitsos, P., et al., 2013. *Design of a precision mass translation system for gravity measurements.* 28th Ann. Mtg. American Society for Precision Engineering, St. Paul, MN, Oct. 20–25, 2013. LLNL-CONF-642832.

Radio-Frequency Noise in Superconducting Devices

Sergey Pereverzev (13-ERD-016)

Abstract

Detection of coherent neutrino scattering from the 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. It will subsequently find applications in infrared single-photon spectrometry for

clandestine reactor detection as well as biosecurity and medical use, such as cell biochemical imaging or detection of trace biochemical impurities. The 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.

Insert and dilution refrigerator to study fundamental noise mechanisms in superconducting devices in high electric fields and at ultralow temperatures.

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 an antineutrino or neutrino transfers a small amount of momentum to an 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.

FY13 Accomplishments and Results

After consultation with Laboratory and outside experts we identified radio-frequency noise in superconducting detectors as a technical hurdle that should to 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. Specifically, we (1) designed and built a crane and earthquake safety structures to operate an existing dilution refrigerator safely; (2) designed, built, or purchased missing parts for this refrigerator, and attained a base temperature less than 15 mK, according to specifications; and (3) developed a high-voltage feed-through to successfully bring voltages greater than 100 kV into the vacuum chamber of a low-temperature cryostat without voltage breakdown. This feed-through can withstand about 50% higher voltages at the same dimensions as commercial designs.

Proposed Work for FY14

In FY14 we will (1) design and build a cryogenic sample holder for superconducting gigahertz resonators that can be filled with liquid helium to allow radio-frequency measurements on these devices in high electric fields without voltage breakdown; (2) purchase and install the holder and associated radio-frequency components to characterize the resonators bandwidth relative to its center frequency in the dilution refrigerator at temperatures less than 20 mK as a function of radio-frequency power, applied voltage (up to 100 kV), and sample surface preparation; and (3) compare our experimental results with simulations on Lawrence Livermore supercomputers.

Biological Sciences

Laboratory Directed Research and Development

Annual Report FY2013

A Rapid Response System for Toxin Removal

Michael Malfatti (11-ERD-012)

Abstract

Our goal is to develop a therapeutic system that mitigates the consequences of exposure to biotoxins and chemical warfare agents. We will package specific naturally occurring cytochrome proteins of the CYP450 group—a large and diverse group of enzymes that catalyze oxidation of organic substances—into nanometer-scale lipoprotein particles (NLPs) to create an intravenously administered drug that will enhance the body's ability to metabolize certain chemical and biowarfare agents, converting them into inactive compounds. This effort will contribute to the development of new technology that could lead to a rapid response system for therapeutic countermeasures. We will administer the CYP450–NLP complex to rodents to enhance their capability to detoxify the highly toxic trichothecene mycotoxin T-2, a potential biological warfare agent.

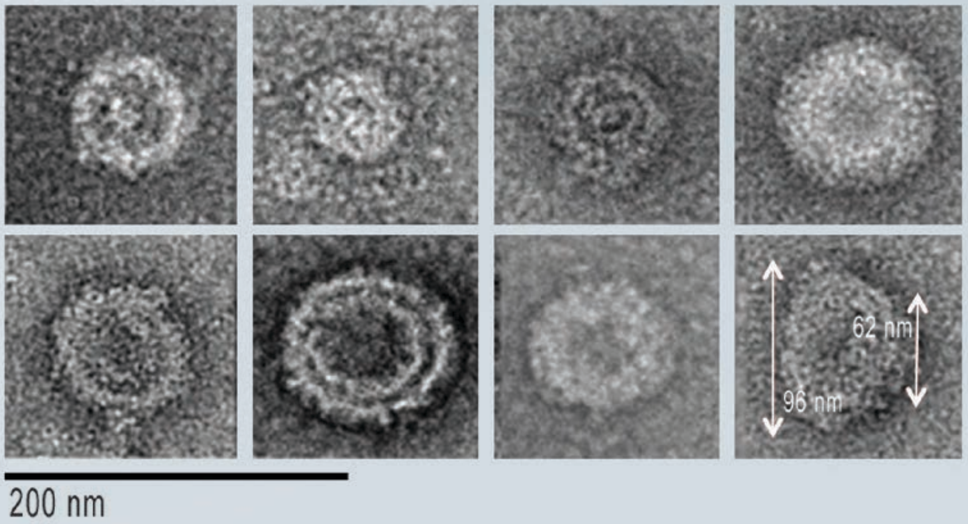
If successful, the CYP450–NLP complex will significantly reduce the blood concentration and biological half-life of the target toxin and increase clearance of the CYP450-mediated metabolites in the treated animals. This proof-of-principle study will lead to a new technology for rapidly treating exposure to biotoxins and other chemical warfare agents. Because its effectiveness lies in bolstering metabolizing enzymes—which deactivate both endogenous and exogenous substrates—this approach will provide a means for responding to many different exposure scenarios, such as treating overdoses of prescription, over-the-counter, or illegal drugs.

Mission Relevance

This project supports LLNL's biosecurity mission by developing new therapeutic countermeasures to biotoxins and other biological and chemical agents that could potentially be used for bioterrorism.

FY13 Accomplishments and Results

In FY13 we (1) utilized telodendrimer chemistry (a unique class of polymers recently developed for nanometer-scale drug delivery) to optimize reaction conditions to maintain stability and functionality of a CYP3A4 nanometer-scale disk complex (CYP3A4 is an important member of the CYP450 enzyme family that is involved in the metabolism of xenobiotics in the human body), (2) demonstrated this complex's ability to metabolize a model CYP450 substrate in vitro, (3) characterized the shape and size of individual complex nanodisks with cryo-electron microscopy imaging and dynamic light scattering, and (4) assessed stability and activity of the nanodisks in rat plasma as a prelude to in vivo studies. The activity observed in rat plasma was similar to that observed in the positive control samples. The ability of the CYP3A4 nanodisks to maintain functionality in plasma and metabolize a substrate in vitro demonstrates their potential utility as a novel way to metabolize and detoxify toxic chemicals or drugs.



Individual cryo-electron micrographs of telodendrimer nanodisks, with an average diameter of 130 nm, containing cytochrome P4503A4 proteins (CYP3A4). Telodendrimers are a unique class of polymers recently developed for nanometer-scale drug delivery. Samples were centrifuged to remove material that had not reacted, and the supernatant was used as the final assembled product.

Project Summary

This project demonstrated the potential utility of protein-embedded nanodisks as a novel approach to detoxify chemical compounds after exposure. We have developed a simple procedure for embedding membrane-bound proteins into lipid nanodisks without the need for protein purification. Our data show that the nanodisks are of a uniform size and shape and are stable under physiological conditions. We have shown that the nanodisks maintain protein functionality and stability in a biological matrix for a sufficient amount of time (eight hours) to be a potentially effective treatment for detoxification of chemicals in vivo. Additional studies are required to assess the effectiveness of the protein and nanodisk complex to metabolize xenobiotics in vivo. The results generated as part of this project have garnered external interest in this approach, and we will investigate these and other potential sponsors to continue our research.

Publications and Presentations

He, W., et al., 2013. "Controlling the diameter, monodispersity, and solubility of ApoA1 nanolipoproteinparticles using telodendrimer chemistry." *Protein Sci.* **22**(8),1078. LLNL-JRNL-643862.

Innate Immunity for Biodefense: Targeted Immune-Modulation to Counter Emerging Threats

Amy Rasley (11-ERD-016)

Abstract

New natural or man-made pathogenic threats cannot presently be countered quickly enough to protect the public, and therefore can potentially result in great loss of life

and billions of dollars in medical costs and lost productivity. Responding rapidly to new and emerging threats requires innovative approaches that do not require pathogen identification or a detailed understanding of that pathogen's biology. Our approach is the targeted modulation of the host's innate immune system that enhances resistance and increases protection from a broad range of pathogens. We will use a combination of LLNL's unique resources, such as accelerator mass spectrometry analyses and bioresearch facilities, to determine the ability of innate immune modulation to protect against the onset of disease.

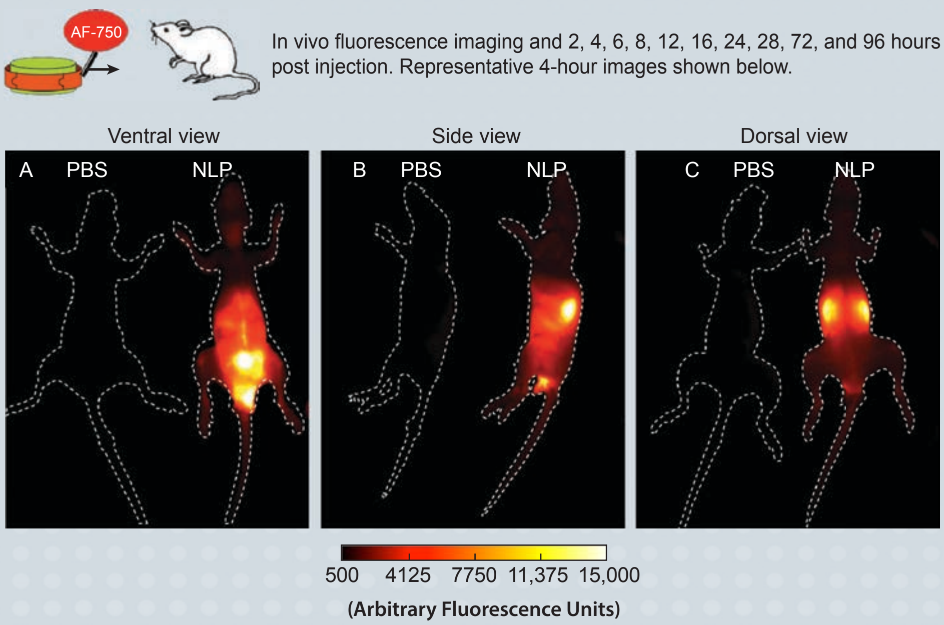
If successful, this project will produce the first-ever description of targeted immune modulation as a means to prevent or delay the onset of an infectious disease and will establish a novel platform for immune-modulation methodologies. This platform will provide the basis for a new biodefense strategy aimed at targeting host immune defenses. Success would lead to the development of new therapeutics that could be administered alone or in combination with other countermeasures—such as those targeting specific host and bacterial proteins involved in pathogen invasion—to kill the pathogen and enhance the natural host immune response.

Mission Relevance

By developing ways to improve innate immune response through targeted immune modulation, this effort is directly aligned with Livermore's biosecurity mission and would support efforts to develop countermeasures against biological threats.

FY13 Accomplishments and Results

We successfully tested two constructs of nanometer-scale lipoprotein (NLP) and agonists (chemicals that bind to a cell receptor and trigger a response by that cell) in vivo using a mouse influenza model. We chose to focus on influenza because of the



Nanometer-scale lipoprotein (NLP) constructs for targeted immune modulation to prevent or delay the onset of an infectious disease exhibit broad tissue distribution profiles depending, in part, on route of administration, as compared with a phosphate buffered saline (PBS) control.

overlap between biosecurity and public health—specifically, influenza represents a model emerging pathogen, ideally suited for assessing the feasibility of this host-based approach to therapeutics. We also characterized the immune response in vitro to an NLP conjugated with two innate immune agonists and demonstrated that co-delivery of multiple agonists on a single NLP significantly enhanced inflammatory responses compared with single agonist formulations.

Project Summary

We investigated the impact of Toll-like receptor agonists and NLP complexes on innate immune responses in vitro and in vivo. This work demonstrated that a variety of innate immune agonists can be readily incorporated and accurately quantified within the NLP platform. We further demonstrated that administration of NLP and agonist constructs resulted in significant enhancement of multiple hallmarks of innate immune activation compared with administration of a free agonist alone. Importantly, NLP conjugation enhanced cytokine production in response to both agonists not only in mice, but also in primary human dendritic cells. Finally, utilizing a mouse model of influenza, we demonstrated that prophylactic administration of complexes of CpG oligodeoxynucleotides (single-strand synthetic DNA molecules that act as immunostimulants) and NLP provided complete protection from an otherwise lethal infection. The efficacy of these complexes at ameliorating infection, coupled with the observation that they enhance innate immune responses not only in mice but also within primary human dendritic cells, indicate potential for future clinical applications of the NLP platform as an innate immune modulator. We have established key relationships with researchers in both academia and industry and plan to submit proposals to the National Institutes of Health centered around the conjugation of innate immune agonists to the NLP platform.

Publications and Presentations

Alam, S., et al., 2013. *Nanolipoprotein delivery enhances immunostimulatory properties of innate immune agonists and provides protection against lethal influenza challenge.* LLNL-PRES-635887-DRAFT.

Blanchette, C.D., et al., 2012. *In vitro and in vivo characterization of nanolipoproteins (NLPs) conjugated with innate immune agonists: Implications for host-based therapeutics*. LLNL-POST-552851-DRAFT.

Blanchette, C. D., et al., 2013. *Nanolipoprotein development and applications: Designer platforms to support antigen presentation.* LLNL-PRES-642147.

Fischer, N. O., et al., 2013. " Co-localized delivery of adjuvant and antigen using nanolipoprotein particles enhances the immune response to recombinant antigens." *J. Am. Chem. Soc.,* **135**(6), 2044. LLNL-JRNL-523576.

Fischer, N. O., et al., 2013. *Evaluation of nanolipoprotein particles (NLPs) as an in vivo delivery platform.* LLNL-JRNL-641712.

Weilhammer, D., 2013. “Nanolipoprotein delivery enhances immunostimulatory properties of innate immune agonists and provides protection against lethal influenza challenge (P4218).” *J. Immunol.* **190**, 48.19. LLNL-JRNL-642038-DRAFT.

Weilhammer, D. R., and A. Rasley, 2012. “Genetics approaches for understanding virulence in Toxoplasma Gondii.” *Brief. Funct. Genom.* **10**(6), 365. LLNL-JRNL-482651-DRAFT.

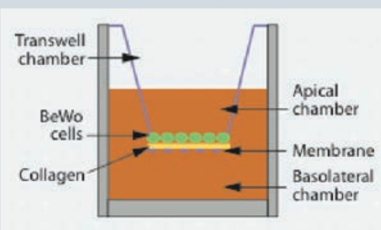
Prenatal Exposure to Endocrine-Disrupting Compounds in the Water Supply

Heather Enright (11-LW-018)

Abstract

Population growth is placing increasing demands on fresh water supplies. However, endocrine-disrupting compounds found at extremely low levels in the water supply are threatening water quality. The need to follow the environmental fate of these chemicals and determine the effect they may have on human development is necessary to protect public health. We will develop methods to determine the extent to which low-levels of endocrine-disrupting compounds in drinking water are transferred from mother to fetus, and to explore whether they have an effect on fetal development. For model development, we will use triclocarban (TCC), an antimicrobial agent that has been found in the water supply at very low levels. In a cell culture model of the human placenta and with the use of accelerator mass spectrometry, we will demonstrate that small amounts of this chemical cross the placental barrier. We will also develop custom tools to study the transfer of low doses of TCC from mother to offspring in a mouse model, along with their effects on development and reproduction.

We expect to see the transfer of TCC labeled with carbon-14 through a polarized cell line in a Transwell assay, which would demonstrate the potential of this compound to be transferred from mother to fetus at the low concentrations that have been found in the water supply. We will investigate the kinetics of TCC placental transfer and explore the developmental effects of TCC exposure using a mouse model. We expect greater amounts of TCC to be transferred through lactation than just through prenatal exposure. It is also likely that TCC will exert developmental effects on the offspring and potentially interfere with their ability to reproduce successfully. These studies will lay the groundwork for further studies of other endocrine-disrupting compounds that have also been reported in the water supply.



Immortalized human trophoblastic BeWo b30 cells are cultured on polyester Transwell inserts with pores coated with human placental collagen. The compound of interest is added to the apical chamber, representing the maternal circulation. Samples are taken from the basolateral chamber, representing the fetal circulation, and measured by atomic mass spectrometry to determine if the compound is transferred across the placental barrier.

Mission Relevance

Studying endocrine-disrupting compounds at environmentally relevant concentrations requires very high sensitivity, and the biological accelerator mass spectrometry facility at LLNL is the only location where this work could be performed. By understanding the effects of these compounds, we will determine whether further research is needed on alternative water treatment strategies that could protect our water supply and public safety, in support of the Laboratory's mission in biosecurity.

FY13 Accomplishments and Results

In FY13 we optimized and completed exposure studies to investigate TCC transfer from mother to offspring. Specifically, we (1) determined, upon successful validation of our custom-made water bottles, 100-nM TCC transfer to offspring through both gestation and lactation periods; (2) found detectable quantities of TCC in utero in fetal and placental tissue; and (3) observed an increase in accumulation of TCC through lactation (about 6-pM TCC/g) compared to the gestation-only exposure group (about 2-pM TCC/g), as expected.

Project Summary

In this project we have made numerous technical accomplishments. We first established and verified a Transwell cellular assay of the placental barrier for endocrine disruptor transfer studies. This cellular assay was utilized to investigate the transfer of three environmentally relevant concentrations of TCC. Custom-made water bottles necessary to accurately mimic chronic exposure from the water supply were made to extend this work to animal studies. Using these bottles, we investigated the transfer of 100-nM TCC from mother to offspring during both lacation and gestation. In summary, our project has established the framework to test further endocrine-disrupting or potentially harmful chemicals across the placental barrier. The highly sensitive nature of our analytical technique—accelerator mass spectrometry—allows for investigation of environmentally relevant concentrations of these chemicals, which until now has not been possible. We are preparing manuscripts on the findings with the Transwell cellular assay and on the in vivo TCC transfer from mother to offspring.

Development of an Economically Viable Biological Hydrogen-Production System

Yongqin Jiao (11-LW-019)

Abstract

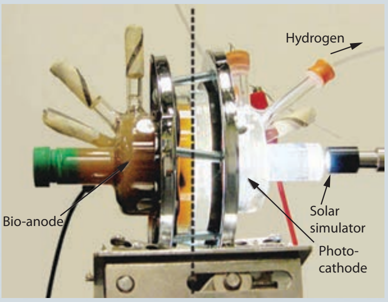
Rising energy demands and the imperative to reduce carbon dioxide (CO_2) emissions are driving research on biofuels development. Biologically derived hydrogen is one of the most promising of these fuels and is seen as a future energy source. We propose to implement a simple and relatively straightforward strategy for hydrogen production by photosynthetic microorganisms using sunlight, sulfur- or iron-based inorganic substrates, and CO_2 as the feedstock. Carefully selected microorganisms with bioengineered beneficial traits will act as the biocatalysts for the process and will be

designed to both enhance the system efficiency of CO_2 fixation and the net hydrogen production rate. We will apply metabolic engineering approaches guided by computational modeling for the chosen model microorganisms to enable efficient hydrogen production.

We hope to demonstrate, for the first time, the ability to produce hydrogen using inorganic substrates coupled with CO_2 fixation by photosynthesis. This would be of great significance, not only eliminating the economic and agricultural burden of producing and growing the organic substrates, but also greatly reducing greenhouse gas emissions commonly associated with their production and usage. The use of sulfur or iron-based inorganic substrates is likely to make hydrogen production more economically viable, because iron is both cheap and abundant and sulfur-containing iron minerals are moderately common. Our ultimate goal is to lead the way towards economically viable biological hydrogen production on a scale sufficient to supply a substantial portion of the hydrogen fuel market.

Mission Relevance

This project will enable a multidisciplinary group of scientists at LLNL to collaborate in achieving a basic understanding of critical microbial metabolism directly relevant to DOE missions in energy security, cleaner biomass energy conversion, and carbon sequestration. Our proposed work directly supports a core Laboratory mission and strategic priority in enhancing energy security. This research also positions Livermore to play an expanded role in a scientific focus area of the DOE Genomic Science Program—understanding microbial community functions in hydrogen production.



Our experimental setup based on a straightforward strategy for hydrogen production by photosynthetic microorganisms using sunlight, sulfur- or iron-based inorganic substrates, and carbon dioxide as the feedstock. The goal is to demonstrate, for the first time ever, the ability to produce hydrogen using inorganic substrates coupled with carbon dioxide fixation by photosynthesis.

FY13 Accomplishments and Results

In FY13 we (1) used transcriptional analysis to test the relative importance of the three nitrogenase enzymes used for hydrogen production in the photosynthetic bacterium *Rhodopseudomonas palustris*, whose genome consists of three versions of nitrogenase enzymes including vanadium, iron, and molybdenum cofactor-containing enzymes; (2) used quantitative real-time polymerase chain reaction analysis to show that the molybdenum-containing nitrogenase was the only one induced under nitrogen-fixing conditions, indicating its vital role for hydrogen production in *R. palustris*; (3) developed a genome-scale model of metabolism in *R. palustris* that accounts for differences in cellular biomass under different environmental conditions to examine the electron economy of the system and link it to different metabolic processes; and (4) used this model to assess the robustness of *R. palustris* metabolism against genetic and environmental perturbations.

Project Summary

We successfully demonstrated that hydrogen can be produced from photoautotrophic growth of the photosynthetic bacterium *R. palustris* with thiosulfate as the sole source of electrons and CO_2 as the sole carbon source. This method may serve as a possible alternative to biomass-based hydrogen production. We showed that hydrogen was produced through the action of molybdenum-containing nitrogenase and that nitrogen-fixation conditions are required for hydrogen production. While the growth and hydrogen production rates were lower with cells grown on thiosulfate when compared to organic substrates, the electron conversion efficiency from thiosulfate to hydrogen was significantly higher compared to that of acetate to hydrogen. In addition, we developed a genome-scale model of metabolism in *R. palustris*. Our results show that for anaerobic and light conditions, the growth rate is not energy- or carbon-limited, but is instead limited by the availability of electron acceptors. Production of hydrogen can serve as an electron sink and alleviate this situation—however, a number of other pathways can also serve as electron sinks.

Unraveling of Assembly and Structure–Function Relationships of Poxviruses

Alexander Malkin (11-ERD-027)

Abstract

Strong interest in poxviruses persists because of their unique replication cycle and assembly, the profound insights they provide into strategies to combat the host immune response, and the potential for deliberate release of poxviruses as a bioterrorist weapon. We aim to elucidate the architecture and assembly pathways for vaccinia virus, which is a cowpox virus that is used to vaccinate against smallpox. We will determine the roles played by individual viral genes in virion assembly and host–pathogen interactions and determine forensic signatures using a multidisciplinary approach. This approach will include a unique combination of nanometer-scale physiochemical methods as well as genetic and biochemical analysis techniques. This improved knowledge will be pivotal for elucidating mechanisms of pathogenesis and developing countermeasures against viral agents.

We will identify and map target protein locations in the vaccinia virion and establish relationships between viral assembly, chemistry, proteomic structure, and replication cycle. In addition, this information will be used to further develop molecular-scale models of the vaccinia virion's complex architecture. This work will support future structural, genetic, and biochemical analysis of the functional repertoire of human viruses and will link viral molecular-scale structural and elemental attributes to production conditions. Our research will provide important insights into the viral replication cycle, physical and chemical properties, and will serve as an enabling platform to identify protein targets for development of vaccines, therapeutics, viral detection, attribution, and bioforensics technologies.

Mission Relevance

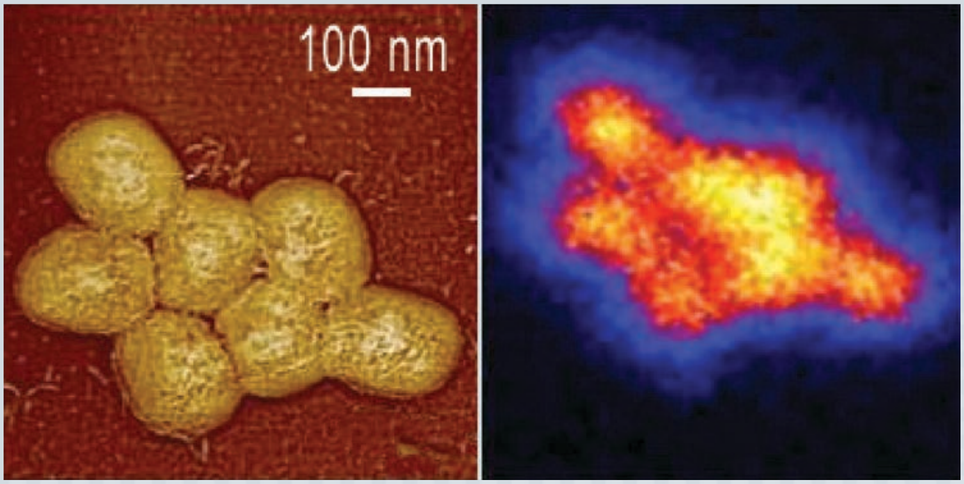
This project supports the Laboratory's national security mission by elucidating key events in the life cycle of a virus with potential bioterror applications, as well as by improving our fundamental understanding of the mechanisms of host–pathogen interactions for applications in human health and biodefense. Developing an improved approach for assessing and characterizing viral structure for pathogen forensics and attribution contributes to more efficacious preventive and therapeutic measures for emerging diseases and biodefense-related agents.

FY13 Accomplishments and Results

In FY13 we (1) completed characterization of viral and subviral structures of selected mutants with atomic force microscopy, (2) correlated specific topographical structures of vaccinia virion with specific viral gene products, (3) compiled data on the functional repertoire of selected mutants representing phenotypic classes with distinctive defective replication properties, (4) produced an improved model of the vaccinia virion's architecture and roles played by selected individual viral genes in the viral assembly, (5) completed analysis of the spatial localization and depth distribution of specific isotopic-labeled constituents in the viral structure using nanometer-scale secondary-ion mass spectrometry, and (6) applied the nonequilibrium sputter rate and densification models to describe observed nonlinear sputtering rates of vaccinia virion.

Project Summary

The successful conclusion of this project resulted in a new orthogonal methodology for use in structural virology. Our approach enabled for the first time physical and chemical analysis of single virions at the nanometer scale, providing direct correlations between structural, chemical, and isotopic viral attributes. High-resolution atomic force microscopy of subviral structures of the vaccinia virion (both wild type and selected mutants) was combined with an analytical method employing isotopic labeling coupled with nanometer-scale secondary-ion mass spectrometry to determine spatial localization and depth distribution of specific viral constituents. This approach allowed us to unravel the detailed architecture and assembly of the vaccinia virion and to improve the fundamental understanding of vaccinia virus assembly and



Orthogonal characterization of the viral structure and compartmentalization of lipids in single virions of the vaccinia virus by atomic force microscopy (left) and by nanometer-scale secondary ion mass spectrometry and carbon-13 labeling of the viral lipid membrane (right).

function. The capabilities developed in this project could contribute significantly to an evaluation of the key events in the viral replication cycle for applications in structural virology and medical pathology. We are planning to seek funding from the National Institutes of Health to study the functional repertoire of vaccinia mutants in addition to seeking support for the development of viral physical and chemical forensics based on atomic force microscopy and nanometer-scale secondary-ion mass spectrometry from a U.S. government sponsor.

Publications and Presentations

Gates, S. D., 2013. *Biophysical analysis of bacterial and viral systems: A shock tube study of bio-aerosols and a correlated AFM/NanoSIMS investigation of vaccinia virus.* LLNL-TH-645412.

Malkin, A. J., 2013. *In vitro high-resolution architecture and structural dynamics of microbial and viral systems.* University of Florida Seminar Series in Structural Biology, Gainesville, Fl, Mar. 18, 2013. LLNL-PRES-562772.

Weber, P. K., et al., 2013. *AFM and NanoSIMS analyses of vaccinia virions.* 2013 Microscopy and Microanalysis Conf., Indianapolis, IN, Aug. 4–8, 2013. LLNL-CONF-624061.

Rapid Development and Generation of Affinity Reagents for Emerging Host–Pathogen Interactions

Matthew Coleman (11-ERD-037)

Abstract

Antibodies are an essential reagent for identification and characterization of proteins. Given the wealth of genomic information in the public database, there is a specific need for robust antibodies for multiple research applications as well as therapeutic use. This is especially true for pathogen and cancer detection as well as for biodosimetry applications. We propose to produce selected antigens such as small molecules, peptides, and membrane proteins for development of a new antibody selection approach where antigens are displayed on nanometer-scale particles for synthetically generating antibodies. Using this approach, we will be able to rapidly select recombinant antibodies generated using cell-free methodologies directed against selected antigens related to host–pathogen interactions.

We expect this project will provide a unique high-throughput laboratory capability for supporting multiple LLNL programmatic needs for detection, mitigation, and in vivo visualization of proteins as well as small molecules of interest. This approach can potentially eliminate the need for use of animals in antibody selection applications.

Mission Relevance

Our proposal focuses on a new avenue of research in advanced molecular tools to fill a serious gap in quantitative biology through development of instrumentation combined with affinity reagents. The technology has the potential to help extend and revitalize LLNL's detection capabilities in support of the Laboratory's mission in biosecurity, including the rapid mitigation of evolving and unknown biothreats, as well as expanding basic research in bioscience to improve human health.

FY13 Accomplishments and Results

In FY13 we (1) applied our novel three-color fluorescent correlation spectroscopy instrumentation to characterize in-solution biological molecules, incorporating techniques that increase the yield and solubility of nanolipoprotiens by 50% and that could be used for generating novel affinity reagents; (2) produced and purified proteins focused on the secretion-III mechanism, including the *Yersinia pestis* outer membrane protein YopB, translocator of Yersinia pseudotuberculosis YopD, nanolipoprotein APOA1, and secretion III protein LcrV; (3) produced functional Pla surface protease (an enzyme in *Y. Pestis* that cleaves arginyl bonds in proteins) using cell-free methods in sufficient quantities to support recombinant antibody generation and production, in collaboration with the University of Illinois at Chicago; (4) demonstrated antibody-binding to LcrV and LcrV-GFP proteins; and (5) demonstrated that an antibody-encoded DNA sequence could be rapidly converted for robust cell-free production.

Project Summary

We were able to develop a novel three-color fluorescent correlation spectroscopy system for characterizing protein and protein–antibody interactions. Furthermore, cell-free expression was used to engineer antibodies in a high-throughput format. This represents the first demonstration of cell-free production of single-chain antibodies to detect a protein (LcrV) associated with a pathogen. Based upon these initial results, we are seeking future funding from commercial and government entities such as Synthetic Genomics Vaccine, Defense Threat Reduction Agency, and the National Institutes of Health to continue our research.

Publications and Presentations

Gao, T., et al., 2012. "Characterization of de novo synthesized GPCRs supported in nanolipoprotein discs." *Plos One* **7**(9), e44911. LLNl-JRNL-618672.

Gao, T., et al., 2011. "Characterizing diffusion dynamics of a membrane protein associated with nanolipoproteins using fluorescence correlation spectroscopy." *Protein Sci.* **20**(2), 437. LLNL-JRNL-618652.

He, W., et al., 2013. " Controlling the diameter, monodispersity, and solubility of ApoA1 nanolipoprotein particles using telodendrimer chemistry." *Protein Sci.* **22**(8), 1078. LLNL-JRNL-643862.

Ly, S., et al., 2011. "Stoichiometry of reconstituted high density lipoproteins in the hydrated state determined by photon antibunching." *Biophys. J.* **101**(4), 970. LLNL-JRNL-459318.

Targeted Drug Delivery for Treating Traumatic Bone Injury

Nicole Collette (11-ERD-060)

Abstract

Significant injuries currently suffered by U.S. troops are fractures and other bone injuries. These injuries result in considerable hospitalization costs and in permanent disability in up to 13% of all fractures, which never heal. Most existing bone-injury interventions are mechanical, designed to stabilize the break and facilitate natural repair. The objective of this project is to enhance the healing of bone fractures, reducing time to union and load-bearing quality in difficult fractures. This work will utilize nanometer-scale lipid particles coupled with therapeutic agents in conjunction with combinatorial drug therapy to promote healing during normal fracture repair with an initial focusing on the inflammatory response in the beginning phases of repair.

We will deliver a fully functional, tissue-specific drug system tested in vivo with synergistic bone-healing therapy that is ready for clinical settings. This therapy will utilize currently approved therapies in a novel way and will also enable the development of novel therapies for bone repair and regeneration. This therapeutic system will provide much-needed effective, nonsurgical treatments for traumatic bone injuries and other difficult fractures. In addition to defense applications, the resulting bone therapy system will also have applications in biodefense and public medicine in general, especially in the areas of slow-healing osteoporotic fractures and fractures complicated by diseases such as diabetes.

Mission Relevance

This work has applications that address Department of Defense research needs and supports LLNL's missions in national security and biodefense by providing a solid foundation—including materials design and novel approaches—for future research in the areas of infection treatment and prevention.

FY13 Accomplishments and Results

In FY13 we (1) identified a novel and potential beneficial genetic mutation in mice that results in a thick cortical shell around the healing bone; (2) determined that the cells affected in these mutants appear to be stem cells, which has other implications for healing and recovery from illness or injury; (3) examined fracture repair in diabetic mice, and dosed them with a potent therapeutic molecule to counter the impaired healing; and (4) used histology to provide insight into the novel mouse mutant.

Unfortunately, we were unable to identify a clear effect or immune-related response when the nanolipid particles were used during fracture repair.

Project Summary

The successful conclusion of this project has led to the discovery of a mutant mouse with a novel gene responsible for a unique fracture repair mechanism. The current data suggests that this phenotype is driven by stem cells, leading to potential new avenues of study. In addition, we have utilized a potent therapeutic molecule in fracture repair studies to facilitate repair in a diabetic fracture model. We successfully developed surgical and histological techniques for this study, and have strengthened our collaborations with industry by utilizing a proprietary therapeutic treatment for our experiments. In summary, our studies have developed a system for in vivo testing of other potentially useful therapeutic interventions for difficult-to-repair fractures, while also providing insight into molecular and cellular mechanisms of repair that can be exploited for future drug target development. The novel nature of our mutant mouse model will generate data important to the scientific field on behavior and control of stem cells in vivo.

Publications and Presentations

Chahine, N. O., et al., 2013. *Biocompatibility of hydrogel-carbon nanotube composites or chondrocyte growth and cartilage tissue engineering.* LLNL-JRNL-636373-DRAFT.

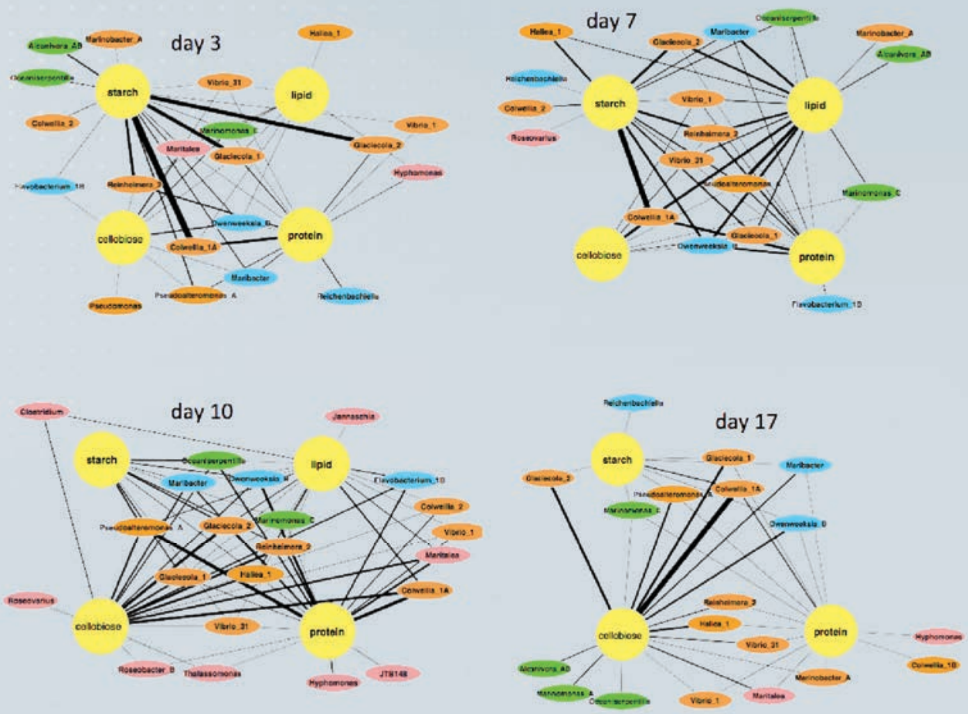
Collette, N. M., et al., 2013. *Loss of SOSTDC1, a paralog of SOST, results in decreased trabecular bone and impaired fracture healing, in vivo*. The American Society for Bone and Mineral Research (ASBMR) 2013 Ann. Mtg., Baltimore, MD, Oct. 4–7, 2013. LLNL-ABS-633635.

Accelerator and Secondary-Ion Mass Spectrometry for Analysis of Coastal Carbon Flux

Xavier Mayali (11-ERD-066)

Abstract

Little is known about the mechanisms of organic carbon use in the sinking particles that control, through the biological pump, the size of the long-term marine carbon sink. In this project, we will determine the effect of simulated climate change, including elevated temperatures and carbon dioxide concentrations on the degradation of coastal marine particles. Using novel methods such as secondary-ion mass spectrometric analysis of microarrays and accelerator mass spectrometric analysis of radiotracers, we will link microbial community structure and function to direct measurements of carbon fluxes into and out of degrading marine particles. We will conduct this work for medium-sized biological communities using both stable-isotope and radioisotope tracers.



Network diagrams from analysis of RNA microarrays linking different bacteria (ellipses, color-coded by species) and the type of organic matter they were consuming (yellow circles) as they attached to marine particles over time, part of an effort to determine the effect of simulated climate change on the degradation of coastal marine particles.

We will perform direct measurements of carbon fluxes into and out of degrading particles and identify the microorganisms responsible for these biogeochemical activities. In addition, we expect to elucidate the currently unknown effect of climate change on these processes. These results will increase our understanding of the potential effects of climate change on the ocean's biological pump, which controls the long-term natural sequestration of carbon. In addition, we will determine a powerful combination of techniques to link microbial structure and function with direct flux measurements, which will enable biogeochemical studies in other ecosystems.

Mission Relevance

This project will advance LLNL's mission in energy security and climate change by determining the carbon-fixing biogeochemical functions of near-shore marine bacteria, which is a cutting-edge area of carbon sequestration research and will help shape efforts to model carbon-drawdown mechanisms in the ocean under realistic climate scenarios.

FY13 Accomplishments and Results

In FY13 we (1) conducted analyses of our climate-effect experiment, which showed that elevated temperatures increased the metabolic breakdown of marine particles, as we hypothesized; (2) determined, from discussions with experts in this field, that examining the effect of light on particle breakdown may be a more relevant field of study because recent evidence suggests that photoheterotrophic organisms capable of utilizing dissolved organic materials and harvesting light energy are more important than previously thought and play a major role in global carbon cycling; (3) performed, as a result of these discussions, an experiment with scientists at the University of Southern California to examine the impact of light on the breakdown of marine particles—we directly labeled the particles with nitrogen-15 and carbon-13, collected sequence data, and performed analyses using a micro-array slide combined with stable isotope probing; and (4) examined the effect of climate (specifically, increased temperature) on carbon and nitrogen incorporation of coastal marine bacteria.

Project Summary

We have investigated the patterns of particle breakdown and bacterial assimilation of carbon eroded from sedimentary rocks using a model system of freeze-thawed diatom cells incubated in laboratory microcosms with bay and coastal natural microbial communities. Dark-incubated particles labeled with carbon-13 were collected over two weeks and analyzed for biomass and carbon-13 isotopic enrichment with isotope-ratio mass spectrometry. Particle-attached bacteria from the coastal site were incubated with labeled carbon substrates (proteins, lipids, and two types of polysaccharides) for taxon-specific isotopic incorporation analysis with micro-arrays and nanometer-scale secondary-ion mass spectrometry. Our subsequent study investigated the effect of light on the breakdown of marine particles and whether particle-attached bacteria provide dissolved organic matter to fuel the growth of bacteria unable to attach to particles. A third study examining the effect of temperature on marine bacterial activity was also accomplished. Building on this project's work, we will continue work on coastal marine organic matter degradation sponsored by the Gordon and Betty Moore Foundation. We are also seeking sponsorship from the DOE's Biological and Environmental Research Program for the study of algal biofuels, specifically examining bacterial attachment to algae.

Publications and Presentations

Mayali, X., P. K. Weber, and J. Pett-Ridge, 2012. "Taxon-specific C/N relative use efficiency for amino acids in an estuarine community." *FEMS Microbiol. Ecol.* **83**(2), 402. LLNL-JRNL-553798.

Mayali, X., et al., 2012. *Bacterial hydrolysis of marine particles: Identifying temporal breakdown patterns and species-specific carbon substrate assimilation using stable isotope labeling, microarrays and NanoSIMS.* ISME14—The Power of the Small, 14th Intl. Symp. Microbial Ecology, Copenhagen, Denmark, Aug. 19–24, 2012. LLNL-ABS-545311.

Mayali, X., et al., 2013. *Degradation and taxon-specific microbial carbon utilization of diatom-derived particles quantified by stable isotope mass spectrometry.* LLNL-JRNL-641031.

Mayali, X., et al., 2011. "High-throughput isotopic analysis of RNA microarrays to quantify microbial resource use." *ISME J.* **6**, 1210. LLNL-JRNL-470805.

Mayali, X., et al., 2011. *Linking identity and biogeochemical function of estuarine microbial communities by analysis of 16S microarrays with secondary ion mass spectrometry.* LLNL-PRES-451282.

Mayali, X., et al., 2010. *Linking phylogenetic identity and biogeochemical function of estuarine microbial communities by analysis of phylogenetic microarrays with secondary ion mass spectrometry.* 13th Intl. Society for Microbial Ecology Conf., Seattle, WA, Aug. 22–27, 2010. LLNL-CONF-428120.

Mayali, X., et al., 2013. *Redefining the microbial niche based on taxon-specific response to differing substrate concentration.* LLNL-JRNL-565076.

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 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 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 and the ability to respond to emerging bio-threats.

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.

FY13 Accomplishments and Results

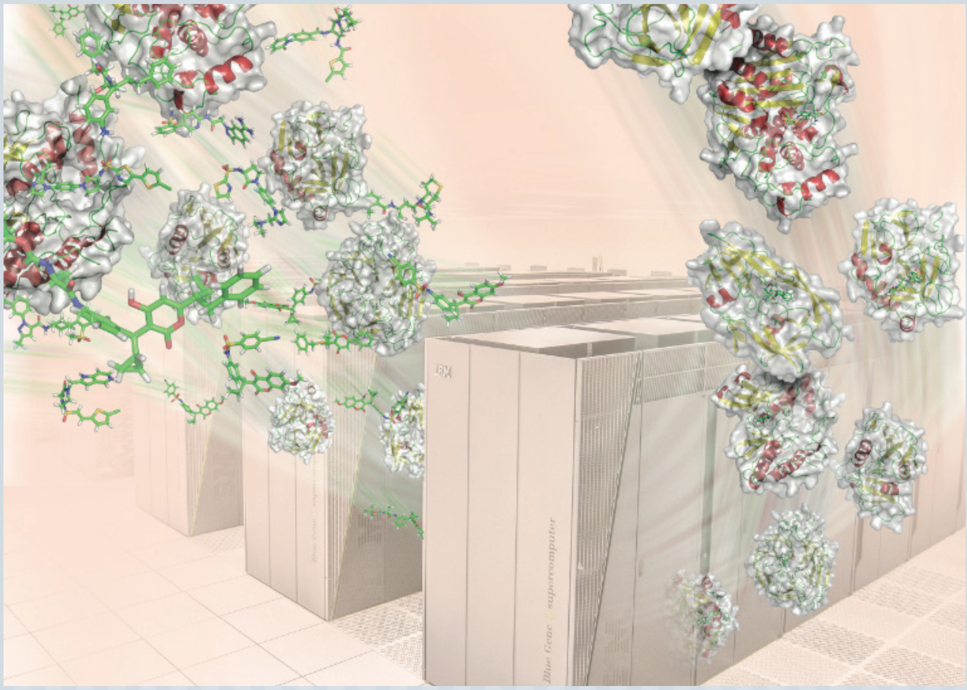
In FY13 we (1) continued to develop each of the modules that will ultimately predict pharmacokinetic properties and adverse side effects of drug candidates, (2) completed a physiologically based pharmacokinetic model of acetaminophen pain reliever, (3) created a correlation between known drug compounds and specific adverse drug reactions, (4) continued to automate the protocol to better evaluate off-target interactions by drug-like molecules, and (5) computed kinetic parameters for small-molecule binding to proteins and membrane permeability.

Proposed Work for FY14

In FY14 we will (1) continue to develop each of the modules that will ultimately predict pharmacokinetic properties and adverse side effects of drug candidates; (2) extend our physiologically based pharmacokinetic model to new compounds and build kinetic modules using parameters from the kinetic parameter-generation module; (3) continue work on neural networks, associating off-target inputs to adverse drug reactions, and linking drugs to targets, pathways, and adverse reactions; (4) develop and implement algorithms to compute absolute binding free energies of small molecules to proteins, continuing our automation protocol, and expand our off-target library for the human genome; and (5) expand the current framework to predict the kinetic parameters for the P450 enzyme metabolism of small molecules.

Publications and Presentations

Carpenter, T. S., E. Y. Lau, and F. C. Lightstone, 2013. "Identification of a possible secondary picrotoxin-binding site on the GABAA Receptor." *Chem. Res. Toxicol.,* **26**(10), 14. LLNL-JRNL-611112.



Parallel molecular docking of large databases on Sequoia, a petascale IBM BlueGene Q supercomputer at Lawrence Livermore National Laboratory. A mixed parallel scheme that combines message passing interface and multithreading is implemented in the Vina molecular docking program named VinaLC. Parallel performance analysis show that the code scales up to more than 15,000 central processing units.

Kirshner, D. A., J. P. Nilmeier, and F. C. Lightstone, 2013. "Catalytic site identification—A web server to identify catalytic site structural matches throughout PDB." *Nucleic Acids Res.* **41**, W256. LLNL-JRNL-618954.

Navid, A., et al., in press. "Quantitative in silico analysis of transient metabolism of acetaminophen and associated causes of hepatotoxicity in humans." *In Silico Pharmacol.* LLNL-JRNL-640155.

Nilmeier, J. P., et al., 2013. "Rapid catalytic template searching as an enzyme function prediction procedure." *PLoS ONE* **8**(5), e62535. LLNL-POST-564184.

Yang, Y., S. E. Wong, and F. C. Lightstone, 2013. "Approaches to efficiently estimate solvation and explicit water energetics in ligand binding: The use of WaterMap." *Expert Opin. Drug Discov.* **8**, 277. LLNL-JRNL-582572.

Yang, Y., S. E. Wong, and F. C. Lightstone, 2013. *Understanding a substrate's product regioselectivity in a family of enzymes: A case study of acetaminophen binding in cytochrome P450s.* LLNL-JRNL-615272.

Zhang, X., S. E. Wong, and F. C. Lightstone, 2012. "Message passing interface and multithreading hybrid for parallel molecular docking of large databases on petascale high performance computing machines." *J. Comput. Chem.* **34**(11), 915. LLNL-JRNL-568309.

Zhang, X., S. E. Wong, and F. C. Lightstone, 2013. *Towards fully automated high performance computing drug discovery: A massively parallel virtual screening pipeline for docking and MM/GBSA rescoring to improve enrichment.* LLNL-JRNL-642881.

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 which 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 National Nuclear Security Administration 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. In addition, a greater understanding of biofuel synthesis proteins could facilitate engineering of new biofuel production processes, in support of the energy security mission.

FY13 Accomplishments and Results

In FY13 we (1) expressed, through recombination, a number of proteins that include bacterial virulence factors, and grew micrometer-scale crystals that were measured at LCLS and with synchrotrons—one new structure resulted from this work; (2) continued work on membrane proteins, including bacteriorhodosin (the light-driven proton pump) and, with collaborators, receptors coupled with a guanosine nucleotide-binding protein in lipidic cubic phase—we performed experiments on two- and three-dimensional crystals of such proteins at LCLS; (3) prepared low-density lipoprotein-particle microcrystals and measured them with a synchrotron—however the quality was not sufficient to warrant experiments at LCLS; (4) continued to improve and demonstrate a fixed-target delivery system for two- and three-dimensional protein crystallography at LCLS; and (5) successfully measured two-dimensional crystals of bacteriorhodopsin retinal protein and other proteins using pump–probe experiments and fixed targets at LCLS.

Proposed Work for FY14

In FY14 we will execute and participate in several LCLS beam experiments to measure two-dimensional protein crystals and three-dimensional protein micrometer- and nanometer-scale crystals using the ultrafast optical pump and x-ray probe experiments on rhodopsin retinal-binding proteins. In addition, we will explore previously unknown structural changes of protein crystals at picosecond-to-microsecond timescales.

Publications and Presentations

Feld, G., 2013. *Structural-genomics approach reveals function of novel virulence factors from* Francisella tularensis. LLNL-PRES-628312-DRAFT.

Hunter, M. S., 2012. *Femtosecond x-ray crystallography using protein microcrystals.* LLNL-PRES-540151.

Loh, N., et al., 2013. "Fractal morphology, imaging and mass spectrometry of single aerosol particles in flight." *Nature* **486**, 513. LLNL-JRNL-556614.

Omattage, N., et al., 2012. *Study of the structure and function of a novel bacterial virulence factor isolated from* Francisella tularensis. LLNL-POST-568415.

Omattage, N., et al., 2012. *Study of the structure and function of a novel bacterial virulence factor isolated from* Francisella tularensis. LLNL-TR-572332.

Pedersoli, E., et al., 2013. "Mesoscale morphology of airborne core–shell nanoparticle clusters: X-ray laser coherent diffraction imaging. *J. Phys. B Atom. Mol. Opt. Phys.* **46**, 164033. LLNL-JRNL-642901.

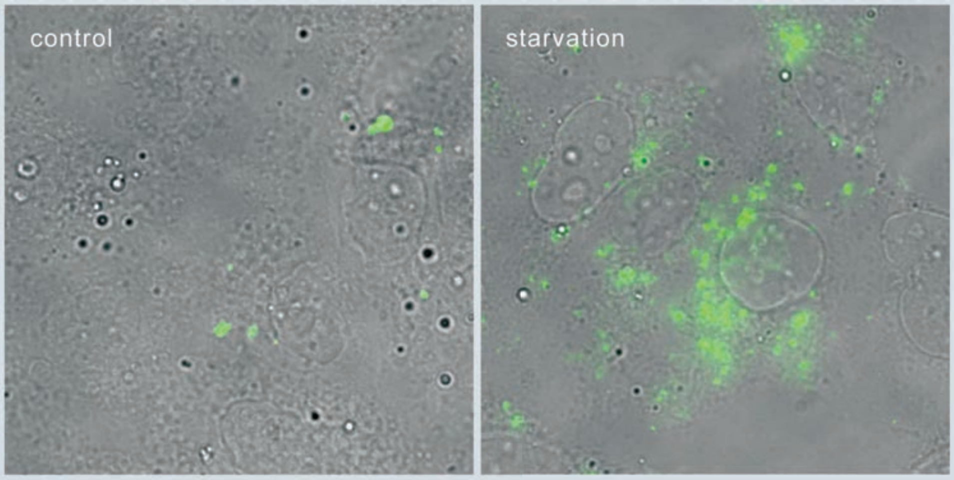
Modulating Cellular Autophagy to Combat Bacterial Pathogens

Catherine Lacayo (12-ERD-049)

Abstract

The continued rise in drug-resistant bacteria and new bio-threats demands the development of alternative strategies to combat infection. The modulation of autophagy, which literally means "self-eating" and is a conserved cellular recycling process, has the potential to fulfill this need by serving as a line of defense against pathogenic bacteria instead of current antimicrobial drug therapies. However, before development of autophagy-modulating therapeutics can realistically begin, we first require an in-depth understanding of this process. We plan to provide, for the first time, a detailed spatiotemporal model of autophagy using living cells infected with bacteria to determine how this mechanism can be fully harnessed as a therapeutic alternative.

We expect to create a comprehensive and quantitative model of the autophagy process during bacterial infection and demonstrate that a pharmaco-modulatory approach can induce significant killing of invading bacteria. In addition, we expect to



Human osteosarcoma bone cancer cells expressing GFP-LC3, a marker for autophagosomes (green). In this example, autophagy was modulated by starving cells (right), which causes a significant increase in autophagosome accumulation compared to control cells (left).

determine the efficiency of autophagy in the degradation of invading bacteria by non-phagocytic cells (which are not involved in directly targeting and killing invading bacteria) and understand how autophagy affects survival of the host during infection. These questions have important implications when considering that all cells in our bodies have the potential for clearing infection using their inherent autophagic machinery, and they could use this mechanism to survive, instead of die, as a means to clear infection.

Mission Relevance

The proposed work directly aligns with the Laboratory's biosecurity mission and the science and technology foundation in host–pathogen interactions by generating knowledge crucial to the development of novel and alternative medical countermeasures against infection. Our proposal seeks to validate autophagy as a novel therapeutic approach against microbial infection and establish a live-imaging platform that can be applied to the evaluation of potential autophagy modulators.

FY13 Accomplishments and Results

A key goal of this project is the specific modulation of autophagy to effect killing of intracellular bacteria so that infection can be controlled. Therefore, in FY13 we (1) focused on understanding how the bacteria *Staphylococcus aureus* and infected host cells respond to various treatments known to modulate autophagy; (2) utilized additional analytical tools, such as flow cytometry to generate quantitative information about host cell cytotoxicity and the infectivity of different pathogenic strains, including some that are resistant to certain antibiotics; (3) successfully modulated autophagy using specific pharmacological agents and nutrient deprivation; and (4) measured corresponding changes in bacterial and host cell survival, with studies performed in human cells, instead of the proposed mouse cells, because human cells are more relevant to public health.

Proposed Work for FY14

In FY14, to complete our analysis of bacterial and host cell survival in response to the pharmaco-modulation of autophagy, we will (1) combine different pharmacological agents, in addition to treatments using individual drugs, and temporally modify their action during infection to ultimately effect optimal bacterial killing through autophagy; (2) continue to measure certain parameters of infection and autophagy using flow cytometry; and (3) visualize the progression of autophagy using live-cell microscopy to determine the co-localization of autophagy markers.

Carbon Nanometer-Scale Membrane Channels

Aleksandr Noy (12-ERD-073)

Abstract

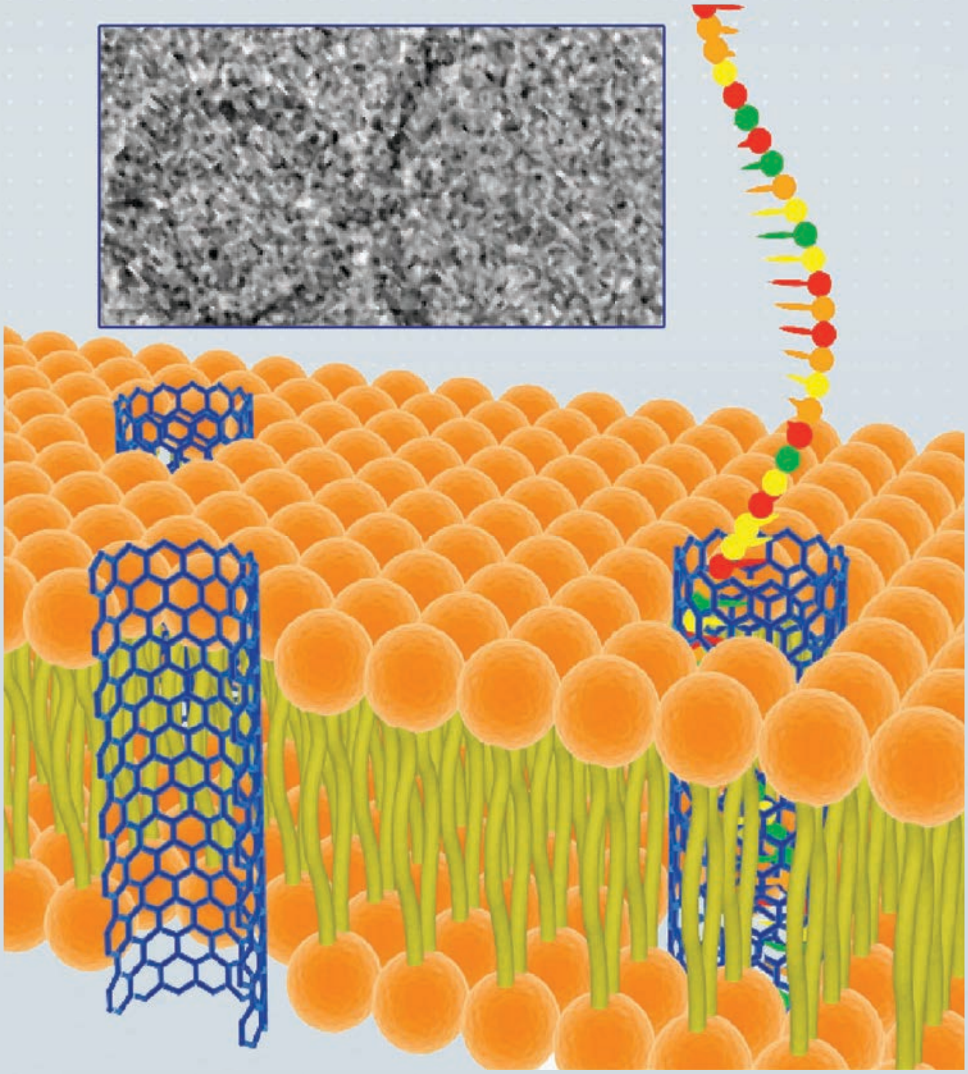
Living cells depend upon the flow of molecules across membranes for essential processes such as sensing, signaling, and energy production. Yet the cell membrane presents a formidable barrier to the transport of these molecules because they cannot cross the membrane unaided. As a result, living systems 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 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 through development of a membrane-penetrating structure that uses a completely



Carbon nanometer-scale tube channel inserted into a lipid bilayer membrane with a single-strand DNA molecule passing through one of the channels. The inset shows a cryogenic electron microscopy image of a carbon nanotube channel connecting two lipid vesicles.

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.

FY13 Accomplishments and Results

In FY13 we successfully accomplished the task of building a functional artificial ion channel based on a carbon nanotube scaffold. Specifically, we (1) demonstrated improved cutting of carbon nanotubes, (2) demonstrated insertion of the carbon nanotube channels into the membrane, (3) characterized transport through the channels, (4) detected ion transport through a single carbon nanotube channel using a pico-amp electrical detection setup, (5) discovered frictionless ion transport and stochastic gating phenomena in those channels, and (6) performed cryo-transmission electron microscope imaging to visualize channels in the lipid membranes.

Proposed Work for FY14

In FY14 we will (1) concentrate on determining ion selectivity of the ion channels, studying transport of more complicated molecules such as DNA through that channel; (2) perform experiments on the interactions of carbon nanotube channels with live cells; and (3) refine the fabrication procedures to increase yield and fidelity of the carbon nanotube channel synthesis.

Publications and Presentations

Geng, J., et al., 2013. *Stochastic transport and gating in carbon nanotube porins in lipid membranes*. LLNL-JRNL-641663.

Noy, A., 2012. "Kinetic model of gas transport in carbon nanotube channels." *J. Phys. Chem*. **117**(15), 7656. LLNL-JRNL-574053.

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. 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.

FY13 Accomplishments and Results

In FY13 we (1) tested the electrical stimulation and recording functions and chemical recording functions of the device in vitro, (2) tested the devices in vivo using a rat model to investigate the different regions involved in deep brain stimulation for depression, and (3) modified our design and fabricated a new set of devices based on our earlier in vitro and in vivo data.

Project Summary

The successful conclusion of this project resulted in the first-ever study of deep brain stimulation for depression in an animal using thin-film polymer electrode arrays capable of stimulating and recording neurons from multiple regions of the brain simultaneously. The neural interface developed under this project serves as a platform technology for future brain studies, and resulted in intellectual property for the multifunctional neural device, peer-reviewed publications and presentations, and a new collaboration with an industrial partner. We hope to collaborate with the biomedical company Medtronic to continue to develop our work towards a human-quality neural prosthetic.

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 EMBS, Osaka, Japan, July 3–7, 2013. LLNL-PRES-639756.

Tooker, A. C., et al., 2013. *Neural interface for deep brain stimulation.* LLNL-CONF-638803.

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, but 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 (TLRs) and immunoglobulin M (IgM). The genomes captured 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 subclinical 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 RNA bound to IgM and TLRs, respectively, from pathogen-infected mice will allow 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 RNA 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 applied to human biological-surveillance efforts such as testing serum and nasal samples from military personnel returning from overseas deployment for the presence of exotic microbes.

Mission Relevance

In support of the Laboratory's mission focus area of biosecurity, this project will enable, for the first time, the potentially pre-symptomatic detection of emerging and bioengineered 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.

FY13 Accomplishments and Results

In FY13 we (1) established an additional laboratory to accommodate workload, and infected 50 mice with the Sindbis virus and collected blood for analysis; (2) designed and tested primers and probes for Taqman real-time assay with the polymerase chain reaction system, and quantitated viral loads in blood of infected mice via real-time polymerase chain reaction at different times after infection; (3) assayed capture of virus bound to IgM using IgM-binding columns and beads, as well as nuclease treatment for additional reduction; (4) measured host RNA and viral RNA present before and after treatment using quantitative polymerase chain reaction, and the most sensitive assay for detection of host RNA was identified; (5) developed three quantitative polymerase chain reaction assays for viral RNA detection; (6) tested two cell lines of mouse macrophage for infectivity with Sindbis virus; and (7) generated lysate from the most productively infected cell line for analysis of the third version of TLRs.

Proposed Work for FY14

In FY14 we will (1) continue to develop methods to rapidly isolate pathogen genetic material captured by host TLRs and IgM; (2) develop techniques to recover the Sindbis virus from infected mice through purification of IgM, recover Sindbis viral RNA from infected mice macrophages through the purification of TLR molecules, and recover Sindbis viral RNA from infected mice through purification of the third version of TLR molecules; and (3) use quantitative real-time polymerase chain reaction and meta-genomics to measure recovery and purity of yield, respectively.

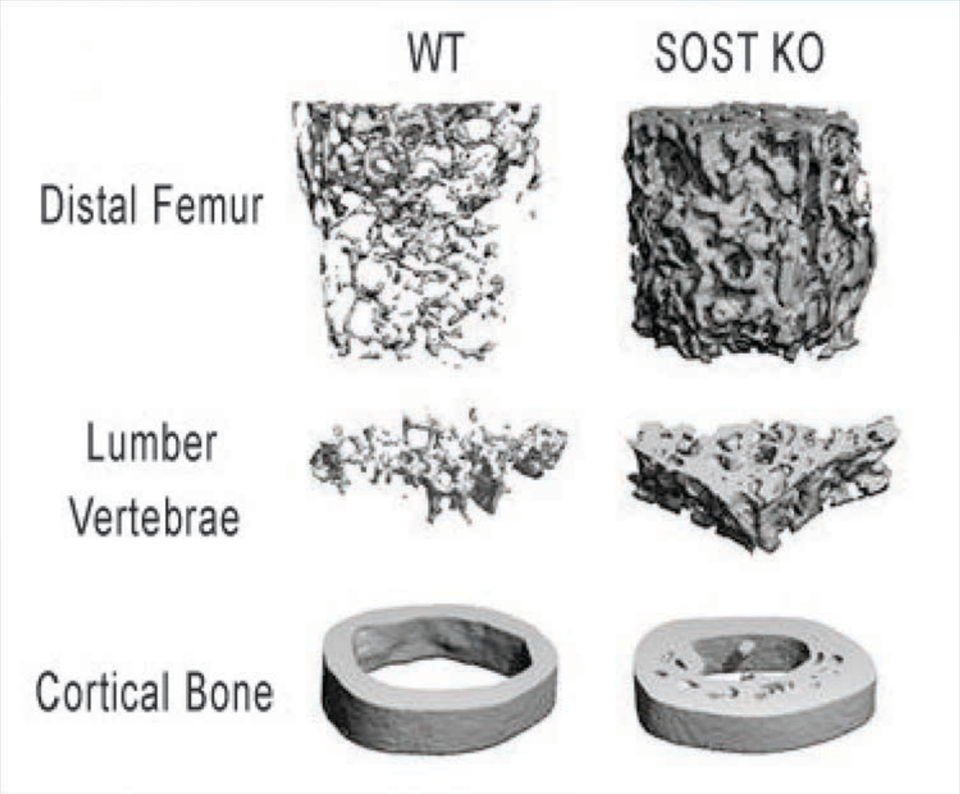
Optimizing Drug Efficacy through Pharmacogenomics-Driven Personalized Therapy

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 make-up 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 before 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



Calcium metabolic profiling can distinguish genetically mutant mice with high bone mass. Mice with an inoperative (knockout or KO) SOST gene (the gene provides instruction for producing sclerostin, which inhibits bone formation) have three to four times more bone mineral density than normal mice as visualized by micro-computed tomography of femurs and vertebrae (left). The SOST gene knockout mice released significantly different amounts of calcium-45 for one to three days post administration, which reflects the elevated metabolic rate from this mutation (right).

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 preparation for a chemical, biological, or radiological terror attack. Lawrence Livermore is the world leader is 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.

FY13 Accomplishments and Results

In FY13 we (1) established a highly sensitive isotope assay for calcium-41 and calcium-45 measured by accelerator mass spectrometry and liquid scintillation, respectively, and measured bone formation in wild-type and inoperative (knockout) SOST-gene mice (the SOST gene provides instruction for producing the protein sclerostin, which inhibits bone formation); (2) examined bone formation in wild-type mice treated with parathyroid hormone daily, and determined that calcium-45 can be used to quantify bone formation; (3) optimized sample preparation for highest sensitivity to determine bone formation as a function of drug response in wild-type and SOST knockout mice; (4) optimized drug response in cell culture and RNA isolation for the RNASeq application that generates a comprehensive, quantitative view of the entire RNA portion of the transcriptome; and (5) determined the normal pharmacokinetic profiling of parathyroid hormone drug metabolism in wild-type mice using iodine-125 labeled parathyroid hormone.

Proposed Work

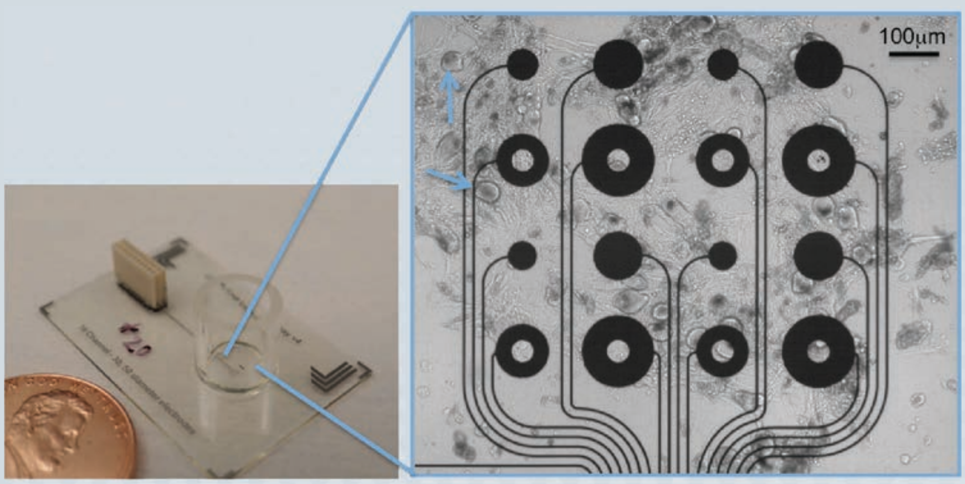
In FY14 we will (1) dose osteoblast bone synthesis cells isolated from three strains of mice with parathyroid hormone and Wnt3A signal transduction protein, isolate RNA, and sequence it; (2) computationally analyze the RNA sequence data to identify novel targets of Wnt protein and parathyroid hormone signaling; (3) predict therapeutic targets; and (4) using pharmacokinetic parameters determined in FY13, treat six strains of mice with parathyroid hormone and dose with calcium-41 and calcium-45.

In Vitro Chip-Based Human Brain Investigational Platform (iCHIP)

Satinderpall Pannu (13-ERD-045)

Abstract

The safety, efficacy, pharmacokinetics, and pharmacodynamics of medical countermeasures to biological or chemical attacks must be characterized in pre-clinical trials that include lengthy protocols involving animal testing. Frequently, the animal models do not accurately predict the clinical safety, efficacy, and tolerability in humans. With a multidisciplinary team of engineers, chemists, and biologists, we propose to develop and demonstrate a novel system that combines primary human cells, tissue engineering, and novel microfluidics to create the first in vitro platform to



Experimental wells for our In vitro chip-based human brain investigational platform (shown at left) contain live human tissue. Dorsal root ganglion cells inside a well are attached to microelectrodes (black circles at right) that stimulate and record tissue response.

reproduce in vivo physiological response of human exposure to neurological toxins. This work will lay the foundation for the ultimate objective of highly integrated, multiple-organ, human-relevant, tissue-based assays for rapidly assessing and predicting the toxicity, safety, and efficacy of new drug entities with the goal of accelerating both development and regulatory approval of medical countermeasures against chemical, biological, and radiological agents.

We intend to essentially create a human brain investigational platform on a chip (iCHIP). We are motivated by the need to rapidly develop medical countermeasures for chemical and biological attacks—currently, the timeline to develop such responses is lengthy. Successfully demonstrating a microfluidic device focused on the long-term survival and functionality of human dorsal root ganglions (part of the peripheral nervous system) is a crucial first step towards building a multiple-organ, fully integrated platform that mimics human response to exposures of chemical, biological, and radiological agents, allowing for the rapid and efficient development of countermeasures. We expect that these accomplishments will ultimately enable a platform to include fully integrated tissue constructs of the lung, liver, heart, and circulatory system, creating a much-needed tool for rapid toxic countermeasure development.

Mission Relevance

Our research addresses LLNL's biosecurity mission in the area of medical countermeasures for rapid mitigation of evolving and unknown threats by enabling timely research into medical countermeasures for response to a terrorist attack employing chemical or biological agents.

FY13 Accomplishments and Results

In FY13 we (1) completed the delivery of an in vitro platform capable of maintaining and repeatedly interrogating human and rodent neuronal tissue, with a modular platform that incorporates tightly controlled conditions in the cell growth chamber to optimize cell growth, a microelectrode array for noninvasive electrophysiology recording, high-resolution bright-field and fluorescent microscopy to monitor cell health and response to chemical exposures, and microfluidics for automated fluid exchange; (2) developed methods to increase the viability of both human and rat dorsal root ganglions on the platform from 2 days to greater than 18 days by modifying the standard cell-plating surface coating, adjusting cell seeding density, modulating the population of support cells, and tightly controlling media concentration of the osmotic solution; and (3) assessed neuron health by exposing cells to various chemicals known to elicit neuronal response—action potentials are clearly visible on the electrodes where the cells are located, and analysis of the response shows the expected action potential.

Project Summary

The preliminary demonstration of a microfluidic device for the long-term survival and functionality of human dorsal root ganglion cells was successful and is a crucial first step towards building a multiple-organ, fully integrated platform that mimics human response to exposures of chemical, biological, and radiological agents, allowing for the rapid and efficient development of countermeasures. The viability and phenotype of the human dorsal root ganglion cells were verified using four chemicals that elicit neural responses.

Publications and Presentations

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

Jennifer Pett-Ridge (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. 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 documentation will be 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.

FY13 Accomplishments and Results

In FY13 we made significant progress towards understanding the metabolic flexibility of Thaumarchaea. Specifically, we (1) designed 1,952 probes targeting 80 groups of amoA, and printed this probe set onto a microarray for use in mRNA Chip-SIP; (2) collected 80 L of Pacific surface water and performed 8 experiments with amino acids labelled with $H^{13}CO_3^-$(hyperpolarized bicarbonate) and nitrogen-15; (3) designed experiments, with collaborators at the University of Aberdeen, amending soil with $H^{13}CO_3^-$; (4) designed 1,127 probes targeting 56 groups of Euryarchaea, 2,053 probes targeting 101 groups of Thaumarchaea, and 691 probes targeting 28 groups of ammonia-oxidizing bacteria; (5) printed these, along with the amoA probe set, onto microarrays and performed mRNA and rRNA Chip-SIP on these samples; and (6) collected and began analysis of geochemical samples.

Proposed Work For FY14

In FY14 we will (1) submit a manuscript for publication with results of the pure-culture whole-genome Chip-SIP performed in FY13; (2) perform carbon-13 and nitrogen-15-substrate incubations with California grassland soils and marine samples; (3) design and print rRNA and mRNA microarrays specific to these communities; (4) perform mRNA and rRNA Chip-SIP on these samples to determine if and when Thaumarchaeota in the environment grow autotrophically, heterotrophically, or with a mix of both; and (5) collect geochemical data in parallel that documents the production of nitrous oxide in these samples during incubation, to determine the effect of carbon utilization on its production.

Publications and Presentations

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

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

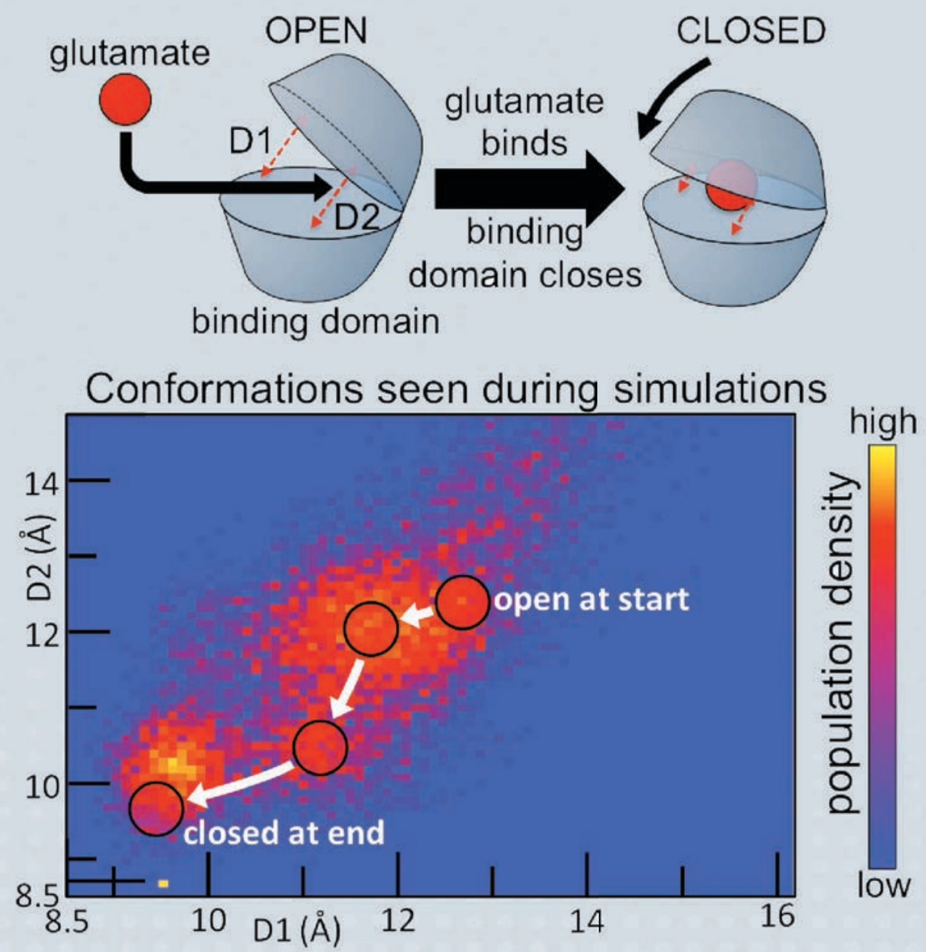
Timothy Carpenter (13-LW-085)

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 propagates 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, we will observe the opening mechanism of NMDAR and provide a biologically relevant



The glutamate-binding domain of the AMPA receptor involved in synaptic transmission exists in an open state, with a binding site present between the two "lobes" of the domain. Upon binding of glutamate, the two lobes move down around glutamate and reach a closed conformation. We have been able to demonstrate the pathway through which this closing occurs by carrying out extensive atomistic simulations, whereby glutamate was added to the open conformation. The closing mechanism appears to be nonlinear, and instead progresses via one or two intermediates. This illustrates that the glutamate binding and activation process is more complicated than previously thought.

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 a membrane protein ever performed.

Mission Relevance

As well as 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.

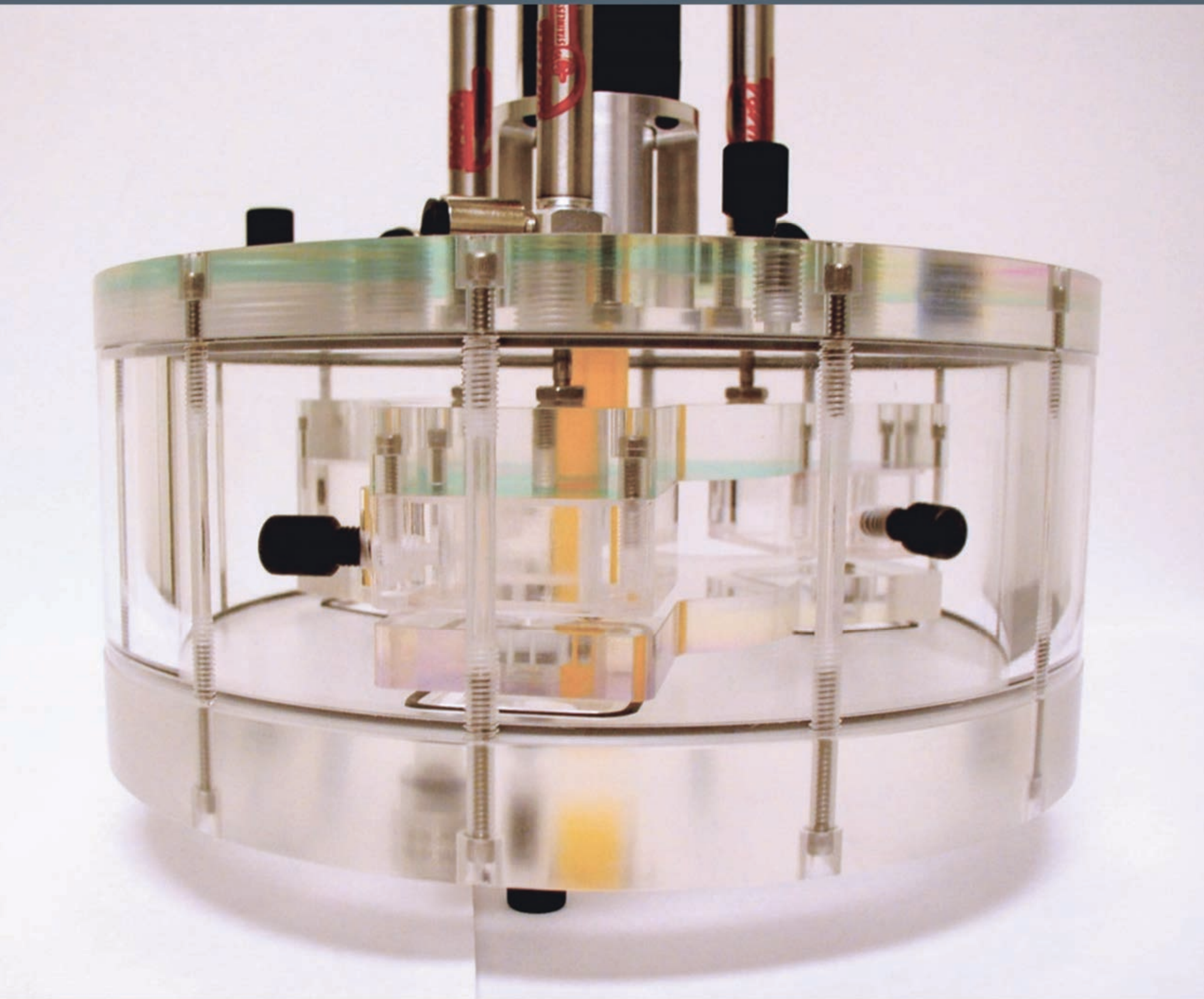
FY13 Accomplishments and Results

In FY13 we (1) built a complete model of the closed-state glutamate receptor AMPAR (which also plays a key role in synaptic transmission) and extensively tested model stability; (2) performed comprehensive testing and simulation on the AMPAR and NMDAR ligand-binding designs to optimize simulation conditions; (3) employed various computational methods to close both ligand-binding designs, revealing two distinct possible mechanistic activation pathways; and (4) optimized the nudged elastic-band code on Livermore computing resources to determine which pathway is lower energy or physiologically relevant. During the course of the research it became apparent that deduction of this pathway will provide many intermediate steps with which to drive channel opening in a much smoother fashion, thereby establishing the opening pathway and open model simultaneously.

Proposed Work for FY14

In FY14 we will (1) complete the nudged elastic-band pathways for closing or activating the AMPAR and NMDAR clamshell ligand-binding designs—the intermediate steps from these low-energy (physiologically relevant) pathways will be used for a much more detailed, comprehensive, stepwise activation of the entire AMPAR and NMDAR structures using targeted computational techniques; (2) generate the open state of the receptor via the more physiologically relevant pathway; (3) use additional computational techniques to relax and minimize this pathway for the whole protein system; and (4) employ docking of potential open-channel blockers for various structures in the opening pathway to identify possible binding sites.

Chemistry



Laboratory Directed Research and Development

Annual Report FY2013

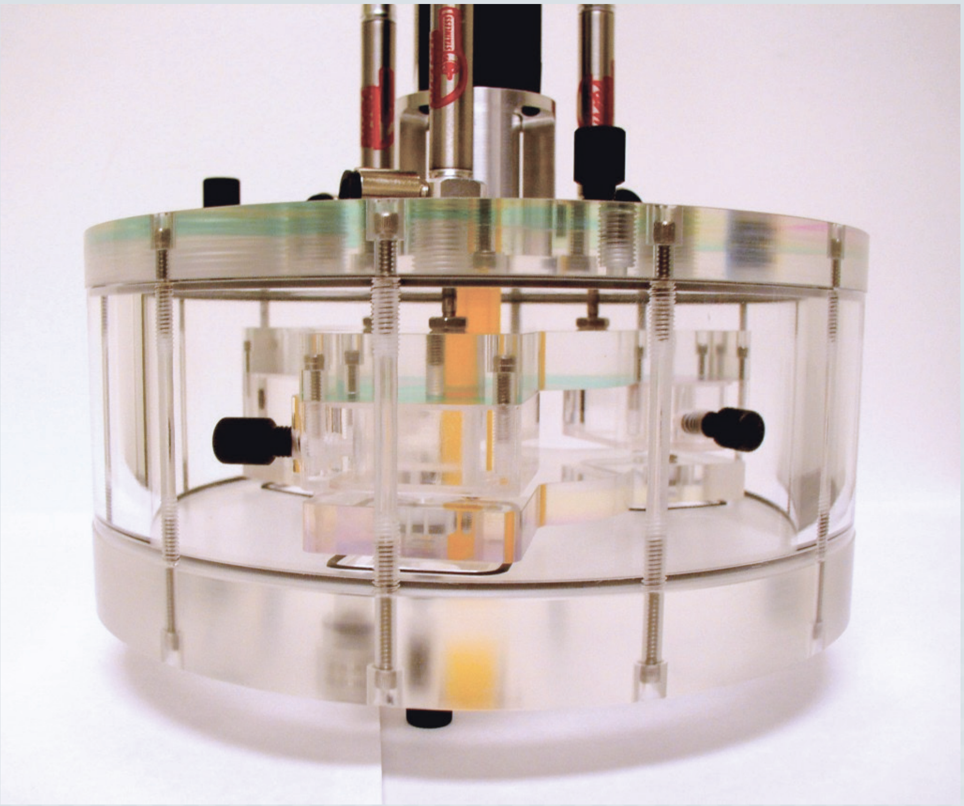
Fundamental Chemical Behavior of Superheavy Elements Through Applications of Online Isotope Production and Automated Chemical Systems

Dawn Shaughnessy (11-ERD-011)

Abstract

Our goal is to study fundamental chemical properties of the heaviest elements. In particular, we will explore reported modifications, caused by relativistic effects, of group behavior in the periodic table for certain transactinides (elements 104–106), and observe the unknown behavior of element 114 in aqueous media. We propose to develop two chemical separation methods for heavy elements with atomic numbers 104 to 106 and 113 to 115, as well as implement our previously developed rapid automated chemistry prototype at various accelerator facilities for fully integrated isotope production, sampling, and analysis. Studies of the lighter homolog elements will be performed at Livermore's Center for Accelerator Mass Spectrometry (CAMS), and heavy-element experiments will be done at the Flerov Laboratory of Nuclear Reactions in Dubna, Russia or the GSI Helmholtz Centre for Heavy Ion Research in Germany.

We expect to develop chemical separations for transactinide elements. The use of both new materials and previously established resins will be investigated for applicability



The GLITTAR (gas–liquid Interface for transferring transactinides to automated radiochemistry) is an interface device that transports reaction products from an accelerator to an automated radiochemistry apparatus. GLITTAR can be adapted to several different accelerators and will be used at the cyclotron at Texas A&M University for heavy-element homolog chemistry experiments.

to heavy elements. We will implement an interface between the CAMS beam line and our automated chemistry system so the chemical methods we develop can be used for online separation of lighter homolog elements, which will establish trends in the behavior of chemical groups. Then we expect to perform long-term runs on the transactinides and evaluate how their chemistry compares to that of their lighter homologs. If modified behavior is observed, relativistic effects will be confirmed. This would also result in the first reported aqueous chemistry of element 114.

Mission Relevance

This research advances the Laboratory's strategic mission of nuclear threat reduction by addressing the challenge of developing autonomous, real-time forensic methods designed for field deployment. This project will also serve as the foundation for future opportunities in such areas as medical isotope production, chemical purification, inertial-confinement fusion diagnostics, and isotope harvesting. The study of heavy elements is a key component of fundamental research in nuclear chemistry and radiochemistry, which are core competencies at LLNL.

FY13 Accomplishments and Results

In FY13 we (1) participated in an experiment at GSI to search for element 119—although no events were detected, a limit on the cross section was established; (2) worked with Texas A&M University on a location to site on-line chemistry facilities and participated in experiments to measure the transfer efficiency of the recoil transfer chamber; (3) developed separation methodologies for elements 113, 114, and 115 using crown ether extractants; and (4) continued setup of online chemistry at CAMS, which included target chamber testing and programming of the chemical interface.

Project Summary

By the conclusion of this project, we made advances toward online chemistry of the heaviest elements. We have constructed a target chamber for use at CAMS and a chemistry interface that will take reaction products from an accelerator to our automated chemistry apparatus. The interface has been programmed and successfully tested off-line. Chemical separation development has progressed as well, with separation schemes determined for elements in groups 13, 14, and 15 that have kinetics rapid enough to separate short-lived heavy element atoms from background materials. While delays at Texas A&M did not permit us to run integrated chemistry experiments, we have all of the tools ready to perform these experiments at a wide variety of facilities. We participated in experiments at GSI that have confirmed discovery of elements 115 and 113, which will allow the collaboration to name these elements in the future. We will continue to collaborate with the University of Nevada, Las Vegas, and Texas A&M University on the chemistry of superheavy elements through scholar programs.

Publications and Presentations

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Tereshatov, E. E., et al. 2012. "Procedures for Db chemical characterization in off-line experiments." *J. Radio and Nuc. Chem.* **293**(1), 331. LLNL-JRNL-519233.

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Detonation Performance of Improvised Explosives via Reactive Flow Simulations and Diamond Anvil Experiments

Sorin Bastea (11-ERD-067)

Abstract

In this project we propose to establish and test a methodology for elucidating the detonation behavior of nonstandard, or improvised, explosive mixtures of fuel plus oxidizer that couples reactive flow simulations with targeted diamond anvil experiments. Although the properties of such mixtures were first considered more than 50 years ago in connection with rocket propulsion and mining operations, numerous questions remain regarding their exo-energetic potential. Theoretical studies of improvised explosives are rare, and their experimental study is expensive and technically challenging. Such explosives are currently of great interest, however, because of security concerns. We plan to develop a thermodynamics and kinetics model for the behavior of hydrogen peroxide and nitromethane mixtures, based on available as well as new experimental data on hydrogen peroxide. The model will be employed in reactive flow simulations to determine detonation capability and critical charge size as a function of composition and confinement.

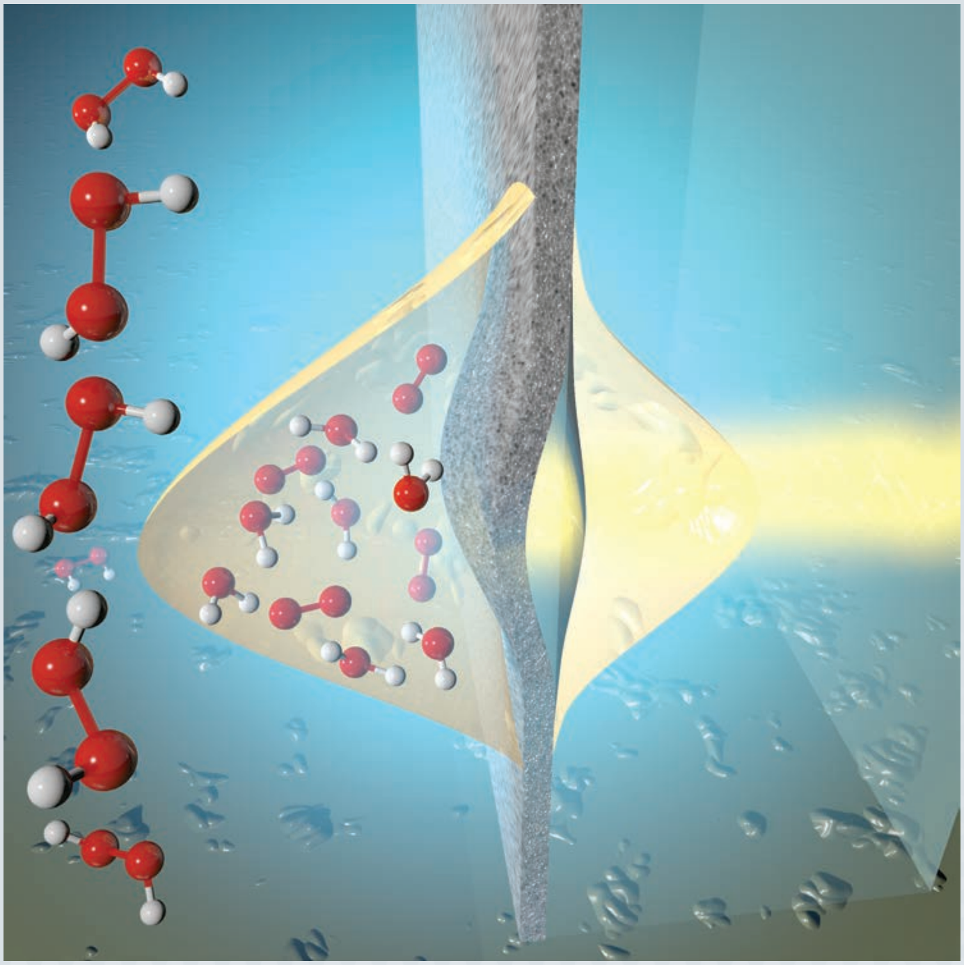
Hydrogen peroxide is a common oxidizer that has been shown to yield powerful energetic formulations when mixed with organic fuels. We expect to provide a thermodynamic and kinetic description of this material and a model for its behavior under detonation conditions. We will perform reactive flow simulations to validate this model in oxidizer and fuel mixtures, and establish such simulations as a tool for determining detonation capability and performance for improvised explosives. Further analysis of the results may yield criteria for detonation capability and performance applicable to a wide range of mixtures and may thus facilitate rapid screening. The Cheetah thermochemical model will be expanded to include potassium and sodium, thus enabling calculations for a variety of solid oxidizer and fuel mixtures.

Mission Relevance

Hidden bombs or improvised explosives have caused over half of all combat casualties in Iraq and Afghanistan. Our proposed project will constitute a step forward in filling the current knowledge gap for these explosive mixtures and expand thermochemical modeling to a new class of energetic materials, in support of the Laboratory's central mission to reduce or counter threats to national and global security.

FY13 Accomplishments and Results

In FY13 we (1) performed new ultrafast shock experiments on an oxygen-balanced hydrogen peroxide and nitromethane mixture aimed at understanding its behavior at low- and moderate-particle velocities; (2) fully validated the experimental method—using the original laser system because of technical difficulties in installing the new laser—in the unreacted regime and provided valuable data for modeling this mixture at detonation conditions and understanding strain effects at high pressures; (3) completed quantum molecular dynamics simulations on oxygen-balanced and oxygen-rich mixtures that yielded a new chemical kinetics mechanism and reaction rates that help elucidate the detonation behavior of such liquid energetic mixtures;



Ultrafast shock initiation of exothermic chemistry in hydrogen peroxide.

(4) developed unreacted equations of state for hydrogen peroxide and nitromethane based on experiments and simulations leading to consistent modeling of reactant and product mixtures, which is essential for reactive flow calculations; and (5) used the equations of state together with the chemistry rates in reactive flow simulations of pure liquids and the oxygen-balanced mixture, to yield detonation behavior for different charge sizes and an estimate of failure diameter.

Project Summary

The successful completion of this project resulted in the validation of the ultrafast shock compression technique for elucidating the equation of state and chemical kinetic properties of energetic liquid mixtures from the low-pressure, unreacted regime to shock-induced chemistry at high pressures. The experiments conducted on hydrogen peroxide show, for the first time, that chemical kinetics at high pressure proceed on timescales of hundreds of picoseconds and that strain rate plays an important role in the initiation of chemistry. This latter result may enable the development of ultrafast methods for assessing sensitivity and perhaps even performance in such energetic formulations, and could open new avenues for the shock synthesis of materials. Quantum molecular dynamics simulations provided a direct window into the chemical kinetic processes evidenced by the experiments, and suggested a novel, slow nitrogen–oxygen kinetic mechanism in oxygen-balanced energetic mixtures. Implementation of experimental and simulation data enabled reactive flow simulations of hydrogen peroxide and nitromethane mixtures and estimation of the failure diameter. By integrating experiments, microscopic simulations, and reactive flow calculations, this project developed important experimental data and simulations for characterizing and modeling the detonation behavior of explosives. The Livermore Advanced Simulated Computing and the joint Department of Defense and DOE Munitions Technology Development programs will benefit from these advances and plan to provide support for their future development and implementation.

Publications and Presentations

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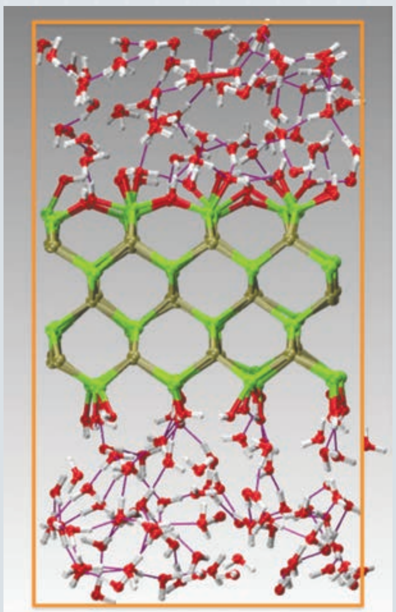
Ab Initio Study of the Water–Semiconductor Interface for Photo-Electrochemical Hydrogen Production

Tadashi Ogitsu (11-ERD-073)

Abstract
Photo-electrochemical hydrogen production from water represents one of the most promising emerging technologies for the production of chemical fuel from sunlight. However, a lack of understanding of the processes governing the hydrogen evolution reaction and photo-corrosion behavior has impeded practical implementation. We propose using ab initio simulations based on density-functional theory to obtain a detailed understanding of the microscopic hydrogen evolution reaction and corrosion mechanisms at the electrode–electrolyte interface. By analyzing the chemistry, structure, and dynamics of these systems and through the identification of structure–property relationships, we aim to enable development of practical photo-electrochemical devices.

Successful completion of the project will provide detailed information on microscopic properties of the surfaces of semiconductor elements in groups III through V of the periodic table as well as general structural and dynamical properties of water–semiconductor interfaces. In addition, we aim to gain an understanding of the stability of various surface oxides, the gas-phase water adsorption on those surfaces, and the effects of nitrogen impurities on surface morphology and energetics. This will provide valuable insight into the relevant microscopic mechanisms of photo-catalytic hydrogen evolution reaction and surface corrosion, which has been largely missing in systematic attempts to improve photo-electrochemical device performance.



A snapshot of ab initio molecular dynamics simulations of the water–indium–phosphide interface, where formation of strong hydrogen bonds between surface adsorbates and water is observed. The interface is stabilized by strong hydrogen bonding with water, causing formation of an ice-like structure in which proton hopping is enhanced.

Mission Relevance
Development of efficient, environmentally responsible solar-to-chemical energy conversion has been identified as a strategic DOE goal for a secure, sustainable energy future. This project directly addresses this as well as the Laboratory's mission to enhance the energy and environmental security of the nation, with direct conversion of solar energy to chemical fuel by combining stable photo-electrochemical operation with suitable efficiency.

FY13 Accomplishments and Results
In FY13 we (1) continued the theoretical investigation of the chemical environment of nitrogen implanted in a galadium–indium–diphosphide semiconductor using the nitrogen low-energy bombardment method at about 550 eV, including developing a rational way to deduce the chemical environment of nitrogen and estimating the modulation of nitrogen 1s electron-shell core level in the chemical environment; (2) calculated x-ray emission spectra using code provided by The Molecular Foundry user program; (3) identified the chemical environment of nitrogen impurities in the periodic groups III to V semiconductors by comparing the theoretical and

experimental spectra; and (4) determined that nitrogen is not solely responsible for the reported improvement of corrosion resistance—it is most likely because of unintended metal impurities as well as nitrogen.

Project Summary

The successful conclusion of this project resulted in the elucidation of the nature of hydrogen-bond network dynamics at the water–indium–phosphide and water–galadium–phosphide interfaces. Based on these results, together with recent experimental results on hydrogen spillover on a silicon-based photo-electrochemical device, it was suggested that proton transport at the water–electrode interface might have a significant impact on the hydrogen evolution reaction. In addition, we were able to identify the relevant chemical environments for the nitrogen-bombarded galadium–indium–diphosphide samples that were prepared by the National Renewable Energy Laboratory, based on x-ray energy spectroscopy measured by the University of Nevada, Las Vegas. As a result of this work, DOE's Energy Efficiency and Renewable Energy photo-electrochemical hydrogen production program has begun supporting our future research in this area. Our most recent results have also led to the submission of another proposal to DOE to continue our investigation of the corrosion resistance work and we will pursue upcoming calls from the DOE in related research areas. We are also in the process of establishing a cooperative research and development agreement with Japan's National Institute of Advanced Industrial Science and Technology to further advance photo-electrochemical hydrogen production technology.

Publications and Presentations

Choi, W., et al., 2013. "Site-dependent free energy barrier for proton reduction on MoS_2 edges." *J. Phys. Chem. C.* **117**(42), 21772. LLNL-JRNL-639087.

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Predicting Weapon Headspace-Gas Atmosphere for Modeling Component Compatibility and Aging

Elizabeth Glascoe (12-ERD-046)

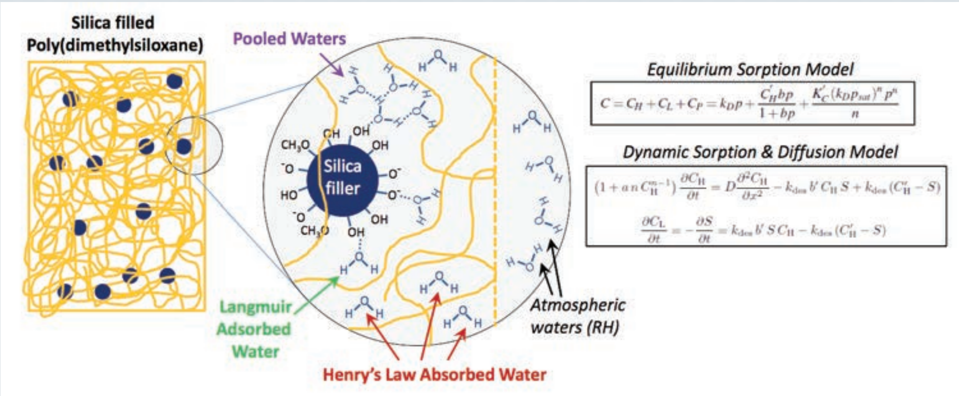
Abstract

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. 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 will enable greater predictive foresight in selecting replacement materials for stockpile life-extension programs.



Predicting the aging and compatibility of materials requires detailed modeling of sorption, diffusion, and chemical reactions of vapors within the materials. The dynamic sorption and diffusion model developed in this work is the cornerstone of our reactive transport model.

FY13 Accomplishments and Results

In FY13 we (1) characterized the diffusion and sorption parameters for water vapor in three sealant and emulsion materials (RTV-732, Halthane 88-3, and Celanese-VAE) and prepared LX-16 polymer-bonded explosive samples for upcoming experiments; (2) developed a continuum-scale model and a computer code for describing the vapor diffusion in a heterogeneous material and utilized the model to investigate sorption and diffusion in poly(dimethlysiloxane) polymers; (3) designed and executed validation experiments of moisture outgassing and diffusion through the Zircar RS1200 refractory composite and Sylgard 184 elastomer in various arrangements; (4) initiated chemical kinetic experiments of the moisture induced-condensation reaction of RTV-732, which produces acetic acid as a volatile product; and (5) modeled the kinetic mechanisms of moisture reactions with ethylene-vinyl acetate polymers and poly(dimethlysiloxane) polymers using molecular dynamics simulations.

Proposed Work for FY14

In FY14 we will (1) execute complex validation in which multiple materials are aged together in a sealed assembly and characterized based on headspace-gas constituents, (2) validate the compatibility model against validation experiments, and (3) further characterize sorption, diffusion, and chemical kinetics of materials if discrepancies exist between validation experiments and models.

Publications and Presentations

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Harley, S. J., et al., 2013. *Investigation of competitive degradative mechanisms in ethylene-vinyl acetate copolymers.* Service Life Prediction of Polymeric Materials: Vision for the Future, Monterey, CA, Mar. 3–8, 2013. LLNL-ABS-618973.

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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 (NIF) 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 NIF. 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 NIF by helping to interpret current and proposed ramp-

compression experiments using carbon-based foams, and by making predictions for future NIF capsule experimental designs.

Mission Relevance

This project supports the Laboratory's missions in national 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 basic science mission.

FY13 Accomplishments and Results

In FY13 we (1) continued our tight-binding parameter development, (2) began to simulate both diamond and the glow discharge polymer analogues under shock compression to timescales close to chemical equilibrium, and (3) began adiabatic expansions of both shock-compressed mixtures.

Proposed Work for FY14

In FY14 we will (1) conclude the tuning of our new density-functional tight-binding models; (2) create a general capability for simulating carbon–hydrogen–oxygen bonded systems under a wide range of pressures, temperatures, and dynamic strains; (3) use our simulations to elucidate the nanosecond timescale behavior of a number of materials under both shock compression and release related to experiments, including carbon and hydrocarbon foams; (4) compute bulk properties of these materials and address how they change with time as the kinetics evolve; and (5) conduct simulations of foams with varying amounts of hydrogen and oxygen to estimate the effects of chemical kinetics on the equation of state for ramp-compression experiments.

Publications and Presentations

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New Energetic Materials

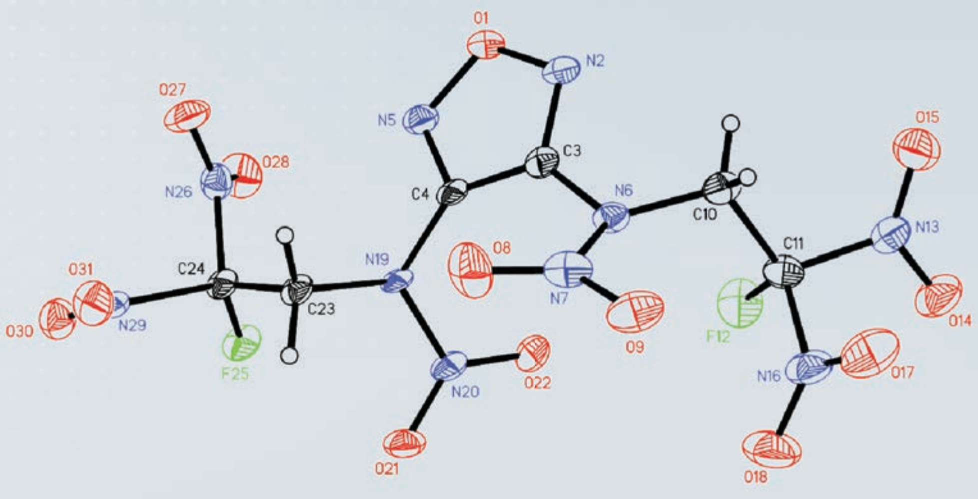
Philip Pagoria (12-ERD-066)

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 the Laboratory's mission in 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.

FY13 Accomplishments and Results
In FY13 we (1) synthesized several new compounds (LLM-204, LLM-208, and LLM-213) consisting of heterocycles substituted with 1,1,1-trinitroethyl and 1,1,1-fluorodinitroethyl moietites; (2) pressed LLM-204 into pellets and measured its heat of combustion at −5364 kJ/mol; (3) characterized LLM-208 and scaled up synthesis to 25 to 50 g; (4) pressed LLM-208 into pellets and measured the heat of combustion at −2891 kJ/mol; (5) prepared LLM-209 with a density of 1.94 g/cm^3, small-scale safety test responses similar to HMX explosive, and predicted performance equal to CL-20 explosive; and (6) began to investigate new potential plasticizer materials with the synthesis of LLM-212, a fairly dense nitrate ester that did not posses the required phase transition properties.



Crystal structure of LLM-209, a new highly energetic compound.

Proposed Work for FY14

In FY14 we will (1) continue to investigate new energetic compounds consisting of highly oxidized, nitro-substituted heterocycles containing pendant 1,1,1-fluorodinitroethyl or 1,1,1-trinitroethyl moieties as possible ammonium perchlorate replacement compounds; (2) continue efforts to synthesize a variety of heterocyclic precursor compounds with functional groups that may be converted to the fluorodinitromethyl, dinitromethyl, and trinitromethyl moiety; (3) investigate more thoroughly the flurodinitroethyl derivatives because of our discovery that they have improved thermal stability; and (4) scale up the synthesis of promising target compounds and perform testing on these new compounds.

Publications and Presentations

DeHope, A., P. F. Pagoria, and D. Parrish, 2013. *New polynitro alkylamino furazans*. 16th Int. Seminar New Trends in Research of Energetic Materials, Pardubice, Czech Republic, Apr. 10–12, 2013. LLNL-CONF-624954.

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DeHope, A., et al., 2012. *Energetic materials synthesis at LLNL.* LLNL-PRES-554571.

DeHope, A., and P. F. Pagoria, 2012. *Synthesis of 1,2,4-oxadiazole derivatives as energetic compounds.* LLNL-POST-560934.

Determining the Coulomb Potential in Electronic-Structure Calculations

Antonios Gonis (12-ERD-072)

Abstract

We propose to develop a code based on an analytic solution of the longstanding problem of self-interaction to determine the potential from Coulomb interaction—that is, the potential acting on an electron because of Coulomb interaction between electrons in a material—whether at a level of atoms, molecules, or bulk solid. We will conduct the research within the Kohn–Sham formulation of density functional theory and develop a single generic coding subroutine for the case of periodic materials, then finalize its development with respect to finite systems such as atoms. We will provide a unique, analytic, and computationally rigorous method for electronic structure calculations that is consistent with strict requirements of quantum mechanics and that is no more difficult computationally than other methods currently in use, which are mostly numerical and invariably approximate.

We expect to produce a well-tested subroutine for calculation of the Coulomb potential portable to multiple codes for electronic structure. We anticipate that development of a generic body of coding will have significant impact on the field of electronic-structure calculations at the Laboratory and in the overall community of researchers studying electronic structure and materials properties. At the Laboratory, for example, this coding will provide definitive answers applicable to actinide materials.

Mission Relevance

Our project will enable the study of electronic structure and its effects on materials of intense interest to Lawrence Livermore, such as the actinides, in support of LLNL's central mission in stockpile stewardship.

FY13 Accomplishments and Results

In FY13 we focused on extending the formalism of calculating exchange potential derived from the Coulomb repulsion in Kohn–Sham density functional theory to periodic systems. Specifically, we developed the code for determining density within a Green's function formalism through integration in the complex energy plane, and developed the formalism for determining the exchange energy through complex integration.

Project Summary

With this project, we determined the viability of a newly proposed methodology for calculation of the Coulomb potential in electronic-structure calculations based on the Kohn–Sham formulation of density functional theory. Our calculation subroutine avoids completely the so-called self-interaction error. We applied the method to all atoms in the periodic table and have formulated it for applications to solids. The project was terminated because of funding cuts as we were beginning to implement the methodology to extended systems (i.e., periodic solids).

Publications and Presentations

Daene, M., et al., 2013. *Computationally simple, analytic and closed form solution of the self-interaction problem in Kohn–Sham density functional theory.* LLNL-POST-638881.

Daene, M., et al., 2013. *Solving the self-interaction problem in Kohn–Sham density functional theory: Application to atoms.* LLNL-JRNL-635885-DRAFT.

Gonis, A., et al., 2012. "Computationally simple, analytic, closed form solution of the Coulomb self-interaction problem in Kohn–Sham density functional theory." *Solid State Comm.* **152**(9), 771. LLNL-JRNL-548640-DRAFT.

Gonis, A., et al., in press. "Self-entanglement and the dissociation of homonuclear diatomic molecules." *Mol. Phys.* LLNL-JRNL-636417-DRAFT.

High-Explosive Components Using Advanced Manufacturing Methods

Alexander Gash (13-ERD-051)

Abstract

We propose to develop the methodology and expertise required for 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.

FY13 Accomplishments and Results

In FY13 we (1) optimized particle size distribution to enable continuous and fine-feature direct ink writing of mock high explosive; (2) studied the influence of two micro-reactor designs on the crystallization behavior of high-explosive surrogate material; (3) demonstrated the synthesis of a mock-explosive high-solids-loaded ink (greater than 74% by weight solids); and (4) procured and received a direct ink writing system for dedicated use with high explosives.

Proposed Work for FY14

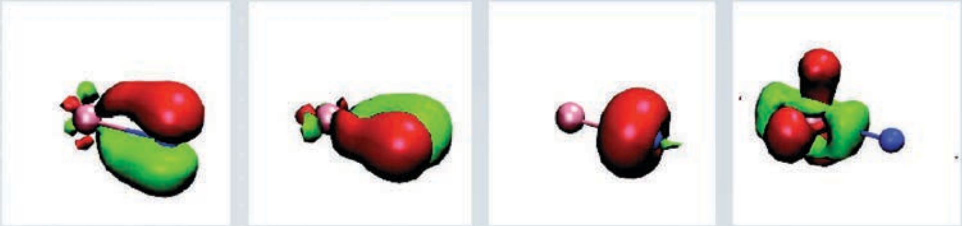
In FY14 we will (1) complete a parametric study on the direct ink printing of greater than 80% solids-loaded mock high explosive; (2) optimize the synthesis of LLM105 energetic in a flow chemistry system; (3) study the influence of two new micro-reactor designs on the crystallization behavior of high-explosive material; (4) integrate the synthesis and crystallization steps to a continuous process; (5) install and demonstrate the operation of a direct ink writing system with a high explosive; (6) demonstrate synthesis of high-explosive, high-solids-loaded ink; and (7) demonstrate printing by direct ink writing of the high-explosive material developed with micro-reactors.

Experimental and Ab Initio Theoretical Search for Lanthanide Covalency for Enhanced Rare Earth Separations

Patrick Huang (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



Natural orbitals (shown in red and green) for the uranium and nitrogen bond in tris(imido) complexes. Uranium is represented as a pink sphere, nitrogen as blue. The interaction is made up of two covalent pi-type bonds (left two panels), plus a sigma-type dative interaction involving the donation of a nitrogen lone pair (third from left) into an unoccupied uranium orbital (right panel).

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.

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 elements. 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 at Lawrence Livermore.

FY13 Accomplishments and Results

In FY13 we (1) concluded that single-reference perturbation theory yields a good description of lanthanide bonding to ligands as compared to the high-level methods such as the multiple-reference perturbation theory, based on benchmark studies involving a series of model lanthanide–cytosine complexes; (2) explored ligands involving second-row main group atoms (p and s shells), finding an increase in covalent character compared to the *n*-atom ligands; and (3) performed experimental work to focus on s-atom ligands and third-row atoms such as selenium.

Proposed Work for FY14

For FY14 we will (1) continue experimental efforts to synthesize and characterize lanthanide–ligand complexes involving lanthanide–sulfur and lanthanide–selenium bonds, (2) examine the effects of spectator ligands such as cyclopentadienyl in the first coordination sphere, and (3) investigate systematic bonding trends based on our combined experimental results and theory strategy.

Earth and Space Sciences

Laboratory Directed Research and Development

Annual Report FY2013

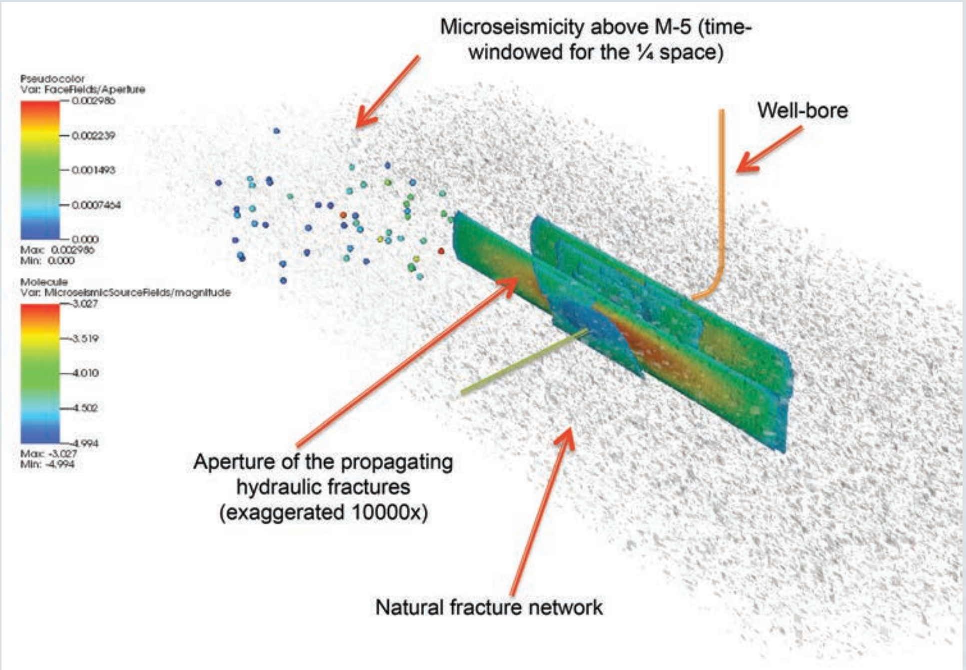
Creating Optimal Fracture Networks for Energy Extraction

Frederick Ryerson (11-SI-006)

Abstract

The technology required to extract the energy resources contained within the Earth`s crust is constantly evolving. Resources 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 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



Our GEOS code simulation of fracture aperture growth and microseismicity produced during hydraulic stimulation of a horizontal well drilled in a shale-gas reservoir. Seismicity associated with the initiation of new fractures has been suppressed to accentuate seismicity associated with reactivation of pre-existing fractures from stimulation-induced variations in the stress regime. Scale bars give seismic magnitude and fracture aperture.

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.

FY13 Accomplishments and Results

We (1) developed a mesh-splitting approach in which fractures pass through elements rather than along element boundaries, allowing realistic models of fracture surface geometry; (2) ran combined problems using both time-implicit and time-explicit solvers for the efficient modeling of fracture events and longer-term events in which implicit solvers are used for matrix deformation, while explicit representations are used at the fracture front; (3) successfully validated a mechanics model of multiple fractures against available experimental data; (4) developed a preliminary model of rate-dependent fracturing high-strain rates transmitted through the fluid phase; (5) obtained improved depth location and seismic moments for events with magnitude scale of 3 to 4 with adjoint-wave seismic data from the Geysers geothermal area; (6) created a synthetic micro-seismic, three-dimensional data set using a fourth-order finite wave propagation code (SW4) to test detection and location algorithms; and (7) implemented coarse-scale three-dimensional fracturing criteria based on a virtual crack closure method to calculate the stress intensity factor.

Proposed Work for FY14

In FY14 we will (1) complete the implicit solver implementation, allowing computationally efficient simulations to be performed at reservoir stimulation times; (2) implement ground motion representations and visualization, which will augment micro-seismic processing, provide complete source-to-station behavior, and allow for direct comparison with field data; (3) add new field-scale fracture criteria to GEOS, along with a deflagration model to simulate pressure pulses associated with energetic materials; (4) develop methodology to simulate transport of so-called proppants (particles driven into created fractures to hold them open during gas production); and (5) model the simulated porous flow of hydraulic fluid or gas in the rock matrix adjacent to fractures.

Publications and Presentations

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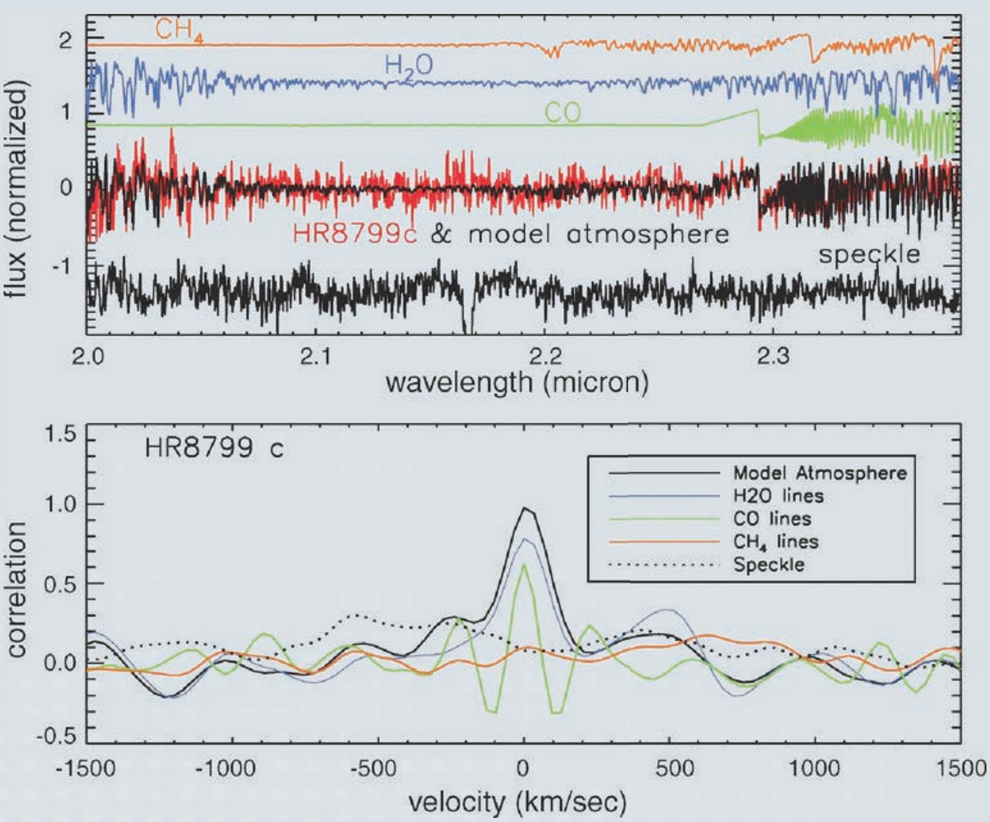
Images and Spectra of Extrasolar Planets from Advanced Adaptive Optics

Bruce Macintosh (11-ERD-048)

Abstract

The newest frontier in studies of planets outside our solar system ("exoplanets") is imaging, which is key to understanding whether solar systems such as our own are common or rare. We propose to study extrasolar planets using adaptive optics and spectroscopy on the Keck and Gemini telescopes. We will characterize the HR8799 multiple-planet system with astronomical and spectroscopic measurements to determine composition and evolution of the planets. In addition, we will develop tools for the unbiased estimates of planetary spectra from noisy hyper-spectral data. We will also lead a 100-night survey campaign on the new LLNL-built Gemini Planet Imager (GPI) adaptive optics system. This new capability will be an order-of-magnitude more sensitive than any to date and could enable the discovery and characterization of as many as a hundred other solar systems.

We expect to produce infrared spectra of the atmospheres of the planets HR8799b and HR8799c—making these the lowest-temperature exoplanets ever spectroscopically characterized—and use the results to adapt models of the planets' complex atmospheres. We will use precision adaptive optics astrometry and computational simulation to find orbital parameters that are consistent with observations and with



Portion of a high-resolution near-infrared spectrum of the extrasolar planet HR8799c. High-pass filtered spectrum compared to molecular species expected in the atmosphere (top). Cross-correlation of the spectrum with molecular templates and model atmosphere is shown at bottom. This is the highest quality spectrum of an extrasolar planet ever obtained.

formation and evolution scenarios. Next, through our large-scale GPI project to survey 600 stars, which is the most sensitive and complete imaging search for planets to date, we will produce detailed characterizations of the atmospheric structure and composition of previously inaccessible classes of exoplanets.

Mission Relevance

This project will advance LLNL's adaptive optics capabilities, which have applications in such fields as internal laser aberration correction, remote sensing, and directed energy, in support of the Laboratory's missions in advanced lasers and national security. Techniques we will develop for extracting faint planetary signals from hyper-spectral data with highly correlated noise will also be applicable to remote sensing applications.

FY13 Accomplishments and Results

In FY13 we (1) obtained high-resolution spectra of exoplanets HR8799b and c, which are the best-quality spectra ever obtained of an extrasolar planet and clearly show individual molecular absorption lines of carbon monoxide and water, allowing the abundance of carbon and oxygen to be measured; (2) determined that these abundances in turn show that HR8799 likely formed through accretion of a solid core, as we think the planets in our solar system formed; (3) obtained very high spectral resolution data, though preliminary analysis shows these are not sensitive enough to detect the rotation rate; (4) worked to initiate first light of GPI, which was slightly delayed because of issues with individual subsystems—the instrument shipped to the observatory in August 2013; and (5) completed the plan for the GPI science campaign and prepared the instrument for first-light observations.

Project Summary

During this project we carried out the highest sensitivity spectroscopy of extrasolar planets ever obtained and developed innovative observing techniques and data processing to extract planetary signal from stellar noise. Spectra of the young planet HR8799b and c allowed us to measure their atmospheric composition. The absence of methane (expected to dominate in these warm atmospheres) shows that the atmosphere circulates between high and low altitudes, destroying long-lived methane molecules and favoring carbon monoxide. We developed image processing techniques to automate the alignment and calibration of adaptive optics systems and developed the science mission for GPI. It features the world's most advanced adaptive optics system, optimized for extrasolar planet studies and constructed by LLNL. Our team proposed a large-scale survey to directly image young exoplanets and successfully won an international competition in which 890 hours were allocated for telescope time for a project expected to discover 20 to 50 planets. We will seek support from the National Science Foundation and National Aeronautics and Space Administration to continue with our GPI exoplanet survey.

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Land-Use Impacts on Belowground Carbon Turnover and Ecosystem Carbon Dioxide Source Attribution Using Radiocarbon

Karis Mcfarlane (11-ERD-053)

Abstract

Our goal is to determine the short-term effects of land use and management on soil respiration and total ecosystem respiration fluxes in a mature northern hardwood forest, and to attribute changes in the regional biosphere signal of atmospheric carbon-14 dioxide ($^{14}CO_2$) to disturbances from logging operations using radiocarbon as an isotopic tracer. We will compare observations of ^{14}C in CO_2 fluxes and pools of soil organic matter of varying turnover time made before and after a selective harvest to determine which belowground carbon pools contribute to changes in ecosystem respiration. Our objectives are to partition carbon sources to CO_2 loss via soil and total ecosystem respiration before and after harvest, as well as determine the relationship between plot-level, ecosystem-level, and regional CO_2 fluxes.

We hypothesize that logging disturbances will cause a shift in carbon sources for the ecosystem and soil respiration, and specifically that disturbances will cause a shift in carbon sources used by soil microbes from primarily labile—or changeable—pools toward increased use of stable soil carbon. We also conjecture that plot-level differences in respiration rates and carbon sources will be detected at both the ecosystem and regional scales, altering the biosphere signal for CO_2 partial pressure.

A Lawrence Livermore scientist measures soil respiration from a survey plot at the Willow Creek study site in northern Wisconsin. Soil respiration is a critical pathway for carbon that is taken up and stored by forests to return to the atmosphere as carbon dioxide. The radiocarbon signature of this carbon dioxide can indicate whether most of the carbon was recently taken up by plants or if it has resided in the soil for many years.

We expect that our work will demonstrate the utility of ^{14}C for partitioning ecosystem and soil respiration fluxes, quantifying shifts in microbial carbon sources, and the sensitivity of regional carbon fluxes and ^{14}C patterns to land use. In addition, we will generate data that can be used to develop improved process-based coupled climate models and improve our ability to predict regional patterns of CO_2 fluxes.

Mission Relevance

This work addresses a central Laboratory mission in energy and climate. Specifically, this research will make valuable contributions to our ability to predict regional climate change by improving our understanding of terrestrial processes affecting regional biosphere and atmosphere carbon exchange. In addition, our research supports LLNL's foundational science in measurement science and technology and will provide data for coupled carbon–climate modelers.

FY13 Accomplishments and Results

In FY13 we (1) completed measurements from air sampled at the Willow Creek eddy-covariance tower and the WLEF-TV regional tall tower in northern Wisconsin to evaluate the influence of forest CO_2 emissions on regional atmospheric $^{14}CO_2$; (2) completed data analysis for $^{14}CO_2$ at the two towers and determined that growing season emissions of CO_2 from forests influence atmospheric $^{14}CO_2$ at the regional scale; (3) completed soil respiration flux measurements and $^{14}CO_2$ measurements and found that over the growing season, soil-respired $^{14}CO_2$ decreases from above current atmospheric values in the early spring to current atmospheric values by mid-summer as root activity increases, and remains at current atmospheric values into fall when root activity is minimal and microbes are consuming current-year root carbon inputs; and (4) completed soil fractionation and associated sample analyses and found no relationship between soil fraction ^{14}C and respired ^{14}C, indicating that microbes selectively consume recently assimilated carbon in this hardwood forest.

Project Summary

This project demonstrated that (1) a relatively large seasonal fluctuation in soil-respired $^{14}CO_2$ is driven by changes in the relative importance of root and soil heterotrophic respiration as well as shifts in the source of carbon accessed by soil microbes, (2) the ^{14}C isotopic end member for soil and ecosystem respiration changes seasonally, and (3) CO_2 from soil respiration can impact regional CO_2 isotopic signatures, reflecting changes in process and carbon source, in addition to impacting regional CO_2 concentrations. The latter impact is an important consideration for top-down quantification of anthropogenic, fossil-derived carbon emissions. This project also provided the first coupled, long-term, high-density $^{14}CO_2$ data sets for ecosystem-to-regional scaling. This work demonstrated the utility of using ^{14}C as an isotopic tracer to provide critical information on carbon source attribution, and is needed for predictive modeling of climate and land use change impacts on future biogenic CO_2 emissions to the atmosphere. Our work established a presence at the Willow Creek research site, now a core site for the AmeriFlux measurement network, and a collaborative relationship with the University of Wisconsin team. As a result of this project, we have received funding from the DOE Biological and Environmental Research Program to support continued research in carbon-cycle science.

Publications and Presentations

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Selecting Better Models for Climate Change Detection and Attribution

Katherine Marvel (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 issues 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.

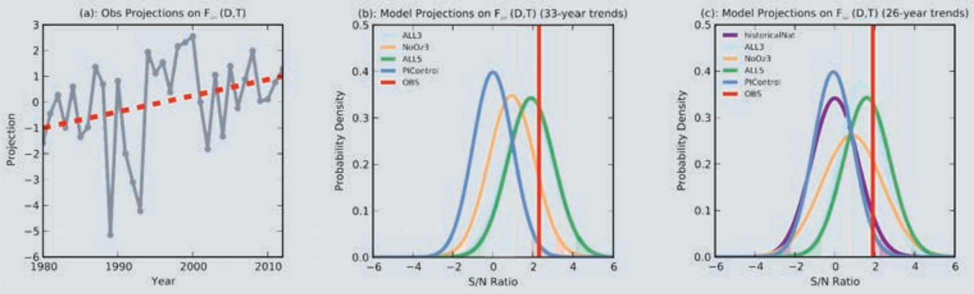
We will develop a suite of metrics to evaluate spatial and temporal variability at different scales and use these metrics to evaluate the performance of the climate code CMIP-5 (Coupled Model Intercomparison Project, Phase 5). We will also use these measures to evaluate the sensitivity of existing detection 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 mission focus area of climate change by advancing the state of the art in computational models for detecting climate change and attributing its cause.

FY13 Accomplishments and Results

In FY13 we (1) used new metrics to evaluate climate model simulations of current and future precipitation, demonstrating that climate models systematically overestimate global precipitation at multiple scales; (2) developed an innovative method to identify physical effects that are robust across models, even in the presence of model errors; (3) applied this method to CMIP5 simulations of forced and unforced changes in climate, which led to the first-ever detection of changes in global rainfall and the attribution of these changes to external, primarily anthropogenic, factors; and (4) developed methods to investigate the roles of various external climate forcing factors on dynamic and thermodynamic changes in climate variables.



Observed projections onto the "fingerprint" of climate change determined from multiple climate models (left). The increasing trend indicates that climate change is becoming more apparent over time. We compare the observations to trends in climate models (middle) that include external forcing components like greenhouse gases and the ozone hole (cyan and green), greenhouse gases only (orange), and internal climate variability only (dark blue). The observed trends are typical of forced model behavior and are unlikely to arise from internal climate variability alone. Comparison of observed trends to models that contain only natural forcing factors such as volcanoes and changes in solar variability (right) indicates that observed trends are very likely to be caused by human influences.

Proposed Work for FY14

In FY14 we will (1) perform detection and attribution studies on other variables of interest, particularly those related to the hydrological cycle, including possibly low cloud cover or ocean salinity changes; (2) continue to develop new measures of model performance; and (3) evaluate the sensitivity of detection and attribution results to these metrics.

Publications and Presentations

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Miniaturized Satellite Constellations for Intelligence, Surveillance, and Reconnaissance Applications

Willem De Vries (13-ERD-059)

Abstract

Currently, the U.S. does not have the operational capability to provide worldwide, on-demand, medium- to high-resolution space-based imaging to the military and intelligence community. We propose to investigate the use of miniaturized milk-box-sized satellites known as nanosatellites for intelligence, surveillance, and reconnaissance applications. Imaging payloads will be integrated into Pathfinder satellites powered by solar- and fuel-cell systems that are scheduled for launch in the coming year. A detailed test plan for characterization of these satellites will be developed to cover metrics such as sensitivity, astrometric measurement accuracy, and orbital refinement quality. The results from this characterization campaign will be compared against specific model predictions of the Space-based Telescopes for the Actionable Refinement of Ephemeris (STARE) space surveillance system, and through

this we will validate the orbital refinement concept. In addition, we will develop a high-resolution, large-sensor payload for space surveillance purposes.

We expect timely delivery of our two payloads, resulting in the successful launch of two LLNL imaging satellites. Data collected as per our test plan will corroborate our model predictions on key STARE mission performance metrics and will validate the concept. A successful STARE demonstration will show the utility of nanosatellites for on-orbit refinement of potentially colliding space objects, and is a crucial step to enabling a fully operational system.

Mission Relevance

This project is closely aligned with the Laboratory's cyber, space, and intelligence strategic mission focus area to provide national security in a highly networked world. It also builds on Laboratory advances in space situational awareness with applications in the intelligence and defense communities.

FY13 Accomplishments and Results

In FY13 we (1) developed an improved Cassegrain reflector design often used in optical telescopes as well as a novel patented monolithic optic design, the latter aligning the primary and secondary mirror surfaces through the glass of the optic itself rather than through metal; (2) developed the necessary sensitive alignment setup that allows for correct assembly; (3) delivered both imagers, which passed strict pre-flight environmental testing, to our payload integrator—however, both spacecraft experienced launch delays outside of our control, with the Cassegrain design launched in November 2013 and the monolithic optic design scheduled for 2014; (4) used the actual Cassegrain-design imaging payload to perform a ground-based orbital refinement campaign, given the launch delays; (5) successfully tested all relevant system components and achieved performance that exceeded expectations; and (6) began work on high-resolution imagers and variants of the monolithic optic design, which has been transferred to industry partners.

Project Summary

We developed two new imaging payloads for our three-nanosatellite orbital-refinement mission demonstration. Each one improved on the previous design. The second nanosatellite improved on the initial sensor design payload including temperature stabilization, and the third carries a novel monolithic optic design that significantly improves both optical quality and robustness. Our imaging payloads passed pre-flight qualification and are scheduled for launch in November 2013 and late 2014. We used the actual Cassegrain-design flight optic to characterize the refinement system performance, and the scheduling and processing infrastructure to capture orbiting objects visible from a test site at LLNL. We validated the refinement approach successfully from the ground, with an on-orbit validation to be done after launch. Lastly, significant external interest on the monolithic optic design has been expressed, resulting in one work-for-others effort and several potential efforts. We will expand the scope of our space effort to include larger optical apertures, novel sensors in different wavelength regimes, and added utility through constellations of small satellites.

Publications and Presentations

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Determining Capabilities of Natural-Abundance Radiocarbon Carbon Dioxide Analyses in Biogeochemistry Research

Thomas Guilderson (13-FS-007)

Abstract

The global carbon cycle is a key biogeochemical cycle that controls Earth's climate and life. Radiocarbon, a rare isotope of carbon that undergoes radioactive decay, is used to answer fundamental questions pertaining to the carbon cycle and biogeochemistry. Typically analyzed by accelerator mass spectrometry, radiocarbon measurements provide the ability to determine biogeochemical process rates and to identify sources and pathways of carbon at the molecular-to-landscape scales. Although routinely used, the application of radiocarbon analyses has yet to realize its full potential. This is in part because of the time-consuming requirement of isolating the carbon fraction to be analyzed and converting that fraction first to carbon dioxide and then to solid graphite, which is the preferred matrix for analysis. Direct injection of carbon dioxide for radiocarbon analysis with accelerator mass spectrometry is a promising avenue that can open up new possibilities for higher-throughput analyses across ecosystem and carbon cycle sciences. We propose to define the mass, accuracy, and precision for natural-abundance radiocarbon samples using a moving-wire gas-hybrid carbon dioxide source and Livermore's Center for Atomic Mass Spectrometry biomedical accelerator mass spectrometer (bioAMS). Although designed for less-precise biological tracer–radiocarbon applications, the instrumentation could be adapted to natural-abundance terrestrial biogeochemistry research. Our empirical experimental results on known natural-abundance carbon-14 and carbon-12 content materials will be used to determine where limiting factors exist. This information will be used in the context

of ion-optical modeling to determine the potential for utilizing this novel system in carbon cycle research.

Our goal is to determine the feasibility of using a moving-wire gas-hybrid carbon dioxide source for the high-sensitivity carbon-14 measurements that are necessary for biogeochemical sample analysis. We expect the results of our empirical experiments will not only allow us to determine the finite analytical limits of the existing system but also where, if any, optical bottlenecks exist in the current configuration of the bioAMS system. We intend to apply these results within the context of ion-optic modeling of the extant system. The combined experimental results and modeling will allow us to develop a strategy for optimizing the optics and transmission of the gas-hybrid carbon dioxide source and bioAMS system for natural-abundance carbon analyses. This translational instrumentation has the potential to broaden the application of radiocarbon accelerator mass spectrometry in carbon cycle and environmental research. Small samples of carbon, even without tracer addition, could be analyzed to elucidate sources—for example, petrochemical-based versus modern biogenic pathways or the source of primary and secondary carbonaceous aerosols.

Mission Relevance

This work has applications in carbon cycle research and novel instrumentation utilized for isotope geochemistry that is in line with LLNL's mission in energy and climate. The Intergovernmental Panel on Climate Change has recognized carbon dioxide as a major driver behind current and future warming of world climate, and the subsequent establishment of international initiatives to curb carbon dioxide emissions has greatly intensified the importance of global carbon cycle research for understanding the changing carbon cycle in the past, present, and future.

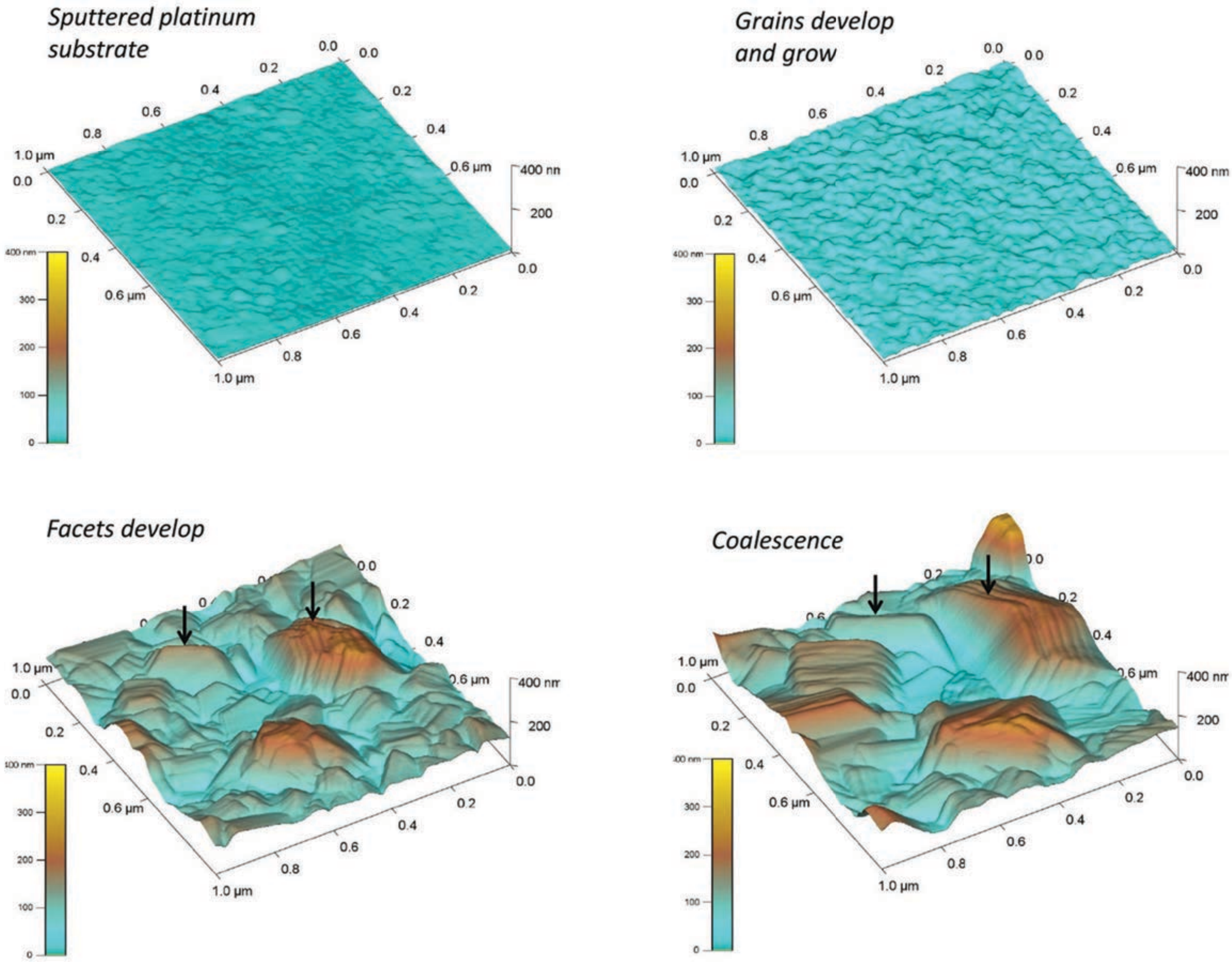
FY13 Accomplishments and Results

In FY13 we (1) developed a strategy for optimizing the compact bioAMS system based on the combined experimental results and our ion-optic modeling for natural abundance radiocarbon samples; (2) used optimized slit settings to reduce the background (detection limit) for full-sized combusted and carbon dioxide samples to about 0.5% of modern carbon levels (effectively 42,000 radiocarbon years before present), which contrasts to about 1% modern background for the initial acceptance tests of the system and slightly higher backgrounds (2–4% modern) during routine analysis of biomedical samples; and (3) performed ion-optic modeling to define an optically optimized low-energy injection system for the compact bioAMS system.

Project Summary

We have concluded, based on both our experimental and modeling work, that it is possible to use a moving-wire gas-hybrid carbon dioxide source to dramatically increase the sample throughput of accelerator mass spectrometer systems while still maintaining the sensitivity required for biogeochemistry-related measurements. Our modeling effort for optimizing the bioAMS system utilized PowerTrace, which is a graphical user interface shell around TRACE 3-D, a first-order optic code for simulating transport of ion beams. We developed the matrix-style code to model transport of high-current bunched beams where space–charge effects may be important. The model allows particle ion-beam current effects on the accelerating voltage gradient and related focusing and electrostatic elements. This process allows for the efficient simulation of einzel charged-particle lenses, accelerating gaps, accelerator columns, and hard-edged magnetic fields. Our model was initialized using measured aspects of the main elements and the ion-beam emittance characteristics for the gas-hybrid ion source. A consequence of optimizing the low-energy optics is that it will define the geometry and physical footprint of the beam line and related elements. Much of the optimization effort that we have proposed is associated with electronics, rewiring, and modifying and reprogramming the vacuum map system, which allows computer control of the vacuum system and interlocks. Experiments with helium as a stripper gas should be performed to reduce scattering and angular loss of ions after dissociation of molecular isobars. We anticipate that with these modifications, our compact system and gas-hybrid source will be suitable for natural abundance biogeochemistry samples.

Energy Supply and Use



Laboratory Directed Research and Development

Annual Report FY2013

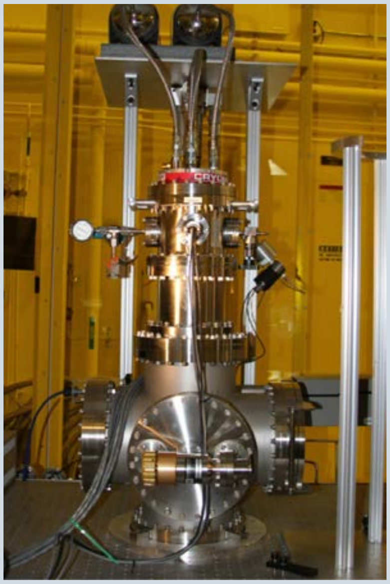
Target Components for Ensuring Survival During Flight into a Laser Inertial Fusion Reaction Chamber

Robin Miles (11-SI-004)

Abstract

We intend to address the materials challenges associated with inertial-confinement fusion targets that are injected into a fusion reaction chamber. The target must be designed to arrive at chamber center in the precise structural configuration necessary for the fusion implosion to occur. This requires understanding a range of fundamental physical properties of deuterium–tritium (DT) and various structural materials under cryogenic conditions and at microscales, and then integrating material, thermal, and hydrodynamic terms in representative test environments. We will achieve these objectives through experimental and modeling efforts to study specific high-risk aspects of the target cycle, including DT layer properties, the fragile support structure of the capsule inside the hohlraum hollow cylinder, and the thermal reflectivity of ultrathin metal films.

Our research will lead to a fundamental understanding of material attributes required to design a structurally sound injected laser fusion target, based upon both measurements and models of requisite materials. This includes the mechanical properties of DT ice and its behavior under acceleration, the properties of liquid hydrogen isotope layers in foams as an alternative to DT ice, static and dynamic mechanical properties of thin films supporting the capsule in a hohlraum at cryogenic temperatures, and thin-film behavior under representative high-acceleration loads. This research will also lead to a fundamental understanding of the material attributes that provide thermal target integrity, including reflectivity of the very thin metalized films at cryogenic temperatures and methods for preventing radiative heating of the capsule.



Cryostat used to measure depressed freezing temperature of hydrogen isotopes in ultralow-density foams used in inertial fusion targets. Specifically, we examined the depressed freezing of deuterium–tritium in foams to qualify a liquid and deuterium–tritium layer with low vapor pressure.

Mission Relevance

This project supports the Laboratory's missions in laser inertial fusion energy, energy security, and climate by addressing key technical challenges in realizing a concept for cost-effective inertial-confinement fusion as a source of clean energy.

FY13 Accomplishments and Results

Our effort to improve the strength of carbon nanometer-scale tube composites for use as a capsule support membrane for targets injected at about 8,000 m/s^2 was successful. Specifically, we (1) determined that ion irradiation increased the strength of composite films from about 300 to about 800 MPa, a twofold factor over that required—strength scaled with ion irradiation, and it is hypothesized that irradiation induces crosslinking between nanotubes first within the bundles and secondly between bundles, resulting in increased strength; (2) determined that beyond a certain point, the membranes become too brittle to handle easily and would be impractical for large-scale production of targets; (3) performed tests of the heat-transfer coefficients that verified our modeling, which suggests that convection currents internal to the target are primarily responsible for the transfer of heat from the laser entrance hole window to the capsule; (4) heated a pseudo-window and measured the thermal and pressure response at various points within the target—measurements were compared with models of thermal conditions to validate thermal predictions; (5) measured the external heat-transfer coefficients for a target with a configuration similar to the lead design, although at much lower velocities and temperatures than expected in the chamber, and determined that the scaled values for the heat-transfer coefficient were consistent with those predicted from models; and (6) completed the effort to determine applicability of low-density foams to liquid DT layers, and verified that at 360 mg/cm^3 density, the energy associated with the solid–liquid interface does inhibit freezing below the normal freezing temperature of the liquid and that the vapor pressure over the super-cooled liquid corresponds to the vapor pressure of the solid at a similar temperature.

Project Summary

We investigated aspects of inertial fusion energy targets related to elements of the target subcomponents and investigated issues related to survival of the target as it was injected into the center of the target chamber. We focused the majority of the effort on the DT layer, the structural survival of the capsule support membrane, and the thermal profile of the target in flight. We examined the depressed freezing of DT in foams to qualify a liquid–DT layer with low vapor pressure. Experiments verified that at 360 mg/cm^3 density, the energy associated with the solid–liquid interface depresses freezing below the normal freezing temperature of the liquid about 1.5 K and that the vapor pressure over the super-cooled liquid corresponds to the vapor pressure of the solid at a similar temperature. Structural analysis indicated that the capsule support membrane is the most vulnerable component to the stresses of acceleration, followed by the DT layer. Carbon composite films exhibit good strength at less than the 0.1-µm thickness required for these films, and ion irradiation increased cross-linking within and between nanotube bundles, increasing the strength of carbon-nantotube-epoxy composite films from about 300 to about 800 MPa, a twofold factor over the required strength. Thermal analysis showed that heat enters the target primarily via the thin laser entrance hole from convection from the hot chamber gases and radiation from the chamber walls. The convective heat from the window establishes convective currents within the helium internal to the target that will transfer heat to the capsule. Experiments were used to validate the models, and analysis shows that the DT layer can be kept cool with careful thermal design. The results of the effort have furthered our understanding of the risks and requirements of inertial fusion energy.

Publications and Presentations

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Forecasting and Uncertainty Quantification of Power from Intermittent Renewable Energy Sources

Wayne Miller (12-ERD-069)

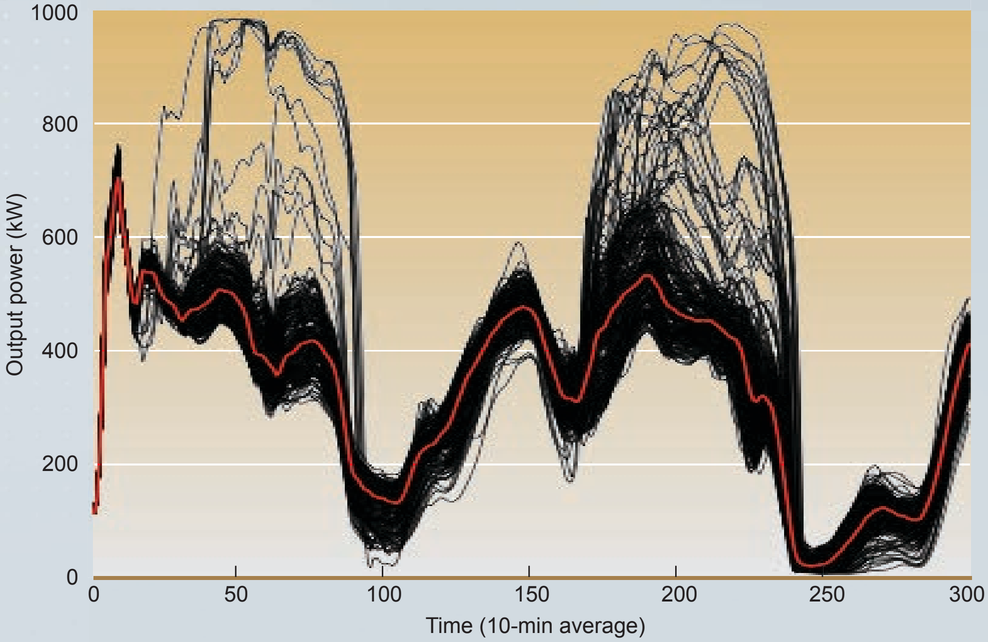
Abstract

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 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 a 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 in line with the LLNL strategic plan relating to energy security, climate change, and our research activities in renewable energy and utility power analysis.



An ensemble of 192 forecasts of turbine power (black curves) surround the theoretical output (red curve) over a 2-day period. This ensemble is based on variations in physical parameters in wind forecasts made with the Weather Research and Forecasting Model, identifying correlations and errors from modeling assumptions.

FY13 Accomplishments and Results

In FY13 we (1) evaluated complementary approaches for reducing forecast uncertainty by ensemble-based forecasts; (2) made significant progress on developing an enhanced power curve for predicting turbine power in complex inflow situations by developing more realistic inflow and power models via statistical analysis; (3) extended our modeling toolkit for more accurate wind simulations over terrain resolved at the wind-farm scale by coupling the Weather Research and Forecasting model to CgWind, adding the lifting-line aerodynamic description of wind turbine rotors to CgWind, and initial field validations studies of the new capabilities; and (4) supported all work with field campaigns for validation data.

Proposed Work for FY14

In FY14 we will employ field campaigns and high-performance computing modeling to (1) perform detailed analysis of forecasting errors and the means to reduce them through statistical analysis of our ensemble-based forecasting methods; (2) develop a best-practice guide for day-ahead wind forecasting methodologies; and (3) complete our dynamic power curve, a model of turbine power production that considers the complexities of real wind inflow at the turbine rotor to improve power production estimates.

Publications and Presentations

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Mirocha, J. D., et al., 2013. *Multiscale simulation of boundary-layer flow over complex terrain using the Weather Research and Forecasting Model*. LLNL-TR-645827.

Simpson, M., S. Wharton, and W. Miller, 2013. *Validation of a WRF short-range ensemble forecast of a significant wind ramp event with LIDAR measurements.* LLNL-TR-645705.

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Increasing the Lifetime of Rechargeable Batteries

Christine Orme (12-LW-030)

Abstract

Our overarching goal is to increase the lifetime of rechargeable batteries by preventing failures from dendrite formation and nonreversible processes. It is a major challenge to design the chemistry, geometry, and charge cycle such that a battery is truly reversible and returns to its original configuration after charging. One of the biggest problems occurs as the oxide converts back to a metal, re-plating onto the anode during recharging. We propose to directly image the cyclic charging and discharging of anode materials using in situ atomic force microscopy, transmission electron microscopy, optical microscopy, and Raman microscopy. All techniques will monitor dynamical changes under electrochemical and temperature control. Together, these techniques will give us the ability to image at higher spatial and temporal resolution than has been reported to date, allowing us to develop an understanding of the parameters and chemistry that control zinc dendrite nucleation and kinetics in alkaline chemistry and ionic liquids. We will also investigate several additives that we believe have the potential to stabilize the zinc interface. We hypothesize that quantifying the dendritic morphological evolution will provide new mechanistic details of how electrolytes and additives impact the stability of battery interfaces.

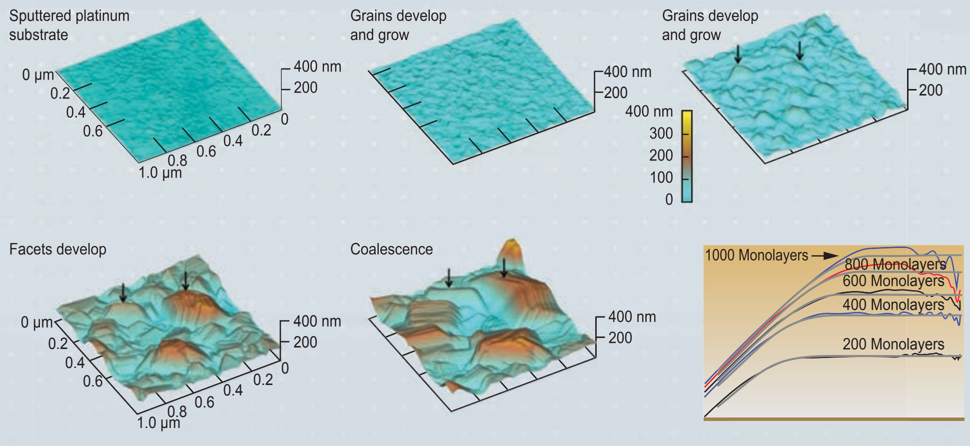
Our objective is to establish a mechanism-based understanding of zinc dendrite formation and prevention at battery interfaces such that the need for barriers at interfaces can be eliminated or at least reduced. The availability and use of additives or electrolytes that prevent dendrite instabilities without compromising battery performance would revolutionize the battery industry. The first step toward this goal is providing hard evidence of how these additives interact with electrified interfaces. The combination of LLNL's imaging capabilities and the team's expertise in crystal growth offer the opportunity to distinguish different mechanistic processes that lead to dendrite formation and thereby suggest ways of controlling it.

Mission Relevance

The development of a program in battery research advances LLNL missions in energy security. For energy security, we must lessen our dependence on fossil fuels and increase our use of sustainable energy such as wind and solar. These needs have triggered a resurgence in battery research, particularly to develop better rechargeable batteries for vehicles and for leveling intermittent energy loads.

FY13 Accomplishments and Results

In FY13 we met our milestones by (1) directly imaging charging and discharging phenomena using in situ scanning probe and scattering techniques; (2) evaluated the effect of additives and electrolyte on crystal growth parameters such as nucleation density, growth rates, and diffusion rates during charging and discharging—we evaluated both alkaline electrolytes and commercial ionic liquid electrolytes over



A sequence of atomic force microscopy 1-µm^2 images showing nucleation and growth of zinc grains during recharging in an ionic liquid electrolyte (1-butyl-3-methylimidazolium trifluoromethylsulfonate). The height–height correlation function quantifies the evolution of roughness and provides statistical metrics that can be used to compare the effect of additives and other electrolytes on the stability of battery interfaces.

a range of conditions pertinent for battery cycling; (3) surveyed several additives and found bismuth to be the most promising; (4) completed a comparison between atomic force microscopy and ultrasmall angle x-ray scattering for an ionic liquid battery electrolyte—results showed that charging in particular ionic liquids leads to an unexpected texture evolution that stabilizes the interface, in marked contrast to traditional alkaline electrolytes; and (5) evaluated the effects of bismuth- and bromide-containing additives, and demonstrated that optimal concentrations of bismuth increases nucleation density and stabilizes basal facets leading to a more compact interface that is less susceptible to dendrite formation. We did not evaluate proprietary ionic liquids because the available quantities are too small for our experiments.

Project Summary

The successful conclusion of this project resulted in a deeper understanding of the kinetics of zinc battery anodes. We completed kinetic measurements of the nucleation and growth of zinc surfaces in alkaline and commercial ionic liquid electrolytes that serve as a baseline for understanding additive interactions. We have found that certain additives (bismuth), and also certain electrolytes (1-butyl-3 methylimidizolium trifluoromethanesulfonate), can alter the fiber texture of the growing interface. Fiber texture is the presentation of a particular crystal orientation perpendicular to the substrate. The result of fiber texture is that the interface grows at a more homogeneous rate, which lessens the propensity for instabilities. We will submit an Energy Frontiers Research Center proposal for support to continue our experimental observations and develop a comprehensive model.

Publications and Presentations

Keist, J. S., et al., 2013. “Coupling in-situ techniques to analyze zinc deposition and dissolution for energy storage applications.” *MRS Proc. Symp. C—Electrocatalysis and Interfacial Electrochemistry for Energy Conversion and Storage*, vol. 1491. Cambridge University Press, New York. LLNL-PROC-603932.

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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 newly created fracture to hold it open during gas production. The water must be viscous in order to move the dense sand particles. The resulting complex mixture of thickeners and friction reducers makes it difficult to treat and reuse the water. If these agents were unnecessary, the fluid would be much easier to treat. The proppant would have to have a similar density as 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 that 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 strong science and engineering in materials that can transport and react under triggered conditions, which could be applied in many other fields. We will also provide solid 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 basic science understanding of proppant physics.

Mission Relevance

This work addresses a core LLNL mission in energy security with applications to natural gas production and geothermal energy production. We will collaborate with the National Energy Technology Laboratory, the dedicated fossil-energy laboratory, which will conduct the imaging of proppant placement in their x-ray tomography facilities.

FY13 Accomplishments and Results

In FY13 we focused on spherical proppant flow, demonstration of reacting encapsulated systems, and initial experimental demonstration. Specifically, we (1) demonstrated an entirely new way to measure spherical proppant flow in fractures by measuring real fractures in rocks, then printing them on a transparent material using additive manufacturing so that an exact replica flow could be observed and compared to our models; (2) applied this method to fractured shale gas source rock; (3) demonstrated triggered proppant capsule permeability and experimented with a variety of encapsulated liquids in our replica fractures; and (4) initiated modeling and proppant chemistry development by identifying requirements and small-scale testing.

Proposed Work for FY14

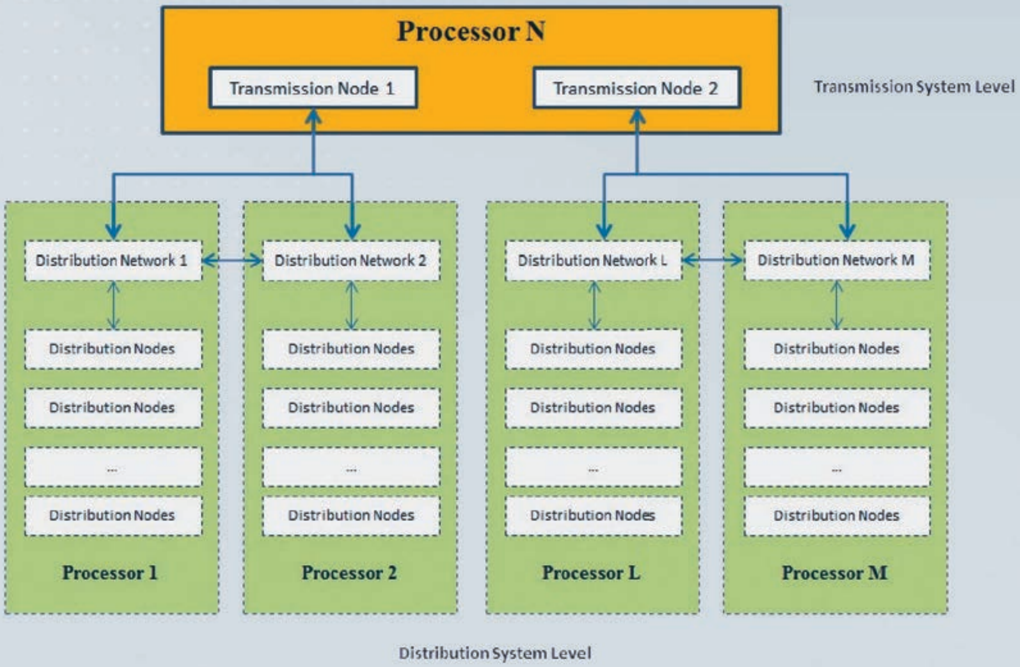
In FY14 we will (1) focus on triggered capsule permeability and forming solid cements inside the capsule by letting water permeate the shell, (2) optimize the specific design of cement materials and their expansion rate, (3) conduct computational modeling to maximize the placement of capsules in fractures and verify the results of experiments with our additively manufactured replica fractures that allow us to observe proppant flow, and (4) implement and test the designs for adjustable density proppants.

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 for 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, this 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.



Our initial parallel framework design focuses on achieving a loose coupling between the existing transmission and distribution software. At each time step, a single transmission mode is advanced, and then parallel instances of the distribution software for each substation on the transmission network are advanced. This loosely coupled model leads to a natural decomposition of the domain—the distribution models are solved in parallel, enabling the use of thousands of processors.

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

FY13 Accomplishments and Results

In FY13 we (1) evaluated existing software packages and decided to develop our own transmission simulator, (2) chose Gridlab-D for distribution simulation and completed the initial parallel framework design for the distribution simulation, and (3) developed test cases to verify the simulation results.

Proposed Work for FY14

In FY14 we will (1) continue to complete the initial parallel framework design and implementation, (2) empirically measure the computational complexity of the algorithms and models using the test case developed in FY13, and (3) evaluate the strength and proximity of couplings and, based on those findings, refine the parallel approach and develop effective solution methods.

Publications and Presentations

Top, P., et al. 2013. *Automated low frequency load cutoff.* North American Power Symp., Manhattan, KS, Sept. 22–24, 2013. LLNL-CONF-638804.

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 from the blanket 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 work is to develop a safe and reliable process for tritium recovery that replaces the 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 problematic lithium halide salts to be replaced 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 a unique position in the event that the promise of a new process for tritium recovery is realized.

Mission Relevance

The development of this next-generation tritium recovery process for a laser inertial fusion energy plant 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.

FY13 Accomplishments and Results

In FY13 we (1) identified two halide-free salts—lithium hydroxide and lithium carbonate—and performed initial insolubility tests between metal and salt; (2) completed the conceptual design for the non-centrifugal contactor, including a distributor plate, an ultrasonic emulsifier to maximize interfacial area between phases, and a proposed hydrocyclone separator; (3) selected candidate surrogate fluids that closely mimic the lithium and lithium-salt systems; (4) developed a nondimensional analysis for the hydrocyclone separator that enables scaling between the lithium and lithium-salt and surrogate fluid systems; (5) constructed a preliminary plastic model for flow visualization to compare to computational fluid dynamics models; (6) created a single-phase model that shows maximum centripetal acceleration for a cylindrical-shape separator; and (7) completed a preliminary design for the electrolyzer.

Proposed Work for FY14

In FY14 we will (1) finalize the design of a system to separate oil and water and demonstrate its performance in a working plastic model, (2) document the detailed design of the electrolyzer, and (3) investigate a promising process based on evaporation and zone refining for separation of tritium and other impurities from lead, including the development of a heat-transfer model of an optimized zone-refining process and the design and construction of a demonstration-scale zone-refining process.

Publications and Presentations

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Optimized Three-Dimensional Electrodes for Energy Storage

Eric Duoss (13-ERD-057)

Abstract

Emerging autonomous microelectronics found in micro-electromechanical systems such as miniaturized biomedical devices, reactors, sensors, 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 nanoparticles 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. 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 economic competitiveness.

FY13 Accomplishments and Results

In FY13 we (1) developed routes to synthesis of nanometer-scale particles and inks for anode and cathode materials with tailored properties; (2) produced three-dimensional micro-battery architectures, based upon interwoven electrode layouts with direct ink writing; (3) characterized these battery architectures; and (4) developed a modeling capability for both two- and three-dimensional battery structures with interwoven electrode layouts.

Proposed Work for FY14

In FY14 we will (1) develop improved nanoparticles and functional inks as electrode and electrolyte materials, (2) create new designs for three-dimensional micro-battery architectures, (3) enhance our predictive modeling capability, and (4) demonstrate additive manufacturing of new three-dimensional micro-battery designs with improved performance versus their planar counterparts.

Dual-Volume Cryogenic Hydrogen Storage for Automobiles

Salvador Aceves (13-FS-010)

Abstract
Cryogenic pressure vessels have been determined to be the best option for onboard automotive hydrogen storage. Today's hydrogen storage technologies (compressed and liquid hydrogen) operate at fixed temperature. Cryogenic pressure vessels, however, drift across several phases depending on the level of use—cooling down and depressurizing when driven and heating up and pressurizing when parked. We propose to investigate a dual-volume system, with an inner high-pressure vessel and a secondary lower-pressure vessel along with a thin vacuum insulation that holds promise for unprecedented storage system density (twofold greater than what has been previously demonstrated), minimum weight and marginal cost, and ultimate safety with full containment even if the inner vessel fails catastrophically.

We expect to determine whether a dual-volume storage system is superior to previously demonstrated cryogenic pressurized storage for hydrogen on automobiles. We will conduct experiments and modeling to determine if it is possible to manufacture very-thin (5-mm) high-performance vacuum insulation and ascertain that vacuum quality in thermal insulation will not degrade because of outgassing from vessel surfaces. Properly insulating a vessel with a thin vacuum space is a major technological challenge that will demand unprecedented approaches to vacuum insulation.

If proven, the resulting storage system will demonstrate maximum safety and range at minimum cost, leading to practical hydrogen vehicles that do not produce carbon dioxide emissions and eliminate reliance on foreign oil.

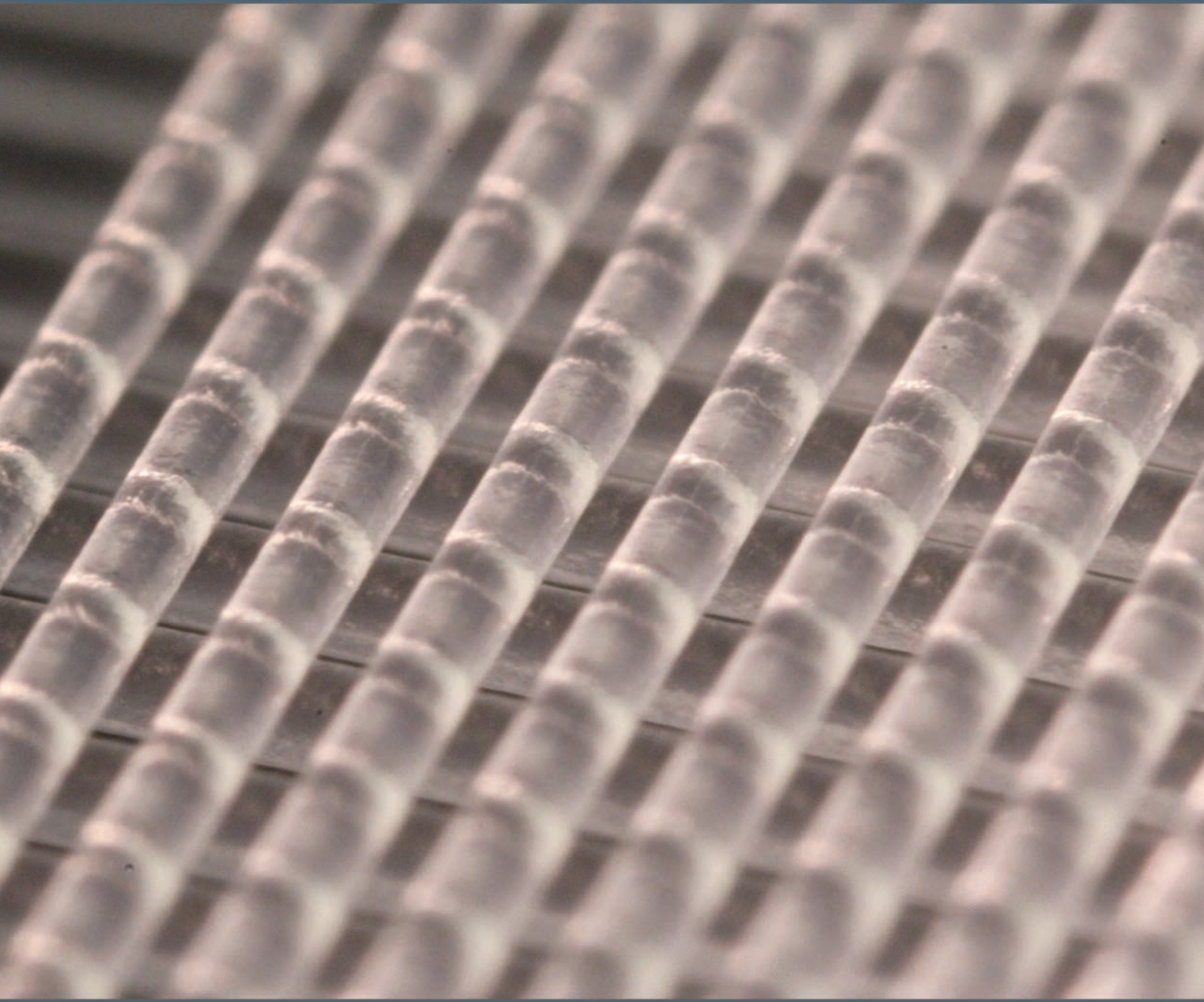
Mission Relevance
Results have important implications for the widespread introduction of hydrogen-fueled transportation by addressing critical barriers such as safety, driving range, and cost. The project builds on important LLNL capabilities in cryogenic systems, hydrogen fuel, and computational modeling. Our research for this project supports the Laboratory's focus area in climate and energy security.

FY13 Accomplishments and Results
In FY13 we (1) conducted experiments in a 10-L subscale vessel, and demonstrated high variability in heat-transfer rate depending on the specific conditions of the experiment such as cryogenic support configuration and radiation shield layers; (2) initiated successive improvements in cryogenic supports and insulation layout that reduced the heat-transfer rate to about 1 W in the best case; and (3) determined that the experimental heat-transfer rate is sufficient to warrant full-scale implementation (100 L), assuming that the experimental conditions can be scaled to a heavier full-size vessel that requires a stronger cryogenic support system.

Project Summary

Cryogenic vacuum insulation is typically thick (~3 cm), thereby occupying space that could otherwise store cryogens, such as hydrogen. Achieving practical hydrogen-based transportation demands efficient packing of hydrogen in the small spaces available onboard a vehicle, therefore thin insulation is key. In a series of subscale experiments, we demonstrated the potential for thin insulation (5 mm) with the low heat-transfer rate (1 W) necessary for long-term hydrogen storage during, for example, long parking periods. While these experiments have to be scaled to full-sized vessels of about 100 L, thin insulation appears to be a promising option for compact hydrogen storage. We expect that our experiments will serve as a basis for future proposed work to DOE, which has expressed interest because of the high potential of this technology.

Engineering and Manufacturing Processes



Laboratory Directed Research and Development

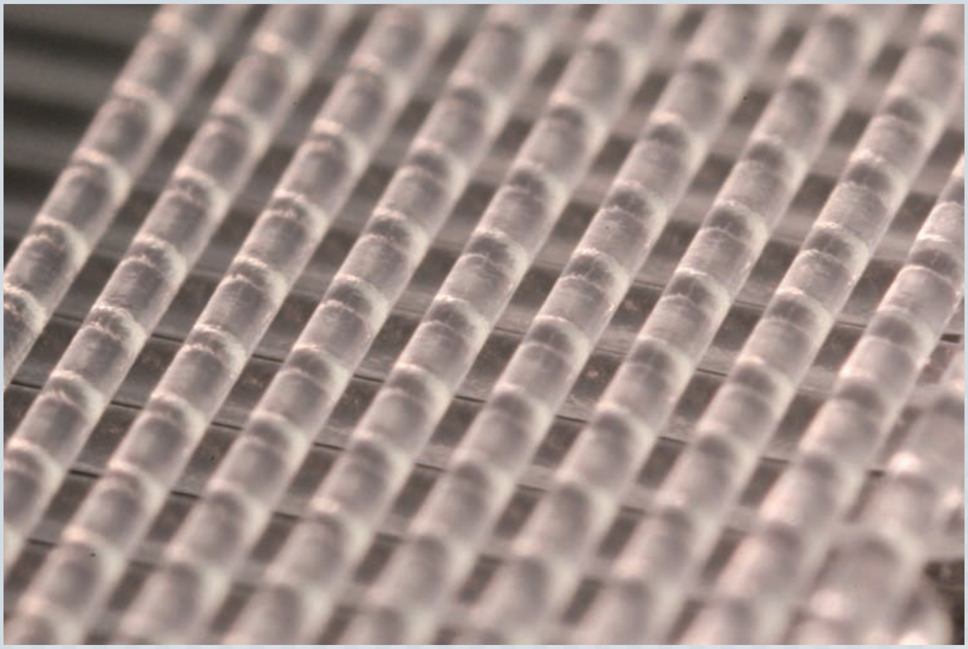
Annual Report FY2013

Scalable High-Volume Micro-Manufacturing Techniques for Three-Dimensional Mesoscale Components

Christopher Spadaccini (11-SI-005)

Abstract
Our goal is to fundamentally understand and to develop, from the ground up, new micro-manufacturing techniques applicable to materials such as polymers, metals, and ceramics. These techniques would enable production of three-dimensional mesoscale geometries with micrometer-scale precision that can be scaled to achieve high manufacturing volumes at low cost. Traditional manufacturing processes start with bulk material followed by a forming or metal-removal process. For mesoscale parts this is time consuming, energy intensive, and generates excess material waste. In addition, grain size becomes problematic and material handling is difficult. This project seeks to revolutionize the way we manufacture parts for several national security applications.

The design of any new hardware component, regardless of the application, is constrained by the materials available and the geometry that can be fabricated using existing manufacturing processes. We expect to overcome both of these limitations through advanced fabrication processes that are capable of achieving arbitrary three-dimensional mesoscale structures with microscale architectures and sub-micrometer precision. These processes will be revolutionary to manufacturing and will have a broad impact because of compatibility with a wide range of materials, rapid translation from computer model to fabricated component, and the ability to scale to large numbers of components. We intend to design and fabricate a new material with specified properties such as thermal expansion versus stiffness outside the bounds



A lattice network of a tri-block co-polymer that was printed using direct ink writing.

of those attainable with bulk materials processed via traditional synthesis methods. Success in meeting our proposed objectives will both illustrate the capabilities of the process and demonstrate novel materials and structures that are relevant to LLNL missions and programs.

Mission Relevance

Precision engineering enables Laboratory programs to field experiments and metrology capabilities to advance science and technology in the national interest. A "bottom-up" three-dimensional manufacturing process combined with multiple materials enables new design concepts that have not been possible and allows rapid turnaround for new designs in support of Livermore missions in stockpile science. These capabilities can also be used to develop novel materials that can be applied to reducing or countering threats to national and global security.

FY13 Accomplishments and Results

In FY13 we (1) designed, fabricated, and measured performance of a negative Poisson's-ratio architecture that has load-carrying capability; (2) designed and partially demonstrated a new wide-area micro-stereolithography system; (3) demonstrated the ability to fabricate heterogeneous structures with several of our processes including micro-stereolithography; (4) fabricated and tested thermite materials with unique architectures including features in three dimensions; and (5) fabricated and tested additional interesting micro-architectures, including an ultra-lightweight, high-stiffness material that has the density of aerogel but is five orders of magnitude stiffer, as well as a material with designed compressive and shear properties that exhibit the unique feature of negative stiffness.

Project Summary

The successful completion of this project has resulted in three, highly utilized, well-understood, robust, and unique additive micro-manufacturing techniques: projection micro-stereolithography, direct ink writing, and electrophoretic deposition. These methods have enabled us to fabricate a wide variety of novel materials and architectures with unique, previously unachievable properties. We have successfully demonstrated control of features at the nanometer-scale for fully three-dimensional microscale parts from a variety of constituent materials. Additionally, this project has explored the concept of using micro-architecture to control bulk-scale properties. Most notably, we have been able to control the energy release rate of thermite materials by controlling the microscale architecture, fabricate soft porous materials with a regular lattice structure that has precisely designed compressive properties, and made a material with the highest stiffness-to-weight ratio in the world. The next steps for our work include continuing the development of designed materials and advances in micro-stereolithgraphy through an ongoing Defense Advanced Research Projects Agency program. We will also integrate materials made with direct ink writing into programmatic applications and create new armor materials for the U.S. Air Force with our electrophoretic deposition technique.

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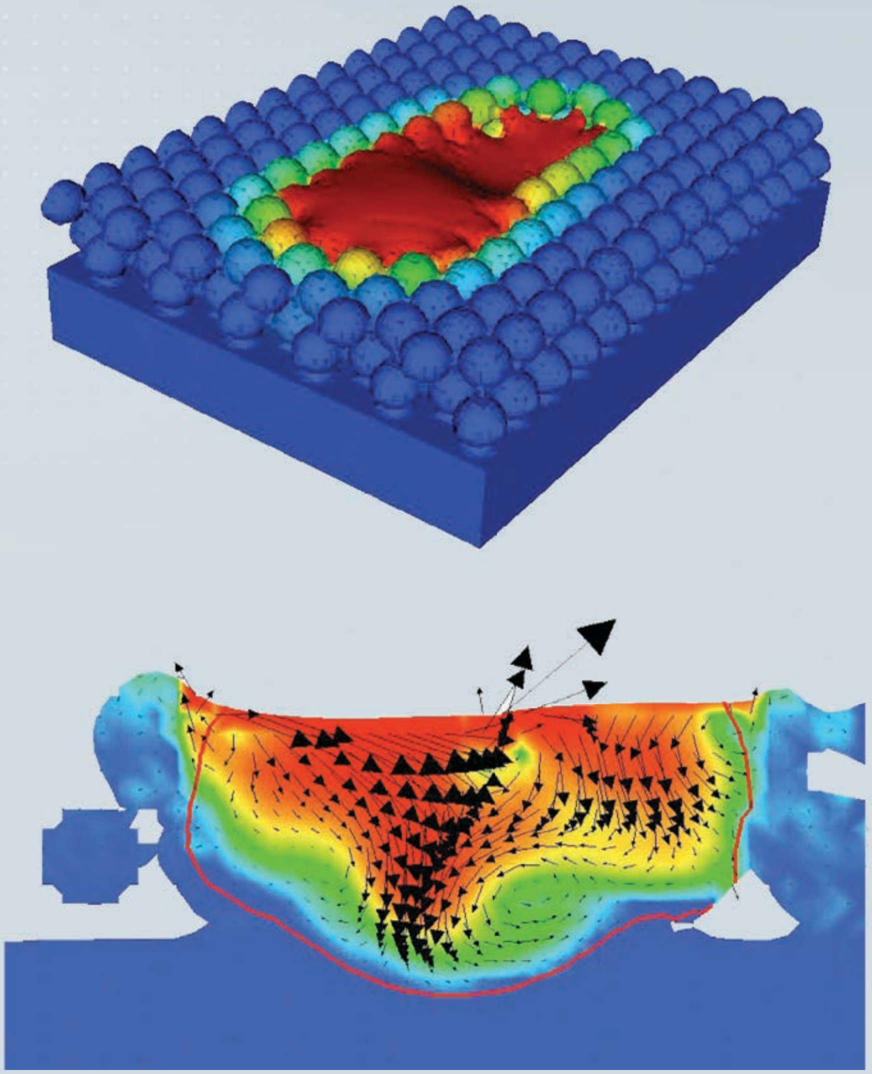
Accelerated Certification for Additively Manufactured Metals

Wayne King (13-SI-002)

Abstract

Stockpile stewardship requires a new manufacturing approach that can speed the development and certification of parts, shrink the manufacturing footprint for a shrinking stockpile, reduce manufacturing waste, avoid the costs and delays normally associated with manufacturing processes, and reduce energy consumption in part fabrication. We propose to employ modeling, simulation, process optimization, experiment design, in situ sensing, and uncertainty qualification 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 simulation, which is used to describe the macroscopic properties of materials, that models the process at the scale of the part, as well as 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 the 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.



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 visible through the arrows in the cross section at the bottom.

Mission Relevance

This project furthers the Laboratory's mission in stockpile certification by developing a substantially improved, science-based approach to rapidly and cost-effectively develop mature manufacturing technologies and systems.

FY13 Accomplishments and Results

In FY13 we (1) processed 600 single-track experiments, yielding data as a function of additive manufacturing parameters, with results revealing the existence of keyhole-mode laser melting that has not been reported in the additive manufacturing literature; (2) verified the effective medium model against a literature example; (3) completed a 1.25-mm by 1-mm by 90-µm powder model simulation with 4,000 powder particles with a uniform size of 27 µm using the Lagrangian–Eulerian three-dimensional multiphysics ALE-3D code on 80 processors in 14 hours; and (4) carried out a residual stress experiment at the Los Alamos Neutron Science Center on an additively manufactured component.

Proposed Work for FY14

In FY14 we will (1) design experiments to accelerate the development of processing conditions in the absence of relevant physics-based models; (2) develop an effective medium model to computationally design complete parts and predict their properties; (3) develop a powder model to computationally model the melting of powder and its resulting densification; (4) build a model to compute solidified microstructures, including grain size and shape and phases formed; (5) determine techniques for accelerating development of new additive manufacturing processes; and (6) devise a means for identifying process, properties, and performance relationships.

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

Jonathan Hopkins (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 at 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 detonating 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-manufacturing 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 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 meta-material manufacturing, in support of LLNL interest in additive fabrication processes using multiple materials.

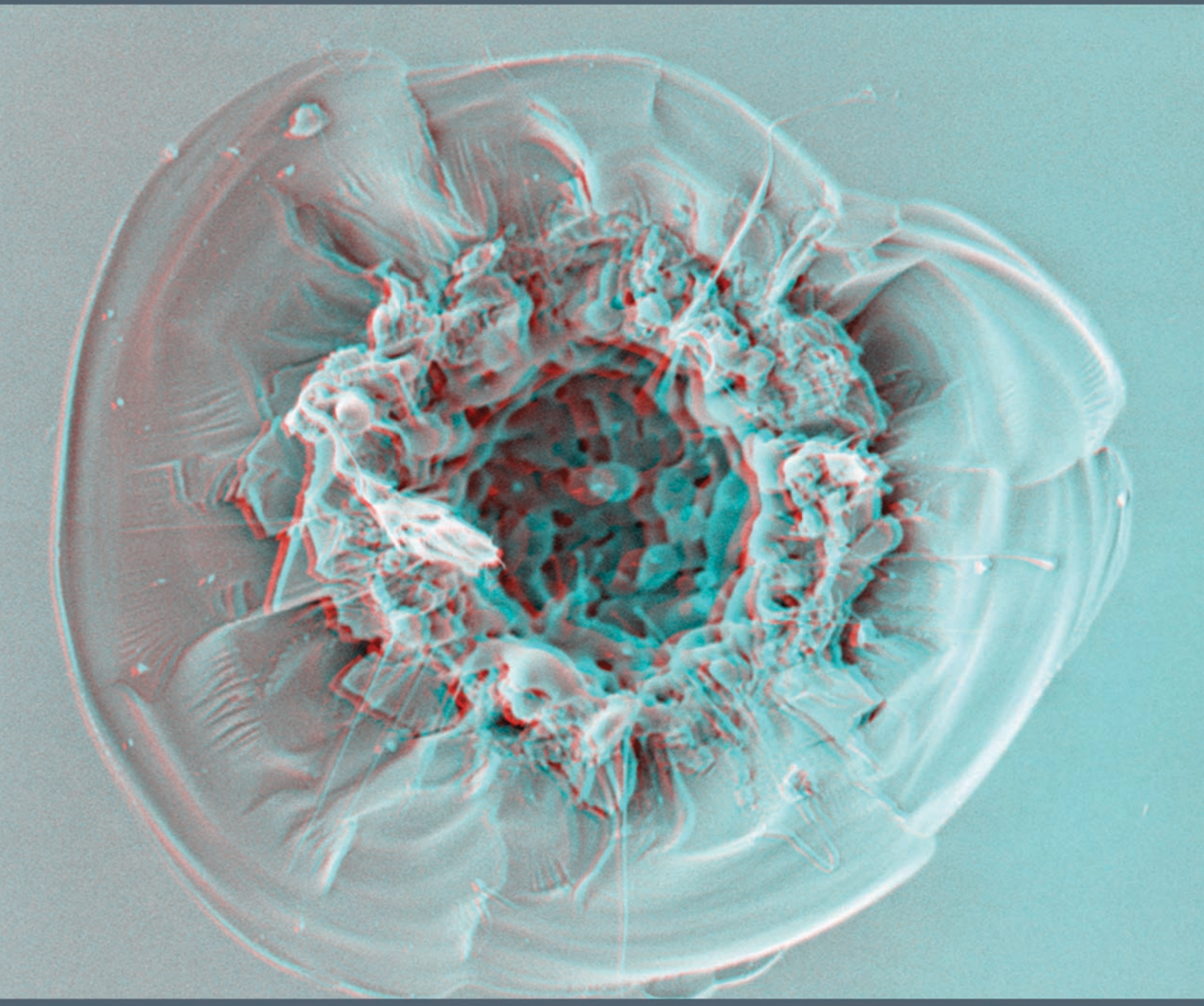
FY13 Accomplishments and Results

All proposed milestones for FY13 were met. Specifically, we (1) thoroughly researched the performance of existing commercially available micro-mirror arrays and demonstrated that our array is expected to substantially outperform the current state of the art; (2) generated comparisons of existing micro-mirror arrays, allowing us to show how our new design offers new capabilities; (3) continued construction of our fabrication demonstration prototype; (4) tested direct ink writing and found it to produce features that were too large for the final structure—however, projection micro-stereolithography was found to be usable and the design was adjusted to account for projection micro-stereolithography fabrication limits; (5) began investigation of coating the hexapod structures to control array rigidity; and (6) fabricated a prototype design to test micro-fabrication issues. These insights were used to plan the next phase of fabrication. The design architecture was demonstrated to be sound by this prototype, with electrical testing carried out to confirm element functionality.

Proposed Work for FY14

In FY14 we propose to (1) complete the rotary comb portion of our apparatus and demonstrate operation as predicted, (2) complete fabrication and testing of our demonstration prototype, (3) continue research into printing hexapod flexures and various coating processes, (4) optimize the design of ribbed mirrors with features that increase adhesion of the hexapods to their backside, (5) explore micro-stamping and other nanoscale mirror-folding techniques as well as approaches for aligning and assembling array parts, and (6) develop control electronics.

Materials Science and Technology



Laboratory Directed Research and Development

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Modeling of Microstructural Processes in Tungsten-Based Alloys for Fusion Applications

Jaime Marian (11-ERD-023)

Abstract

Tungsten is a suitable candidate for the first wall surrounding the central cell of a nuclear fusion device because of its high melting point, good thermal conductivity, and low activation. However, the metal is very brittle and must be alloyed with other elements to improve mechanical properties. With this project, we plan to gain insights into the mechanical properties of tungsten alloys for nuclear fusion applications. We will use a multidisciplinary computational approach to obtain appropriate alloys that are mechanically stable under irradiation and high-temperature loading. Using physics-based modeling, we will propose tungsten-alloy options for structural components in fusion devices.

If successful, we expect this project to deliver science-based design guidelines for first-wall and divertor materials in fusion devices. This is important for inertial-confinement fusion concepts, in which a suitable design for a first wall capable of sustaining high thermal loadings and neutron doses is still lacking. In addition, the design of divertor coatings is crucial to achieve plasma stability and component lifetime in next-generation tokamak fusion reactors.

Mission Relevance

By providing an increased understanding of advanced fusion structural materials and advancing the cause of nuclear fusion as a reliable and clean energy source, this project contributes to two core Lawrence Livermore missions to enhance the energy and environmental security of the nation and to strengthen the nation's economic competitiveness. This work also supports the Laboratory's strategic mission thrust in fusion energy to provide a sustainable and once-through, closed-fuel-cell nuclear energy option.

FY13 Accomplishments and Results

In FY13 we (1) developed a kinetic Monte Carlo code—written in C++ and coupled to graphical output for ease of visualization—to model thermally activated motion of screw dislocations in tungsten, (2) fitted the code to atomistic data obtained via carefully selected semi-empirical interatomic potential atomistic calculations, (3) completed density functional theory needed to characterize the interaction of solutes (rhenium) atoms with dislocations as a function of dislocation type and distance, and (4) used these calculations to further parametrize the kinetic Monte Carlo code to study dislocation motion in a solute field representing a concentrated tungsten–rhenium solution.

Project Summary

The successful conclusion of this project resulted in the development of a first-principles methodology to study dislocation motion in tungsten–rhenium alloys.

These particular alloys are important technologically because they are the main tungsten-based alloy that displays some low-temperature ductility. Our methodology and calculations will enable understanding of the physical mechanisms leading to this ductility and aid in the design of tailored alloys for fusion and other structural applications. The DOE Office of Fusion Energy Science will fund follow-on work to this project through an Early Career Research Award.

Publications and Presentations

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Chemical-Vapor-Modified Laser-Based Damage Mitigation and Surface Shaping of Fused Silica

Manyalibo Matthews (11-ERD-026)

Abstract

Mitigating damage in fusion-class laser optics is critical to maintaining robust and cost-effective laser performance. Previous work indicates that inert-gas-assisted laser machining can significantly increase evaporation rates for greater optics machining flexibility. Leveraging such work, we will develop a modeling and experimental capability to enable the short-pulse infrared-laser micro-shaping of fused silica optics and explore chemistry-assisted enhancements to laser-based damage mitigation. Our primary objectives are to (1) develop methods and predictive models to control the localized surface shaping of silica-based optics using high-irradiance, microsecond-pulsed carbon dioxide lasers; (2) modify laser parameters and local chemical environments to control the evaporation and condensation of material; and (3) explore the use of laser chemical-vapor deposition as a means to plane damaged silica surfaces.

Using optimized short-pulse laser light, we expect to achieve more precise control of laser-machined morphologies by limiting energy absorption dependent on laser parameters. We will establish a method for introducing controlled silica-rich vapor at lower temperatures to fill damage cavities and eventually re-plane the optical surface. By extending precise laser machining and planing to a broader range of surface defect configurations, we should achieve truly scalable laser-based mitigation techniques that would benefit fusion-laser-related research with longer optic life cycles, lower operational costs, and more energetic experiments.

Mission Relevance

This project supports the Laboratory's missions in stockpile stewardship and laser ignition fusion energy by enhancing the fusion-class optics needed for future work in related fields of high-energy-density science and materials in extreme environments. In addition, because this project also involves diagnostics to probe the physics and chemistry of high-intensity infrared laser–matter interactions, our results should also be relevant to laser-related military applications.

FY13 Accomplishments and Results

In FY13 we (1) augmented a laser chemical-vapor-deposition system with real-time interferometry, allowing monitoring and control of deposition processes; (2) demonstrated filling of damaged sites on fused silica windows using both interferometry and laser power feedback control; (3) successfully set up an optical parametric oscillator as a source for sum-frequency generation (nonlinear optical process) spectroscopy to probe surface chemistry, and replaced photo-acoustic measurements with interferometry measurements; and (4) extended our optimized wavelength studies using 100% hydrogen gas and non-oxidizing gas for tetraethyl orthosilicate deposition. Conditions spanned exposure times relevant to rapid ablation-mitigation protocol from about 10 µs up to near steady state of approximately 5 s, where transport rates are comparable and thus expected to affect evaporation rates.

Project Summary

We demonstrated three new methods for laser processing of damaged silica optics to enhance performance, each resulting in at least one patent application and several publications. The first method involved flowing hydrogen gas over laser-heated surfaces to lower the temperature for material removal—an in-house gas-delivery system integrating laser optics was constructed for this purpose. Molecular dynamics simulations were able to relate observed morphology to additional reaction channels and surface energy shifts. Secondly, our gas-assisted laser system was configured for laser-based chemical vapor deposition to demonstrate deposition of optically stable silica in open air. Finite-element simulations captured both rate kinetics and deposition profiles accurately, providing insights into the deposition mechanisms. We demonstrated the use of laser-based chemical vapor deposition for filling damage pits, paving the way for production implementation. Thirdly, a tunable infrared laser was implemented in a custom raster-scanned laser machine to remove damage-prone material with micrometer-scale precision. The wavelength dependence of energy coupling was determined experimentally and through simulation to reveal micro-shaping effects of scientific and practical interest.

We will continue the laser-based chemical vapor deposition in collaboration with the University of California, Berkeley and the development of the infrared laser machining apparatus is currently being explored at Livermore as a test bench for additive manufacturing research.

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Understanding the Stochastic Nature of Laser-Induced Damage

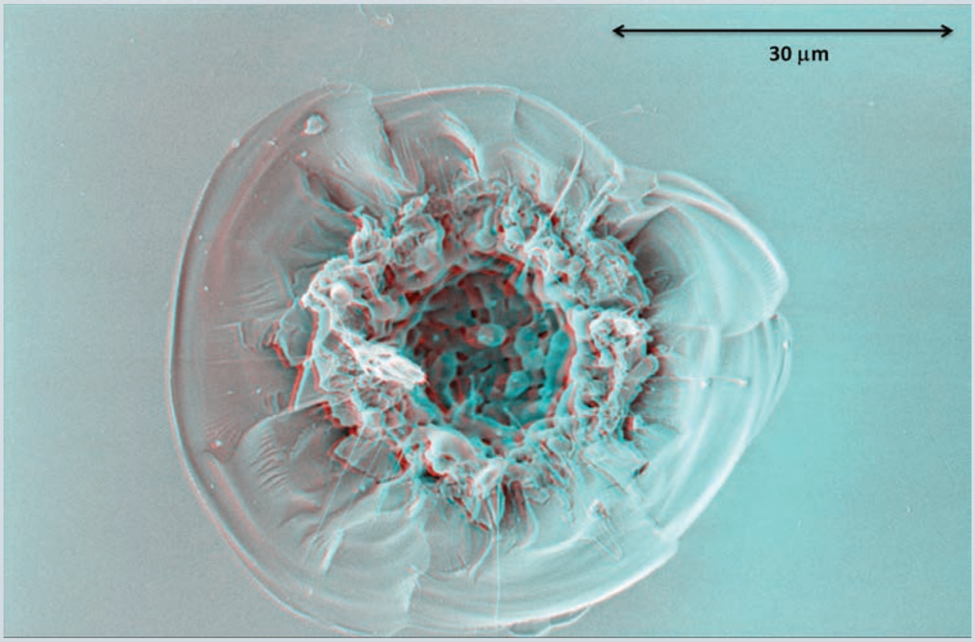
Christopher Carr (11-ERD-030)

Abstract

Laser optic surface damage is of particular concern for laser facilities because even small damage sites initially only a few micrometers in size can quickly grow large enough to destroy the optic, thereby resulting in damage to downstream optics as well. In fact, laser-induced optics damage will remain a key constraint on the operation of inertial-confinement fusion laser facilities for the foreseeable future. Damage growth is widely believed to be statistical in nature, but the internal features of damage sites is still undiscovered. Our goal is to determine the physics behind the stochastic nature of these damage sites. To this end, we will investigate the nature of damage features through experimentation with controlled parameters, new diagnostics for discovering and tracking damage site growth, and advanced analysis techniques. With these findings, we will build long-range predictive models suitable for large-aperture and high-average-power facilities.

If successful, this project will help improve our understanding of how the internal parameters of laser-induced damage sites affect laser energy deposition and how laser profiles affect the final configurations of internal parameters. Improvements in these broad areas will significantly enhance our ability to predict the growth behavior of damage arising over a large number of laser pulses. To this end, we will (1) prepare damage sites to selectively isolate candidate chemical and mechanical attributes, (2) measure energy deposition to locate attributes that drive growth, and (3) examine atypical sites—those that exhibit growth behavior on the extremes of the distribution.

A three-dimensional stereo-pair image of a damage site on the exit surface of a silica optic.



Once the most quickly and the most slowly growing site types are identified, we will study how they can be used to test the sensitivity of various diagnostic techniques and how shot history affects the state of a damage site.

Mission Relevance

Accurately predicting when and how optical components are damaged by laser pulses will allow facilities to operate at energy densities high enough to fully enable research in inertial-confinement fusion and laser ignition fusion energy, in support of the Laboratory's missions in stockpile stewardship and advanced lasers and applications.

FY13 Accomplishments and Results

We have (1) developed integrated growth rules that project the future size of damage sites based both on growth rate and probability of growth; (2) increased our understanding of the internal structure of laser-induced damage sites and used this knowledge to develop a description of growth rate based on fracture mechanics, which is in good agreement with both our statistical description and experimental observations of site growth as a function of laser parameters; and (3) completed experiments on potassium dihydrogen phosphate, which has very different mechanical properties than silica, that support the fracture model of growth but also indicate an additional, previously unknown, contribution to growth by bulk material properties.

Project Summary

The successful conclusion of this project resulted in an improved understanding of how the internal features of laser-induced damage on the exit surface of optical materials effect future growth. We have determined that the amount of sub-surface fracture surrounding a damage site and the presence of organic compounds within a damage site strongly affect growth rate. Conversely, the amount of microstructure on the surface of a damage site pit, the overall integrated photoluminescence, and the bulk stress surrounding a site proved to be weak indicators of future growth rate. These findings were used to produce a growth model based on simple fracture mechanics, which is in good agreement with observed growth rates as a function of fluence on the exit surface of silica.

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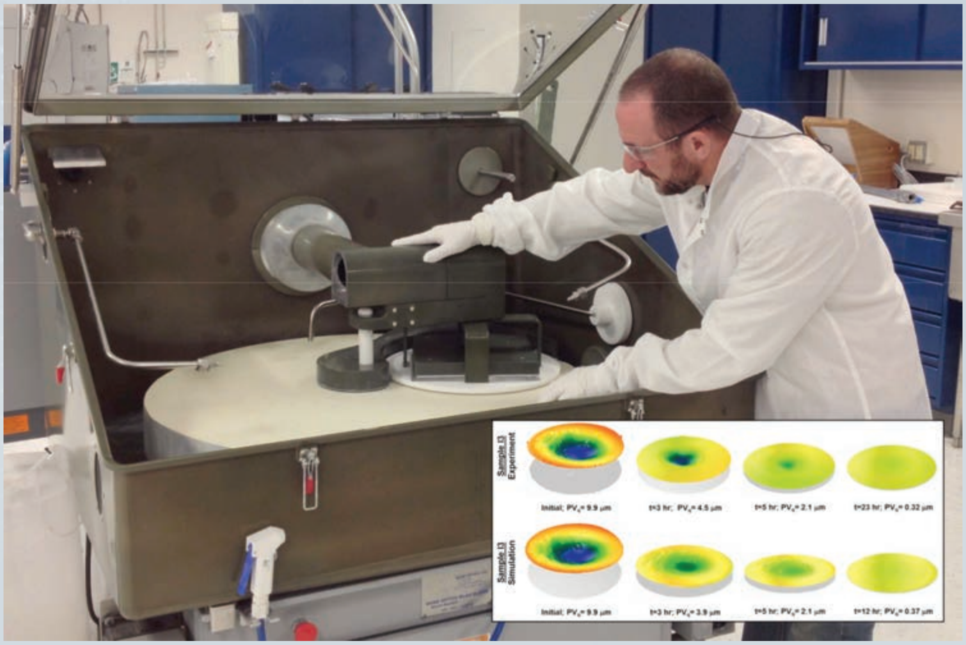
Fundamentals of Figure Control and Fracture-Free Finishing for High-Aspect-Ratio Laser Optics

Tayyab Suratwala (11-ERD-036)

Abstract

We propose to investigate critical phenomena affecting the full-aperture finishing of high-aspect-ratio optical components, conduct full-aperture optical polishing in environments free of rogue particles by understanding and preventing key sources of the particles, and develop methods to enhance the lifetime and reduce the cost of inertial-confinement fusion and laser inertial fusion optics. To this end, we will perform specifically designed polishing experiments and use models to understand the effects



Our convergent polisher enables convergence of a workpiece to its final shape regardless of initial shape and without changes to polishing parameters.

of pad wear, mechanical bending, residual stress, and thermal effects on nonuniform removal. In addition, we will determine methods to control rogue particles, such as using 100% humidity to control particle size distribution. The laser optics that we will target include main debris shields, continuous-phase plate substrates, disposable debris shields, and blue and red blockers for use in high-peak-power laser systems such as the National Ignition Facility and amplifier glass for high-average-power laser systems.

This project will significantly advance our scientific knowledge of optics polishing, which will benefit not only fusion energy but also the precision optical and semiconductor industries. The ability to deterministically finish an optical surface using a full-aperture tool will allow optical glass fabricators and chip manufacturers to achieve figure control of surface profile in a more deterministic manner. Our study will also enhance our understanding of and develop methods to prevent, reduce, or eliminate the influence of rogue particles during polishing, which will lead to optical components with higher laser damage thresholds, greater thermal shock resistance, and improved surface roughness.

Mission Relevance

The project supports the Laboratory's missions in advanced lasers and their applications as well as inertial-confinement fusion by enabling high-value fusion laser optics for the National Ignition Facility and potential applications in laser inertial fusion energy at lower cost through deterministic finishing, while also increasing laser damage resistance, thermal shock resistance, and improved surface roughness. This project also furthers LLNL's mission in strengthening America's economic competitiveness by improving the precision of other optics and semiconductors.

FY13 Accomplishments and Results

In FY13 we successfully (1) developed an understanding of key phenomena affecting the long-term (hundreds of hours) convergence point of the workpiece, including three-body wear, material deposit, temperature, and pad shrinkage, as well as demonstrated stability down to a 0.15-µm peak-to-valley on 100-mm workpieces and a 0.3-µm peak-to-valley on 265-mm workpieces; (2) developed an understanding of and proposed a novel chemical mechanism for stabilizing particle size distributions without altering particle activity; (3) demonstrated improved thermal understanding using a three-dimensional steady-state thermal model during polishing, matching experimental results; (4) demonstrated convergence on flat, concave, and convex spherical laps; (5) developed an ensemble Hertzian gap model probing the microscopic interactions during polishing specifically for relating the tail end of the particle size distribution of the slurry to the resulting workpiece roughness; and (6) developed improved slurry filtration techniques during polishing, resulting in reduced scratch densities on the workpiece surface.

Project Summary

This research project has led to the development of a novel polishing method called convergent polishing, enabling faster and more deterministic optic fabrication. This method allows for a workpiece, regardless of its initial surface figure, to converge to the near final shape in a single iteration without changes in polishing parameters. To accomplish this, we have scientifically investigated the critical phenomena involved in full-aperture finishing of optical components in nonuniform removal, and then determined methods to eliminate them. We have performed designed polishing experiments and used numerical models to understand the effects of pad wear, mechanical bending, residual stress, thermal effects, material deposit, and pad shrinkage. Also, we have explored methods to improve slurry particle size distribution using novel filtration methods and new chemical techniques for reducing agglomerates in slurries. Finally, all of these concepts and accompanying mitigations have been implemented in the design and development of a polishing system, which we successfully used to demonstrate long-time convergence to flat and spherical laps on various round and square glass workpieces up to 374 mm. We have started the process of potential licensing of the patented convergent polishing system and process with polishing equipment manufacturers and finishing vendors.

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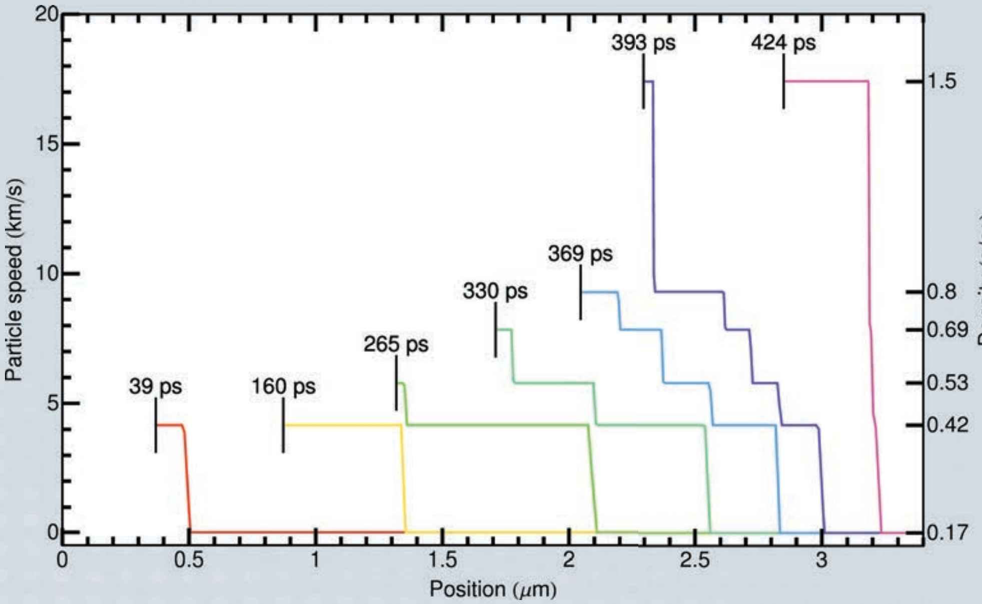
Hydrogen Melting and Metallization at High Density

Michael Armstrong (11-ERD-039)

Abstract

The objective of our project is to experimentally determine phase boundaries in hydrogen–deuterium melts and to examine metallization phase transitions at high density and low temperature. Through a combination of simulations and measurements, we aim to confirm or improve the calculated shock Hugoniot—a valuable tool for analyzing a material's equation of state—of deuterium and hydrogen starting from initial high-density conditions. We will refine equation-of-state models of hydrogen and deuterium under high-density conditions by examining their shock-compressed behavior at high density and low temperature starting under static pre-compression conditions in a diamond anvil cell. Our results are expected to break new ground in examining the fundamental quantum physics of condensed materials and to have multiple applications in planetary science.

We expect to determine scientifically important phase boundaries in the hydrogen–deuterium system at high pressure and density and to map out metallization and other phase transitions. The experimental data we derive will play a fundamental role in the modeling of condensed-phase light materials at high density and low temperature.



This simulation shows multiple shock compressions of cryogenic liquid deuterium on an ultrafast time scale. The small scale of the compressed volume implies that a low-energy laser system could be used to obtain high density in rapidly equilibrating materials like deuterium. Dynamic compression on a sub-nanosecond time scale may reduce the required drive laser energy by as much as a factor of 10,000.

Mission Relevance

This research is aligned with LLNL's strategic objectives in fusion and high-energy-density matter and fundamental research in the disciplines. By refining the equation of state for hydrogen–deuterium mixtures, this work will provide data and insight in support of the Laboratory's core mission in stockpile stewardship science.

FY13 Accomplishments and Results

In FY13 we (1) continued work towards quasi-isentropic compression of a cryogenic liquid, including the build-up of a laser station (and the cryostat) required for these experiments, the design and construction of a sample cell, and the construction of a high-power laser amplifier for use in these experiments; (2) made substantial progress towards the goal of compressing cryogenic deuterium, although we have not yet achieved it; and (3) completed most of the experimental setup.

Project Summary

The primary result of this project was the recognition that, for materials that equilibrate on sufficiently fast timescales, the energy required for dynamic compression experiments may be orders of magnitude smaller than currently used in large-scale dynamic compression experiments. In particular, the energy required for dynamic compression scales as the third power of the compression time. So, if the compression time can be reduced by a factor of 10, the energy required for the experiments may be reduced by a factor of 1,000. This result implies that, for samples like hydrogen that rapidly equilibrate, extreme conditions might be achieved with tabletop (as opposed to facility-scale) laser systems. Rapid equilibration was demonstrated in deuterium itself, and even aluminum, a material with strength, exhibited an ultrafast equilibration time (<100 ps) for compression to more than 30 gPa pressure.

Publications and Presentations

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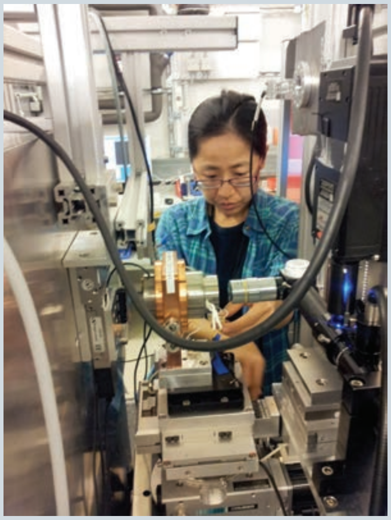
Pressure-Induced Melt, Nucleation, and Growth: Fundamental Science and Novel Technological Materials

William Evans (11-ERD-046)

Project Description

Phase and microstructure have an enormous influence on the macroscopic properties of important materials, including strength, fracture toughness, and radiation damage resistance. The dependence on compression rate of pressure-induced phase transitions is a very sparsely understood but critically important area of high-pressure science. We therefore propose experimental measurements using LLNL's dynamic diamond-anvil cell to measure the influence of compression rates on pressure-induced phase transformations—melt, nucleation, and growth—and on the resultant polycrystalline, amorphous, or other microstructure. Our experiments will combine the dynamic diamond-anvil cell with time-resolved x-ray scattering and imaging to study the dynamics of pressure-induced transitions in the dinitrogen molecule.

We will quantitatively determine transformation kinetics, metastability, and dynamics and potentially discover new metastable materials. Specifically, we will produce (1) time-resolved measurements of structural evolution, nucleation, growth, and possible metastable phases in pressure-induced melt–freeze transitions; (2) new experimental capabilities for time-resolved studies; and (3) improved understanding of pressure-induced phase transitions and compression-rate dependencies. Success will establish LLNL as a pioneer in this new high-pressure science research capability.



Postdoctoral researcher Jing-Yin Chen aligns the Livermore dynamic diamond-anvil cell for x-ray diffraction studies of the pressure-induced solidification of krypton on the Extreme Conditions Beamline at the Petra-III third-generation synchrotron x-ray source at DESY in Germany.

Mission Relevance

This project supports the Laboratory's national security mission by addressing our understanding of pressure-induced nucleation and growth (relevant to modeling system responses under dynamic conditions) and by addressing important technological issues relevant to the nucleation, growth, and melt of components potentially important to the laser ignition fusion energy concept, such as gallium-based heat sinks. Finally, identifying "recipes" for achieving a desired microstructure or phase will dramatically enhance our technological ability to achieve materials by design for defense and other mission-relevant applications.

FY13 Accomplishments and Results

In the project's final year we (1) performed experimental studies of time-resolved phase-transformation dynamics at higher temperatures and compression rates; (2) conducted imaging studies of the liquid-to-solid transitions in bismuth and gallium using monochromatic x-ray radiation for enhanced contrast—the contrast between the liquid and solid was significantly more discernible than using a polychromatic (white) x-ray beam; (3) confirmed that monochromatic x-ray phase contrast imaging is a viable approach for these types of studies; and (4) performed time-resolved x-ray diffraction studies to measure the kinetics of phase transformations in krypton, including pressure-induced liquid-to-solid transformations, and the compression-rate dependence of metastability and growth of hexagonal and face-centered-cubic phases—we determined the pressure-induced nucleation rate of solid krypton.

Project Summary

We have made significant advances in the scientific understanding of the dynamics of pressure-induced transformations. The design of the dynamic diamond-anvil cell has been improved by incorporating a modular configuration that allows rapid setup, high-temperature operation, and access to compression rates exceeding 500 GPa/s. We conducted a series of experiments at synchrotron x-ray sources studying the dynamics of solid–solid and liquid–solid transitions in bismuth, gallium, and krypton, and were able to determine nucleation and growth rates from these studies. In addition, we observed compression-rate dependence of transitions to metastable phases. The dynamic diamond-anvil cell, developed as part of this project, will continue to be used by other fundamental research programs at LLNL and within academia to measure important parameters of pressure-induced transformations. The experimental systems and methods we developed with this project will also be applied to programmatic materials addressing issues of interest to Livermore science campaign and stockpile stewardship missions.

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Materials for Inertial Fusion Energy Reactors

Michael Fluss (12-SI-002)

Abstract

We propose to determine the best materials available for inertial fusion energy reactors from a preselected suite of reduced-activation steels and nanometer-scale dispersed-oxide particle steels using multiple ion-beam irradiation to approximate fusion energy conditions, followed by post-irradiation examination of microstructure and mechanical properties. We will establish a scientific basis for selection of candidate materials to use in the first-wall and blanket of inertial fusion reactors through a careful comparison of the properties of irradiated materials, utilization of data obtained relevant to the physics of materials models, and extension of those models to the actual irradiation conditions of an inertial fusion energy engine.

We expect our project will produce data representing one of the most detailed scientific studies of radiation effects for steels in a fusion energy environment. It will address specific challenging questions about the individual and synergistic roles of hydrogen, helium, and displaced atom production that deleteriously affect the properties of steels used in fusion energy systems. Most importantly, the project will establish the scientific basis for qualification experiments utilizing neutron sources, including both nuclear spallation and fusion sources.

Mission Relevance

Research into materials selection for reactor construction supports Livermore's efforts in advancing laser inertial fusion energy, a central component of the Laboratory's strategic plan to meet vital national needs. In a broader sense, our project directly addresses the fundamental scientific challenges associated with accelerated materials experiments for advanced theory, simulation, and modeling of slow processes associated with materials aging.

FY13 Accomplishments and Results

In FY13 we completed dual- and triple-beam irradiations of the ferritic–martensitic steel EUROFER97 at 500°C using the triple ion-beam facility at the CEA French Alternative Energies and Atomic Energy Commission laboratory in Saclay, France. The irradiations were performed with 40 displacements per atom from 23-MeV iron-ion implants, and the helium-to-displacement ratio was 15 atomic parts per million for dual-beam irradiation. For triple-beam irradiation, the hydrogen-to-displacement ratio was 50 atomic parts per million, with the helium ratio the same as with dual-beam irradiation. The purpose of these irradiations was to compare the effects of hydrogen on the radiation damage accumulation. Hydrogen production from transmugenic reactions is of great concern to the performance of first-wall and fluid containment systems in fusion energy systems. Some characterization work was performed on the dual-beam specimens but not completed before the project was terminated at the end of March 2013.

Project Summary

We irradiated specimens to compare the effects of hydrogen on radiation damage accumulation to determine the role of hydrogen in first-wall and breeder containment materials for inertial fusion energy reactors. However, the project was terminated before reportable results could be achieved.

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 progress in the basic science for the development and qualification of advanced, ultrahigh-burnup nuclear fuel. To achieve this goal, we will couple modern computational materials modeling with modern fabrication, characterization capabilities, and 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 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. Furthermore, the successful accomplishment of this project will augment LLNL's credibility within the nuclear energy community.

FY13 Accomplishments and Results

In FY13 we (1) synthesized samples of uranium–molybdenum, uranium–zirconium, and inert metallic matrix zirconium–iron–copper alloys (three compositions for each class of alloy) and characterized them with x-ray diffraction, transmission electron microscopy, and differential scanning calorimetry analysis; (2) designed sample holders for irradiation experiments and performed two sets of experiments at

450 and 600°C with 4 samples per holder at Livermore's Center for Accelerated Mass Spectrometry; (3) performed post-experiment examinations with x-ray diffraction and transmission electron microscopy on uranium–molybdenum and uranium–zirconium samples—in the case of uranium–zirconium, irradiation damage caused a morphology change in the lamellar microstructure with accompanying coarsening and loss of orientation relationship between the alpha and delta phases of uranium, and for the uranium–molybdenum alloy, detailed transmission electron microscopy work revealed a metastable structure that is different from the commonly known high-temperature body-centered-cubic phase; (4) performed, for the first time, ab initio calculations of the equilibrium properties for ternary uranium–molybdenum–zirconium alloys as function of composition, which revealed the importance of the contribution of the three-body interactions for describing the Gibbs energy of the ternary alloy, an effect that has been largely ignored in past studies (this has consequences on stability and also on kinetics of transformation and microstructure of nuclear fuels); (5) extended the thermodynamic assessment of additional binary alloys to plutonium–molybdenum, plutonium–tungsten, and for the first time to uranium–cesium alloys—in this later case, because of enormous atomic size difference between uranium and cesium, there is a tendency towards phase separation, and we expect the same to be true in the case of uranium–strontium alloys; and (6) extended the LLNL phase-field code to handle multiphase microstructures, and applied the code to uranium–zirconium alloys to study solidification and solid phase-decomposition.

Proposed Work for FY14

We will (1) perform the first-ever ab initio calculations of the equilibrium properties of ternary plutonium–molybdenum–zirconium alloys, along with alloys of plutonium and a body-centered-cubic transition metal, as a function of composition to study the effect of ternary interactions; (2) apply the LLNL phase-field code to the study of microstructure evolution in actinide alloys by extending its applicability to the impact of thermal transport and elastic properties on phase stability; (3) validate the phase-field results with experimental results; (4) characterize the irradiated uranium–zirconium and uranium–molybdenum alloys as a function of temperature, dose, and dose rate; and (5) perform additional irradiation experiments with uranium–molybdenum and newly synthesized zirconium–iron–copper alloys at various compositions and temperatures, followed by post-experiment examination.

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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 advanced world economies, 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 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, substantial 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.

FY13 Accomplishments and Results

In FY13 we (1) employed our new sample growth capabilities to produce poly- and single-crystal magnetic samples with a 2:17 chemical structure, including samarium– and ytterbium–cobalt; (2) pursued nitride versions of these samples; (3) grew poly-crystal samples of 1:5 structure, including lanthanum–cobalt; (4) explored related intermetallic compounds, including growing single crystals of nickel tribismuth doped with various metals (copper, cobalt, and manganese), as well as bismuth–telluride solids and uranium–antimony compounds; (5) characterized the structural and magnetic phase diagrams of samarium–cobalt to 50 GPa at the Advanced Photon Source at Argonne National Laboratory, with both experiment and theory in agreement; (6) began exploring physical properties of new magnetic samples as a function of pressure; and (7) performed neutron scattering on lanthanum–cobalt samples as a function of pressure at the new SNAP beam-line at the Spallation Neutron Source at Oak Ridge National Laboratory.

Proposed Work for FY14

In FY14, using our sample growth capabilities to produce single- and poly-crystalline samples related to the 2:17 structure, we will (1) produce samples of structurally similar samarium–cobalt, which our calculations predict will undergo a volume collapse under pressure; (2) calculate and measure an expected change in magnetic moments; (3) explore the effects of pressure on the samarium–iron–nitrogen system we synthesized and characterized at ambient pressure, seeking a magnetic spin reorientation transition and a local magnetic sweet spot as a function of pressure; and (4) continue to explore the properties of materials produced in FY13 and their variants, as well as work with substitutions of thorium and uranium onto the rare earth sites in these materials.

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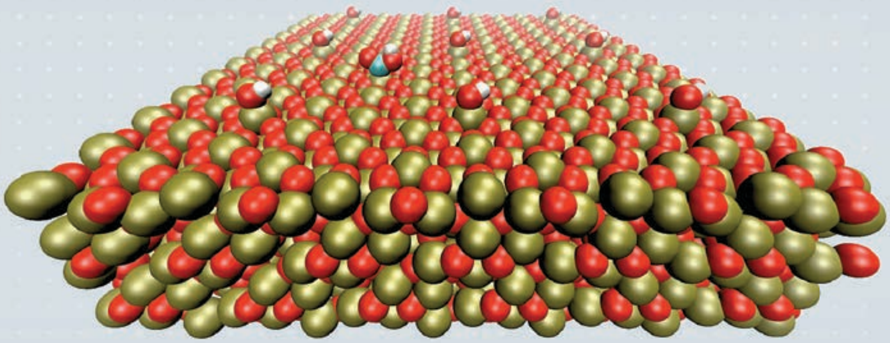
First-Principles Materials Characterization and Optimization for Ultralow-Noise Superconducting Qubits

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. This 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.



Simulations show that different molecules adsorbed onto a sapphire substrate, such as hydroxyl and carboxyl shown here, can induce magnetic instabilities that add noise to devices built on it. (Gold = aluminum, red = oxygen, blue = carbon, and white = hydrogen.)

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 science and technology foundations in materials on demand, high-performance computing and simulation, and measurement science.

FY13 Accomplishments and Results

In FY13 our focus was on developing sapphire substrate passivation strategies to minimize localized paramagnetic sources of decoherence, identifying paramagnetic defects on silica surfaces, and studying defects at the sapphire–aluminum interface. Specifically, we (1) examined how exposure to humidity can change surface hydroxyl termination on sapphire and affect the density of localized paramagnetic defects; (2) studied alternative chemical passivation strategies for sapphire and discovered that, for example, amidogen (NH_2) passivation can displace paramagnetic defects associated with surface hydroxyls; (3) found that surface electric fields may passivate sapphire; (4) built models of crystalline and amorphous silica surfaces and identified structural features associated with oxygen deficiency at the surface that show paramagnetism; (5) constructed atomistic models of the sapphire–aluminum interface and searched for spin-localizing defects therein; and (6) implemented the first version of an analytical circuit-level noise model to enable connecting microscopic properties to macroscopic device characteristics.

Proposed Work for FY14

In FY14 we will (1) conduct experiments to validate and test our predictions of the surface passivation of sapphire, (2) finish assessing paramagnetic defects on silica surfaces, and (3) explore the atomic structure of oxide–metal interfaces by developing a methodology to construct relevant atomistic models of real device interfaces by assessing features of the interfaces—that is, native and impurity-related structures—that contribute to decoherence, and using the sapphire–aluminum interface as a model. In developing our methodology, we will aim to create process options that minimize decoherence from material defects in superconducting qubits fabricated on alumina or silica substrates.

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. 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 missions. Reduction of high-fluence damage on optical glasses will allow fusion-class lasers to operate at higher fluences and with reduced sensitivity to contrast. 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.

FY13 Accomplishments and Results

In FY13 we (1) performed experiments on artificial precursor surface layers that highlighted important properties of precursors, including strong surface adhesion, and in some cases, low thermal expansion; (2) completed the optic transmission imaging system, performed high-repetition rate tests up to 100 million equivalent shots, and began fluence-scaling experiments; (3) established a stable baseline process for silica optics, which reduces damage densities at high fluence by greater than thirty-fold, and identified several sources of important precursors including trace impurities in de-ionized water; (4) successfully etched up to 10 µm with hydrofluoric vapor without introducing any new precursors; and (5) began to test initiation models for the artificial precursors we examined.

Proposed Work for FY14

In FY14 we will (1) perform surrogate precursor studies to link key material properties to damage for high-quality silica, including the importance of precipitate size, optical properties (including absorption mechanisms), and thermal–mechanical properties; (2) quantify the importance of specific precursors in high-quality optic processing and perform experiments to determine limits for certain classes of impurities; (3) explore techniques to continue damage reduction towards an intrinsic minimum; and (4) complete a full scaling study for the multipulse experiment out to a billion-shot equivalent.

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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 a large number 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.

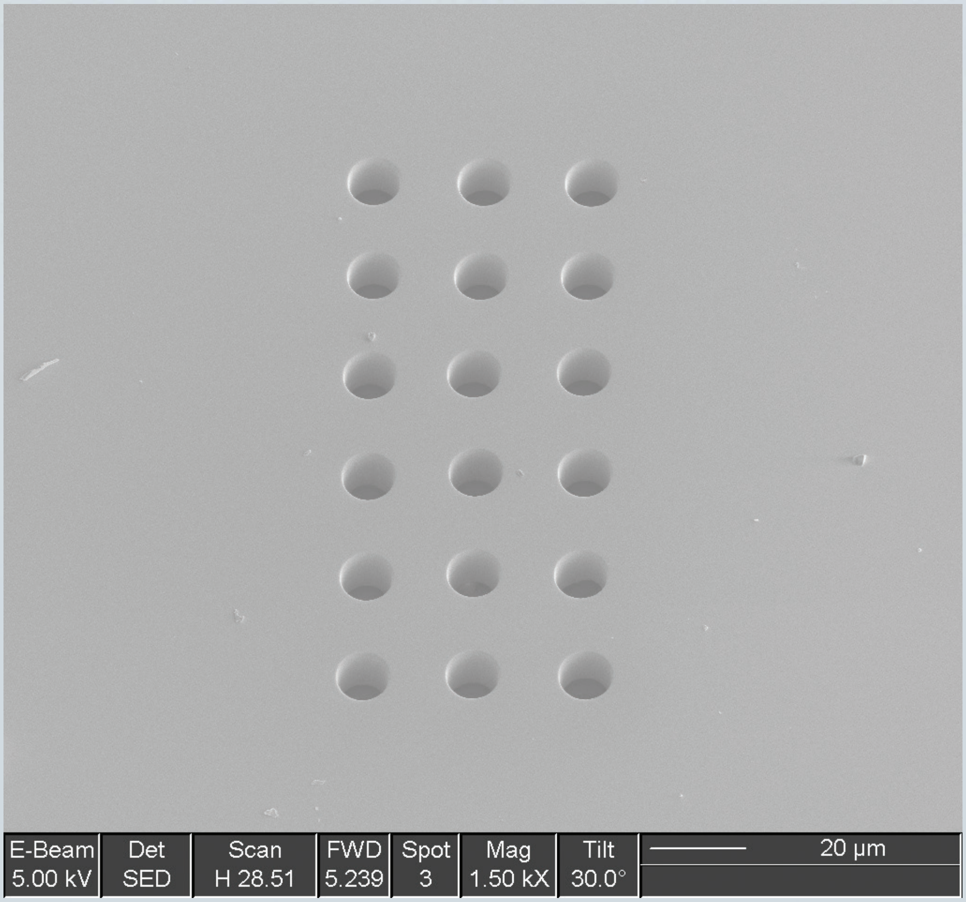
We expect to demonstrate, using a scanning superconducting quantum interference device detector, the creation and measurement of entangled topological states on the surface of a topological insulator containing many quantum bits (the quantum analogue of the classical computer bit) using coherent terahertz 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 the Laboratory's 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.

FY13 Accomplishments and Results

In FY13 we (1) obtained an optical cryostat and outfitted it with an window capable of transmitting infrared light, a system for holding and positioning our telluride samples



An electron micrograph of an array of holes made in bismuth telluride, using a focused ion beam, for research into effects of the holes on a material's surface resistance.

with microscopic precision, and a system for cooling the samples to liquid helium temperatures; (2) installed this cryostat and equipped it with infrared lasers operating at wavelengths of 3.3 and 10 µm; and (3) constructed an optical system for aligning the laser beams and focusing them onto the telluride samples inside the cryostat.

Proposed Work for FY14

In FY14 we will extend our studies of spin-filtered surface photocurrents to longer wavelengths using either a 22-µm quantum cascade laser or a terahertz-pulse generated using a femtosecond laser. A 22-µm laser would allow us to measure directly the surface conductivity of bismuth telluride, while a terahertz source would be useful for both bismuth telluride and bismuth–antimony alloys, which have a higher surface mobility than bismuth telluride. In addition, we will demonstrate, in collaboration with Lawrence Berkeley National Laboratory, that we can use nitrogen vacancy spins in diamond nanometer-scale crystals to observe the spatial distribution of surface currents on topological insulator samples punctured with holes—such an observation will open the door to using these surface currents for quantum computation.

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.

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. Our work 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.

FY13 Accomplishments and Results

In FY13 we (1) performed in situ Raman measurements on free-standing graphene electrodes, (2) developed next-generation graphene electrodes with higher energy density, (3) synthesized boron and nitrogen-doped carbon aerogels, (4) started to study the effect of doping on electronic structure and energy storage capacitance in these structures, (5) demonstrated the concept of electrochemically gated bulk transistor materials and measured gating induced conductivity changes of 400%, (6) performed atomic-scale simulations on potential-dependent electrode and electrolyte interactions that confirm the experimentally observed potential-induced changes of the electronic structure, and (7) achieved performance improvements by surface functionalization.

Proposed Work for FY14

In FY14 we will continue to use a combination of experimental and theoretical tools to improve our fundamental understanding of interfacial charge-transfer phenomena on interface-controlled materials, and apply this information to develop the next generation of energy-storage materials. Specifically, we will (1) study the fundamental mechanism of electrochemically gated resistivity changes in graphene-based electrodes, (2) evaluate nonaqueous electrolytes, (3) begin mesoscale modeling of charge transport, (4) include potential and electrolytes in atomic-scale modeling, (5) explore bottom-up doping with functional building blocks, and (6) further push the limit of energy density by combining surface functionalization with improved hierarchical architectures.

Publications and Presentations

Biener, J., et al., 2012. “Macroscopic 3D nanographene with dynamically tunable bulk properties.” *Adv. Mater.* **24**(37), 5083. LLNL-JRNL-520232.

Kalluri, R. K., et al., 2013. “Partition and structure of aqueous sodium–chloride and calcium–chloride-2 electrolytes in carbon-slit electrodes.” *J. Phys. Chem.* **117**(26), 13609. LLNL-JRNL-610512.

Kalluri, R. K., et al., 2013. “Unraveling the potential and pore-size dependent capacitance of slit-shaped graphitic carbon pores in aqueous electrolytes.” *Energy Env. Sci.* **15**(7), 2309. LLNL-JRNL-571333.

Shao, L. H., et al., 2012. “Electrically tunable nanoporous carbon hybrid actuators.” *Adv. Funct. Mater.* **22**(14), 3029. LLNL-JRNL-601453.

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.

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 1053 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 manufacturing 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 strategic mission thrust in advanced laser and optical materials. 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.

FY13 Accomplishments and Results
In FY13 we (1) characterized three base-glass systems for suitability of doping with iron–tin–carbon and found that the best host would be fused silica—however, the spectroscopy must be studied; (2) continued oxidation–reduction ion-pair studies and developed an understanding of dopant concentrations and effects on the reaction to drive the oxidation state of iron-3+ more fully to iron-2+; (3) collaborated with the University of Missouri to obtain materials and characterizations using Mossbauer spectroscopy and electron spin resonance measurements to detect valence states and other factors that affect molecular coordination; (4) finalized solution studies for studying copper and iron oxidation–reduction effects based on the solubility of starting materials; (5) studied the effects of disorder on the absorption of dopant ions;

(6) completed modelling to benchmark the copper solution work; (7) began work on the feasibility of doping iron–tin–carbon into fused silica; and (8) benchmarked a model for doped solutions—however, it was determined that a more advanced model was required to examine coordination environments and the effect on absorption characteristics.

Proposed Work for FY14

In FY14 we will (1) continue to define the best host material for the dopant ion-pair system; (2) continue dopant ion-pair studies with an emphasis on understanding dopant concentrations and effects on the oxidation–reduction reaction; (3) continue to develop and exercise the most advanced model to simulate optical properties of dopant ions and host materials with the goal to actively predict the absorption position and coordination environment of the dopant ion; (4) characterize the high-fluence behavior of the most promising material, fused silica, to determine suitability as a red blocker optic; and (5) identify a vendor for proof-of-principle tests and scaling to a usable optical filter.

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.* OCEC 2013, London, England, Sept. 22–26, 2013. LLNL-JRNL-598992-DRAFT.

Super-Strained Three-Dimensional Semiconductors

Lars Voss (12-LW-043)

Abstract

The electronic properties of materials can be controlled by applying elastic strain. However, there are limits to the materials that can be used and the volume of material that can be strained using conventional techniques. Conventionally strained semiconductor technology relies on lattice mismatch—to achieve a desired strain, a specific combination of semiconductors must be used that dictates the fundamental properties of the resulting device. We propose a potentially revolutionary new class of strained semiconductor technology that will use thin-film-coated, three-dimensional structures to apply large, volumetric strains to semiconductors, up to the fracture point. By applying the strain externally using thin-film deposition, we will enable an additional degree of freedom when designing advanced devices such as laser and light-emitting diodes, photovoltaics, and sensors. The precise amount of strain applied will be tuned by the choice of thin film, deposition technique, and deposition parameters to enable control over the full range of electronic properties achievable within a given semiconductor.

We expect to enable a new class of highly tunable semiconductor devices that do not rely on hetero-structures of differing materials for control over the electronic

properties. In addition, the external strain achieved through our approach will allow for greater volumetric effect and will decouple the choice of materials from the achievable strain. Thus, the approach will impact devices that rely on both single semiconductors and multiple semiconductor hetero-structures, enabling performance beyond what is currently achievable.

Mission Relevance

This work is relevant to LLNL missions in that it will impact a broad array of semiconductor devices. These include both laser and light-emitting diodes in support of Livermore's strategic mission thrust in inertial-confinement fusion energy; gas, radiation, and other types of sensors in support of stockpile stewardship and nuclear threat reduction; and energy-harvesting technologies such as photovoltaics and thermoelectrics in support of energy security.

FY13 Accomplishments and Results

In FY13 we (1) unambiguously demonstrated our proof of concept using a gallium arsenide substrate with silicon nitride dielectric coatings to achieve a blue shift in the band gap of 51 MeV; (2) built a test system to perform photocurrent measurements using a variety of light sources with varying wavelengths in the infrared, which allowed us to demonstrate a photocurrent response of strained silicon to light at energies below 1 eV, although the results are not completely conclusive because of complicating physics; and (3) demonstrated large strains on various orientations of silicon pillars using conventional dielectrics—for the same coating the strain state differs significantly, with important implications for device design in future work.

Project Summary

With this project we demonstrated large strains over relatively large volumes of semiconductor material, specifically silicon and gallium arsenide. These strains were shown to measurably shift the band gap, particularly in the case of gallium arsenide. This demonstration also required construction of various characterization stations, including a uniform, temperature-controlled electrical station and a photocurrent station utilizing infrared light. Our results have the potential to impact numerous electronic devices for emitting, modulating, transmitting, and sensing light, and will enable the extension of strain engineering of semiconductors to larger volumes of material, with particular import for devices such as photovoltaics and photodetectors, which rely on relatively large thickness. This result is not limited to these material systems and the technology can be applied to any semiconductor of interest. We are exploring potential funding opportunities to continue both the basic science of our technology as well as device applications. We are also preparing a DOE Early Career Research Proposal based on the results generated in this project.

Publications and Presentations

Voss, L. F., et al., 2013. "Analysis of strain in dielectric coated three dimensional Si micropillar arrays." *J. Vac. Sci. Tech. B* **31**(6), 060602. LLNL-JRNL-426150.

Voss, L. F., et al., 2013. *Band gap shift of highly strained, microstructured Si p-n diodes.* 2013 MRS Spring Mtg. and Exhibit, San Francisco, CA, Apr. 1–5, 2013. LLNL-ABS-595632.

Voss, L. F., et al., 2013. "Blue shift of GaAs micropillars strained with silicon nitride." *Appl. Phys. Lett.* **103**(21), 212104. LLNL-JRNL-644638.

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 develop 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 Livermore Valley Open Campus Initiative to address urgent problems in energy-related applications.

FY13 Accomplishments and Results

In FY13 we (1) extended our electrochemical phase-field framework to include anisotropic strain coupling, surface orientation dependence, and basic charge-transfer kinetics for a model being used to study surface phase-segregation dynamics of lithium iron phosphate cathodes under cycling; (2) analyzed microstructure evolution and failure mechanisms in silicon anodes theoretically and experimentally; (3) began ab initio calculations of lithium binding on model graphene-derived anodes;

(4) demonstrated atomic layer and chemical vapor deposition methods for controlled deposition of metal oxides on carbon anodes and on different cathode micro-architectures; (5) collected and began analysis of in situ and ex situ x-ray spectra of carbon anodes and lithium iron phosphate cathodes during cycling; and (6) demonstrated orientational dependence of (de)lithiation of cathode materials.

Proposed Work for FY14

For FY14 we will (1) continue coupling the phase-field models for electrolyte and electrode transport, including a structurally dependent diffusivity model; (2) begin integrating first-principles electrochemical interface parameters into the mesoscale model by mapping microscopic properties of the materials into measurable quantities for experimental validation; (3) continue controlled synthesis of carbon and molybdenum disulfide anode materials via atomic layer and chemical vapor deposition and solution chemistry; (4) collect x-ray, transmission electron microscopy, and electrochemical cycling data of synthesized anode materials to improve the models and identify problems for further investigation; and (5) perform first-principles calculations of voltage-dependent lithium storage and diffusion in model anodes based on realistic features in carbon, silicon, and molybdenum disulfide materials.

Publications and Presentations

An, Y., M. Tang, and H. Jiang, 2013. *Phase field simulation on the doncurrent plastic deformation, phase transformation, and mass diffusion in silicon anode in lithium-ion batteries.* TMS 2013 142nd Annual Mtg. and Exhibit, San Antonio, TX, Mar. 3–7, 2013. LLNL-PRES-622892.

Lee, J. R. I., et al., 2012. *In situ soft x-ray spectroscopy for the vnvestigation of advanced materials for electrical energy storage.* Advanced Light Source Users' Mtg., Berkeley, CA, Oct. 7–9, 2012. LLNL-PRES-591092.

Tang, M., and A. Karma, 2012. "Surface modes of coherent spinodal decomposition." *Phys. Rev. Lett.* **108**, 265701. LLNL-JRNL-513943.

Ye, J. C., et al., 2013. "Unusual lithiation and fracture behavior of silicon mesoscale pillars pre- and post- atomic layer deposition of ultrathin coatings." *J. Power Sourc.*, **248**, 447. LLNL-JRNL-640293.

Multilayer Thin-Film Science for Core Missions

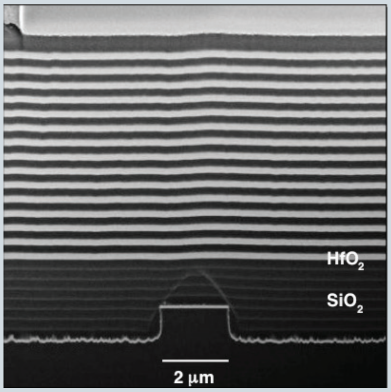
Regina Soufli (12-ERD-055)

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 in Palo Alto 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.



Defect planarization increases multilayer mirror laser resistance to exceed 100 J/cm^2 at a wavelength of 1064 nm and 10-ns pulse length.

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 science, technology, and engineering foundations in materials on demand and measurement science and technology.

FY13 Accomplishments and Results

In FY13 we (1) began elucidating the physics of intermixed aluminum–magnesium corrosion barrier layers via transmission electron microscopy and analysis; (2) optimized magnesium–silicon carbide multilayer designs with corrosion barriers and demonstrated efficient experimental reflective performance at wavelengths of 25 to 80 nm; (3) received one issued U.S. patent; (4) demonstrated, for hafnia–silica multilayers, planarized defects with performance of greater than 100 J/cm^2, which is more than 20-times higher than non-planarized defects; (4) optimized planarization of multiple materials; and (5) demonstrated planarization during multilayer deposition to address coating defects.

Proposed Work for FY14

In FY14 we will (1) conclude studies of the physics of intermixed aluminum–magnesium corrosion barrier layers and of the long-term lifetime (aging) properties of magnesium–silicon carbide multilayers; (2) work with external collaborators toward implementation of the newly developed corrosion-resistant magnesium–silicon carbide multilayer coatings on optics for solar physics, plasma physics, and x-ray laser applications; (3) optimize the smoothing of defects during hafnia–silica multilayer deposition for high laser-damage resistance; and (4) develop external collaborations to scale this technology for mirrors large enough for commercial high-fluence laser systems.

Publications and Presentations

Fernandez-Perea, M., et al., 2012. "Triple-wavelength, narrowband Mg-SiC multilayers with corrosion barriers and high peak reflectance in the 25–80 nm wavelength region." *Optic. Exp.* **20**(21), 24018. LLNL-JRNL-574772.

Soufli, R., M. Fernández-Perea, and J. C. Robinson, 2013. "Corrosion-resistant Mg/SiC multilayer coatings for EUV laser sources in the 25–80 nm wavelength region. *Proc. SPIE* **8849**, 88490D. LLNL-PROC-645327.

Soufli, R., et al., 2012. "Corrosion-resistant, high-reflectance Mg–SiC multilayer coatings for solar physics in the 25–80 nm wavelength region." *Proc. SPIE* **8443**, 84433R. LLNL-PROC-575279.

Soufli, R., et al., 2012. "Spontaneously intermixed Al-Mg barriers enable corrosion-resistant Mg/SiC multilayer coatings." *Appl. Phys. Lett.* **101**(4), 043111. LLNL-JRNL-561992.

Soufli, R., et al., 2013. *Spontaneously intermixed Al-Mg coatings as efficient corrosion barriers in Mg/SiC multilayers.* Optical Interference Coatings, Whistler, Canada, June 16–21, 2013. LLNL-ABS-645491.

Stoltz, C. J., et al., 2013. "High laser-resistant multilayer mirrors by nodular defect planarization." *Appl. Optic.* **53**(4), A291 LLNL-JRNL-642712-DRAFT.

Stoltz, C., et al., *Nodular defect planarization to significantly improve multilayer mirror laser resistance.* OSA Optical Interference Coatings, Whistler, Canada, June 16–21, 2013. LLNL-ABS-620553-DRAFT.

Giga-Shot Optical Laser Damage

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? This knowledge will enable the next-generation lasers critical to Lawrence Livermore's research, and will leverage existing Laboratory expertise in optical system design, damage testing, and simulation.

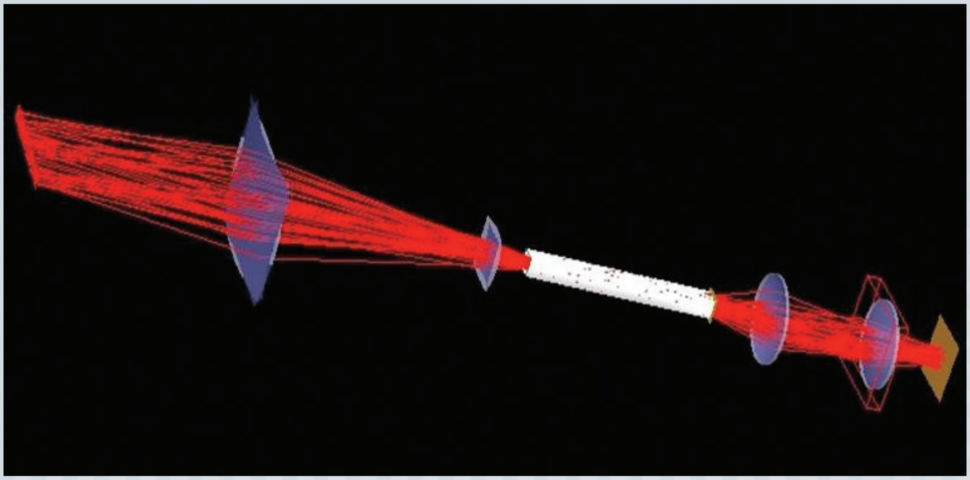
We expect to develop a novel high-energy, high-repetition-rate laser capable of exploring optical materials behavior and optical damage testing for extremely long exposures. A successful project will enable the first optical damage data on extreme timescales of one billion pulses for key laser materials. 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. 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 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.

FY13 Accomplishments and Results

In FY13 we (1) established a laboratory infrastructure and activated the front end of the laser system, with suitable spatial and temporal characteristics; (2) completed design of



Ray tracing simulation of the optics that deliver light from the semiconductor laser pumps to the main amplifier gain medium for use in exploring optical materials damage under pulsed exposure over extreme timescales.

the overall laser system, which included development of a novel amplifier head suitable for high-repetition-rate, high-energy operation with minimal thermal birefringence and strain; (3) completed the two key research and development tasks of pump diode design and simulation as well as a demonstration of a novel amplifier head-bonding technology; and (4) developed and demonstrated the front end of the laser.

Proposed Work for FY14

In FY14 we will continue development of the laser, including activation of the main amplifier of the laser chain. Specifically, we will (1) perform a design review of the experimental campaign to be conducted in FY15; (2) commission the 10-J amplifier, and demonstrate system capability in a damage test exposure of at least 1-million-shot duration; (3) complete the frequency converter required to generate 0.35-μm light from the 1-μm amplifier output; and (4) perform a 10- to 100-million-shot experiment and develop associated analyses and simulations of the small-section fused-silica damage, which will provide data on the growth probabilities of very small damage sites.

Publications and Presentations

Bayramian, A., 2013. *High energy repetition-rate average-power laser driver (HERALD) for the Dynamic Compression Sector (DCS) at the Advanced Photon Source (APS).* LLNL-TR-639459.

Deri, R. J., 2013. *Engineering diode laser pumps for extremely large-scale laser systems.* Conf. Lasers and Electro-Optics, San Jose, CA, June 9–14, 2013. LLNL-ABS-623012.

A Three-Dimensional Radioisotope Battery

Rebecca Nikolic (13-ERD-004)

Project Description

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 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 use of wideband-gap, radiation-tolerant semiconductors and amorphous semiconductors to greatly extend the lifetime of the battery. We will carry out fundamental design work to determine the 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 successful 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 Department of Defense, the Intelligence Community, and beyond.

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 material and actinide sciences, as well as enabling new energy sources for military applications.

FY13 Accomplishments and Results

In FY13 we (1) worked towards developing a portfolio of intellectual property by submitting one patent application, (2) prepared three-dimensional semiconductor structures for template growth of icosahedral boron phosphide and crystal growth of these films, and (3) achieved proof-of-principle current generation in planar semiconductor structures coated with uranium oxide.

Proposed Work for FY14

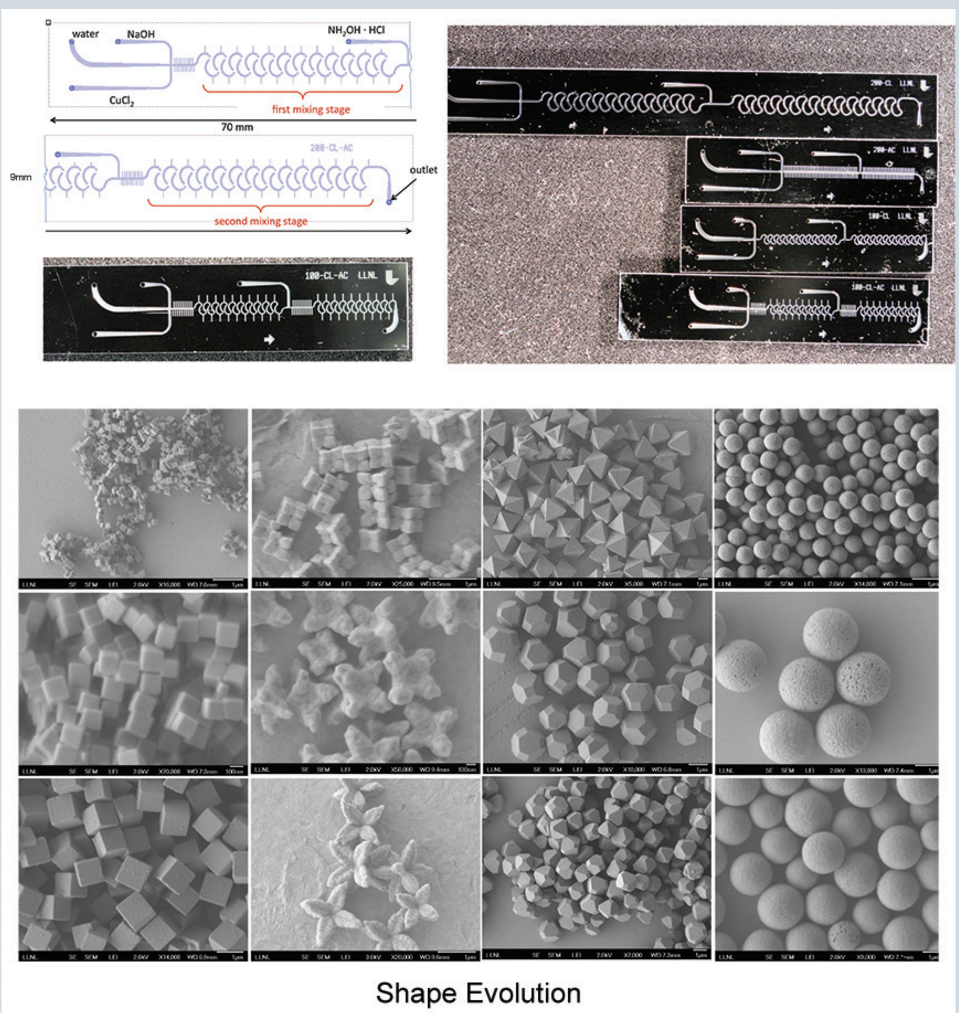
In FY14 we will (1) determine the relationship between alpha and beta dose, displacements per atom, output power, and corresponding battery longevity for silicon and at least one semiconductor with a higher band gap (e.g., gallium arsenide, silicon carbide, or icosahedral boron); (2) model thermal properties of semiconductor structure and develop heat sink approaches; (3) template crystal growth of icosahedral boron and characterize its transport properties and electrical conduction; (4) coat ridge or pillar semiconductor structures with a radioisotope; and (5) demonstrate output power.

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 preprogrammed building blocks that can assemble predictively or as directed into structures with unique functions and properties. These materials would also have the ability to self-regulate or adapt properties such as electrical conductivity, elasticity, or magnetic, optical, or mechanical properties. We propose to understand the governing principles of nanometer-scale particle assembly to create three-dimensional composite materials with multiple components. 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



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 (below).

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 to control and manipulate the emerging properties, providing a paradigm shift in materials science.

We expect that the proposed advances in materials science we propose will result in the rapid synthesis, functionalization, 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 strategic focus area of materials on demand 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.

FY13 Accomplishments and Results

In FY13 we (1) designed and fabricated a simplified synthesis platform; (2) performed preliminary experiments to synthesize nanoparticles with the synthesis platform; (3) synthesized copper oxide particles using bulk synthesis methods, optimizing the process and parameters to achieve desired sizes and shapes; and (4) tested the feasibility of using copper oxide particles synthesized by bulk process as feed materials for additive manufacturing, especially electrophoretic deposition.

Proposed Work for FY14

We will (1) continue developing the continuous-flow synthesis platform by improving the micro-mixer design and fabricating a functionalization and aging chamber with in situ characterization windows, (2) incorporate heating control in the flow system, (3) identify and integrate an in situ characterization tool, (4) synthesize copper oxide particles with size and shape control using our continuous flow synthesis system, (5) demonstrate the capability of creating feedstock for additive manufacturing, and (6) functionalize copper oxide particles with ligands (surfactants or biomolecules) for directed assembly at the mesoscale.

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

Francesco Fornasiero (13-ERD-030)

Abstract

Carbon nanotubes (CNTs) 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 CNT as a flow channel. With this device and synergistic multiscale simulations, we will (1) decouple transport modes in CNTs, (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 CNT ionic diodes, and (5) understand and control the selectivity of single-molecule translocation through local CNT functionalization.

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 biomolecule 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 mission thrust area of biological, chemical, and explosives security 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. This effort in world-class nanofluidics and advanced functional nanomaterials will also bolster LLNL's science and technology foundation in measurement science.

FY13 Accomplishment and Results

In FY13 we (1) successfully fabricated a novel nanofluidic platform—a Coulter counter having one or more CNTs (each less than 5–nm wide) as flow channels—by using a focused ion beam to locally remove material at the surface of a CNT to open a single or a few CNTs to fluid flow, (2) recorded transient current spikes with a single current level after introducing a small polyanionic dye with a size comparable to the CNT diameter in one of the chambers containing potassium chloride solution—these spikes suggest translocation of the dye through a single nanochannel of similar diameter,

(3) developed a multiscale computational platform that combines quantum-mechanical analysis with molecular dynamics simulations and quasi-continuum calculations to examine the transport of aqueous electrolyte solutions in CNTs, and (4) investigated ionic flow in a CNT as a function of solution conditions, pore size, and field strength. Results suggest a power-law dependence of the pore conductance with respect to concentration.

Proposed Work for FY14

In FY14 we propose to (1) fabricate ionic diodes based on a CNT channel with a functional asymmetric entrance, (2) perform the corresponding multiscale simulations of ionic currents in asymmetrically functional CNTs, and (3) decouple transport modes (diffusion, electro-osmosis, electrophoresis, and pressure-driven flow) with both experiments and multiscale modeling of single-molecule translocation through a CNT. Test analytes will consist of both charged and neutral small molecules under different driving forces and in ad hoc solution conditions.

Publications and Presentations

Guo, S., et al., 2014. *A simple, single-carbon-nanotube nanofludic platform for fundamental transport studies.* Biophysical Society Mtg., San Francisco, CA, Feb. 15–19, 2014. LLNL-ABS-644567.

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 with a low probability of error at ports, borders, and airports. 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 the computation of the rare earth electronic structure, in relation to the 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

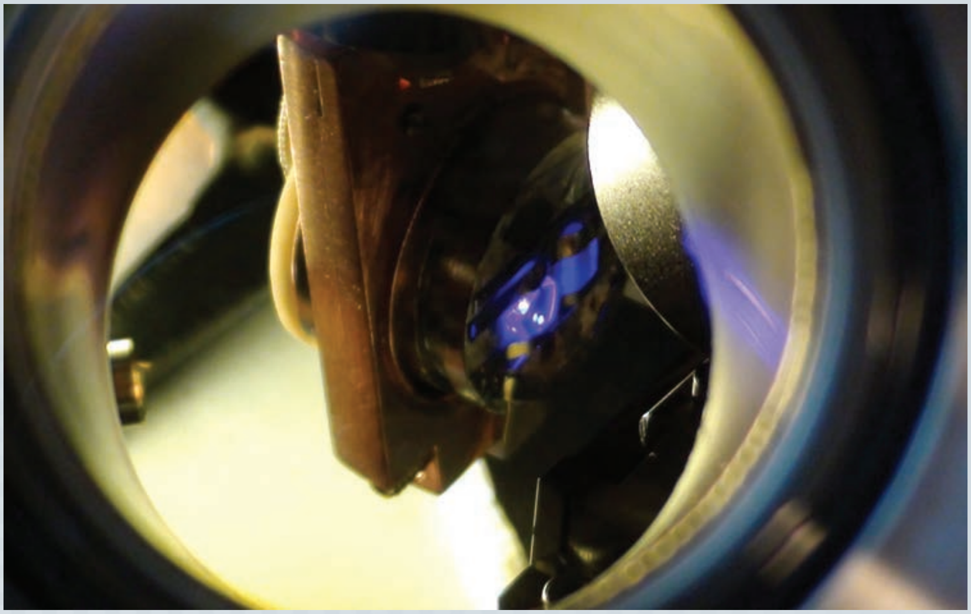
crucial elements in understanding the entire scintillation event from the initial cascade to final emission of a detectable photon. In particular, the capture cross sections determine how far out, from the initially dense track of electron–hole pairs, the electrons can be pushed before trapping occurs. 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.

Mission Relevance

The ultimate goal of this project is to develop unique computational and experimental capabilities at Lawrence Livermore to address limiting factors in scintillator radiation detector functionality. As such, it is well aligned with the Laboratory's strategic mission thrust in nuclear counterterrorism and forensics. 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.

FY13 Accomplishments and Results

In FY13 we (1) began modelling 4f energy levels in yttrium aluminum perovskite using density functional theory—we added this task because it turned out to be necessary to constrain 4f-occupation density-functional theory; (2) obtained a local basis set for cerium-doped lanthanum bromide from density-functional theory and unscreened Coulomb matrix elements; (3) interfaced lanthanide with our atomic code, and found the results agreed perfectly with model Hartree–Fock calculations; (4) determined that the standard projector augmented-wave methodology for ab initio electronic structure calculations is not suitable for the dielectric response function—therefore,



Bright scintillation with a 3-keV electron beam from the cerium-activated yttrium aluminum perovskite scintillator in ultrahigh vacuum. By measuring the emitted x rays, the electronic structures of scintillating materials such as europium-doped strontium iodide and cerium-doped lanthanum bromide are being studied to find correlations between electronic structures and the material's performance as a radiation detector.

we transferred our focus to all-electron techniques with a mixed product basis set used in strongly correlated problems (within the dynamical mean field theory); (5) determined that the activator position in the band can indeed be found with x-ray absorption spectroscopy and x-ray emission spectroscopy only using a non-hygroscopic system of yttrium aluminum perovskite activated by cerium; and (6) began studies on the hygroscopic systems of europium-doped strontium iodide and cerium-doped lanthanum bromide.

Proposed Work for FY14

In FY14 we will (1) continue using the random phase approximation to model screened Coulomb interactions, a challenging task that is in the forefront of what is possible today in dynamical mean field theory of correlated materials; (2) establish a collaboration with the Vienna Ab initio Simulation Package group to advance state-of-the art methods, which will lead to improved spectra; and (3) determine the activator (europium and cerium) positions within the band gaps of europium-doped strontium iodide and cerium-doped lanthanum bromide with x-ray absorption spectroscopy and x-ray emission spectroscopy.

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. In May 2013, the DOE announced establishment of a new Energy Innovation Hub for research on critical materials, including rare earth elements, important for a growing number of energy technologies. 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 materials 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 core competency in computational materials. It also fits closely with the DOE's newly established Energy Innovation Hub and the search for rare earth element substitutes.

FY13 Accomplishments and Results

In FY13 we (1) constructed an ab initio model that accounts for non-collinear magnetic excitations in iron; (2) worked towards a fully ab initio description of coupled magnon–phonon dynamics, which was beyond what we expected to do in the first year, having realized that this development would better position us to achieve our goals in later years; (3) studied the magnetic anisotropy of lanthanum–cobalt and cobalt–platinum and formulated a consistent theory of on-site contributions to magnetic anisotropy; (4) studied the electronic structure of samarium–cobalt systems with limited success because lanthanum–cobalt is proving to be a better system for this work, as it has for experimentalists; (5) computed temperature-dependent magnetic exchanges and magnon spectra with the Korringa–Kohn–Rostoker atomic-sphere approximation; and (6) began investigating the magnetic and structural properties of lanthanum–cobolt under pressure to aid experimentalists in the analysis of recently obtained data as well as to suggest further directions.

Proposed Work for FY14

In FY14 we will (1) further develop the coupled magnon–phonon computations, both for iron and more complex materials, including a fully ab initio spin dynamics capability; (2) use this technique to compute Curie-temperature and temperature-dependent phonon spectra beyond the adiabatic spin dynamics regime; (3) continue the study of magneto–crystalline anisotropy, specifically the dependence on pressure and lattice structure of lanthanum–cobalt, cobalt–platinum, and other systems; (4) begin to study magneto-crystalline anisotropy in the presence of defects; and (5) use Korringa–Kohn–Rostoker techniques, along with a disordered local moment picture, to compute exchange parameters (including spin-wave stiffness) for permanent magnet materials, which is needed for predictive modeling of domain wall energies involved in coercivity.

Transient Loading Effects on Structural Materials for Laser Inertial Fusion Energy

Bassem El-Dasher (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 a laser inertial fusion energy engine. Although the behavior of the steels is reasonably understood in steady-state conditions, the greatest threat to the fusion energy process 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 environments relevant to laser inertial fusion energy, 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 a benchmark to the above data. 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 designs for laser inertial fusion energy technology.

Mission Relevance
This project will improve our understanding of candidate structural materials for achieving a laser inertial fusion energy engine as a potentially revolutionary carbon-free energy technology, in support of the Laboratory's mission in energy and environmental security.

FY13 Accomplishments and Results
In FY13 we (1) built most of the high-temperature fatigue facility; (2) procured, received, and installed the high-rate fatigue tester; (3) designed, procured, and received the glove-box enclosure; (4) designed and fabricated a dome for the test specimens and test fixtures; (5) secured high-purity lithium suppliers; and (6) reviewed our experimental plan with a specialist vendor to verify that geometries of the test specimens were adequate and useful.

Proposed Work for FY14
In FY14 we will quantify the effect of embrittled lithium and high-temperature effects on ductility by fatigue-testing alloy materials, including fatigue-crack growth-rate measurements at room and elevated temperature, with and without lithium, and alternating stress-amplitude fatigue-life curves for the same sets of conditions.

Quantum Monte Carlo Benchmarks for Materials on Demand

Randolph Hood (13-ERD-067)

Abstract

The current grand challenge in condensed matter and chemical physics for development of a predictive foundation for materials on demand is the accurate solution of the Schrodinger 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 for understanding the behavior of materials at the level of the electrons that bind the atoms together. Monte Carlo algorithms rely on repeated random sampling to compute their results. 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 propose 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 capability 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.

FY13 Accomplishments and Results

In FY13 we (1) tested and applied the existing quantum Monte Carlo code base to computations of equation-of-state properties (equilibrium lattice constant, pressure versus atomic volume, bulk modulus, and cohesive energy) of various transition and other metals; (2) assessed the effects of nodal errors and choice of pseudo-potentials on one or more of these systems (i.e., an approximation for the simplified description of complex systems); (3) determined if any new development is required of our existing quantum Monte Carlo capability and incorporated these into our CASINO and QMCPACK computer programs for quantum Monte Carlo calculations; and (4) hired a postdoctoral researcher.

Proposed Work for FY14

In FY14 we will (1) perform extensive calculations of band gaps and spectral data of sodium chloride, magnesium hydride, silicon carbide, and gallium phosphide solids; (2) assess the effects of nodal errors and choice of pseudo-potentials on one or more of these systems; and (3) determine, in one or more solids, phase stability at high pressure predicted by quantum Monte Carlo.

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 dependence on kinetics, 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. We will develop diffraction techniques with our collaborators using an instrument at the Advanced Photon Source (APS) 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 for 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 put Livermore in a leadership role in developing and commissioning the Dynamic Compression Sector beam-line at APS.

Mission Relevance

High-fidelity materials models are a critical piece of the multiphysics simulation codes employed in stockpile stewardship, and this project will enable the rigorous verification and validation of cutting-edge predictive models for strength and phase diagrams of metals under extremes of pressure and strain rate. This work also strengthens the Laboratory's foundations in measurement science and technology.

FY13 Accomplishments and Results

In FY13 we (1) developed dynamic Laue diffraction experiments with online one-metal targets using a reflection geometry on the APS 32-ID-B beam-line, (2) worked with other LLNL collaborators to lay out the arbitrary waveform compression-drive specifications for a 100-J laser system to be fielded at the Dynamic Compression Sector at APS—the approximately 2-J system is ready for measurements in FY14, (3) expanded the scope of the project to include pilot experiments on the Linac Coherent Light Source at the SLAC National Accelerator Laboratory in Menlo Park as well as on the Janus laser at LLNL—the latter involving backlighted diffraction data, (4) performed a feasibility study on using a betatron x-ray source that has been developed at Livermore, and (5) added a full-time graduate student to assist with laser-based experiments and general data analysis.

Proposed Work for FY14

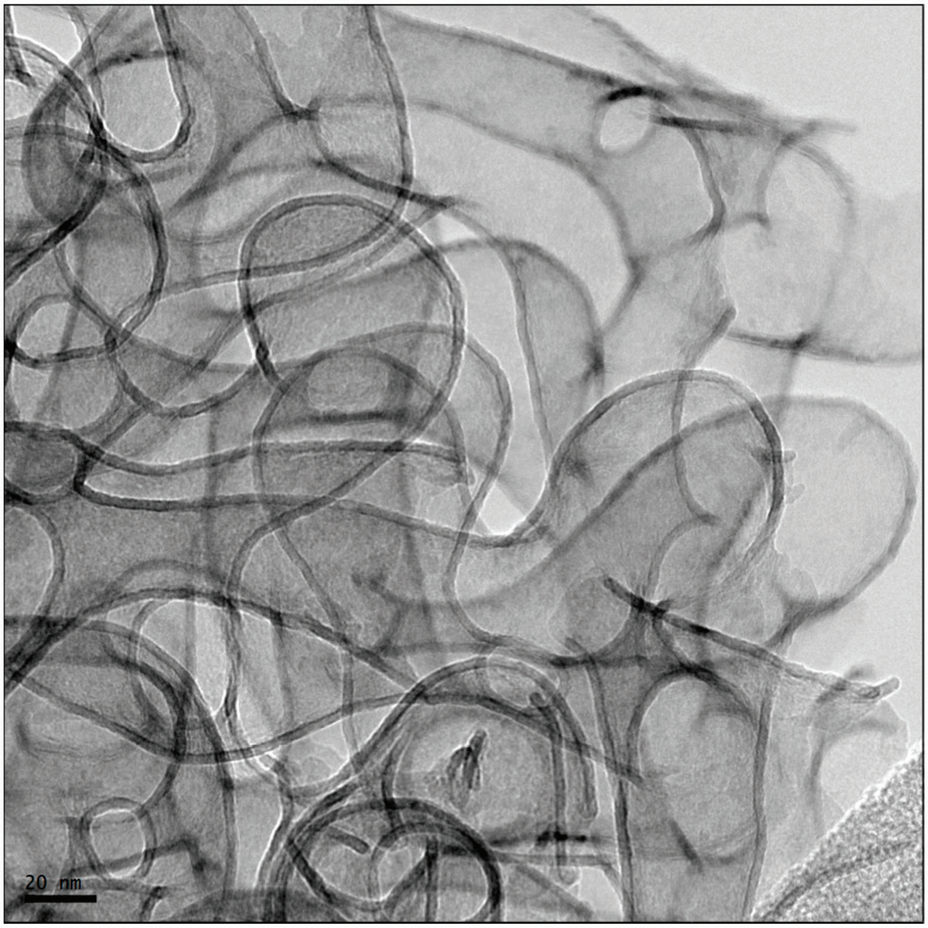
In FY14 we will (1) begin to contribute to the commissioning of the Dynamic Compression Sector at APS, with a large amount of beam-time to refine the dynamic Laue diffraction measurements; (2) explore the use of a Livermore-developed bremsstrahlung electromagnetic radiation source for performing Laue diffraction at the OMEGA laser at the University of Rochester and Livermore's National Ignition Facility; (3) have the new graduate student perform iron–nickel experiments on multiple laser platforms, as well as at APS; (4) perform, contingent on the new K-B mirrors for sophisticated x-ray focusing installed at beam-line 32-ID at APS, our first dynamic Laue diffraction measurements on iron; and (5) use the high-energy monochromatic beam at APS 1-ID to perform dynamic powder diffraction, the results of which will have a direct impact on the final design of the Dynamic Compression Sector.

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



Nanometer-scale tubular freestanding titanium dioxide with a wall thickness of about 2 nm obtained by coating a nanoporous gold template with atomic-layer deposition, and subsequent removal of the gold core by a wet etch process.

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 materials on demand. 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.

FY13 Accomplishments and Results

In FY13 we (1) fabricated and characterized nanoporous gold coated with titania and zinc oxide by atomic-layer deposition, (2) tested the performance of titania-coated nanoporous gold as a lithium-ion battery electrode material and demonstrated its high catalytic potential for exhaust control, (3) demonstrated fabrication of ultralow-density (>5 mg/cm^2) nanoscale tubular titania and zinc oxide bulk materials with precisely controlled density and morphology, (4) optimized the etching process parameters resulting in reliable and reproducible results, (5) began to explore alternative template materials such as polystyrene beads (more cost effective) and micro-trusses (different microstructure), and (6) initiated modification of the chamber for iron oxide atomic-layer deposition and photo-catalytic testing.

Proposed Work for FY14

In FY14 we will (1) continue fabrication and characterization of atomic-layer-deposition functional bulk nanoporous gold; (2) continue to test materials for catalytic, solar energy harvesting (water splitting), as well as energy storage applications; and (3) continue to explore fabrication of ultralow-density nanoscale tubular bulk materials with a focus on improving the etch parameters to remove the template core, and on improving the etch parameters for alternative template structures such as polystyrene beads, block copolymers, and engineered micro-trusses.

Publications and Presentations

Bagge-Hansen, M., et al., 2013. *Phase/catalytic activity correlations of TiO_2 ALD functionalized nanoporous gold*. AVS 60th Anniversary Symp. and Exhibition, Long Beach, CA, Oct. 27–Nov. 1, 2013. LLNL-ABS-636488.

Bagge-Hansen, M., et al., 2013. *Quantitative phase composition of TiO_2-coated nanoporous-Au monoliths by x-ray absorption spectroscopy and correlations to catalytic behavior*. LLNL-JRNL-638539.

Biener, M. M., et al., 2013. "Atomic layer deposition for rapid on demand development of ultra-low density bulk materials with deterministic density and composition." *ACS Appl. Mater. Interface*., **5**(24), 13129. LLNL-JRNL-638994.

Biener, M. M., 2013. *Functional Cellular Bulk Materials via Atomic Layer Deposition*. 3rd Intl. Conf. Multifunctional, Hybrid and Nanomaterials, Sorrento, Italy, Mar. 3–7, 2013, and 13th Intl. Conf. Atomic Layer Deposition (ALD 2013), San Diego, CA, July 28–31, 2013. LLNL-PRES-612174-DRAFT.

Biener, M. M., et al., 2013. *Ultralow density nanotubular bulk materials with deterministic feature size and density*. LLNL-JRNL-645195-DRAFT.

Shin, S. J., et al., 2013. *Robust nanoporous alumina monoliths by atomic layer deposition on low-density carbon-nanotube scaffolds*. LLNL-JRNL-644426-DRAFT.

Wichmann, A., et al., 2013. "Maximizing activity by turning gold catalysis upside down: Oxide particles on nanoporous gold." *ChemCatChem*. **5**(7), 2037. LLNL-JRNL-561433.

Ye, J. C., et al., 2013. *Unusual lithiation and fracture behavior of silicon mesoscale pillars pre- and post- atomic layer deposition of ultrathin coatings.* LLNL-JRNL-640293.

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. 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 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 our ability to detect explosives, chemical agents, and biological threats as well as environmental contaminants, biomedical markers, and more.

FY13 Accomplishments and Results

In FY13 we (1) successfully demonstrated the assembly of micron-sized polystyrene particles onto electron-beam lithographically patterned electrodes with particle-to-particle precision, (2) scaled down hole patterns to accommodate metal nanoparticles where their optical properties will be tuned and optimized for surface-enhanced Raman sensing of a target molecule, (3) submitted a user proposal for the Molecular Foundry at Lawrence Berkeley National Laboratory to obtain optical measurements of the metal nanoparticle assemblies, and (4) conducted numerical simulations of an electric field around a patterned electrode hole and the potential energy experienced by a particle near the patterned hole.

Proposed Work for FY14

In FY14 we will (1) polymerize the nanoparticle junctions to suspend and release assemblies from the substrate, (2) demonstrate optimization of the surface-enhanced Raman signal using assemblies with the desired surface plasmon resonance, and (3) perform numerical simulations for analysis of surface plasmon resonance of the nanoparticle assemblies, which is expected to match experimental results.

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 operation 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. It would enhance the economic and energy security of the nation by reducing imports of energy from foreign sources.

FY13 Accomplishments and Results

During FY13 we (1) fabricated a metal sulfide-coated catalyst and characterized its thermal stability in a catalytic environment, (2) determined the catalyst using the thin carbon-based scaffold showed an order-of-magnitude better durability than conventional carbon scaffolds (7 vs. 70% weight loss), and (3) performed surface area, conductivity, and composition characterizations of the catalyst.

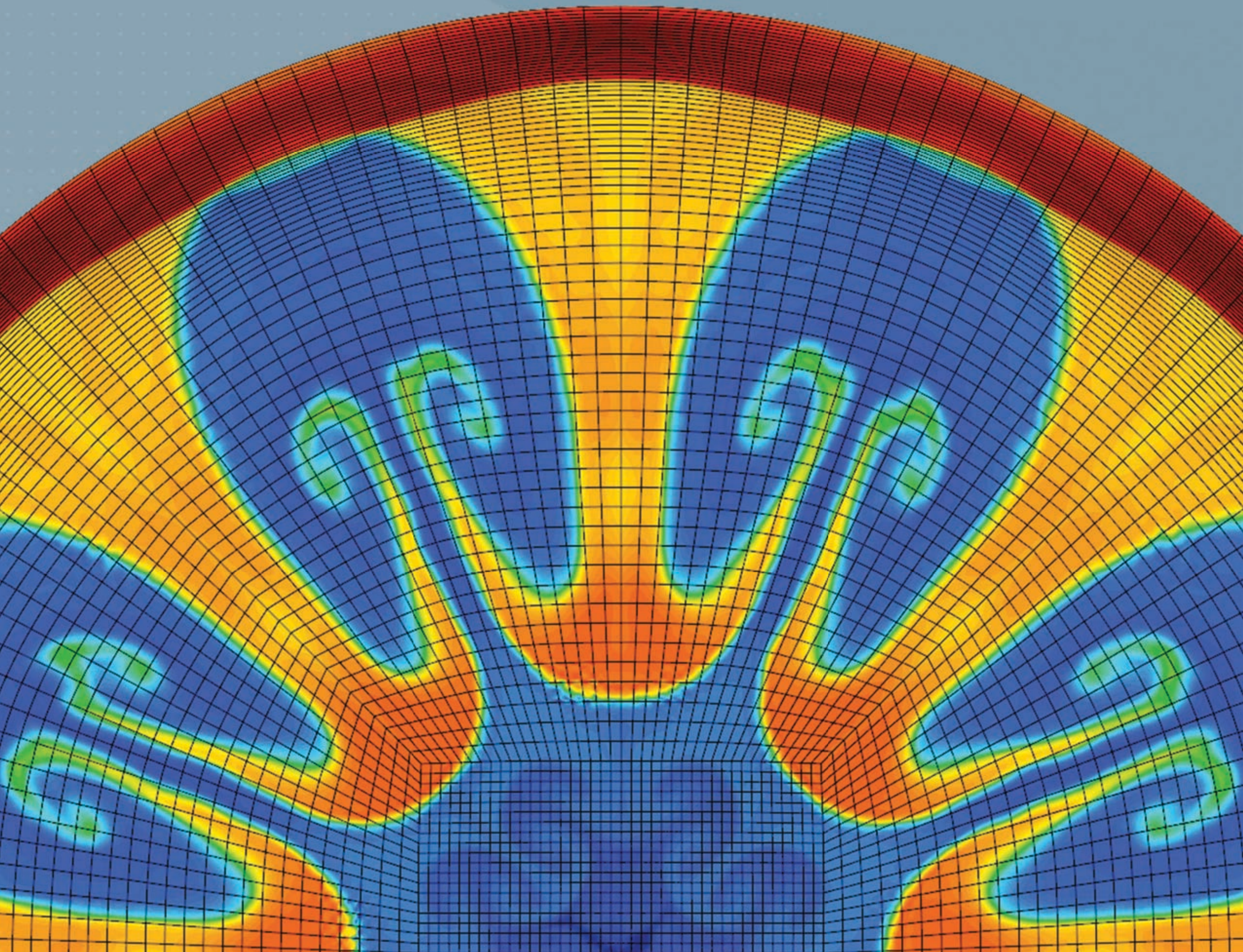
Proposed Work for FY14

In FY14 we propose to (1) determine how properties of the newly developed catalyst impact performance for converting coal and bio-feedstock to refined products, (2) demonstrate improved catalytic performance with the novel catalyst, and (3) demonstrate dynamic control using an applied electrical potential. This added control is an essential tool for improving our understanding of complex reactions with multiple possible reaction paths.

Publications and Presentations

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

Mathematics and Computing Sciences



Laboratory Directed Research and Development

Annual Report FY2013

Compressive Sensing for Wide-Area Surveillance

Paul Kidwell (11-ERD-022)

Abstract

This project will provide a practical solution to the problem of "data explosion" faced by the defense and intelligence communities as larger and larger sensor arrays are developed and readied for deployment. We will leverage capabilities in compressive video sensing—including distributed compressive sensing—to design better data-compression pipelines for color and multispectral video data sets from wide-area surveillance. Our main objectives are to determine how much sparsity is available for color and multispectral video data sets and identify a compressive sensing-based approach to exploit this sparsity. This work will pioneer a next-generation architecture for more cost-effective onboard data-compression and processing methods for wide-area surveillance platforms, helping to manage the huge volumes of data collected by aerial platforms and ultimately to improve quality of the refined intelligence produced.

If successful, this project will help achieve the data-compression factors in multispectral data sets that are necessary to transmit surveillance data with current bandwidth-limited technologies and to reduce the onboard processing costs of wide-area surveillance. Using compressive sensing, we expect to achieve significant compression factors for the large, multidimensional data sets that are collected in computationally scarce environments in which conventional wavelet-based transforms cannot be efficiently computed using onboard processing resources.

Mission Relevance

The innovations achieved in this project will further the Laboratory's mission in cyber security, space security, and intelligence by developing advanced data-management capabilities that will yield higher-quality intelligence in wide-area aerial surveillance.

By carefully designing a projection matrix, both the separation capability of the compressed target classes and compression can be increased. In this example, the objective is to distinguish three vehicle classes: car, truck, and van (shown at top). The middle row represents the projection matrix, while the bottom row is a two-dimensional projection of the compressed signal. The two-dimensional projection (maximizing mutual information) in the right column shows much less overlap in classes than the left column (random sampling).

FY13 Accomplishments and Results

We have achieved all of our goals for FY13. Specifically, we (1) developed a hierarchical algorithm for background modeling and change detection that employs a sequence of hypothesis tests at progressively finer scales operating on a compressed signal; (2) performed an empirical analysis of wide-area motion imagery (<10Hz) captured by a 100-megapixel camera to characterize noise properties, sampling size, and patch size requirements—results indicated comparable detection, localization, and reconstruction to traditional sensors with a data reduction of approximately 85%; (3) coupled the hierarchical change-detection algorithm as a track initiation module for the FY12 compressive particle filter, enabling tracking to be performed entirely in the compressed domain and thereby obviating reconstruction; and (4) designed a linear projection measurement scheme for a vector Poisson signal model, leveraging latent-class information while maximizing the mutual information between observed and projected data.

Project Summary

The successful conclusion of this project has resulted in the development of a sparse modeling infrastructure for wide-area motion imagery. Our main objectives were to determine the degree of sparsity available in video data sets and to exploit this sparsity via compressive sensing. New technologies were developed in four distinct areas and performance was characterized with respect to wide-area motion imagery for each. These technologies include: (1) two strategies for motion prediction—progressive encoding and an iterative method to jointly reconstruct data and refine motion parameters; (2) classifiers coupled with a projection matrix designed to minimize data collection while maximizing separation between classes; (3) background models for compressed imagery providing comparable detection, localization, and reconstruction to traditional methods while reducing data requirements by approximately 85%; and (4) an automated tracking algorithm capable of initiating and tracking directly on a compressed signal. Our project has demonstrated the level of compression achievable on wide-area motion imagery via a compressive sensing paradigm, while enabling algorithms to successfully exploit the signal in its compressed form. We have identified opportunities for elements of this work to be matured in support of Air Force wide-area motion imagery capabilities. In addition, a dictionary-based technique that recovers missing image information by interpolating data from the neighborhood may be incorporated into an image-processing pipeline for Army full-motion video analysis.

Publications and Presentations

Kidwell, P., et al., 2013. *Compressive sensing and dictionary learning: Methods and applications for imagery collected by airborne sensors.* LLNL-TR-645237-DRAFT.

Data Abstractions for Portable High-Performance Computing

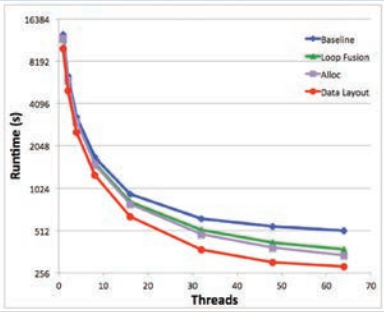
Ian Karlin (11-ERD-028)

Abstract

Almost all large scientific codes at LLNL will need to be modified to effectively utilize the new generation of high-performance computing systems. The most important issue in performance is the effective use of memory. Code developers must determine the optimal data layout for all major data structures in the code. Current compilers force code developers to express data layouts by writing overly specialized code. As a result, codes need major rewriting when a new generation of computing system is introduced. Our goal is to demonstrate how writing codes with higher levels of abstractions will enable codes to be transferred to newer supercomputers without requiring major rewriting, while simultaneously maintaining performance.

This research will result in C++ code library of data abstractions relevant to Livermore Advanced Simulation and Computing codes that, when implemented with the ROSE compiler, will automatically rewrite the code for selected high-performance computing systems. The primary metric for success will be performance of the ROSE-

generated specialized codes on diverse computer platforms. We will demonstrate the ease of writing and maintaining the data abstraction version of a code, the breadth of machines on which that code can achieve high performance, and the level of performance achieved on each of those machines. We will also assess how semi-automated processes using the ROSE system could expedite transforming an existing code to one that effectively uses the data-abstraction approach.



Improvement in compute execution time resulting from changes to data layout. In this example, the LULESH (Livermore unstructured Lagrangian explicit shock hydrodynamics) code is running on the IBM BlueGene/Q system. As parallelism is increased (threads), overall execution time decreases. The data layout option incorporates two optimizations we used (shown in green and purple).

Mission Relevance

By enabling high-performance computing codes to be quickly and efficiently moved to new computing systems as they come online, this project supports Laboratory missions that depend on those codes, particularly stockpile stewardship. To maximize mission relevance and impact, we will restrict the notations and transformations we explore to the actual control and data structures of importance to existing Advanced Simulation and Computing codes. In addition, we will focus on current and explicitly proposed exascale machine architectures.

FY13 Accomplishments and Results

In FY13 we established the foundation for a new approach to supporting portable performance of scientific codes across high-performance computing platforms. Specifically, we (1) developed an upgraded TALC (topology aware layouts in C) multiple-architecture compiler source-to-source transformation tool, which has been delivered to the ROSE compiler repository, and permits application developers to maintain one "neutral" data-layout source code and explore architecture-specific array layouts—the new automated portion of TALC can analyze the original source code based on platform characteristics and produce source code with an array data layout customized to that system; (2) performed experiments with the IRSmk (implicit radiation anisotropic diffusion microkernel) code, SRAD (speckle-reducing anisotropic diffusion) code, and the LULESH (Livermore unstructured Lagrangian shock hydrodynamics) code on four computer platforms (IBM POWER7, AMD Accelerated Processing Unit, Intel Sandybridge, and IBM BlueGene/Q) and benchmarked the performance of these codes using optimized manual data layouts and; (3) determined that our automated algorithm resulted in performance of 95 to 99% of the best manual layout for IRSmk, was within 78% for SRAD, and for LULESH, the automated approach was better than any manual layout for 8 processing threads on the Power7 platform and within 10% of the best layout for almost all other processors.

Project Summary

The successful conclusion of this project resulted in an improved version of the software framework for the TALC multiple-architecture compiler delivered to the ROSE compiler. Using TALC, we were able to explore up to 32 data layouts for a single application on 4 hardware platforms. In addition, an initial algorithm for automatically exploring data layouts was developed that in some cases was better than the best layout picked by hand. We examined the LULESH proxy application and showed that it is important to apply other optimizations to the code first to profit from data layout changes.

Publications and Presentations

Karlin, I., 2013. *LULESH programming model and performance ports overview.* LLNL-TR-608824.

Karlin, I., et al., 2012. *Memory and parallelism tuning exploration using the LULESH proxy application.* LLNL-POST-567121.

Karlin, I., et al., 2012. *Tuning the LULESH mini-app for current and future hardware.* Nuclear Explosive Code Development Conf., Livermore, CA, Oct. 22–26, 2012. LLNL-ABS-564179.

Sharma, K., et al., 2013. *Automatic data layout selection using architectural features.* SC13 Intl. Conf. High Performance Computing, Networking, Storage and Analysis, Denver, CO, Nov. 17–22, 2013. LLNL-CONF-635555-DRAFT.

Sharma, K., et al., 2011. *Data layout strategies for array abstraction.* LLNL-POST-492010.

Sharma, K. et al., 2013. *User-specified and automatic data layout selection for portable performance.* LLNL-TR-637873.

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, 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 the 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 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 it will be accomplished while both minimizing the effects of uncertainty introduced by sub-sampling as well as maintaining mathematical guarantees of estimation consistency.

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 establish the Laboratory's reputation as a leader in analysis of cyber security and intelligence data.

FY13 Accomplishments and Results

In FY13 we (1) extended our data analysis system (particle filters) to increase its data ingestion rates; (2) designed and implemented a Storm topology for distributed processing of streaming data, and re-designed particle filters to work within it; (3) laid the groundwork to process on-the-fly data in parallel in arbitrarily large Storm clusters, addressing problems previously unapproachable with such a nuanced and powerful approach; (4) made numerous novel extensions to the Storm framework itself, facilitating hitherto impossible functionality within the toolkit, potentially beneficial to the entire Storm community; (5) developed and implemented a novel streaming adaptive-sampling technique, which consists of a model that approximates particle filters, but is substantially less computer intensive; (6) implemented tunable parameters that control the tradeoff between model complexity and computational cost; and (7) established a new collaboration with a particle physics project at LLNL—initial results are promising, with nearly perfect prediction accuracy by particle filters.

Proposed Work for FY14

In FY14 we will (1) improve the data processing rates of particle filters by implementing a distributed version of Storm particle filters, augmenting our streaming adaptive-sampling approach to automatically adjust data-processing rates on the fly to optimally match data arrival rates, and integrating streaming adaptive sampling with Storm—this will significantly increase particle filters' speed because the two approaches are complementary; (2) package our tool into filters, performing rigorous benchmarks of different aspects of the model; and (3) by the end of the project, deliver stand-alone optimized and extremely high-throughput particle filters, with capabilities to turn on and off adaptive sampling and Storm, along with thorough documentation.

Publications and Presentations

Challis, C. J., et al., 2011. *Particle learning for probabilistic deterministic !nite automata with application to DNS query classification.* LLNL-TR-499031.

Sales, A., et al., 2012. *"Semisupervised classification of texts using particle learning for probabilistic automata."* Bayesian Inference and Markov Chain Monte Carlo: In Honour of Adrian Smith. Clarendon Press, Oxford, United Kingdom. LLNL-JRNL-513511.

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 the National Ignition Facility.

FY13 Accomplishments and Results

In FY13 we (1) conducted particle-in-cell simulations of high-Mach-number shocks traversing multiple-species fuels such as deuterium and tritium or helium-3, and found significant species separation; (2) performed an assessment of possible anomalous entropy generation, whose results have been applied to gauging the ignition performance margins in low-adiabatic ignition targets for the National Ignition Facility, and were found to show a potential for significant erosion of one-dimensional performance; (3) found another new physical effect in shocks traversing a plasma interface, leading to a potential new source of atomic mix that is strictly kinetic in origin (in contrast to hydrodynamically based); and (4) applied this effect to understanding the behavior of exploding pusher targets on the OMEGA laser facility at the University of Rochester in collaboration with Massachusetts Institute of Technology researchers.

Project Summary

We have developed analytical methods and performed simulations for assessing kinetic and species separation effects in inertial-confinement fusion implosions. We applied the adiabatic lapse rate to a plasma (temperature variation with height) and showed it lead to a novel source of temperature gradient that depends on the concentration gradient and difference in ion charge states in a binary mixture. Application of the model shows significant plasma modifications to the temperature gradient compared with gas dynamics upon which single-fluid radiation-hydrodynamics simulations are based. We intend to collaborate with Livermore weapons researchers in soliciting programmatic funding to assess and develop the kinetic plasma mix phenomenon discovered in FY13.

Publications and Presentations

Amend, P., C. Bellei, and S. Wilks, 2011. *Plasma adiabatic lapse rate.* Anomalous Absorption Conf., San Diego, CA, June 14–19, 2011. LLNL-PRES-489072.

Amend, P., C. Bellei, and S. Wilks, 2012. “Plasma adiabatic lapse rates.” *Phys. Rev. Lett.* **109**(7), 075002. LLNL-JRNL-497396.

Bellei, C., et al., 2013. “Response to comment on ‘Species separation in inertial confinement fusion fuels’.” *Phys. Plasma.* **20**, 044701. LLNL-JRNL-625632.

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 LLNL's mission in energy security.

FY13 Accomplishments and Results

In FY13 we (1) extended the grid model to include mid- and large-scale systems; (2) completed implementation and testing of an alternating-current power-flow optimization algorithm; (3) worked with collaborators from the University of California, Berkeley to demonstrate their stochastic optimization code on larger high-performance computing systems; and (4) worked with IBM to improve performance of their mixed-integer optimization code for solving power-system production scheduling problems to increase computing speed by fourfold. We also set up and exercised a high-performance computing infrastructure for parallel runs of production simulation problems which used a total of 3 million core-hours of time to simulate 3,000 days of operation.

Proposed Work for FY14

In FY14 we will (1) use uncertainty quantification techniques to improve renewable-generation ensemble forecasts, (2) perform uncertainty analysis to estimate the value provided by stochastic grid-optimization techniques that use an ensemble of renewable-generation patterns compared to the deterministic methods currently in use, (3) identify effective system structures that can accommodate high levels of renewable generation by coupling network equilibrium models of alternative structures with stochastic production models, and (4) conduct numerical experiments of grid stability.

Publications and Presentations

Edmunds, T., et al., 2014. *Integrated stochastic weather and production simulation modeling.* IEEE PES 5th Innovative Smart Grid Technologies Conf., Washington, DC, Feb. 19–22, 2014. LLNL-PROC-642353.

Epperly, T. G. W., et al., 2012. *High-performance computing for electric grid planning and operations.* 2012 IEEE Power and Energy Society General Mtg., San Diego, CA, July 22–26, 2012. LLNL-PROC-528131.

Lamont, A., 2013. "Assessing the economic value and optimal structure of large-scale energy storage." *IEEE Trans. Power Syst.* **28**(2), 911. LLNL-TR-599835.

Papavasiliou, A., S. Oren, and B. Rountree, 2013 *Applying high performance computing to multi-area stochastic unit commitment for renewable energy integration.* LLNL-JRNL-645297.

Santiago, C., 2013. *An efficient heuristic for the optimal power flow problem.* LLNL-JRNL-645296-DRAFT.

Secure Virtual Network Enclaves

Domingo Colon (12-ERD-016)

Abstract

The pervasive threat posed by overseas adversaries, industrial espionage, insider threats, denial-of-service attacks, and aggressively spreading malware designed specifically to damage or disrupt a system has substantially increased the risk of conducting business operations on traditional network infrastructures. Military services operating under hostile engagement scenarios face even greater threats to the security of their network-based information systems. We propose to research and develop the capability to dynamically create secure network enclaves consisting of virtual sub-nets capable of operating securely in a potentially compromised network infrastructure. We will leverage a novel combination of internally developed network-generation components, cloud computing, and virtual-capability software packages. Embedded sensor technology will be used to monitor operations in the enclave environment and to apply custom experience-based problem solving (heuristics) to determine the state of the enclave's security. If successful, we would have the first true capability to establish secure virtual enclaves in a way that is practical for military and other operations in the field.

We will deliver a system capable of accepting ad hoc descriptions of secure network enclave configurations and, based on those formal descriptions, dynamically generate a fully virtual representation capable of meeting both the functional and operational security requirements of the master configuration. The descriptions will be encoded in a declarative domain-specific language and will define both the required network resources and mission-specific security policies.

Mission Relevance

This project supports the Laboratory's mission in cyber security by providing the capability to dynamically create secure network enclaves and thereby enable the management of production-level network applications in potentially compromised network infrastructures for defense applications.

FY13 Accomplishments and Results

In FY13 we (1) defined the central goal of a secure virtual enclave to be maximizing the number of segmented zones of the virtual network enclave in a manner that supports the principal of least-privileged access (that is, ability to access only the information and resources necessary for a legitimate purpose); (2) developed an understanding of the data that can be sampled out-of-band from a virtual platform and defined a framework for measurement possibilities—this process represents the foundational building block upon which complex event-processing monitors can be established to support enforcement of required mission invariants (a condition that is often true during execution of a program); and (3) created a configuration-synthesis algorithm to enhance the resiliency of a virtual enclave environment by reenforcing operational reliability and by returning the enclave to a secure state when possible.

Project Summary

We developed the capability to automatically build a virtual enclave environment from a security-focused intermediate representation and explored and identified characteristics that are unique to purely virtual environments, which together form the basis for novel security measurement approaches. We developed a methodology and supporting framework for dynamically provisioning virtual network enclave environments that are capable of adhering to mission-specified security profiles. This capability will allow mission planners to create computing environments on demand with security profiles appropriate for the defined mission. It will also provide mission planners with the capability to formally express a custom definition for each mission, including a methodology for explicitly enumerating the appropriate network and host-based security controls. In addition, we determined that implementing a dynamic trust model, automating a course-of-action matrix, adding machine learning for determining invariants, and developing more-sophisticated moving-target defense heuristics will add to the uniqueness and security of virtual enclave environments.

Network Simulation and Its Applications

Peter Barnes (12-ERD-024)

Abstract

Our proposed 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 the national leader in cyber modeling and situational awareness. Completion of this effort will position the Laboratory to invent new 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.

FY13 Accomplishments and Results

In FY13 we (1) scaled up our laboratory model to complete activity, with all nodes participating in simulated communication; (2) developed a new parallel scheduler for ns-3 (network simulator 3) for Internet systems that should produce significant performance improvements on large simulation models; and (3) demonstrated operation of federated ns-3 simulations by exchanging data through our private-sector partner's internetwork operating system emulation.

Proposed Work for FY14

In FY14 we will (1) perform both large runs and code development, needing approximately 20,000 central processing unit hours per week in parallel; (2) simulate the Laboratory's network model with sufficient detail to enable the LLNL mapper to use the data; (3) complete development of the realistic application traffic mode; and (4) complete parameter studies of the mission execution model.

Publications and Presentations

Barnes, P. D., Jr., et al., 2012. *A benchmark model for parallel ns3*. LLNL-PRES-535536.

Barnes, P. D., Jr., et al., 2013. I*ntegrating ns-3 model construction, description, preprocessing, execution, and visualization*. WNS3 2013, in conjunction with SIMUTools

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Barnes, P. D., Jr., et al., 2012. *Livermore computer network simulation program*. LLNL-POST-538331.

Nikolaev, S., et al., 2013. *Performance of distributed ns-3 network simulator*. SIMUTools 2013 6th Intl. ICST Conf. Simulation Tools and Techniques, Cannes, France, Mar. 5–7, 2013. LLNL-CONF-607992.

An Open Framework to Explore Node-Level Programming Models for Exascale Architectures

Chunhua Liao (12-ERD-026)

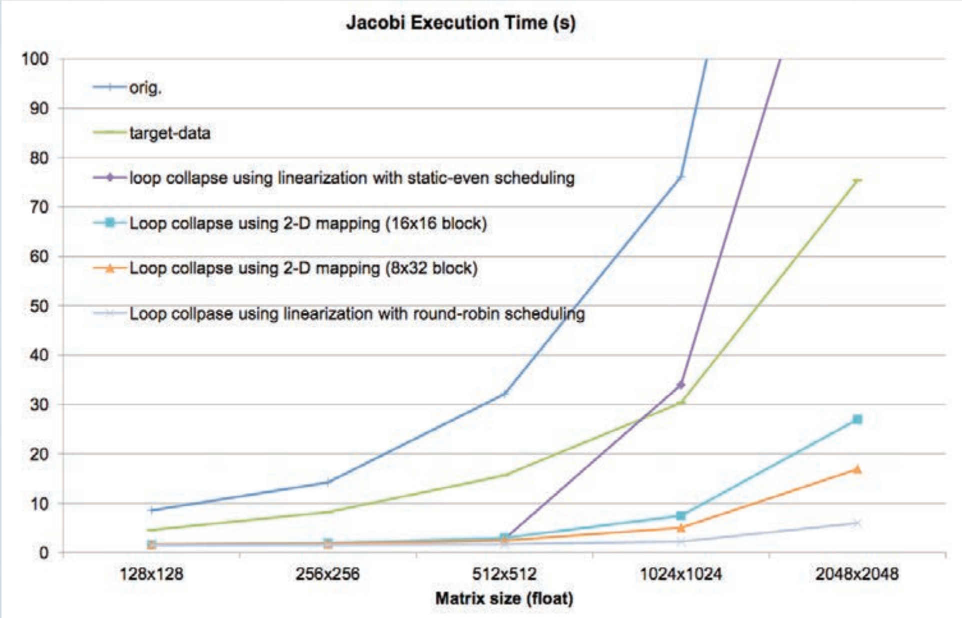
Abstract

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 revolutionize 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



The Heterogeneous OpenMP programming model developed with the building blocks of this project has delivered competitive performance for the important stencil computation kernel, Jacobi. The improvements in execution time are shown by incrementally using more advanced compiler and run-time building blocks, whereas the original version uses a minimum set of building blocks. The target-data version uses data-reuse, whereas the others use loop collapse, two-dimensional mapping, and different scheduling policies.

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 and simulation.

FY13 Accomplishments and Results

We investigated advanced analysis and optimization building blocks to improve the performance and power efficiency of programming models built on our framework. Specifically, we (1) designed and implemented power management building blocks for important computation kernels, (2) optimized data locality to make better use of hierarchical memory in graphics processing units, (3) optimized Laboratory applications for specific domains using stencil operations on structured arrays, and (4) released the first-ever nontrivial programming model, named Heterogeneous OpenMP. Our new model can automatically translate code generated with OpenMP (an interface for multiple-platform, shared-memory multiprocessing programming) with additional accelerator directives into CUDA (compute unified device architecture) codes. With other major features including CUDA kernel generation and data offloading, reduction, and reusing, Heterogeneous OpenMP is a tool for demonstrating how building blocks can be used to address exascale challenges such as heterogeneity and energy efficiency.

Proposed Work for FY14

In FY14 we will continue to explore novel and advanced building blocks to help address exascale challenges by creating one or more example programming models to demonstrate use of our framework to explore the design tradeoff of programming models. Specifically, we will (1) research language, compiler, and run-time support for the programming of multiple graphics processing units in a single computation node; (2) explore building blocks to optimize irregular DOE applications with dynamic or indirect memory access patterns; and (3) compare multiple programming models based on our framework to generate insights about the tradeoffs of designing, implementing, and using exascale node-level programming models.

Publications and Presentations

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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.

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.

High-Order Curvilinear Arbitrary Lagrangian–Eulerian Hydrodynamics

Tzanio Kolev (12-ERD-030)

Abstract

The framework of arbitrary Lagrangian–Eulerian (ALE) computer 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, 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 develop new methods for optimizing curvilinear mesh geometry representations, conservative monotonic high-order field remapping, and handling multiple-material curvilinear zones. This work will leverage previous research at LLNL on high-order curvilinear finite elements for the Lagrange phase.

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

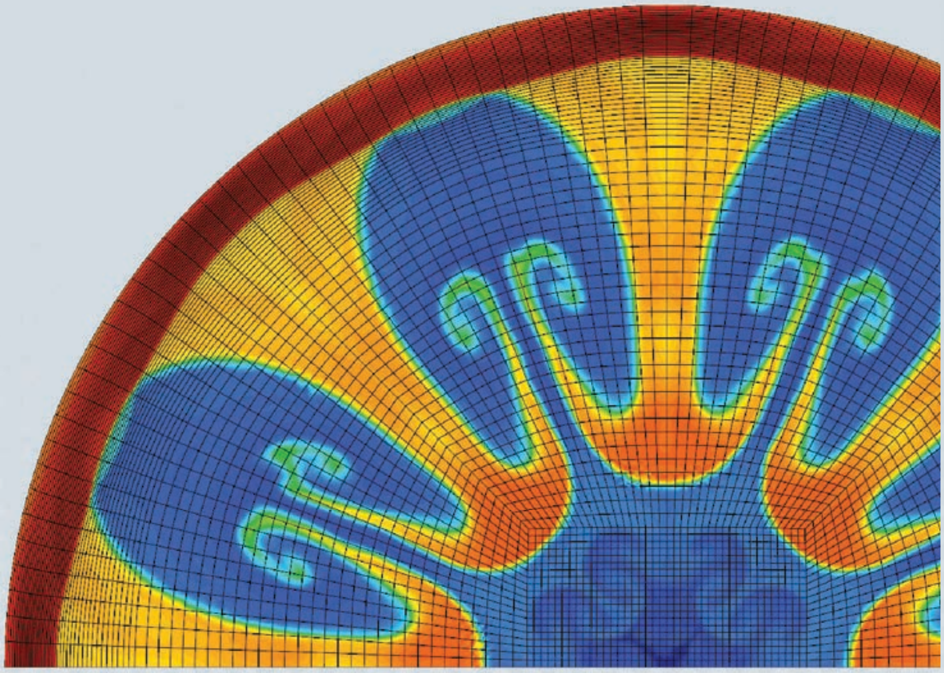
mesh-motion parameters and for manual intervention by users, minimize diffusive errors by running longer in a Lagrange mode, improve accuracy by diminishing mesh imprinting and improving symmetry preservation, and more effectively utilize future multiple-core and graphics processing unit architectures 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 critical importance for stockpile stewardship and inertial-confinement fusion in support of the Laboratory's missions in national and energy security.

FY13 Accomplishments and Results

In FY13 we (1) continued the investigation of our discontinuous Galerkin advection-based, high-order remapping algorithm, which has computationally desirable features such as excellent parallel scalability, geometric flexibility, and conservation properties—we analyzed its conservation properties and proposed and evaluated several new approaches for ensuring high-order monotonicity; (2) demonstrated the high-order accuracy and practical performance of the new methods on model problems and single-material ALE simulations, showing that the order of the error in our high-order remapping matched the original high-order Lagrangian scheme; (3) performed simulations with a simple ALE capability in which material interfaces are kept Lagrangian; (4) initiated research in multiple-material zone treatment based on high-order material indicator functions and material-specific density and energy



Our newly developed high-order arbitrary Lagrangian–Eulerian algorithms enable efficient, accurate, and robust simulations of a variety of challenging shock hydrodynamics problems, including inertial-confinement fusion applications such as a parallel fourth-order computation of a multiple-material configuration with a perturbed material interface. The new high-order discontinuous Galerkin advection algorithm with locally scaled diffusion ensures the conservative and accurate remapping of functions with steep gradients on highly distorted or unstructured curved meshes with sub-zonal feature resolution.

fields; and (5) developed a parallel implementation of the new ALE methods by incorporating them in the high-order research shock hydrodynamics code BLAST, where we extended the high-order ALE algorithms to problems with axial symmetry or elastic–plastic flow and demonstrated favorable comparison with purely Lagrangian simulations.

Proposed Work for FY14

In FY14 we will widen the applicability and improve the efficiency of our high-order finite-element algorithms for ALE hydrodynamics simulations. Specifically, we will (1) explore novel high-order numerical algorithms for multiple-material curvilinear zone treatment and couple them with the high-order Lagrangian and remapping phases, (2) demonstrate and evaluate the performance of the full high-order ALE method on a set of multiple-material ALE hydrodynamics test problems in the BLAST code, and (3) investigate performance-enhancing techniques and document trade-offs between speed and robustness.

Publications and Presentations

Anderson, R., et al., 2013. *High-order curvilinear ALE hydrodynamics.* 9th Intl. Conf. Large-Scale Scientific Computations, Sozopol, Bulgaria, June 3–7, 2013. LLNL-PRES-637772-DRAFT.

Anderson, R. W., et al., 2013. *High-order curvilinear ALE hydrodynamics.* SIAM Conf. Computational Science and Engineering, Boston, MA, Feb. 25–Mar.1, 2013.LLNL-PRES-621512.

Anderson, R., et al., 2013. *High-order discontinuous Galerkin remap methods for curvilinear ALE hydrodynamics.* MultiMat 2013 Intl. Conf. Numerical Methods for Multi-Material Fluid Flow, San Francisco, CA, Sept. 2–6, 2013. LLNL-PRES-642492.

Anderson, R. W., et al., 2012. I*terative relaxation of high-order curvilinear meshes.* 12th Copper Mountain Conf. Iterative Methods, Copper Mountain, CO, Mar. 25–30, 2012. LLNL-PRES-541291.

Anderson, R., et al., 2013. *Recent developments in the high order finite element hydrodynamics code, BLAST.* MultiMat 2013 Intl. Conf. Numerical Methods for Multi-Material Fluid Flow, San Francisco, CA, Sept. 2–6, 2013. LLNL-POST-643018.

Dobrev, V. A., T. V. Kolev, and R. N. Rieben, 2012. "High order curvilinear finite elements for Lagrangian hydrodynamics." *SIAM J. Sci. Comput.* **34**, B606B641. LLNL-JRNL-516394.

Kolev, T. V., et al., 2012. *High-order curvilinear ALE hydrodynamics.* ECCOMAS 2012 6th European Congress Computational Methods in Applied Sciences and Engineering, Vienna, Austria, Sept. 10–14, 2012. LLNL-PRES-579453.

Kolev, T., et al., 2013. *Scalable multi-physics simulations will require new discretization and numerical methods research.* DOE Workshop Applied Mathematics Research for Exascale Computing, Washington, DC, Aug. 21–22, 2013. LLNL-PRES-642533.

Efficient and Accurate Meta-Genomics 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 thus 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 bolstering the Laboratory's core competency in high-performance computing by using low-cost, multicore compute nodes augmented by persistent flash computer memory.

FY13 Accomplishments and Results

In FY13 we (1) demonstrated accurate metagenomic classification in the presence of novel organisms; (2) demonstrated a major improvement in run time performance using a novel database indexing scheme with a persistent memory device, and released the software as open source on the SourceForge source code repository; and

(3) participated in the Defense Threat Reduction Agency algorithm challenge, which focused on using DNA sequence data in identifying pathogens in complex mixtures from clinical samples, and obtained an accuracy score on all challenge data sets with the software running an order of magnitude faster than required by the challenge—we obtained an accuracy rank of 12 among 103 submissions from around the world.

Proposed Work for FY14

In FY14 we will (1) continue with the design of new methods for characterizing novel sample contents by developing new indexing strategies that identify higher rank (genus or family) classifications when species or strain representation is lacking, and accommodate larger numbers of eukaryotic background genomic sequences for an organism to support screening of environmental samples; (2) investigate continued improvements to the accuracy benchmarks by developing new methods that use sample content summary data to refine individual read labels to support more confident taxonomic classification; and (3) characterize and improve algorithm performance using persistent memory architectures.

Publications and Presentations

Allen, J., et al., 2012. *Efficient and accurate metagenomics search using a desktop computer and a large scalable persistent memory device*. LLNL-POST-559075.

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.

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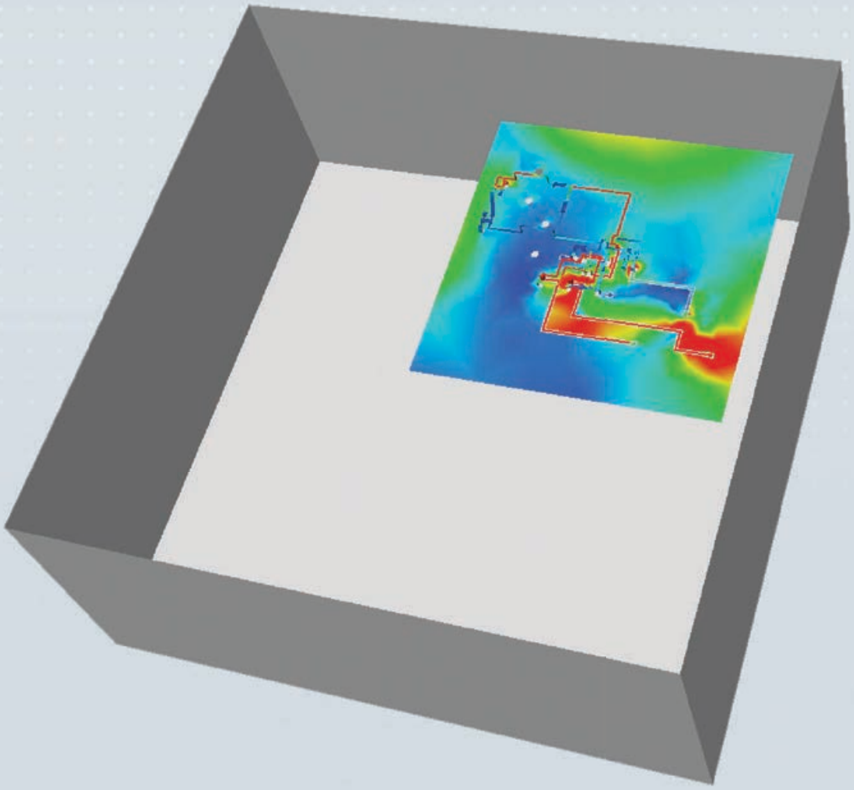
Automatic Complexity Reduction with Application to Electromagnetic Effects Simulation

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 Maxwell's 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.



Oscillator circuit in a metal enclosure. The pseudocolor denotes the amplitude of the induced electric current, which is a measure of electromagnetic interference. The current is computed using a frequency domain boundary-element method. Our automatic complexity-reduction algorithm is used to compute the electromagnetic interference for all circuit locations within the enclosure.

Using a combination of reduced-order models, radial basis functions for interpolating matrix triple products, and parameter adaptivity, we will 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 LLNL strategic missions, including national and international security and energy security.

FY13 Accomplishments and Results

In FY13 we (1) investigated the source of ill-conditioning of interpolatory linear systems—the problem is that as new radial basis functions are added to parameters, they overlap too much; (2) developed a novel algorithm based on optimizing the width of the radial basis function to address this problem; (3) explored empirical interpolation methods (also known as a magic points algorithm), which worked quite well but required an undesirable modification to the partial differential equation solver (we will continue to investigate empirical interpolation for those cases in which radial basis functions are ill-conditioned); and (4) investigated applying our complexity-reduction algorithm to a structural mechanics problem, which ties in nicely with the Laboratory's recent thrust in additive manufacturing.

Proposed Work for FY14

In FY14 we will (1) focus on error estimators, which are a critical component of automatic complexity reduction, and on a large-scale application problem to verify our algorithm's capability; (2) work with a postdoctoral researcher with a background in error estimation to develop possible new approaches for error estimation and achieve a thousandfold reduction in complexity compared to a brute-force approach; and (3) engage various programs and organizations to promote collaborations.

Publications and Presentations

Lange, K. J., and D. A. White, 2013. *A comparison of radial basis functions for MIROM of a boundary element electromagnetics simulation.* SIAM Conf. Computational Science and Engineering, Boston, MA, Feb. 25–Mar. 1, 2013. LLNL-POST-558893.

Lange, K. J., and D. A. White, 2013. *Using proper orthogonal decomposition as a tool for homogenization of PEM fuel cell catalyst layer simulations.* ASME 2012 Intl. Mechanical Engineering Congress and Exposition, Houston, TX, Nov. 9–15, 2012. LLNL-ABS-532532.

Lange, K. J., M. L. Stowell, and D. A. White, 2013. *Microstructure design using matrix interpolation reduced order modeling.* ASME 2012 Intl. Mechanical Engineering Congress and Exposition, Houston, TX, Nov. 9–15, 2012. LLNL-ABS-532671.

White, D. A., et al., 2013. *Application of automatic model order reduction to electromagnetic interactions.* SIAM Conf. Computational Science and Engineering, Boston, MA, Feb. 25–Mar. 1, 2013. LLNL-ABS-579936.

White, D. A., et al., 2012. *Application of model order reduction to multi-parameter electromagnetic compatibility modelling.* LLNL-JRNL-591792.

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 for 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 MGmol (a first-principles molecular dynamics computer code based on density-functional theory) 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

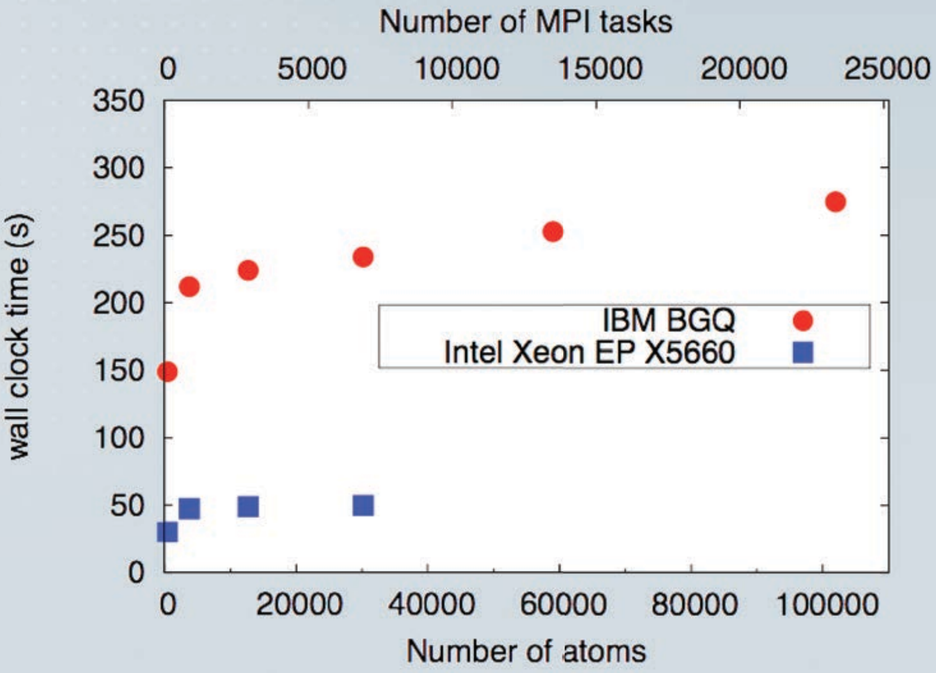
An O(N) complexity algorithm will enable quantum molecular dynamics calculations at an unprecedented scale and accuracy on DOE's next-generation supercomputers, providing insight at the molecular level in various fields such as materials in the extreme environment of fusion energy and the toxicity of chemical agents, in support of the Laboratory's missions in national security, energy security, and basic science.

FY13 Accomplishments and Results

We (1) finished coupling the new algorithm for computing selected elements of the inverse of relevant matrices to the MGmol code, addressing various remaining bottlenecks in parallel scaling and demonstrating excellent weak scaling up to 100,000 atoms, with only a small increase in the real time that elapses from start to end of the computational task, compared to 500 atoms; (2) made major progress in speeding up the code to run less than a minute for a first-principles (density functional theory) molecular dynamics time step on Peloton computer clusters at Livermore (about four minutes on the Vulcan supercomputer platform); and (3) adapted an open-source code in the MatLab language RSDFT (real-space density-functional theory) to replicate the algorithms in our optimized C++ code and to enable the quick, easy testing and development of new solvers.

Proposed Work for FY14

We will (1) continue our weak-scaling studies on the Vulcan supercomputer of density-functional theory applications beyond 50,000 atoms; (2) address further bottlenecks, such as remaining data replication on each message-passing interface task, with the goal of removing all global communications and data replication;



Parallel weak scaling showing time for a single molecular dynamics simulation step on an IBM BlueGene/Q supercomputer architecture and Intel Xeon EP X5660 Linux cluster with a high-speed interconnect as a function of number of atoms and number of message-passing interface (MPI) tasks.

(3) continue new algorithm development to speed up solvers for the MGmol code, including looking into using OpenMP (an interface for multiplatform, shared-memory multiprocessing programming) to speed up code on BlueGene/Q-class supercomputers; (4) work with application scientists to apply our methodology to relevant condensed-matter problems at LLNL; and (5) investigate how our recently developed algorithm could be generalized to treat problems without a band gap—that is, metal-related problems.

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.

Fattebert, J. L., and D. Osei-Kuffuor, 2013. *Short-range* O(N*) algorithm for scalable molecular dynamics simulations from first-principles.* 12th US National Congress Computational Mechanics, Rayleigh, NC, July 22-25, 2013. LLNL-ABS-623418.

Osei-Kuffuor, D., S. Hamel, and J.-L. Fattebert, 2013. O(N*) parallel algorithm for computing selected elements of the inverse of a Gram matrix in electronic structure calculations.* SIAM Conf. Computational Science and Engineering, Boston, MA, Feb. 25–Mar. 1, 2013. LLNL-ABS-580273.

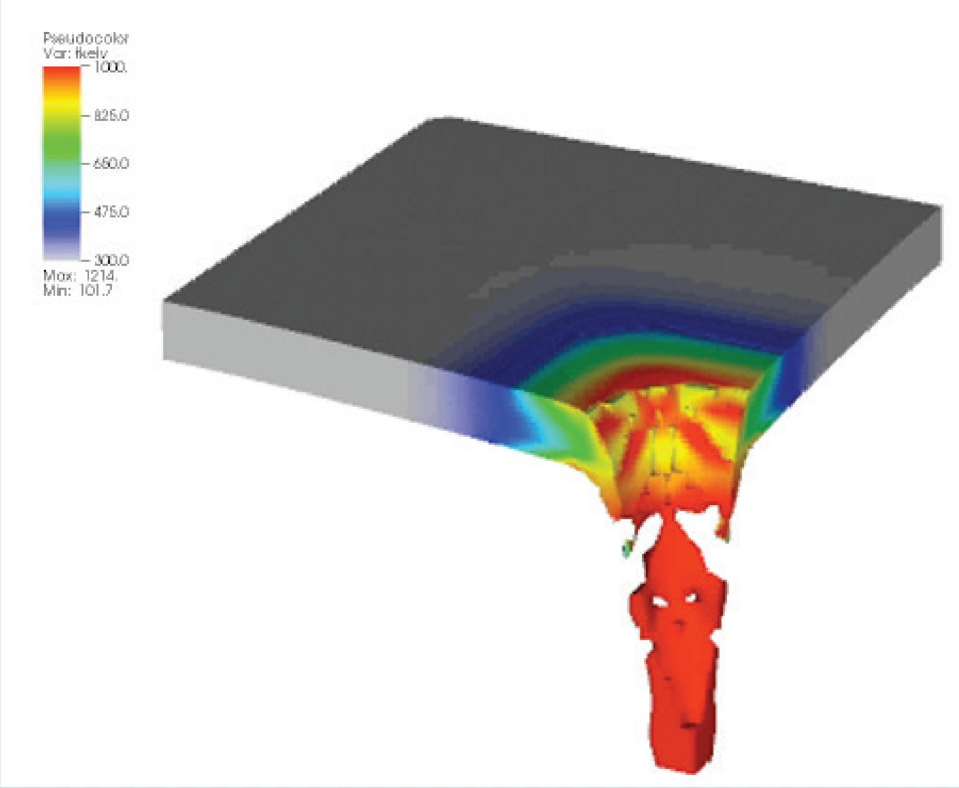
Laser Lethality Experimentation, Modeling, and Simulation Capability

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 select 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



Computer simulation of laser rupture and penetration of a metal plate.

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. 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.

FY13 Accomplishments and Results

We (1) completed measurement of radiation absorption by direct calorimetry on engineering-grade aluminum, steel, and titanium, characterizing effects of polarization and surface modifications under irradiation; (2) developed methods to calibrate a thermal-imaging camera to permit standoff temperature measurement on samples during laser heating; (3) measured full-field strain displacement of aluminum under combined mechanical and thermal load; (4) modeled these same scenarios with ALE3D to evaluate Johnson–Cook and Fisher failure strength and failure models for application-relevant heating and strain rates; (5) verified the ALE3D heat-deposition package with erosion and explicit time integration, addressing erosion criteria and solver issues for implicit time integration; (6) completed installation and testing of equipment, and began laser irradiation experiments on aluminum plates to characterize material rupture and penetration phenomenology as a function of laser power; (7) designed and fabricated test articles for pressurized vessel experiments; and (8) completed initial simulations of material rupture and laser penetration and began to model laser–target interactions.

Proposed Work for FY14

In FY14 we will (1) characterize the phenomenology of laser interaction with stressed plates and pressurized cylinders, beginning with those made of aluminum and then, depending on the results obtained, proceed to titanium and steel plates and pressure vessels; (2) conduct measurements with dual-layer plate and pressure vessel configurations in application-relevant combinations of material, thickness, and stress state; (3) use our experimental results to enhance our ALE3D capability—integrating heat transfer, thermal contact, and element erosion—and determine the ablative pressure and traction boundary conditions for free surfaces; and (4) complete our material breakup model and validate it against laboratory measurements.

Publications and Presentations

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Boley, C. D., and A. M. Rubenchik, 2013. *Modeling of laser interactions with composite materials.* LLNL-PRES-642696.

Florando, J. N., et al., 2013. *Modeling the effect of laser interactions on the strength and failure of high strength aluminum alloys.* LLNL-PRES-642876.

Lowdermilk, W. H., 2011. *Laser–material interaction experiments using a diode array to simulate a DE weapon-class source.* 14th Ann. Directed Energy Symp., La Jolla, CA, Nov. 14–18, 2011. LLNL-ABS-484191.

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.

Rubenchik, A. M., et al., 2013. *Temperature dependent 780-nm laser absorption by engineering grade aluminum, titanium, and steel alloy surfaces.* LLNL-JRNL-635613.

Wu, S. S., et al., 2014. *Laser heating and penetration of aluminum alloys at 0.8-micron wavelength.* 16th Ann. Directed Energy Symp., Huntsville, AL, Mar. 10–14, 2014. LLNL-ABS-642906.

Wu, S. S., et al., 2013. *Near-IR laser interaction with composite materials.* LLNL-PRES-606013.

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 the 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. 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.

Mission Relevance

Our research on laser light propagation and the effects of plasma waves on electron distribution and hard x-ray generation is directly relevant to fusion energy research, which supports a central Laboratory mission of energy security for the nation.

FY13 Accomplishments and Results

In FY13 we (1) completed the extension to our two-dimensional Vlasov LOKI code to multiple species and began simulations, (2) began the Langmuir decay study, (3) developed a plan for implementing collisions in LOKI, (3) updated the SAMRAI (structured adaptive mesh-refinement application infrastructure) software library to enable development of an adaptive mesh-refinement Vlasov code, and (4) collaborated with the University of California, Los Angeles, for benchmarking LOKI with an established particle-in-cell code, and with the Swiss Federal Institute of Technology in Lausanne, Switzerland, for theory and algorithm development.

Proposed Work for FY14

In FY14 we will (1) add, if our approach continues to look feasible, a collisional capability to LOKI; (2) develop new diagnostics for the refactored LOKI code; (3) continue two-dimensional ion acoustic-wave LOKI simulations; (4) consider a 2D+ 2V adaptive mesh-refinement code; (4) study collisional effects on ion acoustic waves; and (5) complete our study of thresholds and the nonlinear state of Langmuir decay instability.

Publications and Presentations

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.

Banks, J. W., et al., 2012. *Vlasov simulations of electron plasma and ion acoustic waves: Self-focusing and harmonics.* APS March Mtg., 2012, Boston, MA, Feb. 27–Mar. 2, 2012. LLNL-ABS-490292.

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Chapman, T. D., et al., 2013. *Nonlinear ion acoustic waves in multi-ion species plasmas at low electron to ion temperature ratios.* 54th Annual Meeting of the APS Division of Plasma Physics, Providence, RI, Oct. 29–Nov. 2, 2013. LLNL-ABS-564171.

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Chapman, T. D., et al., 2013. *The stability of sound waves in plasmas.* LLNL-POST-638197.

Chapman, T. D., et al., 2013. *Vlasov simulation of the parametric instability of ion acoustic waves.* 40th European Phys. Soc. Conf. on Plasma Physics, Espoo, Finland, July 1–5, 2013. LLNL-ABS-623373.

Hittinger, J. A., and J. W. Banks, 2013. "Block-structured adaptive mesh refinement algorithms for Vlasov simulation." *Comm. Comp. Phys.* **241**, 118. LLNL-JRNL-515291.

Strozzi, D. J., et al., 2012. "Threshold for electron trapping nonlinearity in Langmuir waves." *Phys. Plasmas.* **19**, 112306. LLNL-JRNL-574812.

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Predictive Models for Target Response During Penetration

Tarabay Antoun (12-ERD-064)

Abstract

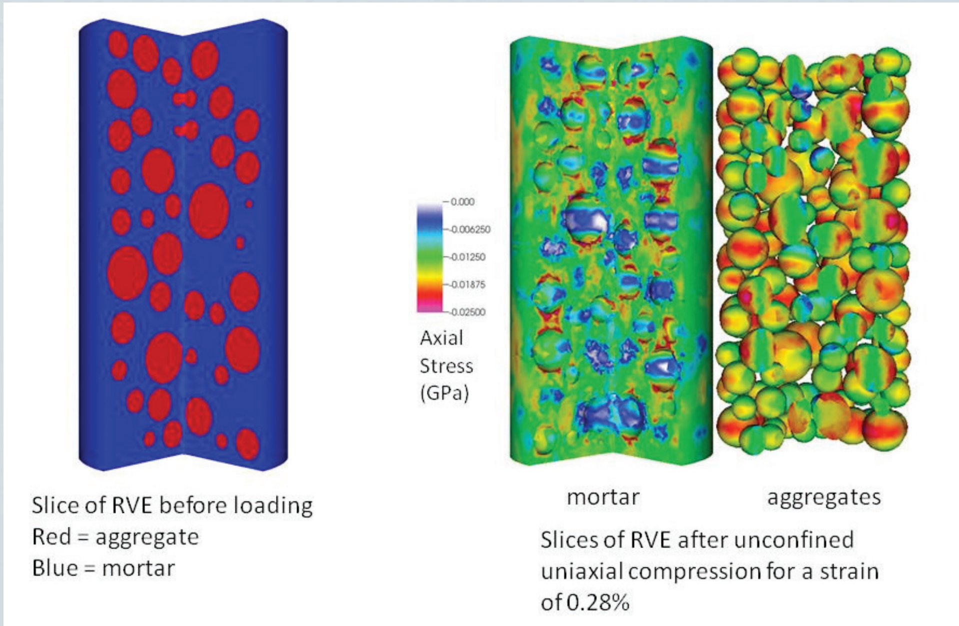
Hardened and deeply buried targets, used by potential adversaries to protect strategic assets, are increasing in number and hardness, making them largely invulnerable to today's conventional weapons. The objective of our proposed research is to develop new, high-fidelity, three-dimensional modeling capabilities 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 heterogeneities 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. We expect that this novel modeling capability will be applicable to the needs of various sponsors, including the Defense Advanced Research Projects Agency, Army Research Laboratory, Defense Threat Reduction Agency, and various branches of the armed services.

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.

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.



FY13 Accomplishments and Results

In FY13 we (1) developed an element decoupling algorithm that maps a tensor damage variable into cohesive contacts inserted around the cell during fracture; (2) completed a parallel algorithm to obtain statistical fragmentation data from our mesoscale simulations to build databases for the DQMOM (direct quadrature method of moments) model, which is used for simulating multiphase flow—this prompted the organization of a working group with the Laboratory's visualization group to provide this capability to other code developers; (3) successfully matched our model results to data from our Department of Defense collaborator; (4) implemented a continuum model for use by the ALE3D (arbitrary Langranian–Eulerian three-dimensional) code, which is used for examining hydrodynamics and structures; and (5) hosted an external collaborator for the summer to help with the final continuum framework for the model.

Proposed Work for FY14

In FY14 we will (1) focus on refining the continuum material model, which will require multiple mesoscale simulations to provide statistical data to develop evolution equations for the fragment size distribution for the DQMOM and constitutive model; (2) develop the DQMOM and continuum model in the GEODYN geological drilling simulation code and then transfer it into the ALE3D code; (3) perform large-scale continuum penetration simulations for validation; and (4) use the model to assess its predictive capability.

Publications and Presentations

Antoun, T., et al., 2012. *Correlation of observed macroscopic response to underlying response mechanisms*. LLNL-PRES-576174.

Applying High-Performance Computing and Simulation to Future Energy Challenges

Clara Smith (12-ERD-074)

Abstract

High-performance computing and simulation has the potential to optimize production, distribution, and conversion of energy. Although a number of concepts have been discussed, a comprehensive research project to establish and quantify the effectiveness of computing and simulation at a scale appropriate for core energy problems has not been conducted. We propose to perform the basic research to adapt existing high-performance computing tools and simulation approaches to two selected classes of problems common across the energy sector. The first, applying uncertainty quantification and contingency analysis techniques to energy optimization, allows us to assess the effectiveness of LLNL core competencies to problems such as grid optimization and building-system efficiency. The second, applying adaptive meshing and numerical analysis techniques to physical problems at fine scale, could allow immediate impacts in key areas such as efficient combustion and fracture or spallation. By creating an integrated project team with the necessary expertise, we can efficiently address these issues, delivering both near-term results as well as quantifying developments needed to address future energy challenges.

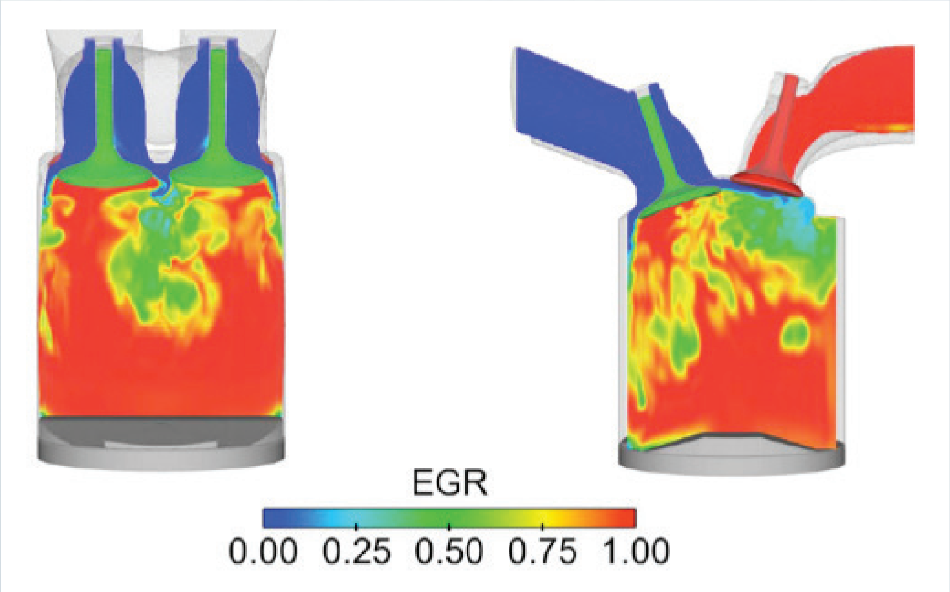
We expect to provide fundamental progress in the basic scientific challenges of applying high-performance computing and simulation to energy issues. Partnering with the private sector, we have identified representative near-term research topics and goals. Specifically, we intend to demonstrate application of uncertainty quantification and contingency analysis to the solution of large energy-grid problems such as electricity production and distribution. We will also provide computer simulations related to improved engine combustion and efficient fracture or spallation for oil and gas drilling. In addition, we will assess the capabilities that must be developed to make longer-term progress in applying computing and simulation to energy issues. Peer-reviewed publications as well as significant intellectual property are expected as a result of this research.

Mission Relevance

Applying LLNL's competency in high-performance computing and simulation expertise to energy production, distribution, and use directly supports the Laboratory's strategic mission focus in energy security and could allow the Laboratory to optimize a potential expansion of its efforts in these areas.

FY13 Accomplishment and Results

In FY13 we (1) analyzed, in collaboration with GE Research, the Cornell fuel spray code and demonstrated that it captured physics consistently compared with experiments; (2) compared, in collaboration with Bosch, large-eddy simulation computational fluid-dynamics codes to experiments and demonstrated the viability of using high-performance computing to model complex combustion mechanisms in a time frame



A simulation in collaboration with Bosch using high-performance computing and large-eddy simulation to understand the transient dynamics for novel multiple-mode combustion engines.

relevant to industry; (3) performed, in collaboration with Potter Drilling, a sensitivity study of spall drilling simulations for a range of borehole conditions, as well as microstructural, thermal, and mechanical properties—we determined the factors most crucial to damage extent and spall size; (4) performed, in collaboration with GE Energy, parallel contingency analysis for the electricity-distribution-grid code PSLF (positive sequence load flow), and reduced the run time from 23.5 days to 23 minutes—we improved matrix-reordering algorithm solution times through optimization and threading by over sevenfold; (5) completed, in collaboration with ISO New England, numerous electrical grid simulations to demonstrate approximately 2% cost saving by employing "robust" unit-commitment algorithms, and developed statistical clustering techniques to model weather effects; and (6) conducted, in collaboration with United Technologies Research Center, high-performance computing uncertainty analysis to understand parameter interactions, and identified the 50 most-impactful parameters.

Project Summary

Our project provided six key demonstrations on how high-performance computing could be used to impact applied energy research for industry. We gained valuable experience and insight into the computational challenges facing industry and have shown that high-performance computing is a viable approach to solving difficult problems. A concrete example is the collaboration with ISO New England, in which Lawrence Livermore has been cited in their press releases and annual report for exploring new algorithms for electric grid operations. Finally, in two of the six projects, we either acquired or developed new modeling and simulation technology that will be used to benefit other programs. We are exploring cooperative research and development agreement opportunities and joint proposals with three of our industry partners.

Publications and Presentations

Hansen, R., 2013. "Scaling up energy innovation through advanced computing." *Sci. Tech. Rev.* June 2013, 4. UCRL-TR-52000-13-6.

Heller, A., 2012. "Incubator busy growing energy technology." *Sci. Tech. Rev.* June 2012, 24. UCRL-TR-52000-12-6.

Walsh, S. D. C., and I. Lomov, 2013. "Micromechanical modeling of thermal spallation in granitic rock." *Int. J. of Heat Mass Tran.* **65**, 366. LLNL-JRNL-637634.

Walsh, S.D.C., et al., 2013. *Size dependent spall aspect ratio and its effects in thermal spallation.* LLNL-JRNL-642106-DRAFT.

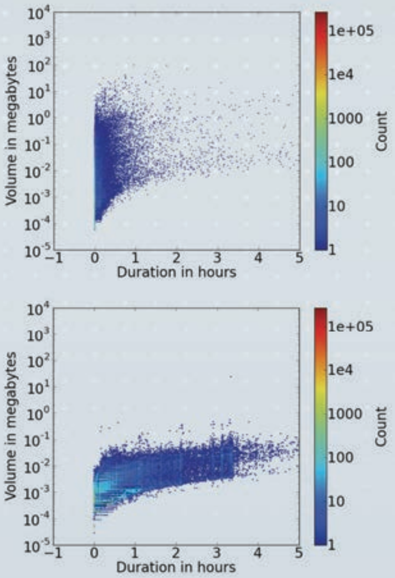
Continuous Network Cartography

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-mapping capabilities and analytics that apply machine-learning and statistical methods for understanding network activities. In addition to continuous mapping and analytics, 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.

This project will establish the foundations of future infrastructure for 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 the continuous maps



Continuous network mapping enables dynamic analysis, which reveals important temporal patterns in computer networks. For example, continuous mapping led to a novel technique for detecting peer-to-peer botnets (distributed networks of malicious computer programs) based on traffic patterns over time. Normal network traffic (top) is compared to peer-to-peer botnet traffic (bottom), which tends to be longer duration and lower volume than normal traffic.

for understanding 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 complex adversarial intrusion, denial-of-service attacks, or deception tactics.

Mission Relevance

This project is strategically aligned with the mission focus area of cyber security, specifically, cyber situational awareness, and will enable LLNL to develop more sophisticated, robust techniques for national security and homeland security applications.

FY13 Accomplishments and Results

In FY13 we accomplished all proposed tasks. Specifically, we (1) procured and deployed a continuous network mapping platform, implemented continuous mapping capability, and collected data; (2) developed three models (dynamic role-based, generative network incorporating structure and attributes, and individual communication channels and protocols) of real-world network characteristics and behaviors with network structure, communication patterns, connection-level, traffic, and host features; (3) developed novel approaches to address network change detection including hypothesis testing, attribution or explanation of network changes, and an axiomatic approach to network similarity; and (4) continued to explore ways to apply user feedback for improvements.

Proposed Work for FY14

In FY14 we will (1) develop metrics for determining a timescale for updating continuous maps and an approach to determine areas for prioritized mapping; (2) integrate various change-detection components into a comprehensive approach; (3) test the scalable implementation of change detection on a mapping cluster; (4) develop an approach for incorporating a human analyst in the change-detection loop; (5) perform initial mission mapping and infer the importance or function of collections of hosts, with regards to resilience; (6) develop algorithms to quantify the importance of hosts and links to a given mission; (7) develop an initial mobile network-mapping capability; and (8) create new capabilities to best display the results of these continuous maps.

Publications and Presentations

Berlingerio, M., et al., 2013. *Network similarity via multiple social theories.* ASONAM 2013, The 2013 IEEE/ACM Intl. Conf. Advances in Social Networks Analysis and Mining, Niagara Falls, Canada, Aug. 25–28, 2013. LLNL-CONF-645771.

Gilpin, S., T. Eliassi-Rad, and I. Davidson, 2013. *Guided learning for role discovery (GLRD): Framework, algorithms, and applications.* KDD2013, The ACM SIGKDD Conf. Knowledge Discovery and Data Mining, Chicago, IL, Aug. 11–14, 2013. LLNL-CONF-645813.

Hang, H., et al., 2013. *Entelecheia: Detecting P2P botnets in their waiting stage.* IFIP Networking 2013 Conf., Brooklyn, NY, May 22–24, 2013. LLNL-CONF-645814.

Rossi, R. A., et al., 2013. *Modeling dynamic behavior in large evolving graphs.* 6th ACM Intl. Conf. Web Search and Data Mining, Rome, Feb. 4–8, 2013. LLNL-CONF-603132.

Tong, H., et al., 2012. *Gelling, and melting, large graphs by edge manipulation.* CIKM: 21st ACM Intl. Conf. Information and Knowledge Management, Oct. 29–Nov. 2, 2012. LLNL-CONF-645812.

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 weapon scans to airport security to images of National Ignition Facility target capsules. While each area 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 for nondestructive evaluation to advance the state of the art of computed tomographic segmentation. We will 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 significant new capabilities that can form the basis for 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. This project will enhance LLNL's capabilities in the area of airport security. This project will also significantly advance the Laboratory's capabilities in x-ray imaging for nondestructive evaluation in such areas as biology and experimental physics.

FY13 Accomplishments and Results

In FY13 we (1) developed new reconstruction techniques that provide a better beam-hardening correction, interface with various segmentation algorithms, and potentially provide uncertainty information for variance and covariance terms per grid points; (2) explored and evaluated various segmentation schemes and decided to proceed with a bottom-up approach; (3) started implementing a novel multiple-hierarchy graph structure to create fuzzy segmentation for use in semantically based threat detection and target recognition techniques; (4) developed new localized dimension estimation techniques; and (5) continued the development of our NDDAV (*N*-dimensional data analysis and visualization) toolkit, which is now deployed in an alpha release to Livermore computing clusters.

Proposed Work for FY14

In FY14 we will (1) develop reconstruction techniques that return a mean value as well as standard deviations and/or local covariance information to better compensate for unavoidable artifacts; (2) develop region-based active contour segmentation algorithms able to segment multiple-valued volumes and store the results in a region-adjacency graph; (3) develop techniques to process the region-adjacency graph into robust, high-quality segments; (4) extract the alignment of objects from the segmentation to enable reconstruction on a optimal rotated grid; and (5) integrate threat-detection techniques with the region-adjacency graph to complete an initial prototype of the computational data-processing stages for the detection pipeline.

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 because of 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, 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 LLNL 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 science, technology, and engineering foundation in high-performance computing and simulation.

FY13 Accomplishments and Results

In FY13 we (1) engaged in active discussions with DOE Advanced Scientific Computing Research vendors concerning integration of logic and compute nodes with three-dimensional memory stacks to be used in exascale architectures; (2) initiated the design of a data-intensive active memory architecture based on the Hybrid Memory Cube and measured a 25% decrease in required memory bandwidth for a mesh simulation benchmark; (3) completed a streaming bioinformatics benchmark and additionally demonstrated performance improvement of almost an order of magnitude for a key-value storage application in an emulated storage controller based on a field-programmable gate array; (4) designed a new algorithm to optimize a high-performance, data-intensive memory map device driver for mixed read and write workloads; (5) conducted an initial study of data structures for dynamic, time-varying graphs stored in NAND flash memory; (6) completed initial implementation of streamline tracing in the data-intensive memory map; and (7) implemented a two-level index for a bioinformatics database.

Proposed Work for FY14

In FY14 we will (1) continue design, implementation, and evaluation of data-intensive active memory hardware architectures using simulation and emulation based on a field-programmable gate array; (2) design and prototype the system software interface between active memory and application; (3) evaluate the data-intensive memory map driver on new data-intensive workloads and design optimized buffer management algorithms for the driver; (4) complete design and implement data structures optimized for dynamic, time-varying graphs; (5) begin advanced algorithm (e.g., community analysis) design for time-varying graphs stored in NAND flash memory; (6) replay historical Twitter data on new algorithms and data structures; and (7) experiment with flow exploration and seeding strategies to determine data access patterns for streamline tracing.

Publications and Presentations

De, A., et al., 2013. *Minerva: Accelerating big data applications using next-generation non-volatile memory-based SSD.* 21st IEEE Intl. Symp. Field-Programmable Custom Computing Machines, Seattle, WA, Apr. 28–30, 2013. LLNL-CONF-609932-DRAFT.
Gokhale, M., and S. Lloyd, 2013. *Memory integrated computing.* LLNL-MI-632592-DRAFT.

Pearce, R., M. Gokhale, and N. Amato, 2013. *Scaling techniques for massive scale-free graphs in distributed (external) memory.* IEEE Intl. Parallel Distributed Processing Symp. (IPDPS), Boston, MA, May 20–24, 2013. LLNL-CONF-588232-DRAFT.

Van Essen, B., et al., 2013. *DI-MMAP: A scalable memory map runtime for out-of-core data-intensive applications.* Conf. Cluster Computing, Lviv, Ukraine, June 12–14, 2013. LLNL-JRNL-612114-DRAFT.

Wong, D., G. S. Lloyd, and M. B. Gokhale, 2013. *A memory-mapped approach to checkpointing.* LLNL-TR-635611.

Fast-Running Codes via High-Fidelity Reduced-Order Models

Kyle Chand (13-ERD-031)

Abstract

We propose to develop practical 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.

FY13 Accomplishments and Results

In FY13 we (1) developed new automated and incremental reduced-order model construction algorithms that are time-adaptive and controlled using estimates of the projection error; (2) developed approaches for the application of parameter-dependent, time-varying, boundary conditions in reduced-order models built from simulations based on partial differential equations—this work is critical for application of reduced-order models to LLNL-relevant applications; (3) tested our research on model problems designed to guide further research and development; and (4) commenced initial design and prototyping of a reduced-order model software framework that can interface to simulation tools, which will form the basis of future work.

Proposed Work for FY14

In FY14 we will (1) improve our time-adaptive and incremental algorithms and extend them to parameter-dependent reduced-order models; (2) begin to explore spatially adaptive reduced-order models, another research goal of this project—our work will involve further testing of the algorithms on model problems as well as their application to some selected simulation tools, initially from the Overture framework for representing problems involving complex domains with moving components; (3) continue design and development of high-performance computing software; and (4) exercise our software via the adaptive construction of a time-dependent reduced-order model from a representative Overture-based simulation.

Whole-Heart Modeling on High-Performance Computing Systems

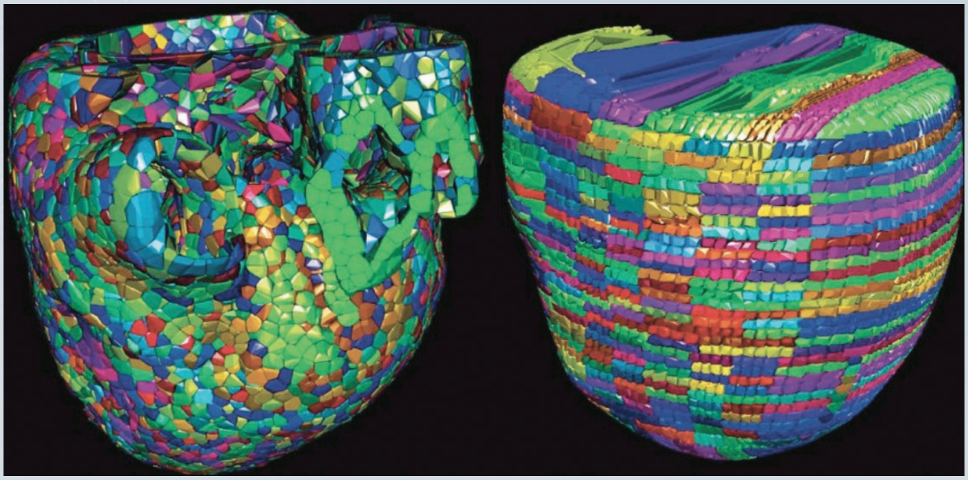
David Richards (13-ERD-035)

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 also will 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 is a Livermore core competency. Furthermore, it supports a new strategic thrust in bioscience and biotechnology 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.

FY13 Accomplishments and Results
Because of increased interest in our electrophysiology capability, we have added an FY13 milestone to investigate mechanisms of arrhythmia, or irregular heartbeat.



A heart simulation computationally divides the human heart into numerous subdomains.

In FY13, therefore, we (1) generated a sustained arrhythmia in the presence of an arrhythmogenic drug, with an electrocardiogram showing a Torsade de pointes signature, which causes a sudden drop in arterial blood pressure; (2) established software collaboration agreements with the University of Rochester and the Federal Drug Administration; (3) initiated a comprehensive whole-heart model with various components and their interactions, including drug effects; (4) determined that our mechanics code agrees with the standard Chaste (cancer, heart, and soft-tissue environment) library results for computational biological simulations; (5) implemented a new finite-element procedure better suited to the incompressible regime; and (6) began coupling electrophysiology to the mechanics, with an initial step being computation of the electrocardiogram directly from our electrophysiology code.

Proposed Work for FY14

In FY14 we will (1) perform a systematic study of drug effects on arrhythmia; (2) compare our mechanics code results to those of others and ascertain benefits of high resolution (e.g., whether papillary muscle representation improves results); (3) complete the coupling of mechanics to electrophysiology through a contractile model; (4) improve scalability of the electromechanical code, particularly that of the mechanics solver, and carry out cardiac resynchronization therapy studies using our electromechanical capability; (5) complete scoping of a whole-heart model; and (6) institute, time permitting, mechanical and electrical feedback into our electromechanical code.

Publications and Presentations

Richards, D. F., and J. J. Rice, 2013. *Simulation of drug-induced arrhythmias in the human heart using over 1.5 million CPUs*. Current Challenges in Computing Conf., Napa, CA, Sept. 3–5, 2013. LLNL-ABS-642634.

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.

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 videos 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 partition 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 comprising the proposed data structures and algorithms, and 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 fields of application.

Mission Relevance

This project supports the Laboratory's national security mission and the mission focus area of cyber, space, and intelligence 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.

FY13 Accomplishments and Results

In FY13 we (1) explored current state of the art in video segmentation and developed an approach combining the best of interest-point homography-based methods (which are efficient but the coverage can be uneven) with pixel-based optical flow methods (which are exhaustive but computationally expensive as typically applied); (2) leveraged Spark, a new high-performance architecture for big data, to develop a more flexible deep-learning approach to visual features using the unsupervised kernel k-means clustering technique; (3) established a collaboration with the University of California, Berkeley International Computer Science Institute and imported their state-of-the-art audio analytics capabilities—their audio feature hierarchy of low-level features, to intermediate-level percepts, to high-level concepts is inspired by neuroscience and has strong analogs with the deep-learning approach—as well as their new tagged video data sets; and (4) dropped the proposed mosaic-generation task in favor of more emphasis on unsupervised feature learning (e.g., exploring the relationship between the International Computer Science Institute audio feature and that obtained via deep learning) and fusion of multiple modal features.

Proposed Work for FY14

In FY14 we will (1) construct a large feature video layer tag graph using Flickr (social media) data; (2) establish, by leveraging earlier segmentation and feature extraction work, both a performance baseline for learning (e.g., tag propagation) and query tasks as well as a foundation for beginning relevance-feedback work; (3) explore early, late, and hybrid audio–visual fusion strategies in collaboration with the International Computer Science Institute that exploit motion and scene dynamics, going beyond the current "bag of features" approach, and measuring performance against our baseline; and (4) define a high-performance computing grand-challenge problem in which we map this framework to a high-performance computing platform and produce ensembles of learners using large sparse-basis sets (such as spectro-temporal Gabor feature extractions for audio, which are useful for automatic speech recognition).

Publications and Presentations

Elizalde, B., G. Friedland, and K. S. Ni, 2013. *Is what you hear is what you get? Audio-based video event detection and audio concept classification.* LLNL-POST-644334.

Ni, K. S., and R. J. Prenger, 2013. F*eatures in deep learning architectures with unsupervised kernel k-means.* GlobalSIP: IEEE Global Conf. Signal and Information Processing, Austin, TX, Dec. 3–5, 2013. LLNL-CONF-644296.

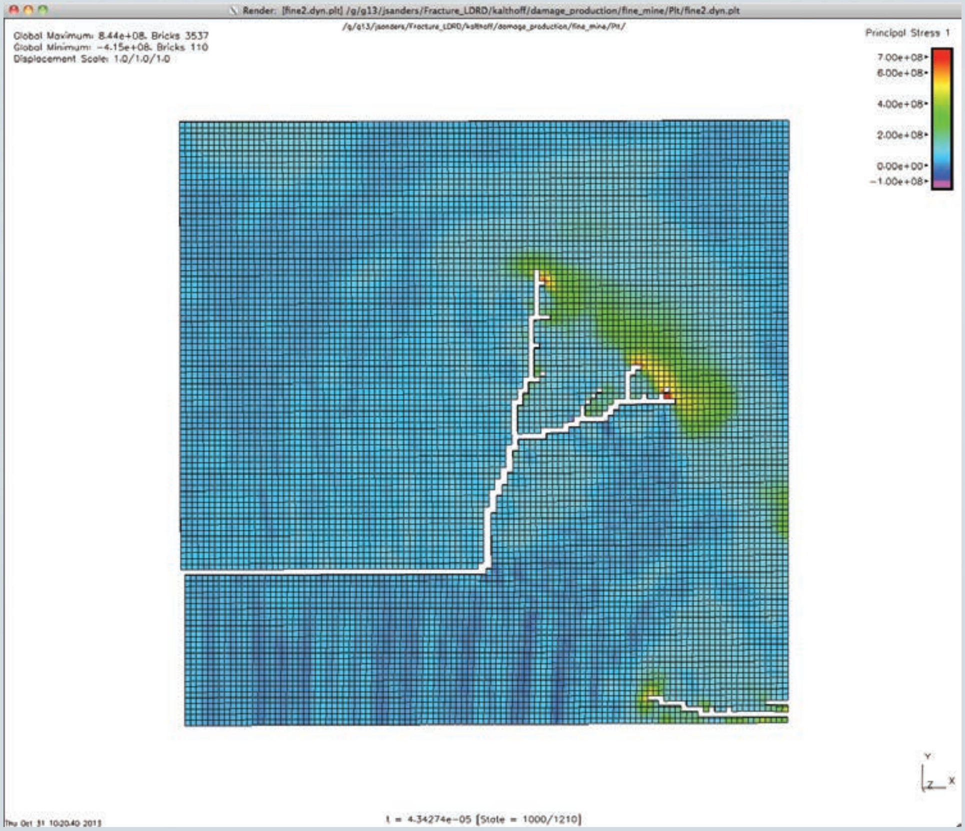
Vesom, G., 2013. *Towards hybrid motion-cued region-based video segmentation.* LLNL-POST-639605-DRAFT.

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



Simulation of brittle fracture of a poly (methyl methacrylate) plate under impact loading.

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 a technology 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

The fracture of structural materials is relevant to many national security applications. For stockpile stewardship science, true virtualization of the nuclear weapon development cycle and materials is needed. Defense applications include prediction of structural survivability for transport security and the 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.

FY13 Accomplishments and Results

Our focus for the first year was to demonstrate a numerical fracture method for a cracked body, without re-contact. Specifically, in FY13 we (1) developed the discrete mathematical framework for our version of nodal enrichment adapted to the generalized finite-element method, as well as for crack tracking; (2) created a prototype in two dimensions in the MATLAB technical computing language and extended it to three dimensions with a DYNA3D mechanical deformation code prototype; (3) chose an initial model for brittle failure and performed the first three-dimensional test problems; (4) reproduced a classic experimental benchmark in brittle failure in poly (methyl methacrylate) plates; and (5) proposed, for the problem of element-local discrete crack tracking in three dimensions, an algorithm different from that presented in the current literature—specifically, continuity of the discrete crack is neither assumed nor enforced in an ad hoc manner, such that the material model is given full control over the fracture orientation, and cracked elements are still able to separate.

Proposed Work for FY14

In FY14 we will (1) consider numerical treatment of multiple cracks passing through a single element; (2) consider advanced failure and material models, including extending the method to inelastic behavior such as the rupture of a steel plate as well as issues for the more difficult problem of ductile tearing; and (3) develop a test suite to demonstrate these capabilities.

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.

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.

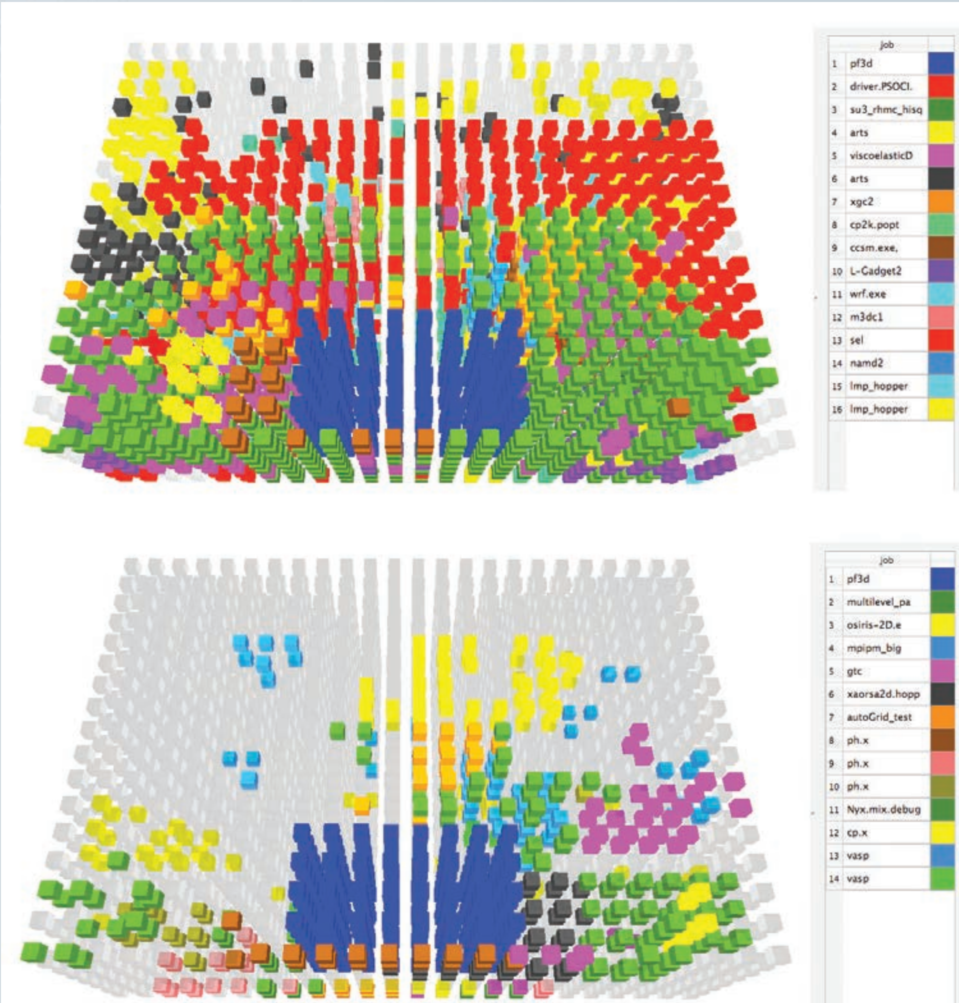
The proposed tools will help code-development teams to decide whether the communication in a computer application can benefit from better mapping. The tools will also help us in identifying phases in the application that are most severely affected by congestion. We expect this work will lead to development 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 to 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 science, technology, and engineering foundation in high-performance computing and simulation. 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.

FY13 Accomplishments and Results

In FY13 we (1) developed a prototype tool that can measure the types and sizes of messages sent in a parallel application and began adding the functionality to obtain effective minimum latency and peak bandwidth for a network; (2) used hardware counters to quantify contention and used machine-learning algorithms to



The placement of pF3D, a laser–plasma interaction code (shown in blue), and other conflicting computational jobs on Hopper, a Cray XE6 system at the National Energy Research Scientific Computing Center in Berkeley, California, for two separate short runs. The first job (top) yielded a messaging rate nearly 25% below that of the second job (bottom). The two jobs had the same node placement, but the slower job was surrounded by nodes of several other jobs, including a large communication-heavy job (green).

predict application performance for different task mappings; (3) characterized the communication of pF3D, a laser–plasma interaction code, and began performance characterization of the SAMRAI (structured adaptive mesh refinement application infrastructure) software library and AMG (algebraic multigrid method); (4) helped implement a framework, Chizu, which will be used by Laboratory programmatic codes for task mapping, and implemented a tool to determine the computer network topology on Livermore's Blue Gene/Q supercomputer; and (5) implemented a fast network simulator to predict the execution time of bulk synchronous structured codes.

Proposed Work for FY14

In FY14 we will (1) use the tools developed in FY13 to characterize applications, (2) continue studying network contention and create models for a torus network topology, (3) explore and evaluate mapping and graph-partitioning algorithms with Laboratory programmatic codes, and (4) develop generic mapping algorithms that can be used for different application classes.

Publications and Presentations

Bhatele, A., and T. Gamblin, 2012. *OS/runtime challenges for dynamic topology-aware mapping.* U.S. DOE Exascale OS/R Workshop, Washington, DC, Oct. 3–4, 2012. LLNL-PRES-587572.

Bhatele, A., and T. Gamblin, 2013. *Placing communicating tasks apart to maximize bandwidth.* SIAM Conf. Computational Science and Engineering, Boston, MA, Feb. 25–Mar. 1, 2013. LLNL-PRES-621732.

Bhatele, A., et al., in press. "Network simulations of a future supercomputer with half-a-million processor cores." *IEEE Trans. Parallel Distr. Syst.* LLNL-JRNL-619172-DRAFT.

Bhatele, A., et al., 2013. *There goes the neighborhood: Performance degradation due to nearby jobs.* Intl. Conf. High Performance Computing, Networking, Storage and Analysis, Denver, CO, Nov. 17–22, 2013. LLNL-CONF-635776.

Jain, N., et al., 2013. *Predicting application performance using supervised learning on communication features.* Intl. Conf. High Performance Computing, Networking, Storage and Analysis, Denver, CO, Nov. 17–22, 2013. LLNL-CONF-635857.

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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 types. 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 arise from 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, practical algorithms, and codes that will enable the analysis of diverse relational data at a scale heretofore not 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 who are working on extremely large data sets.

Mission Relevance

This project is closely aligned to the Laboratory's cyber, space, and intelligence mission. Data mining and analysis using large-scale graphs and hypergraphs is fundamental to the analysis of Internet traffic, network intrusions, financial networks, funding flow, 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 applications areas.

FY13 Accomplishments and Results

In FY13 we (1) developed a vertex affinity measure, which is used for determining similarity between arbitrary pairs of vertices, by employing weighted path information that generalizes to hypergraphs; (2) designed a random projection technique to factor general distance matrices, which also applies to nonsymmetric distance matrices and directed graphs; (3) designed and implemented a family of graph generators using community decompositions with extensions to directed graphs and hypergraphs in development; (4) built a hypergraph clustering approach based on an algebraic multigrid; (5) analyzed factorization techniques to determine their fidelity in reproducing matrices of affinity distances; and (6) continued to enhance numerical linear algebra and eigensolver techniques through our collaborations.

Proposed Work for FY14

In FY14 we will (1) implement serial and parallel versions of data-mining codes that incorporate vertex affinity measures combined with novel factorization techniques; (2) apply the codes to important data-mining tasks including clustering, community identification, and community detection from known members (seed-set expansion); and (3) create multilevel approaches for data-mining tasks, verifying that the added information developed in this process is useful to accomplish the tasks and that the multilevel approach provides the extra efficiency necessary for successfully handling extremely large data sets.

Publications and Presentations

Boman, E., K. Devine, and S. Rajamanickam, 2014. *Scalable matrix computations on large scale-free graphs using 2D graph partitioning.* SC14: HPC Matters, New Orleans, LA, Nov. 16–21, 2014. LLNL-JRNL-645609.

Breuer, A., G. Sanders, and A. Lumsdaine, 2013. *Restarted minimal Krylov subspaces for low-rank approximation.* LLNL-JRNL-643736.

D'Ambra, P., and P. S. Vassilevski, 2013. *Adaptive AMG with coarsening based on compatible weighted matching*. LLNL-TR-613612.

De Sterck, H., 2013. "Steepest descent preconditioning for nonlinear GMRES optimization." *Numer. Lin. Algebra Appl.* **20**; 453. LLNL-JRNL-645775.

De Sterck, H., and K. Miller. 2013. "An adaptive algebraic multigrid algorithm for low-rank canonical tensor decomposition." *SIAM J. Sci. Comput.* **35**, B1. LLNL-JRNL-645774.

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Hu, X., 2013. *Fast multi-level co-clustering.* Master's Thesis, Waterloo University, Waterloo, Canada. LLNL-TH-645773-DRAFT.

Sanders, G., and V. Henson, 2013. *Eigenvectors of matrices associated with scale-free graphs.* 16th Copper Mountain Conf. Multigrid Methods, Copper Mountain, CO, Mar. 17–22, 2013. LLNL-PRES-628160.

Sanders, G., et al., 2013. *Maximum principles and decay rates for extremal eigenpairs of scale-free adjacency and modularity matrices.* LLNL-JRNL-644341.

Leveraging Computer Log Analytics to Understand Application Input and Output

George Todd Gamblin (13-FS-002)

Abstract
Parallel input/output performance and reliability are critical for large-scale high-performance computing applications. However, there are currently large gaps in our understanding of the causes of parallel input/output performance problems and failures. We propose to explore the feasibility of developing techniques to explain input/output reliability issues for the Lustre file system for computer clusters when applications write checkpoints. Lustre servers generate voluminous diagnostic and error logs. However, because of the lack of effective analysis tools, there is no way to associate particular failures and performance problems with the application and system events that caused them. We will deliver off-line tools to bridge the gap between applications and file system logs. This work will demonstrate the feasibility

of future scalable online tools that continuously monitor, analyze, and optimize the Livermore Computing facility. Our long-term goal is to use data analytics in conjunction with existing LLNL tools to automate the diagnosis of problems at Livermore Computing.

For this study, we will develop a set of scalable queries and associated libraries to correlate log events with application performance and resilience data. This will provide Livermore Computing with valuable information about the failure rates of Lustre file systems. It will help developers of system configuration repositories to coordinate checkpoint writing, increasing the performance of codes using the repository. This will be the first attempt to bridge the gap between system-level data such as Lustre file system logs and user application data, which is the first step towards a holistic understanding of Livermore Computing.

Mission Relevance

This project is relevant to LLNL's strategic science, technology, and engineering pillar in high-performance computing and simulation. Developing expertise in data analytics will provide a foundation for future analytics projects and demonstrate the feasibility of gathering and analyzing Livermore Computing system and facility data. We will leverage the results of log analytics to benefit high-performance computing by increasing the efficiency and reliability of Livermore Computing's file systems. This has the potential to drastically increase simulation performance, which directly benefits LLNL's mission thrusts.

FY13 Accomplishments and Results

In FY13 we applied statistical and machine-learning techniques to historical performance data and system logs from simulations. Specifically, we studied a massively parallel code that simulates laser–plasma interaction (pF3D) and CESM (community earth system model) as executed over several months, and analyzed system logs to correlate performance degradation with root causes. In addition, statistical analysis of the long-term network data collected for these long-running simulations can explain large performance variability or degradation and allowed us to isolate the cause to particular variables in the input data.

Project Summary

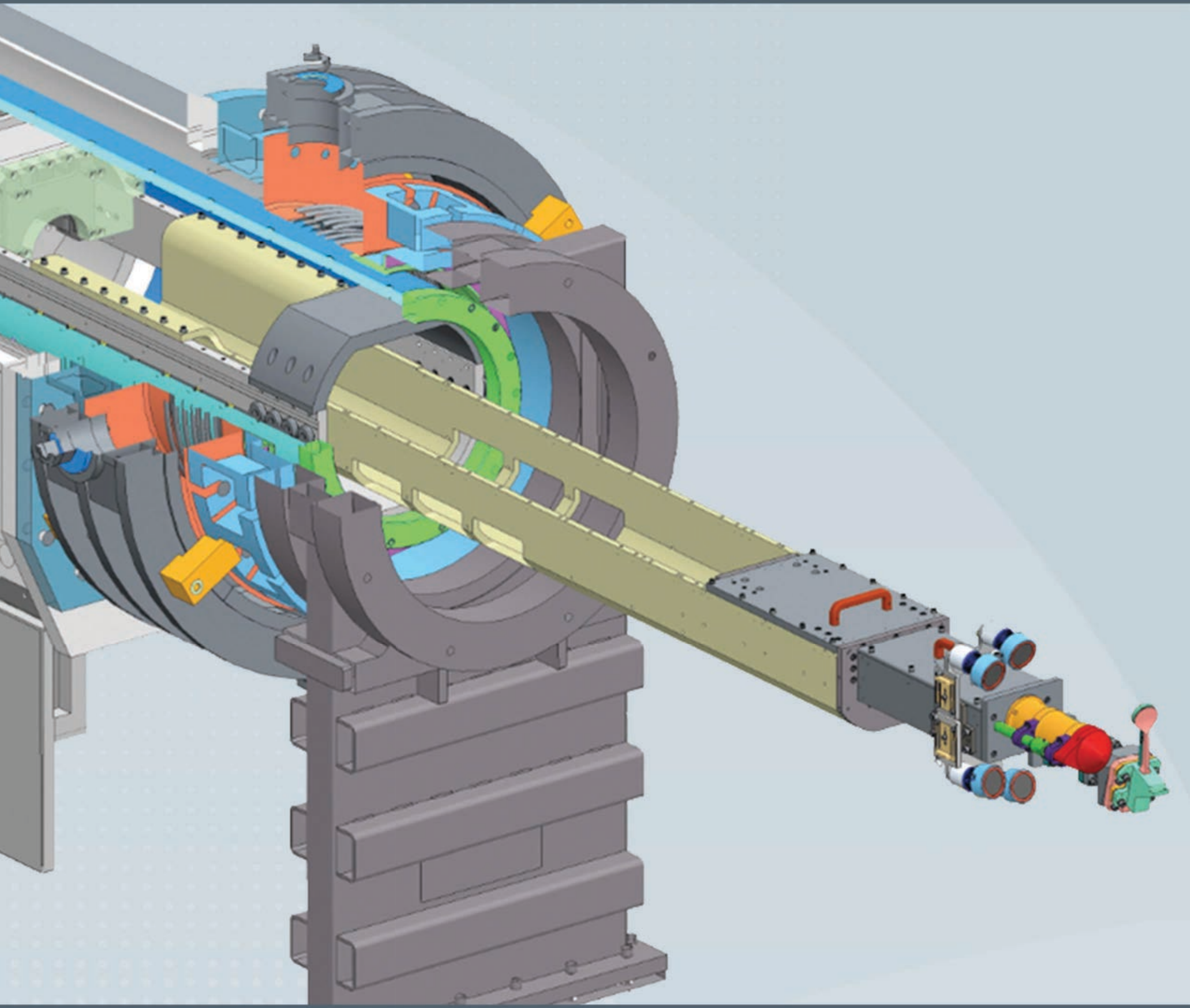
The successful completion of this project has shown that using machine-learning and statistical techniques to analyze historical log data is a promising direction, and that it could be applied in the future to automatically determine the causes of performance problems. Because of administrative and security concerns, we were unable to analyze the input/output data for the Lustre file system as originally intended. We were, however, able to show that it is possible to apply these techniques to network data and correlate application performance degradation to job queue information and system allocation information. This demonstrates that the approach is feasible and worth pursuing. Pending funding, the next steps for this research are to begin to store performance data from many more simulations run at LLNL and apply statistical and machine-learning techniques to historical job data.

Publications and Presentations

Bhatele, A., et al., 2013. *There goes the neighborhood: Performance degradation due to nearby jobs.* Intl. Conf. High Performance Computing, Networking, Storage and Analysis, Denver, CO, Nov. 17–22, 2013. LLNL-CONF-635776.

Gamblin, T., and K. Mohror, 2013. *Leveraging log analytics to understand inter-job interference.* LLNL-TR-645337.

Nuclear Science and Engineering



Laboratory Directed Research and Development

Annual Report FY2013

Advanced Inertial Fusion Target Designs and Experiments for Transformative Energy Applications

Peter Amendt (11-SI-002)

Abstract

Inertial fusion energy research is gaining momentum in the scientific world and the energy arena. A key challenge for realizing fusion energy production is to develop a viable and experimentally validated "point design," which is an integrated simulation used to assess overall performance for the laser target within the larger framework of a fusion energy system. This project will develop central hot-spot ignition and fast-ignition target designs for inertial fusion energy, validated with experiments on the OMEGA laser facility at the University of Rochester and on the National Ignition Facility (NIF) at Livermore. This planned research, founded upon the extensive experience of Laboratory scientists and engineers who developed the point design for NIF, will deliver a suite of inertial fusion energy target designs developed with radiation-hydrodynamic simulations and tailored to test several key physics issues on the OMEGA laser facility and NIF.

We expect to deliver an ensemble of central hot-spot ignition target designs for inertial fusion energy that leverage the current National Ignition Campaign point design. A collection of techniques to significantly increase the coupling efficiency of these targets for moderate energy gain (less than a hundredfold) through innovative measures—such as low drive temperature, rugby-ball-shaped hohlraum target capsules, and radiation shields—will be applied and tested at NIF. In parallel, we will strive to develop a suite of point designs for high-gain (greater than a hundredfold) fast ignition using the high-efficiency hohlraums developed for central hot-spot ignition. Experiments at NIF to test the key physics uncertainties in the fast-ignition concept are also planned.

Mission Relevance

The proposed research could have a groundbreaking, transformative impact on fusion energy and high-energy-density matter research. The research will thus help enhance America's national security by developing proliferation-resistant advanced energy technologies, economic security by improving energy efficiency, and energy security by reducing energy imports and greenhouse gases.

FY13 Accomplishments and Results

In FY13 we (1) fielded, for the first time, two rugby-shaped hohlraums at NIF—the second shot was at full laser energy and produced a highly asymmetric implosion in contrast to mainline simulation predictions; (2) adopted a mix model to ultimately provide fairly good agreement between modeling and simulations—results are consistent with the presence of mix between the high-atomic-number wall and the confining helium gas fill; (3) determined that the rugby hohlraum platform provided a new test of our simulation capabilities and may lead to an improved understanding of hohlraum dynamics in general in the pursuit of ignition; and

(4) tested high-density carbon ablators for the first time at NIF with favorable results—the ablators have become one of two mainline paths to demonstrating ignition on NIF.

Project Summary

Our research supported early successful experimental testing of high-density carbon ablators and lead hohlraums on the OMEGA laser for inertial fusion energy applications. The successful conclusion of this project resulted in the adoption by Livermore's National Ignition Campaign of the high-density carbon ablator for ignition studies, and continued testing in FY14 of the rugby-shaped hohlraum as an alternative platform for simplified hohlraum dynamics and description.

Publications and Presentations

Amendt, P., et al., 2011. "LIFE pure fusion target designs: Status and prospects." *Fusion Sci. Tech.* **60**(1), 49. LLNL-PROC-463711.

Amendt, P., et al., 2013. *Rugby hohlraum campaign on the National Ignition Facility: Status and comparison with modeling.* 8th Intl. Conf. Inertial Fusion Science and Applications (IFSA 2013), Nara, Japan, Sept. 8–13, 2013. LLNL-PROC-645277.

Ross, J. S., et al., 2013. "Lead (Pb) hohlraum: Target for fusion energy." *Sci. Rep.* **3**, 1453. LLNL-JRNL-588175.

Shay, H. D., et al., 2012. "Implosion and burn of fast ignition capsules—Calculations with HYDRA." *Phys. Plasma.* **19**(9), 092706. LLNL-JRNL-550171.

Imaging for Use in a Verification Regime

Mark Cunningham (11-ERD-051)

Abstract

Current U.S. national security policies clearly call for moving past Cold War arms control towards verifiable stockpile reductions. The focus is likely to shift to determining whether or not an item is a nuclear warhead, and if so, whether it is most probably in an operationally deployed configuration. Imaging may play an important role in these verifications. Our objective is to establish the utility and optimization of a new concept for a gamma-ray imaging system appropriate for nuclear treaty verification and on-site inspections. To do so, we will demonstrate an innovative science-based approach for the decomposition of a gamma-ray image of an object, such as a nuclear warhead, into a set of unclassified metrics. These metrics must be sufficiently accurate to identify a warhead from a gamma-ray image without divulging secret information.

We expect a successful project will establish the utility of a new type of gamma-ray imaging system for future arms-reduction treaty applications. This will be performed by determining the efficacy of a set of metrics derived from gamma-ray images, such as the density and isotopic profile of materials that are used in warhead construction, which is a new concept that has not been realized for objects such as a nuclear warhead. In addition, we intend to develop a prototype gamma-ray imager design.

Mission Relevance

This research supports the central Laboratory strategic missions of ensuring national security. Specifically, the concept will assist in reducing the threat of nuclear attack by a rogue state or terrorists through technologies that support nuclear nonproliferation treaties.

FY13 Accomplishments and Results

In FY13 we (1) optimized filtering of two-dimensional gamma-ray images using a two-step threshold method to account for low-statistics pixels and obscured views, which resulted in a significant improvement in three-dimensional image-reconstruction error; (2) modified the three-dimensional image reconstruction algorithm to handle multiple, noncontiguous radioactive objects by moving from a centralized ray-based reconstruction to a space-carving technique employing three-dimensional scan conversion; and (3) developed a conceptual design using the MCNP Monte Carlo transport code for simulating nuclear processes of a two-sensor and coded-aperture gamma-ray imager that has a much higher photo peak and imaging efficiency compared to previous imaging sensors.

Project Summary

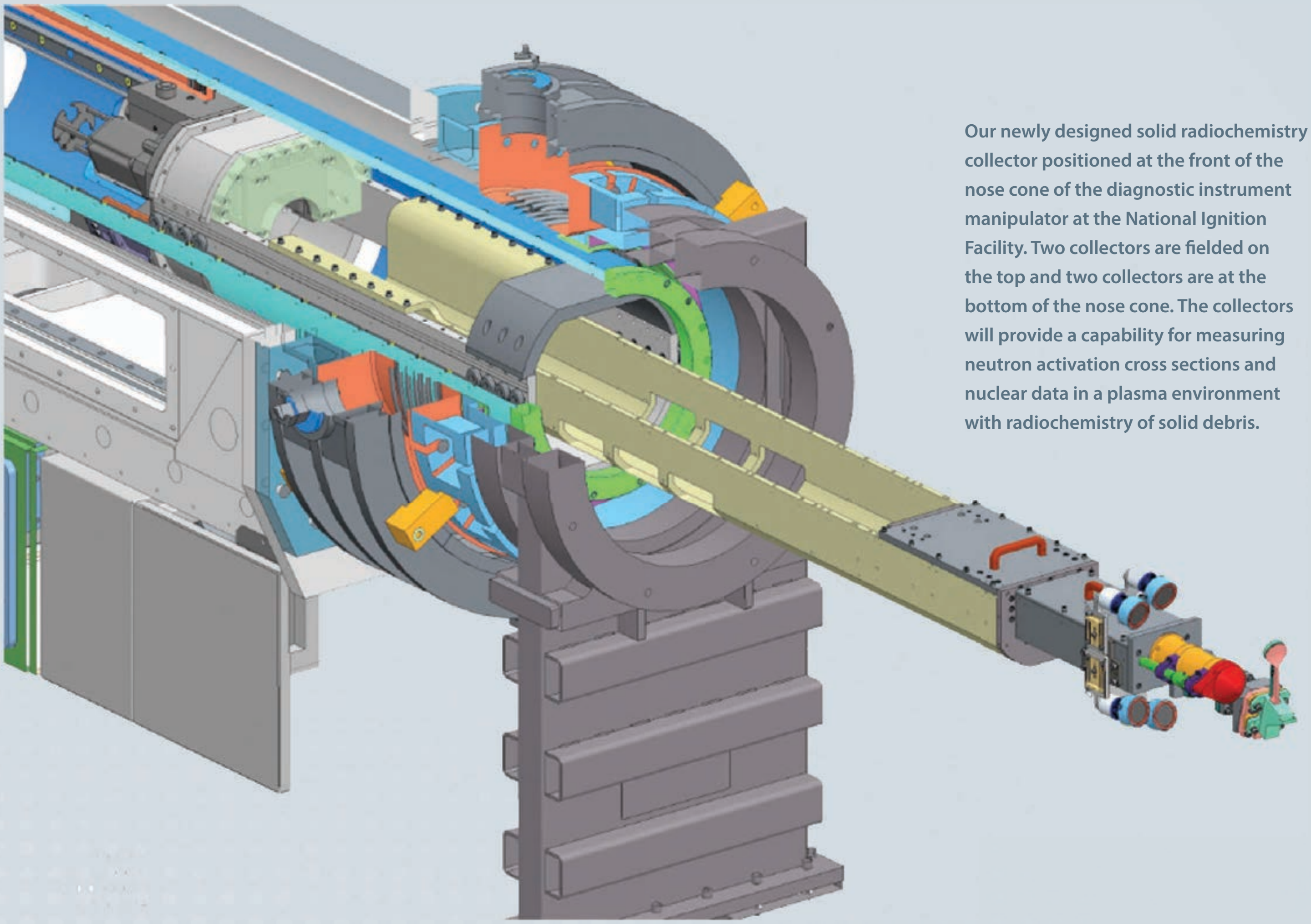
The goal of this project was to establish the efficacy of gamma-ray imaging for use in arms control by (1) simulating the radiation transport of multiple, noncontiguous radioactive objects through shielding material into a coded-aperture gamma-ray imaging system; (2) reconstructing the three-dimensional arrangement of the objects from a set of two-dimensional gamma-ray images; and (3) quantifying the reconstruction error based on the size and relative distance of the objects, the number of camera views, and with the assumption of symmetry to minimize the number of camera views. We demonstrated with gamma-ray imaging, using as little as three images, the ability to accurately reconstruct multiple radioactive objects and determine size and relative distances to 1-cm accuracy. The new image reconstruction code base is ready to be tested on multiple, noncontiguous radioactive objects in a laboratory or field measurement. Our next step is to prepare a successful proposal to acquire the necessary gamma-ray images and quantify the image reconstruction of these objects.

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 the National Ignition Facility (NIF) in high-compression deuterium–tritium capsules. They have been observed through radioactive gold isotopes produced during neutron interactions in the hohlraum cylinder housing the target capsule. Kiloelectronvolt-neutron reactions have been modeled with uncertainties up to 300%, and experiments at NIF offer a unique opportunity to measure these quantities. This project builds on the existing solid-debris radiochemistry diagnostic at NIF. Metal atoms will be added to the hohlraum or



Our newly designed solid radiochemistry collector 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.

incorporated into the capsule ablator. 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 target material into the NIF capsule assembly and hohlraum, and (3) measure neutron capture cross sections on 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 target 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, the availability of measurements of neutron activation cross sections will significantly improve the ability to interpret archival radiochemical data from nuclear tests. This work also helps to promote growth in several Laboratory core competencies, most notably radiochemistry and nuclear science, extreme measurements, and materials properties.

FY13 Accomplishments and Results

In FY13 we (1) completed designs for fielding solid-debris radiochemistry collectors within the target chamber, which includes the capability to place collectors at three different distances from the center of the target chamber (the collectors themselves will be fabricated and deployed in FY14); (2) identified alternate locations for the collection hardware in the NIF chamber, including possible dedicated lines of site; (3) began design of a large-area collector with at least ten times the collection area and the ability to field collectors at different distances; (4) identified two hohlraum platforms for fielding material on the outside of the hohlraum for nuclear measurements, which allows up to 300 mg of material to be used and enough for a 10% measurement—this included design of foil placement; and (5) created a target doping apparatus that is a duplicate of the one currently used for adding foams to capsules, but with the ability to handle radioactive materials. This system will be tested next fiscal year and capsules have already been obtained for testing purposes.

Proposed Work for FY14

In FY14 we will (1) field collectors on the diagnostic instrument manipulator, (2) complete design of a large-area collector for the diagnostic instrument manipulator or other identified location that has the capability for fielding at different distances from the target chamber center, (3) fabricate at least two hohlraums with stainless steel for use with deuterium–tritium capsules, (4) schedule shot dates for at least two hohlraum material shots, and (5) accelerate adding material to the capsule ablator with a high atomic-number material.

Publications and Presentations

Shaughnessy, D. A., et al., 2012. *Radiochemical measurements of neutron capture products at the National Ignition Facility.* Nuclear Physics Processes in Dynamic High Energy Plasmas, London, United Kingdom, Oct. 15–17, 2012. LLNL-ABS-573456.

Dynamic Predictive Analytics Approach to Comprehensive Nuclear Forensic Analysis

Simon Labov (13-ERD-062)

Abstract

New techniques are needed to improve the speed and accuracy of post-detonation nuclear forensic analysis. Determining design information from measurements is challenging even when the weapon performance, measurement conditions, emplacement configurations, and environmental factors are known. A current Laboratory challenge is to develop analysis tools that combine some subsets of the measurements under a limited range of conditions. We propose an exploratory effort to take all the measurements and data into account with all the variations that are likely to be encountered, to provide a range of new analysis capabilities to assist in deducing weapon design information and the optimal design of measurement systems. This will require a unique intertwining of several physics-based analysis techniques with machine-learning application development (programming that enables systems to automatically learn and improve). We will design our framework to enable automated analysis usable by non-experts, yielding a quick assessment and confidence in that assessment, and also to provide detailed explanatory information to assist nuclear forensic experts in analyzing and optimizing measurement and response plans.

Our analysis framework will provide five capabilities to assist in post-detonation nuclear forensic analysis including (1) classification probabilities, including broad and narrow weapon-design classes; (2) quantitative evaluation of confidence of the classification; (3) projection of predicted classification confidence over time based on typical data collection, and how that will change if various measurements are accelerated or delayed; (4) explanation of how conclusions were reached, including estimates of performance factors and emplacement parameters, which measurements were most critical to the current assessment, and which additional measurements are most likely to improve confidence; and (5) assessment of the impact, capabilities, and optimal implementation of current and future measurement systems.

Mission Relevance

Development of more post-detonation nuclear forensic analysis capabilities is a high priority in the Laboratory's strategic mission focus area of nuclear counterterrorism and forensics. The proposed research is designed to provide a physics-based, dynamic machine-learning capability to analyze a wide range of post-detonation nuclear forensics measurements and information to extract weapons design and performance information.

FY13 Accomplishments and Results

In FY13 we (1) obtained more than 3,000 samples of nuclear detonation outputs from detailed models created for other projects; (2) modeled the propagation of gamma-ray, seismic, and acoustic signals to distant sensors and used a number of techniques to estimate the accuracy and errors in the measurements; (3) began developing tools to estimate the reaction history, yield, and placement from these measurements; (4) established coordination with other projects to provide a similar capability for additional signal types; (5) performed preliminary feature extraction and machine-learning analysis; and (6) demonstrated that source-type classification can be conducted with even a crude estimate of the gamma-ray emission evolution.

Proposed Work for FY14

In FY14 we will (1) complete algorithms to reduce multiple gamma-ray measurements from multiple positions into a single-reaction history profile and a feature vector for machine learning; (2) use each of the 3,000 detonation output samples to create possible detector measurements with different source emplacements and detector locations; (3) create feature vectors, including uncertainties, for seismic, acoustic, and gamma-ray measurements, with optical and electromagnetic pulse measurements added as they become available; (4) use machine learning to create predictive models that include the impact of measured uncertainties; and (5) develop dynamic machine learning by creating a sequence that indicates the features available at specific times after detonation.

First Measurement of Low-Energy Nuclear Recoils in Liquid Argon—Enabling a New Technique for Nuclear Reactor Monitoring

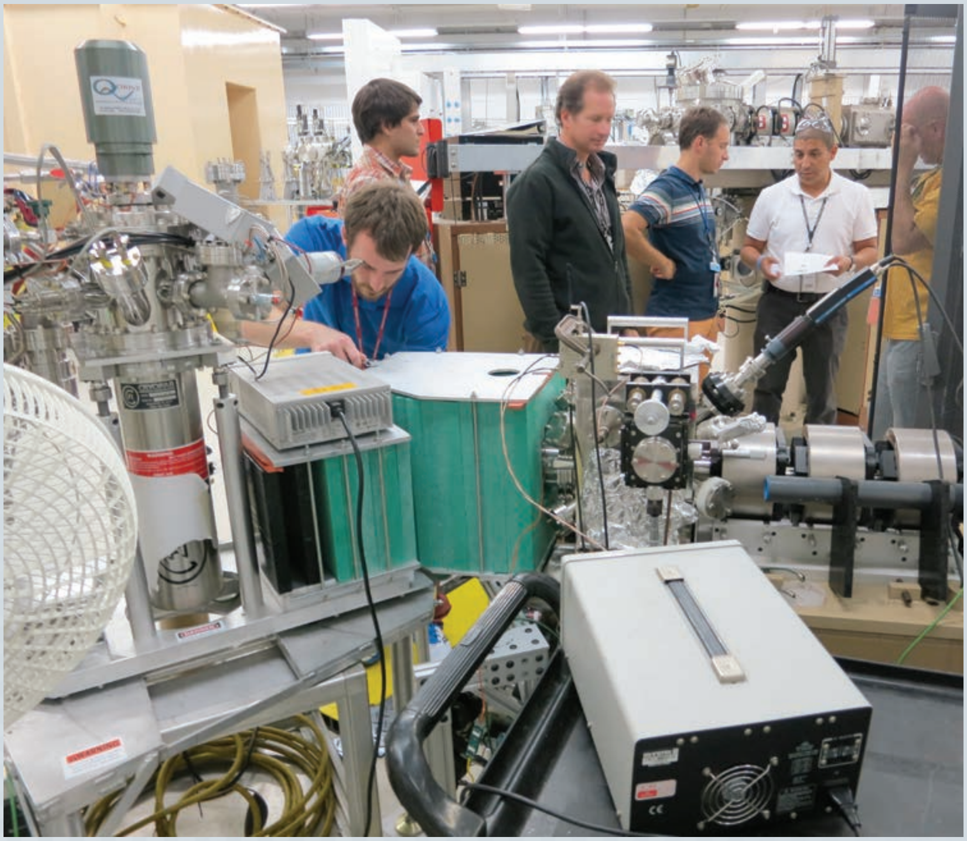
Peter Sorensen (13-FS-005)

Abstract

A new method of remote nuclear reactor monitoring, using particle recoils of coherent neutrino nucleus scattering, is well underway. In coherent neutrino scattering, a neutrino interacts coherently with all the nucleons in a target nucleus, transferring a small amount of its kinetic energy to the nucleus before continuing on its way. This happens much more often than inverse beta decay in which the neutrino collision with a nucleon results in a complete energy transfer and the neutrino is destroyed. The appeal of utilizing coherent neutrino nucleus scattering is that the interaction cross section is calculated to be about a hundredfold larger than that for inverse beta decay used in current neutrino detectors. This could enable significantly increased detector

sensitivity, a smaller (possibly portable) detector, or both. However, to successfully use coherent neutrino nucleus scattering for remote reactor monitoring, a critical first step is demonstrating sensitivity to the very-low-energy (sub-kiloelectronvolt) nuclear recoils that will result from this process. We propose to explore the feasibility of measuring the quenching factor of nuclear recoils in liquid argon. Knowledge of this quenching factor would enable the higher sensitivity reactor measurements as well as related experiments that aim to detect galactic particle dark matter.

We expect to obtain the first measurement of the quenching factor for ionization in liquid argon. We plan to accomplish this through measurement of the number of ionized electrons that result from 7-keV argon recoils, which will be created by the elastic scattering of 73-keV neutrons in argon. We intend to perform the research at Lawrence Livermore's Center for Accelerator Mass Spectrometry. In addition to the possibility of a new remote nuclear reactor detection technique, there is a strong basic science motivation for this work. The low-energy nuclear recoils characteristic of coherent neutrino nucleus scattering are very similar to the expected signal from elastic scattering of galactic particle dark matter. The search for direct proof of the existence of dark matter is a central outstanding problem in modern cosmology and particle physics. A coherent neutrino nucleus scattering detector, though not designed as such, has the potential to perform a very sensitive search for dark matter.



Researchers review a liquid argon scintillation detector installation at Livermore's Center for Accelerator Mass Spectrometry in an effort to obtain the first measurement of the quenching factor for ionization in liquid argon created by the elastic scattering of 73-keV neutrons in argon. Ultimately, the effort may enable higher sensitivity in nuclear reactor measurements as well as related experiments that aim to detect galactic particle dark matter.

Mission Relevance

Our results will be an important step toward deploying a liquid argon detector for remote reactor monitoring in support of the Laboratory's core mission of limiting or preventing the spread of materials, technology, and expertise relating to weapons of mass destruction.

FY13 Accomplishments and Results

In FY13 we (1) completed construction of a novel source of neutrons at the LLNL Center for Accelerator Mass Spectrometry and verified that it generates a useful flux of quasi monochromatic neutrons; (2) deployed a small 100-g liquid argon proportional scintillation detector at this neutron source, which was operated to provide primarily 70-keV neutrons; (3) made a first demonstration of the sensitivity of liquid argon to such low-energy neutrons, which deposit a maximum of 6.7 keV in the detector; and (4) measured the number of electrons extracted from the interaction vertex as a function of the applied electric field.

Project Summary

The successful conclusion of this feasibility study resulted in a demonstration of a highly useful source of neutrons for characterizing rare-event search detectors. The next steps include further optimization of the neutron source, exploration of different notch filters (to provide different neutron energies), and study of the response of our liquid argon detector to lower-energy neutrons. In particular, we would like to demonstrate sensitivity to neutrons with energies as small as 10 keV, which would be useful for a range of applications from detection of special nuclear materials to reactor monitoring.

Publications and Presentations

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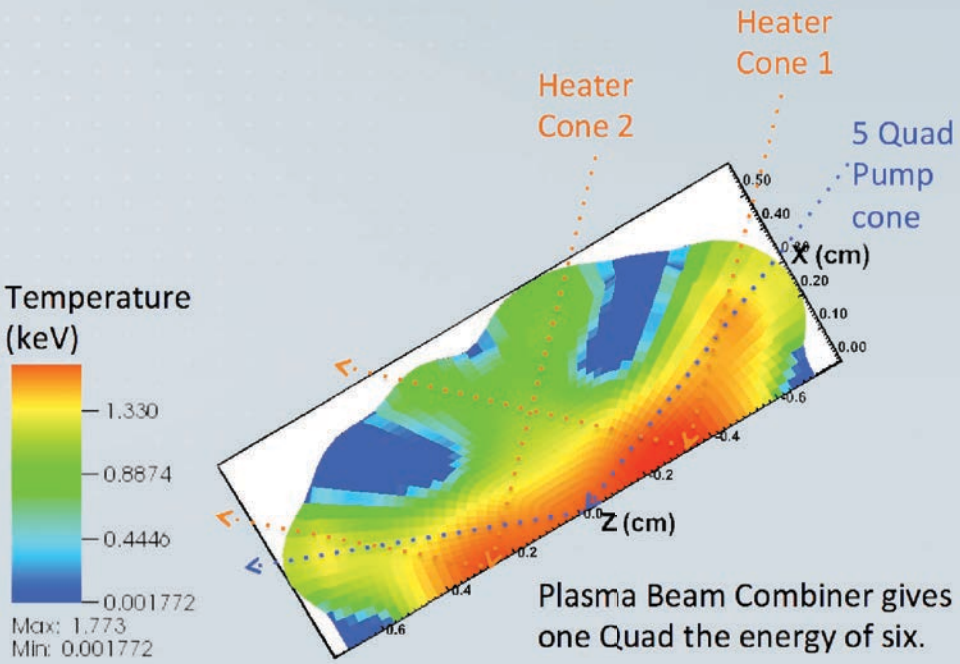
Demonstrating a Laser Beam Combiner for Simulating High-Yield Nuclear Events

Robert Kirkwood (13-FS-008)

Abstract

The Energy-Partitioning Energy-Coupling (EPEC) program is simulating the blast and shock produced by nuclear events with a single quad of laser beams (group of four beams) at Livermore's National Ignition Facility (NIF). We propose to test and perform simulations to examine the feasibility of a beam combiner designed to increase the available energy range for EPEC and allow events as large as 10 kt to be simulated. We propose to combine beams at best focus via scattering from stimulated ion waves in a



A 6-quad beam combiner (equivalent to 24 individual laser beams) designed to enhance experiments for simulating the blast and shock from nuclear events at Livermore's National Ignition Facility.

plasma, which is an adaptation of the ion-wave beam process presently in use for the NIF ignition target. We intend to experimentally test the first beam combiner with six quads combined into a single beam incident on a plate target.

We expect our target design, which will combine multiple quads of NIF beams into a single beam, will allow simulation of nuclear events up to 10 kt, making the capability applicable to a much broader set of problems. A larger energy range will enable an expanded data suite for use with more accurate weapons physics codes. We intend to finalize the target design of a six-quad combiner and perform a demonstration experiment to optimize the beam combiner. In addition, we will perform simulations of future EPEC experiments at high energy and develop a concept for operating the EPEC chamber at higher energy.

Mission Relevance

The EPEC six-quad laser-beam combiner is expected to enable a wide range of weapons effects and high-energy-density science experiments, as well as potential applications for bright back lighters, in support of the Laboratory's central mission in stockpile stewardship science and the strategic science, technology, and engineering foundation in controlling fusion and high-energy-density matter.

FY13 Accomplishments and Results

In FY13 we (1) successfully prepared the motivation and shot plan for a demonstration of the beam combiner at NIF, including an initial and scientific review—our proposal was identified as having sufficient scientific merit and feasibility to receive NIF shots;

(2) studied target details, including designing witness plates and diagnostic setups to measure the spot size and energy of amplified and pumping beams; (3) evaluated the feasibility of manufacturing the gas balloon target at Luxel in Friday Harbor, Washington—which supports x-ray and extreme ultraviolet research programs—and acquired a target; and (4) performed simulations of optical signals from EPEC targets emulating 10-kt events.

Project Summary

We have successfully prepared the NIF shot plan, obtained appropriate approvals, and specified the targets for three shots to demonstrate a beam combiner capable of performing EPEC experiments that emulate devices with a yield of about 10 kt. We designed the diagnostic witness plates and instrument setups to allow the amplified beam energy and spot size to be determined from NIF standard x-ray diagnostics, and completed the needed reviews to be allocated NIF shots. The redirection of NIF activities in the summer of 2013 did not give our demonstration shot priority, and our remaining resources were used to perform simulations of experiments that can study critical optical outputs of devices in the 0.1- to 10.0-kt range in support of nuclear forensics and nuclear detection missions.

Publications and Presentations

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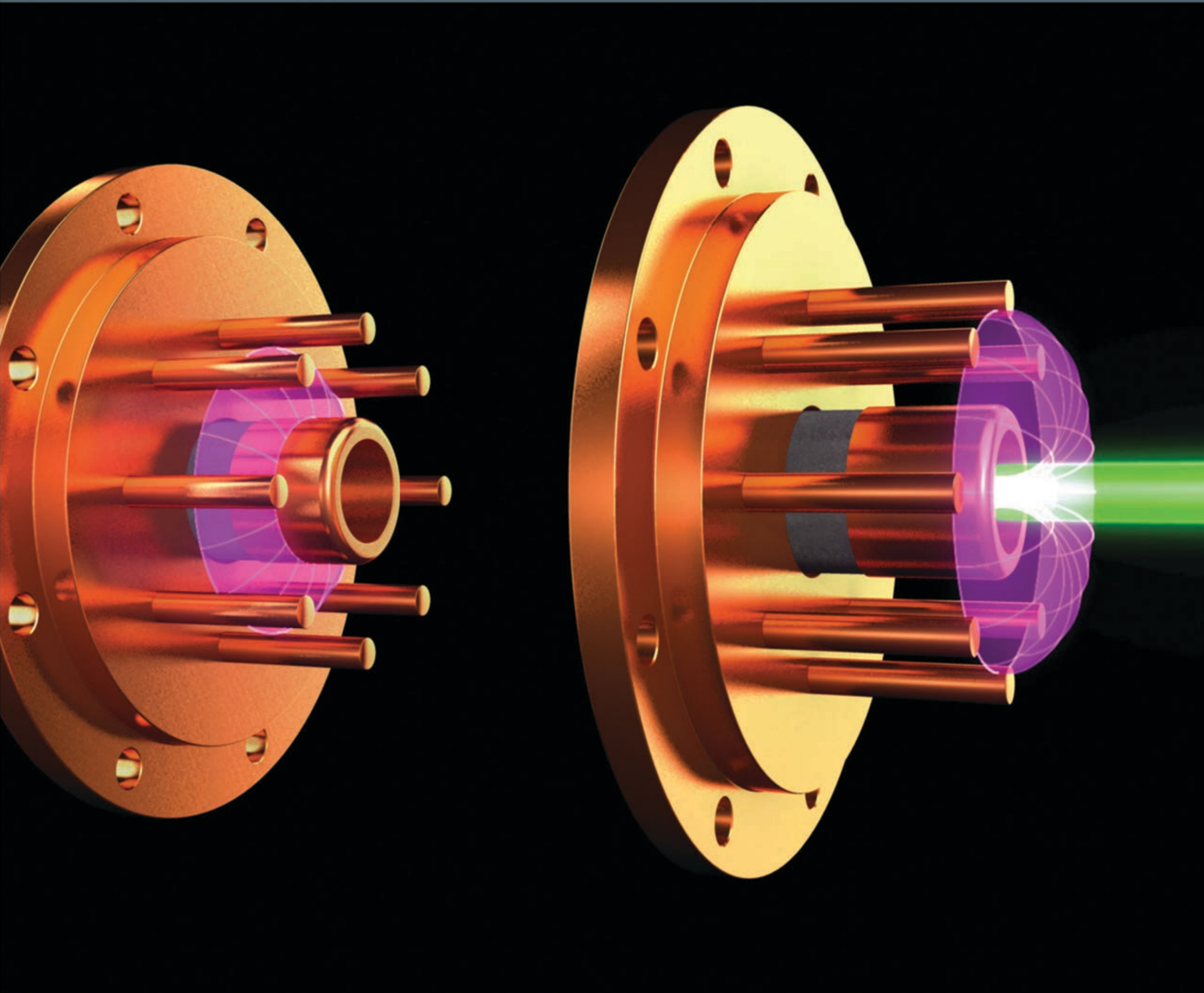
Physics



Laboratory Directed Research and Development

Annual Report FY2013

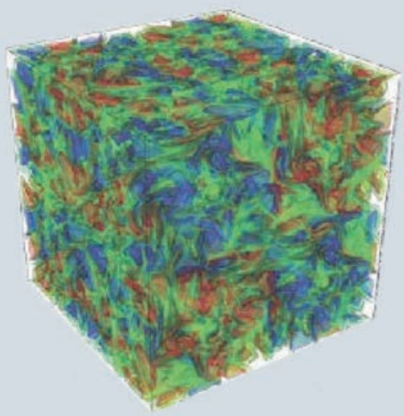
Multiscale Polymer Flows and Drag Reduction

Todd Weisgraber (10-ERD-057)

Abstract

Reducing drag in bounded turbulent flows by adding long-chain polymers is a well-established phenomenon. However, despite decades of research, the fundamental mechanisms of drag reduction are not adequately understood. We believe that a complete description of the coupled polymer and flow dynamics for drag reduction must incorporate wall roughness, a coarse-grained molecular representation of the polymer, and hydrodynamic fluctuations at the length scale of the polymer. We propose to develop new algorithms—including an unconditional, fluctuating lattice-Boltzmann solver coupled with molecular dynamics—to enable fully turbulent, multiscale simulations of drag reduction that will increase our understanding of the underlying physics.

We expect to perform a series of large-scale simulations to provide valuable insight into a fundamental hydrodynamics problem, including a more detailed understanding of the complex dynamics of polymers and turbulence. Additional knowledge of drag reduction could conceivably make it possible to optimize this effect in more practical applications. A modest decrease in drag would dramatically improve the deployment efficiency of naval vessels and reduce expenditures in the civilian sector. In addition to the fundamental science, we will also develop a high-performance computing capability applicable to biosecurity and Rayleigh–Taylor instability research.



A simulation, conducted with 1,500 processors, of a vortex structure in a turbulent flow as part of our research into the fundamental mechanisms of drag reduction.

Mission Relevance

Our research will advance the high-performance computing and simulation foundations with which the Laboratory can develop new, mission-relevant capabilities. For instance, we will employ Livermore's high-performance computing to address fundamental scientific questions in hydrodynamics. The interaction between flow and polymer physics is also relevant to developing the next generation of pathogen detection and analysis systems, which are part of the LLNL mission focus area of biosecurity.

FY13 Accomplishments and Results

To close out the project, in FY13 we (1) demonstrated that a single roughness element with an amplitude one order of magnitude smaller than previously studied can initiate turbulence, (2) determined that the disturbances initiated by the wall evolve into large-scale parallel vortices whose interaction destabilizes the flow and generates turbulence, and (3) incorporated hydrodynamic fluctuations into the adaptive mesh framework.

Project Summary

This project resulted in a novel code for fluid flow simulation based on the lattice-Boltzmann method with improved algorithms for adaptive mesh refinement. We demonstrated that our approach, which provided rigorous conservation of mass and momentum, produced smaller errors and converged to second order on all levels of refinement when compared to other mesh-refinement algorithms. With this capability, we applied the code to long-standing problems in turbulent flows. Specifically, we simulated the onset of turbulence and demonstrated for the first time that small-amplitude wall topology can initiate turbulence without any other flow perturbations. We also showed that the wavelength of flow structures induced by the wall is a function of the Reynolds number, which characterizes the degree of turbulent flow, and these structures ultimately destabilize the flow and generate turbulence. Our work has attracted the attention of a potential industrial sponsor interested in simulating high-Reynolds-number flow through nuclear fuel rod arrays, and we anticipate establishing a work-for-others contract in the near future.

Publications and Presentations

Guzik, S. M., et al., 2012. *Interpolation methods and the accuracy of lattice-Boltzmann mesh refinement*. LLNL-JRNL-594615.

Weisgraber, T. H., and B. J. Alder, 2012. *Computer experiments on the onset of turbulence.* 28th Intl. Symp. Rarified Gas Dynamics, Zaragoza, Spain, July 9–13, 2012. LLNL-CONF-575632.

Weisgraber, T., and B. J. Alder, 2010. *Polymer flows in channels with roughened walls.* 19th Intl. Conf. Discrete Simulation of Fluid Dynamics (DSFD 2010), Rome, Italy, July 5–9, 2010. LLNL-PRES-439178.

Fundamental Research in Advanced Quantum Simulation Algorithms

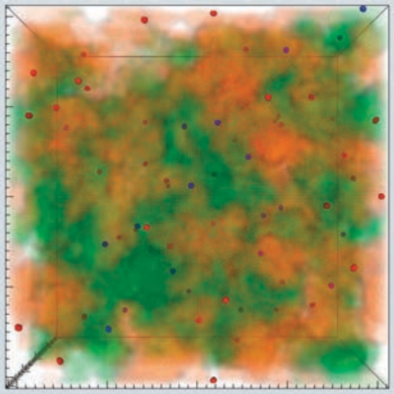
Jonathan DuBois (10-ERD-058)

Abstract

First-principles simulation of materials properties has played an essential role in Livermore's science and technology for the last half-century. Mission-critical applications range from quantum chemistry to plasma physics. Broadly, the most significant challenge for these simulations today is the ability to efficiently measure and reduce systematic errors. While first-principles simulations continue to be applied, current bottlenecks will ultimately limit their usefulness, regardless of available computing power. We propose a research program aimed at estimating and reducing systematic errors within the framework of ground-state and finite-temperature quantum Monte Carlo codes, in which results are calculated from repeated random sampling.

We intend to develop a robust tool set based on released-node quantum Monte Carlo for estimating the systematic error introduced by fixed-node approximation for large

systems, and thus increase the accuracy of state-of-the art calculations by two orders of magnitude, so that the typical error lies within the chemical accuracy bound. We will also develop a constant-pressure diffusion Monte Carlo algorithm in which the proton and electron degrees of freedom, along with the cell degrees of freedom, can adjust. Finally, we will implement several novel enhancements to the standard path-integral quantum Monte Carlo method using the knowledge garnered in ground-state quantum Monte Carlo calculations to extend the path-integral quantum method to lower temperatures.



Snapshot of a full many-body path-integral quantum Monte Carlo simulation of a strongly coupled degenerate plasma mixture of lithium (red spheres) and hydrogen (blue spheres). The instantaneous delocalized electron distributions are represented as orange (spin up) and green (spin down) point clouds. This calculation is representative of a new first-principles simulation capability.

Mission Relevance

Accurate first-principles simulations of materials are essential for current and future LLNL goals for materials on demand, uncertainty quantification, and stockpile stewardship science, especially where experiments are not feasible. Currently available methods are unable to meet these demands with acceptable accuracy. Development of a high-accuracy first-principles simulation capability is therefore well aligned with the Laboratory's strategic missions.

FY13 Accomplishments and Results

In FY13 we (1) completed work on the finite-temperature homogeneous electron gas; (2) continued work on a high-fidelity treatment of the low-temperature properties of hydrogen; (3) continued work on the formulation of a general simulation approach to exact finite-temperature, many-body fermion systems capable of efficiently treating inhomogeneous systems, focusing efforts on exact treatment of low-atomic-number ions at finite temperature; and (4) developed and tested a mathematical expression for optimizing nodal constraints in these systems.

Project Summary

This project resulted in several significant technical advances in the exact path-integral quantum Monte Carlo treatment of materials properties in extreme environments. A significant highlight is the first exact calculation of the exchange correlation energy of the homogeneous electron gas as a function of temperature and density. This benchmark result will be essential for development of accurate temperature-dependent density functionals for simulation of materials in the regime of warm dense matter. Additional fundamental developments provided for derivation and application of a method for reducing the computational cost of evaluating the exact fermion partition function for many-body quantum systems from exponential to polynomial scaling for a wide class of systems. Much of the algorithmic and code infrastructure developed in the course of this work has already resulted in contributions to programmatic efforts, including development of new program focus areas.

Publications and Presentations

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Dubois, J. L., D. Aberg, and V. Lordi, 2010. *Diffusion Monte Carlo calculations of formation energies of charged self interstitial defects in silicon.* APS March Mtg. 2010, Portland, OR, Mar. 15–20, 2010. LLNL-PRES-426109.

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Morales, M. A., et al., 2013. "Nuclear quantum effects and nonlocal exchange-correlation functionals in liquid hydrogen at high pressure." *Phys. Rev. Lett.* **110**, 065702. LLNL-JRNL-578753.

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Coherence-Preserving X-Ray Adaptive Optics

Lisa Poyneer (11-ERD-015)

Abstract
We propose to conceive, design, build, test, and optimize the first prototype grazing-incidence, adaptive x-ray optics suitable for use at high-intensity, high-coherence DOE light sources such as the Linac Coherent Light Source at the SLAC National Accelerator Laboratory and the National Synchrotron Light Source II at Brookhaven National Laboratory. This new class of x-ray optics will unleash the full scientific potential of these new national facilities. Our research builds on recent LLNL efforts to develop the primary x-ray mirror systems for the Linac Coherent Light Source and the adaptive-optic-based Gemini Planet Imager to be initially deployed on the Gemini South telescope in the Chilean Andes. Livermore's acknowledged world leadership in extreme adaptive optics with state-of-the-art design, coupled with fabrication expertise developed for the Linac Coherent Light Source, provides the foundation necessary for our novel research in adaptive x-ray optics.

We expect to produce a prototype system, including the x-ray adaptive optic, a wave-front sensor, control and sensing algorithms, and a detailed performance model. It will be developed and tested at an existing light source. This research will establish a national capability that all DOE facilities and labs can draw upon to build new beam lines or end stations. By enabling delivery of coherent or nanometer-scale focused x-rays, this research will enable advances in physics, chemistry, and biology. If prototype systems can be developed and proven effective, this pioneering research could serve as the basis for many instruments at future facilities that require x-ray imaging.

Mission Relevance
This research supports the Laboratory's foundations in energy manipulation and advanced measurements, enabling fundamental scientific discoveries in diverse areas relevant to LLNL and DOE. In particular, DOE is investing heavily in next-generation

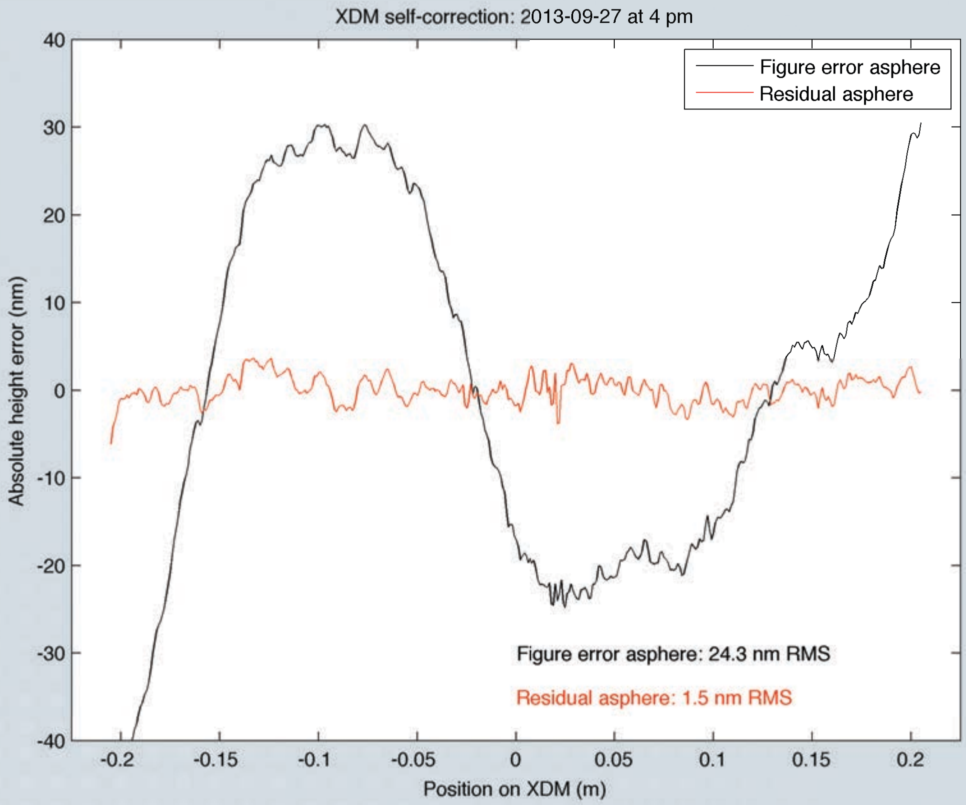
x-ray light sources with unprecedented brilliance and coherence. Revolutionary advances in the quality of x-ray optics will be required to take full advantage of these capabilities. Adaptive x-ray optics offers one possible path for meeting the extremely challenging engineering specifications.

FY13 Accomplishments and Results

In FY13 we completed fabrication of our x-ray deformable mirror, a 45-cm-long super-polished single-crystal silicon bar with 45 actuators along its length to control the surface figure at centimeter spacing. In addition, we calibrated the mirror using high-precision visible-light metrology and flattened it to 1.5-nm root-mean-square height figure error. Unfortunately, because of time constraints, we were not able to perform at-wavelength testing at a light source.

Project Summary

Our central achievement was the successful development of our x-ray deformable mirror with 7 internal temperature sensors and 45 strain gauges to provide position feedback. The mirror was designed to be flattened to a higher order than the existing hard x-ray offset mirrors at the Linac Coherent Light Source, and our demonstrated flattening is as good or better than three of those four mirrors. We have not only demonstrated that our fundamental technological approach is sound, but we have also done so with an x-ray deformable mirror that is significantly longer and with more actuators than the handful of extant mirrors. An x-ray deformable mirror requires



We have flattened the surface of our 45-cm x-ray deformable mirror to just 1.5-nm root-mean-square height error (red). Though originally polished to 3.5-nm root-mean-square, the actuator bonding process resulted in 24-nm root-mean-square height error (black). Flattened surface quality is as good as or better than three of the four hard x-ray offset mirror system mirrors at the Linac Coherent Light Source at the SLAC National Accelerator Laboratory. Results are absolute height error as measured and stitched together with a calibrated 12-in. Zygo interferometer.

highly accurate models and sensors for proper control. However, important physical effects are not captured by commonly used ray-tracing simulations. We developed tools that use wave optics and correctly handle propagation of the electric field on the tilted surfaces of grazing-incidence x-ray optics. These simulations also include accurate models of wave-front sensors and our mirror. Using these tools, we have uncovered two previously unknown performance limitations for adaptive x-ray optics. We have found that for some common configurations, phase-amplitude mixing occurs—a simple phase-conjugation approach for control will not produce the best correction. We also found that the commonly used Hartmann sensor is highly susceptible to aliasing, which corrupts phase measurements.

Publications and Presentations

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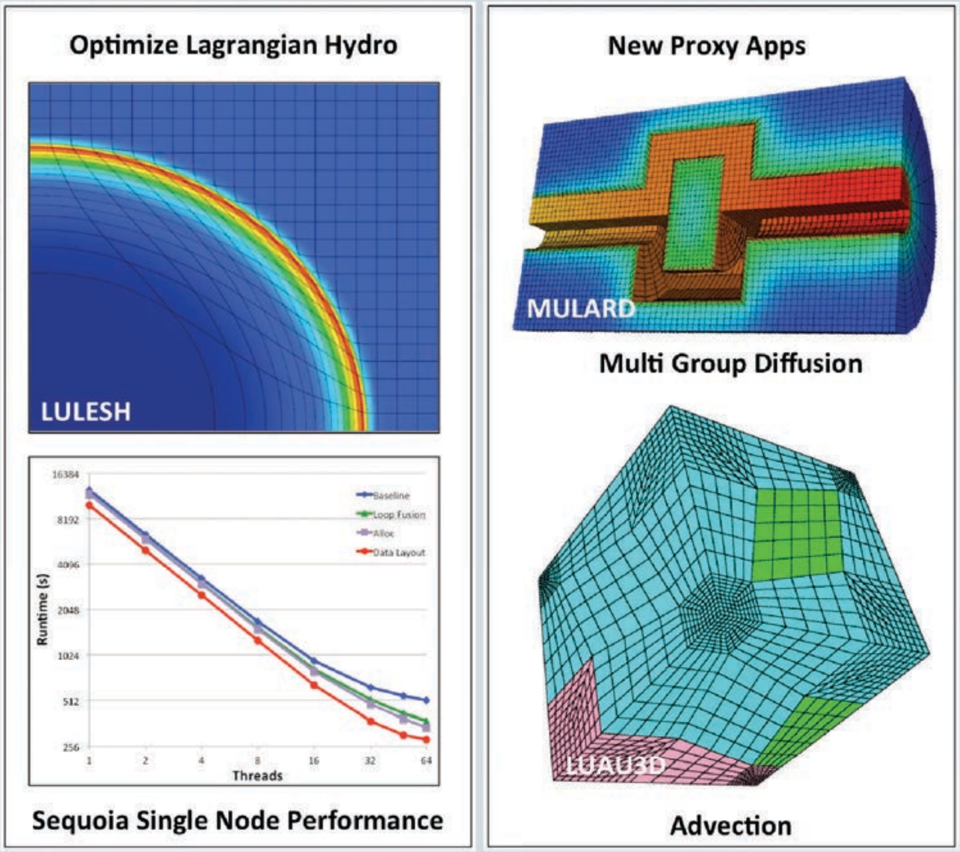
McCarville, T. J., M. J. Pivovaroff, and R. Soufli, 2011. *LCLS x-ray mirror measurements using a large aperture visible light interferometer.* ATOP11: 4th Workshop Adaptive and Active X-ray and XUV Optics, Oxfordshire, United Kingdom, Apr. 4–5, 2011. LLNL-ABS-510933.

Advanced Algorithm Technology for Exascale Multiphysics Simulations

Charles Still (11-ERD-017)

Abstract

We propose to develop computational algorithms that will enable multiphysics simulation codes to efficiently utilize exascale computers. Within the next decade, NNSA plans to site an exascale computer at LLNL with a thousandfold performance increase and only a tenfold increase in the electricity requirement. The system architecture is a dramatic departure from hardware of today, and codes will have to



We delivered an optimized Lagrangian hydrodynamics proxy application (LULESH) and new proxy applications for multigroup radiation diffusion (Mulard) and advection (LUAU3D) advanced simulation and computation codes.

change drastically to utilize it. We will characterize which multiphysics algorithms extend to exascale hardware, which do not, and how to determine this differentiation in general. If successful, this project will result in metrics and characterization techniques for others to use and an exascale-tuned hydrodynamics and diffusion reference code as proof of principle on exascale surrogate hardware.

Current multiphysics codes were designed to meet criteria that are very different from the demands of exascale systems. Power constraints and performance targets are driving those future systems to have more than a thousand cores per computational node, leading to restricted memory and message-passing bandwidth per node. Algorithms must exhibit significant intra-node concurrency while limiting data motion to take advantage of these systems. Using a coupled hydrodynamics–diffusion reference code as an example, we will develop computational algorithms that will enable multiphysics simulation codes to efficiently utilize exascale machines.

Mission Relevance

There is a pressing need to ensure that Livermore's current suite of advanced simulation and computation codes for defense applications can successfully migrate to the exascale computers coming to LLNL. Our research will provide the performance metrics and methodology needed to transition multiphysics codes to exascale systems, along with a coupled arbitrary Lagrangian–Eulerian hydrodynamics and diffusion reference code. This work furthers the Laboratory's stockpile stewardship science mission and will bolster Livermore's scientific and technological foundation in high-performance computing and simulation.

FY13 Accomplishments and Results

In FY13 we (1) explored metrics identifying characteristics for memory motion, instruction mix, single-instruction multiple-data vectorization, and thread-level parallelism; (2) delivered the proxy applications Mulard (an application that solves the multigroup diffusion equations coupled to a material energy equation) and LUAU3D (material advection on an unstructured hexahedral mesh), which complement the Lagrangian hydrodynamics proxy-application code, LULESH; (3) created code and data transformations applicable to optimizing hydrodynamics, advection, and diffusion using the metrics identified; and (4) developed a lessons-learned document that will guide node-focused optimizations in radiation-hydrodynamics applications. Research into programming models led to a best-paper award at the 27th International Parallel and Distributed Processing Symposium sponsored by the Institute of Electrical and Electronics Engineers ("Exploring Traditional and Emerging Parallel Programming Models using a Proxy Application").

Project Summary

The successful completion of this project resulted in (1) the identification of metrics for obtaining improved efficiency and performance on future computer architectures; (2) code and data transformations (based on fine-grained parallelism techniques like loop-level threading) that will enable current arbitrary Lagrangian–Eulerian hydrodynamics and radiation diffusion methods to obtain higher performance; (3) the definition of an abstraction layer that would separate the computational science from the architectural details, thus enabling the transformation of large-scale applications codes; and (4) an understanding of the limitations that current methods have in exploiting future capabilities. As a result of this project, there are two avenues for future work. The first is to extend the results to physics problems not studied within this project, and to apply these lessons learned to large-scale applications codes, which will be carried out as part of Advanced Simulation and Computing programmatic work at Livermore. The second is to investigate the computational characteristics of higher-order arbitrary Lagrangian–Eulerian hydrodynamics and radiation diffusion using the methodology outlined in this project.

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Temperature-Dependent Lattice Dynamics and Stabilization of High-Temperature Phases from First-Principles Theory

Per Söderlind (11-ERD-033)

Abstract

We propose to extend our current capability in quantum-mechanical equation-of-state and materials investigations by developing and implementing a new finite-temperature self-consistent phonon calculation scheme for large-scale, first-principles electronic-structure calculations on massively parallel computer platforms. By combining state-of-the-art electronic structure theory with the proposed self-consistent phonon approach, we aim to dramatically improve our capability for predicting high-temperature phase diagrams of materials and provide a complementary approach to the more time-consuming quantum molecular dynamics simulation method. Specifically, we will focus on the temperature-induced body-centered-cubic phases all actinide elements adopt prior to melt.

Our results will predict and explain the occurrence of the body-centered-cubic crystal structure as the prevalent melting phase in many metallic systems, including the actinides. This will be of significance because, for the first time, the high-temperature portion of the phase diagrams for actinides (and other metals) can be computed from first principles with the reliability that already exists for the low-temperature region. Our results are important for fundamental scientific reasons and are also an extremely important technical component in describing equation-of-state properties that are now very difficult to assess, either with current theory or experiment. Hence, our capability to address an essential region of many metals phase diagrams can be improved dramatically with this research.

Mission Relevance

The properties of actinides and other metals must be well understood and predictable to ensure reliability of the nation's nuclear stockpile. Here we present a new idea that, together with our current expertise and capabilities, can substantially improve the reliability of our actinide phase models—particularly at high temperatures—relevant to LLNL's mission in stockpile stewardship science.

FY13 Accomplishments and Results

In FY13 we (1) concluded two exhaustive studies of alloy systems, the titanium–vanadium system and a high-pressure vanadium metal and vanadium–chromium alloy system; (2) took advantage of the technique to calculate phonons for determination of elastic constants, which are important for strength models; and (3) used this approach to calculate elastic constants for uranium metal under high pressure and temperature. We have fulfilled our goals in that we have established a new approach, validated it, and proved its efficacy for studying phase stability and computing elastic constants. These properties are fundamental for equation-of-state and strength programs.

Project Summary

With this project, we have calculated body-centered-cubic phonon dispersions for uranium metal that compares well with existing experimental phonon density of states. No such property has previously been calculated from first principles for a high-temperature phase of an actinide metal. We have further expanded the technique to include studies of alloy systems. We have shown that the body-centered-cubic stability phase line in the titanium–vanadium system can be reproduced by our models. Also, we have applied the scheme to high-pressure vanadium metal at pressures where the well-known rhombohedral phase is stable. We show that very high temperatures are required to remove the rhombohedral distortion, restoring the body-centered-cubic phase. We have furthermore applied the methodology to calculate high-temperature and high-pressure elastic constants for uranium and molybdenum metals. Elastic constants can otherwise only be obtained from very computationally intense, quantum molecular-dynamics simulations that are also fraught with technical challenges. Lastly, we have utilized the computation of forces, required for the methodology, for interatomic potential developments.

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Dynamics of Ultrafast Heated Matter

Tilo Doeppner (11-ERD-050)

Abstract

We propose to use x-ray Thomson-scattering techniques to measure the kinetics of novel materials created in the high-energy-density regime. Working from first principles, we will determine the properties of matter heated by spherical compression, fast-particle beams, and ultrafast x-ray pulses based on data obtained from experiments at advanced laser facilities around the world. The microscopic physics data collected will be compared with observations of macroscopic quantities—size, velocity, compression, and temperature. This research can only be performed at facilities with combined ultrashort-pulse laser heating and probing capabilities. The experimental techniques as well as the results will benefit efforts at the National Ignition Facility (NIF) and the Laboratory as a whole by elucidating materials science, equation-of-state, and high-energy-density physics conditions. We will collaborate with three University of California campuses—Berkeley, San Diego, and Los Angeles.

We expect to characterize the dynamics of ultrafast heated matter over a broad range of heating mechanisms and timescales, ranging from implosion experiments at 500 ps down to ultrashort heating with a free-electron laser at 0.1 ps or less. Primary new findings will be direct measurements of temperatures, densities, and collective collisional effects with x-ray Thomson scattering. In the non-collective scattering regime, we will determine the temperature and density of free electrons, which in turn determine the width and absolute intensity of the scattering spectrum. We will also determine the collective (forward) scattering regime of the plasmon shift, which determines collective electron Langmuir oscillations and provides insight into the quantum correlations of dense matter.

Mission Relevance

Ultrafast heating of matter is an important phenomenon for understanding physical processes relevant to astrophysics and high-energy-density science Laboratory programs. This newly proposed application of x-ray scattering would provide data important to several LLNL missions. For instance, equation-of-state and collision processes are important for successful modeling of experiments on NIF, in support of the Laboratory's missions in stockpile stewardship, climate and energy, and ignition fusion energy.

FY13 Accomplishments and Results

In FY13 we (1) conducted high-energy-density experiments and used x-ray Thomson scattering to characterize states of matter, with highlights being the accurate measurement of the ion–ion correlation peak in about 10-eV aluminum, shock-compressed to threefold solid-state density at the OMEGA laser at the Laboratory for Laser Energetics in Rochester, New York, and subsequently with higher resolution in a series of experiments at the SLAC National Accelerator Laboratory in Menlo Park, California; (2) found that, in order to explain the shape of the angular dependence of the elastic scattering signal, an additional short-range repulsive potential has to be invoked in the models, which contributed to improving our understanding of modeling warm dense-matter states; and (3) continued our work on shock-compressed deuterium and isochorically heated hydrogen droplets, demonstrating the first high-energy-density free-electron laser pump–probe measurement.

Project Summary

We demonstrated the scaling of the ion–ion correlation peak in shock-heated aluminum as a function of mass density in single-shot experiments by correlating these measurements with spectrally resolved plasmon scattering as an independent density reference diagnostic at the SLAC National Accelerator Laboratory. This establishes a new, highly accurate and highly sensitive density diagnostic for high-energy-density plasmas. Our experiments have made an important contribution towards improving modeling of warm dense matter, in particular the inner atomic potential in dense aluminum, and improving the understanding of lowering the ionization potential in compressed carbon–hydrogen plastic from capsule implosion experiments on the OMEGA laser, which is highly relevant for modeling inertial-confinement fusion implosions on NIF. We hope to continue this research through collaborations with colleagues from SLAC; University of California, Berkeley; Laboratory for Laser Energetics; and the DESY national research center in Hamburg, Germany. One of the next challenges is to apply this method to discover novel phases—such as electrides, where electrons occupy interstitial atomic states—when matter transitions from the solid to the plasma state. In addition, we envision transferring these x-ray scattering techniques to characterize extremely high-density states of matter obtained in indirectly driven inertial- confinement fusion implosions at NIF.

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Astrophysical Collisionless Shock Generation by Laser-Driven Laboratory Experiments

Hye-Sook Park (11-ERD-054)

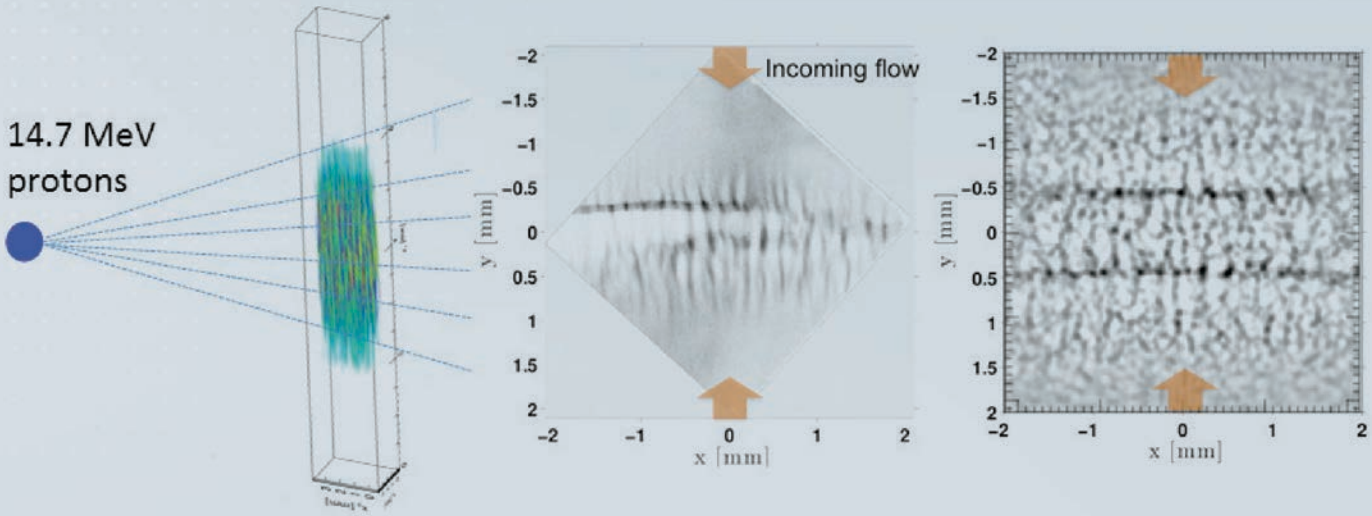
Abstract

The exact mechanism of ultrahigh-energy cosmic-ray creation is a longstanding mystery. Collisionless shocks are a possible mechanism for creating, through Weibel instability, magnetic fields that may reveal shock acceleration dynamics that amplify cosmic-ray particle energies beyond 10^{15} eV. We seek to observe and measure the creation and amplification of magnetic fields from collisionless shocks—specifically, high-Mach-number shocks created by the counter-streaming laser-produced plasmas associated with dynamic astrophysics processes. Using the OMEGA Laser Facility at the University of Rochester, we will develop a new experimental technique—applicable to LLNL's National Ignition Facility—to observe the creation of collisionless shocks and generation of magnetic fields.

If successful, this project will generate a wealth of knowledge about the physics of collisionless shock dynamics. Our observations and experimental results will be important for understanding plasma processes occurring in astrophysical collisionless shocks, such as the generation of magnetic fields and acceleration of cosmic-ray particles, which remain open questions of great interest in modern astronomy and astrophysics. The experiments we develop will also add new diagnostic capabilities for measuring magnetic fields, electron spectra, and the density and structure of plasmas' capabilities that will be applicable to advanced fusion-class laser experiments. This project will also attract the world's foremost experts in this field to participate in these experiments.

Mission Relevance

This project aligns well with LLNL's mission of cutting-edge science while creating expertise and understanding for follow-on applications of fusion-class laser facilities



Filamentary magnetic field structure observed in a counter-streaming plasma flow experiment. The field structure is visualized via protons generated by an imploding capsule filled with deuterium–tritium fuel. The experimental data (center) and three-dimensional particle-in-cell simulation and proton-ray trajectory from the experimental angle are shown at right. The magnetization (magnetic energy conversion from kinetic energy in the system) is about 1%. This observation has a significant role in understanding the mechanism of magnetic seed-field generation in astrophysics.

for the Laboratory's core mission in stockpile stewardship. In addition, the project will leverage investments in the National Ignition Facility to further missions in energy security.

FY13 Accomplishments and Results

In FY13 we (1) performed five experiments at the OMEGA laser facility to understand the interactions of high-velocity counter-streaming plasma flows under conditions that could produce collisionless shocks, (2) commissioned an imaging Thomson-scattering detector on OMEGA to measure the plasma parameters at different spatial locations, (3) used imploding capsules filled with deuterium and tritium to create mono-energetic 14.7-MeV protons for imaging the magnetic field structures created by counter-streaming plasmas, and (4) clearly observed filamentary structures created by the Weibel electromagnetic instability according to our three-dimensional particle-in-cell simulation, with results indicating that approximately 1% of the plasma kinetic energy is converted into magnetic energy for our experimental conditions.

Project Summary

The successful conclusion of this project resulted in the development of a new platform for studying astrophysical collisionless shocks. We used laser-driven, counter-streaming, interpenetrating, supersonic plasma flows to understand astrophysical electromagnetic plasma phenomena in the laboratory. In our OMEGA experiments, the counter-streaming flow plasma state is measured using a Thomson-scattering diagnostic. We observed surprising electron and ion heating attributed to ion drag force and electrostatic instabilities. Using protons from a deuterium–tritium implosion to image the magnetic fields, we observed unexpectedly large, stable, and reproducible electromagnetic field structures that can be explained by recompression of the Biermann battery magnetic field after advection along the electron flows. The images showed very clear filamentary structures—three-dimensional particle-in-cell simulations indicate that they are generated by Weibel instabilities. These findings have significant astrophysical relevance and implications. We have received funding from the High Energy Density Laboratory Plasmas program by the DOE Office of Fusion Energy Sciences to continue this research.

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Control of Impulsive Heat Loads in Tokamaks: Measurements and Modeling

Max Fenstermacher (11-ERD-058)

Abstract

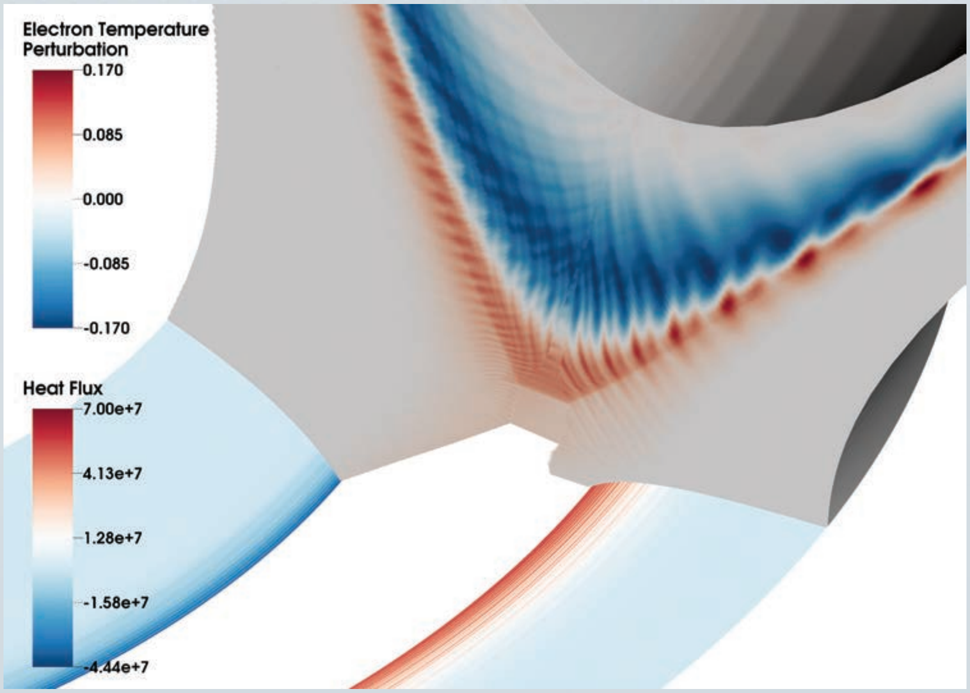
Hot, magnetically confined fusion plasmas in tokamak nuclear reactors generally exhibit periodic impulsive edge instabilities called edge-localized modes (ELMs). The impulsive nature of the ELM plasma heat load on wall materials in such devices indicates that unacceptably high erosion and damage will occur unless ELM loss fluxes are reduced. We propose to develop a physics capability for understanding and predicting the damaging heat and particle fluxes from ELMs through a unique combination of experimental diagnostics and validated simulation tools. This includes measuring and analyzing ELM behavior with new and existing diagnostics on the two largest U.S. tokamaks to understand surface heat-deposition patterns and more benign ELM regimes, both naturally occurring and those induced by mitigation techniques. We will extend the capability of our BOUT++ plasma turbulence code to include the full ELM cycle. We will conduct experiments on both the DIII-D reactor at General Atomics in San Diego to understand ELM control by three-dimensional non-axisymmetric perturbation fields, and on the National Spherical Torus Experiment at Princeton University to understand ELM control by novel two-dimensional divertor geometries, then use these experimental data to validate our simulation models.

We expect to produce (1) simulations of fundamental nonlinear ELM phenomena, including magnetic field transport with a full scrape-off layer, yielding time-resolved wall-heat profiles; (2) a predictive ELM simulation capability applicable to fusion research; and (3) analysis of ELM mitigation, including a predictive capability. The resulting validated model should provide detailed information on plasma heat and particle fluxes at reactor device walls and the plasma response to hydrogen and impurity fluxes from the walls.

Mission Relevance

This project will enable the Laboratory to play a major role in next-step fusion experiments, including the ITER international fusion research reactor in southern France, in support of the energy security mission. The project also supports Livermore's science and technology foundations in fusion and high-energy-density science, as well as high-performance computing and simulation.

Plasma electron temperature perturbation (top) and heat flux on divertor floor (bottom) from BOUT++ edge-localized mode simulation shows plasma structure and the increased heat flux impinging on target surfaces during an edge-localized mode crash event.



FY13 Accomplishments and Results

In FY13 we (1) calculated resonant magnetic perturbation penetration in rotating plasmas using BOUT++ in a simplified geometry; (2) completed the fundamental self-consistent perturbed-magnetic-field determination with plasma shielding; (3) compared DIII-D mid-plane density profile, divertor heat flux, and energy loss data with BOUT++ simulations, which showed reasonable agreement, and identified that the ratio of radial electron heat flux driven by turbulent convection versus stochastic diffusion depends on the parallel heat-flux-limiting coefficient; (4) completed BOUT++ and data validation dependence on plasma triangularity and collision frequency using a new small ELM case compared with our previous work on a large ELM; and (5) used UEDGE (a two-dimensional edge-plasma transport code) to map the BOUT++ solution to target surface heat-flux profiles.

Project Summary

This successful project resulted in substantial improvement to the predictive capability of nonlinear ELM simulations with the BOUT++ code. We validated multiple aspects of the BOUT++ models by comparing simulations of two very different ELM cases from the DIII-D tokamak against high-temporal-resolution data from multiple diagnostics. The evolution of mid-plane density profile, divertor heat flux, and energy losses are in reasonable agreement with two-fluid, six-field simulations for the high-collision-frequency, small-ELM case. Simulations show that turbulent convective fluxes are 100-times larger than conductive fluxes, but the fraction of stochastic radial heat flux depends sensitively on the electron parallel heat-flux-limiting coefficient. Models for ELM control using resonant magnetic perturbation coils were extended analytically and numerically.

Plasma response to resonant magnetic perturbations is predicted to be sensitive to pedestal electron rotation and yields outward particle flux, in agreement with experiment. The BOUT++ simulations with self-consistent plasma shielding were verified in simplified geometry, and realistic ELM-control coil models were extended to multiple-machine geometries. The use of BOUT++ to analyze ELM dynamics and resonant magnetic perturbation effects in the current generation of international tokamak devices, as well as to predict ELM events and resonant magnetic perturbation ELM control in future tokamaks, will continue through programmatic support and international collaboration funding from the DOE Office of Fusion Energy Sciences.

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Investigation of Fast Z-Pinches for Scalable, Large-Current and High-Gradient Particle Accelerators

Andrea Schmidt (11-ERD-063)

Abstract
Our objective is to obtain the first detailed understanding of the physics of extremely high (greater than 100-MV/m) acceleration gradients in hot, dense Z-pinch plasmas, which are confined through the use of an electrical current to generate a magnetic field that compresses, or pinches, the plasma. We will perform unique, first-ever probe-beam experiments that will measure the gradients directly and develop state-of-the-art fully kinetic plasma simulations. Our motivation is twofold. First, at the megavolt level, dense plasma focus (DPF) devices optimized for beam production and acceleration could serve as the basis for compact, intense radiological sources such as

directional neutron sources, and also as unique high-current ion injectors. Second, the DPF device could, if scaled to higher levels, lead to multistage, very-high-gradient plasma-based accelerators notably simpler than current laser or electron-beam systems, thus revolutionizing accelerator technology and applications.

We expect to produce a fundamental understanding of acceleration gradients in hot, dense Z-pinch plasmas, and use that knowledge to examine how these plasmas can be systematically exploited for accelerator applications such as a new class of high-current injectors and a fundamentally new kind of high-gradient plasma accelerator using multiple staged Z-pinch devices. We will also experimentally demonstrate, for the first time, the use of a DPF device as a very-large-current injector source for an induction linear accelerator. Success in this project could potentially be a revolutionary breakthrough in accelerator technology that would lay the groundwork for enabling mission-relevant applications such as portable devices for the very-large-standoff detection of special nuclear material using gigaelectronvolt protons.

Mission Relevance

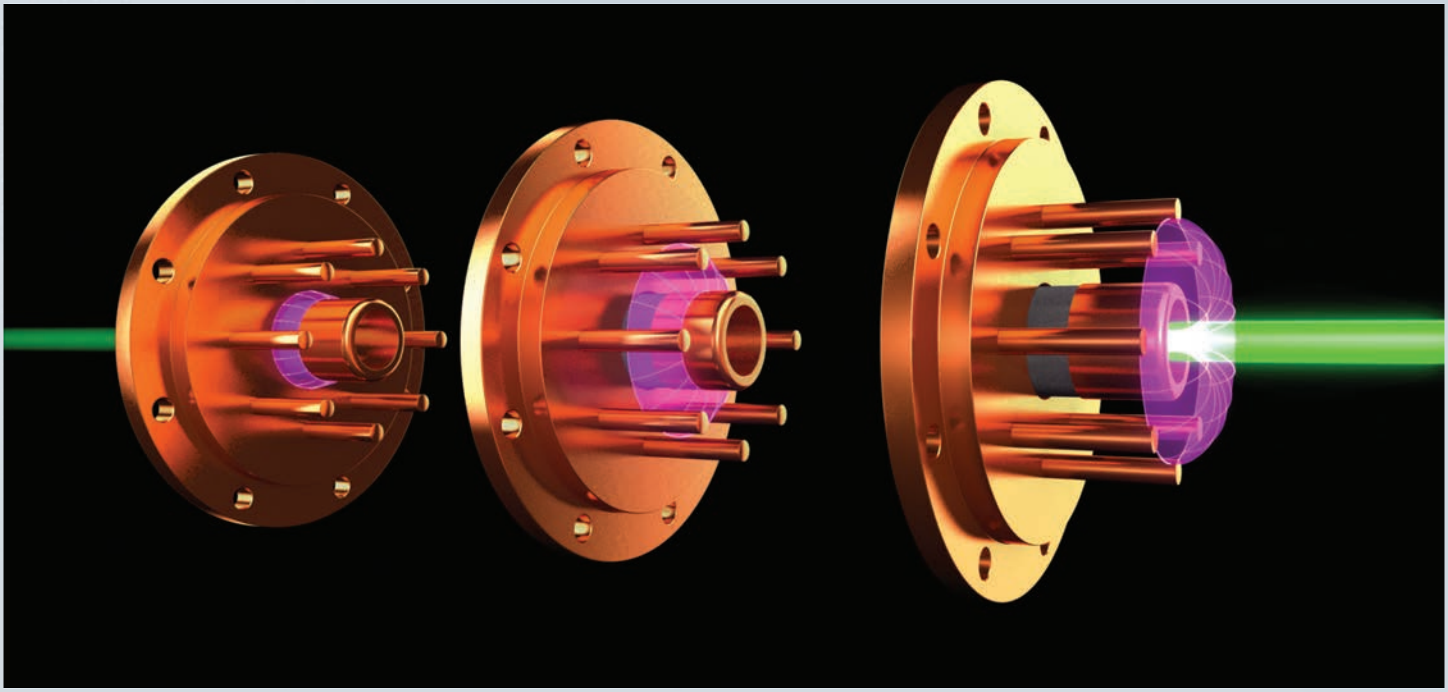
The work supports LLNL's national security missions—specifically homeland security, nonproliferation, and accelerator science—by providing the technological and scientific basis for compact alternative radiological sources and next-generation accelerator technology for active interrogation and radiography. Successful resolution of the physics behind the Z-pinch plasmas formed in a DPF device would resolve longstanding questions in plasma physics, a core Livermore competency.

FY13 Accomplishments and Results

In FY13 we (1) completed the first-ever probe-beam experiments and compared the results to predictions from our fully kinetic code; (2) investigated the feasibility of using our DPF Z-pinch device as an injector for an induction linear accelerator; (3) developed the capabilities to model staging DPF devices for a next-generation gradient accelerator, but did not examine the feasibility of staging the devices because of hardware cost considerations relative to the funded scope of this project; and (4) produced the first-ever demonstration of an ion beam injected into a high-gradient Z-pinch plasma along with predictive DPF simulations to study the feasibility of using DPF devices as a next-generation accelerator technology.

Project Summary

This project concluded successfully with the first-ever fully kinetic simulations of a DPF device and first-ever measurements of an ion beam injected into a Z-pinch plasma. The kinetic simulations correctly predicted, for the first time, neutron yields and ion beam energies observed on these devices that have not been reproduced in past fluid simulations. Additionally, we used an electromagnetic pick-up probe to make the first measurements of oscillations indicative of a plasma instability long suspected to be the cause of the high gradients inside a DPF, and also observed these oscillations for the first time in our kinetic simulations. We performed extremely challenging beam-into-plasma experiments using a deuteron probe beam sent through a hollow DPF



A plasma sheath (purple) runs down the length of a dense-plasma focus gun and then implodes, creating a hot, dense Z-pinch plasma with high gradients. We inject a probe beam (green) into the dense plasma focus during the Z-pinch phase to sample the electric fields in that region.

during the Z-pinch phase. We used a spectrometer and radiochromic film to characterize the steering and acceleration of the beam by the DPF device. This project allowed us to greatly increase our understanding of DPF and demonstrate injection of a beam into a DPF device for the first time. Follow-on external projects and feasibility studies through the Defense Advanced Research Projects Agency, Defense Threat Reduction Agency, and NNSA will be established to develop next-generation beam-driven sources. Additional government agencies have expressed interest and have requested proposals that would leverage the modeling and experimental capabilities we developed. We are pursuing multiple applications of our new capabilities in the areas of nuclear forensics, x-ray and neutron radiography, and high-energy-density physics.

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Nuclear Plasma Physics

Dennis McNabb (11-ERD-069)

Abstract

To understand the evolution of the universe and fundamental underpinnings of high-energy-density plasmas, as well as fully realizing the potential of inertial fusion energy, we need to understand nuclear reactions in burning plasmas. We propose to characterize nuclear effects important to stellar evolution, nucleosynthesis, burning plasmas, and nuclear reactions with excited nuclei that occur under high-energy-density plasma conditions. We will conduct near-term experiments to explore some of these issues and develop the theoretical underpinnings to motivate and execute a longer-term scientific program that engages the basic nuclear and weapons physics communities.

We expect our research will help improve understanding of neutrino production in the Sun as well as enable a new diagnostic of plasma temperatures in National Ignition

Facility (NIF) experiments via measurement of the energy spectrum of scattered, charged particles. This work will also address the question of how electron screening in plasmas differs from screening in beam interaction with target experiments. The results from our modeling and measurements of neutron capture reactions, if successful, will enable a new capability to measure neutron capture rates for small quantities of highly radioactive isotopes generated in NIF shots. Neutron capture rates for short-lived isotopes are important to a detailed understanding of the production of elements in stellar evolution as well as nuclear chemistry analysis of historical nuclear test data for stockpile stewardship. We expect to develop a model for how nuclear plasma and neutron interactions populate excited states and how those excited states change the outcome of nuclear processes.

Mission Relevance

By improving our understanding of nuclear reactions in burning plasmas, we will enable new science capabilities important to stockpile stewardship, a key mission area at Lawrence Livermore, as well as further research in inertial fusion energy, in support of the Laboratory mission to enhance the nation's energy security. In addition, our research supports and enhances Livermore's foundations in high-energy-density matter.

FY13 Accomplishments and Results

In FY13 we (1) performed new, more precise measurements and calculations of the neutron-spectra emitted in the tritium–tritium reaction for gas-filled, inertial-confinement fusion capsule implosions at both NIF and the OMEGA laser facility at the Laboratory for Laser Energetics in Rochester, New York, to obtain data at several different temperatures; (2) developed new calculations that account for exchange symmetry and interference effects between the outgoing neutrons of tritium–tritium and deuterium–tritium that reproduce the data quite well; (3) modified our approach for observing plasma–nuclear coupling in plasma experiments to address problems we encountered in separating meta-stable atomic excitations from nuclear excitations; (4) performed calculations of the distribution of nuclear states in a hypothetical ignition capsule with high yield (10^{18} neutrons); and (5) found that in some cases, high-spin-state lifetimes of 10 to 100+ ps are predicted to be populated with significant cross sections at up to 2 to 3 MeV of excitation energy—however, the uncertainty in the lifetime of these states is at least a factor of two with our current models.

Project Summary

Over the course of this project we (1) demonstrated viable techniques for measuring fundamental nuclear processes, (2) provided a detailed understanding of tritium and helium-3 reactions at thermonuclear conditions relevant to understanding energy and neutrino production in our Sun, and (3) elucidated fairly convincingly that ion species separation is occurring in exploding pusher experiments and might be important in other inertial-confinement energy experiments. In addition, we developed techniques and capabilities for modeling and performing experiments to look for how nuclear plasma and neutron reactions populate excited states. Finally, we helped to significantly improve neutron time-of-flight capabilities at NIF and OMEGA that will be of broad benefit to high-energy-density science communities. We have established collaborations with Indiana University, Massachusetts Institute of Technology, and

Notre Dame in recognizing the compelling nuclear science that can be performed at inertial-confinement fusion facilities. The techniques and experiences gained here will also lead to new diagnostic capabilities that will support future inertial-confinement fusion and stockpile stewardship science campaigns at Lawrence Livermore.

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Transport Properties of Dense Plasmas and a New Hybrid Simulation Technique for Matter at Extreme Conditions

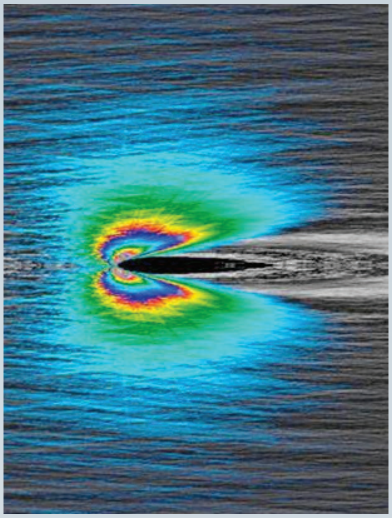
Frank Graziani (12-SI-005)

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 codes' 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 the behavior of these quantities 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.



Wake potential of a fast charged-ion projectile in a dense plasma background.

Mission Relevance

Strategically, this project will improve the predictive capability of Livermore's Advanced Simulation and Computing codes and 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.

FY13 Accomplishments and Results

In FY13 we (1) extended ddcMD's capability to include Yukawa inter-ionic potential models and applied this to species diffusivity problems in dense plasmas; (2) performed an exhaustive study of stopping power for various target–plasma coupling and target ionization parameters; (3) implemented a simple version of kinetic theory molecular dynamics as part of the thesis work of one of our students; (4) began theoretical developments in quantum hydrodynamics, which will be a major focus of FY14; and (5) performed significant theoretical and computational work on conductivity, although questions remain in the computational results relating electrical and thermal conductivities.

Proposed Work for FY14

In FY14 we will (1) develop diffusivity models for programmatic application and validate existing models, (2) implement a new capability based on quantum hydrodynamics for electrons and apply it to the diffusivity for warm dense matter, and (3) expand the application of thermal and electrical conductivity simulations to inertial-confinement fusion efforts.

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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 from 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 the 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, and advanced laser optical systems and applications.

FY13 Accomplishments and Results

In FY13 we (1) measured the equation of state of ramp-compressed iron to 2.5 and 8.0 Mbar on OMEGA and NIF, respectively, and demonstrated multishock compression on diamond to above 100 Mbar on NIF; (2) performed three gigabar NIF shots, obtaining the stress-density Hugoniot (valuable for analyzing a material's equation of state) of carbon–hydrogen to over 500 Mbar, and conducted one diamond diffraction shot on NIF; (3) improved the signal-to-background noise ratio on our x-ray diffraction shots at both OMEGA and NIF; (4) explored the phase diagram of ramp-compressed iron oxide and molybdynum by x-ray diffraction at OMEGA, and performed OMEGA experiments on pre-compressed helium, hydrogen, and deuterium in the region of the plasma phase transition; (5) performed OMEGA shots on stishovite, a form of silicon dioxide, thereby extending the melt line of silicon dioxide to over 4 Mbar; (6) characterized the dissociation of molecular nitrogen into an atomic fluid at OMEGA; (7) explored the elastic–plastic and beta-tin phase transition damage response of silicon using a high-spatial-resolution two-dimensional VISAR (velocity interferometer system for any reflector) at Livermore's Janus laser; and (8) observed and simulated compression-rate-dependent response equation-of-state data on the iron alpha–epsilon phase transition over five orders of magnitude in total strain rate.

Proposed Work for FY14

In FY14 we will (1) perform additional NIF experiments on compression of iron for equation-of-state studies, terapascal diffraction of diamond, gigabar Hugoniot of carbon–hydrogen, and cryostatic deuterium equation-of-state measurements; (2) continue our OMEGA work on Earth-related materials and fundamental science diffraction experiments on samples such as molybdenum, iron oxide, magnesium, aluminum, and iron; (3) perform Hugoniot experiments on ambient and pre-compressed water; (4) continue to develop two-dimensional experiments using the high-resolution VISAR, including measurements of material viscosity; (5) perform absolute Hugoniot measurements of carbon–hydrogen and aluminum using x-radiography, and continue our pre-compressed hydrogen–helium experiments; (6) perform field diffraction experiments at the Linac Coherent Light Source at the SLAC National Accelerator Laboratory in Menlo Park on water, body-centered-cubic omega-phase transition mechanisms, and liquids; and (7) field a range of experiments on the Janus laser.

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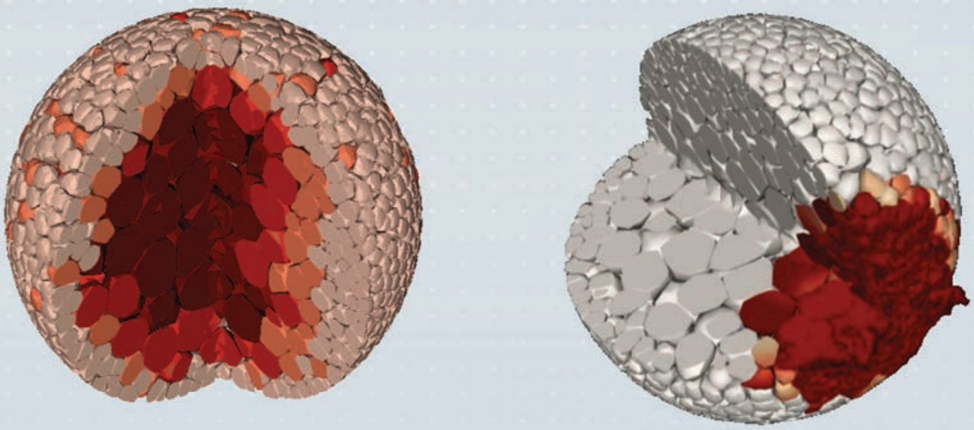
Asteroid Deflection

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

Model of a 500-m-diameter "rubble pile" asteroid comprised of 2,380 individual boulders (left). A simulation of the asteroid receiving 5.4 kt of energy shows the behavior inside each boulder as well as interactions with their neighbors (right). Material in the vicinity of the energy deposition vaporizes, melts, or fails, contributing to blowoff. The blowoff imparts momentum to the assembly of boulders.



with a range of threat compositions, sizes, dynamics, and times to impact, and optimize parameters such as the height of burst and the 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 to evaluate and optimize a variety of nuclear approaches, develop an asteroid breakup modeling capability, and increase the expertise 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 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.

FY13 Accomplishments and Results

In FY13 we (1) refined our range of scenarios and their simulation, (2) continued the use of test problems to verify our code results, (3) completed an error analysis of a dispersed object and further developed orbital-dispersal simulations, (4) conducted validation of the code through simulations of the Stickney crater on Phobos (a moon of Mars) as an integrated validation, and (5) participated in a joint National Aeronautics and Space Administration and Federal Emergency Management Agency tabletop exercise on an asteroid-impact scenario.

Proposed Work for FY14

For FY14 we will (1) continue to refine and expand our range of scenarios and their simulation within our suite of codes; (2) continue the use of our test problems to validate code results; (3) complete our assessment of U.S. capabilities, using our best current methodology; and (4) further develop our collaborative interactions with a wide range of institutions.

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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 in Menlo Park 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 basic 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 diagnostic 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. In addition, this high-profile project will attract top postdoctoral researchers and allow us to engage in international collaborations.

FY13 Accomplishments and Results

In FY13 we (1) evaluated the scattering data obtained in our previous LCLS experiment and found that the high-intensity LCLS beam imparted damage to our crystals over a much larger area than expected—post-exposure sample analysis is required before comparisons to our models can be made, (2) performed interferometric and electron microscopy analysis of the samples, (3) continued development of our x-ray and matter interaction models, (4) collaborated on LCLS experiments on ultrafast processes in optical materials and on damage in iron-doped samples, and (5) designed and demonstrated LCLS diffraction samples that are less prone to damage.

Proposed Work for FY14

In FY14 we will (1) complete the post-exposure analysis of our samples irradiated by the LCLS beam, (2) design and propose a new LCLS experiment based on our improved sample design, (3) perform a first round of experiments on the improved samples, and (4) continue development of an x-ray interaction model.

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Computational Gyro-Landau Fluid Model for Tokamak Edge Plasmas

Xueqiao 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 Atomic's 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.

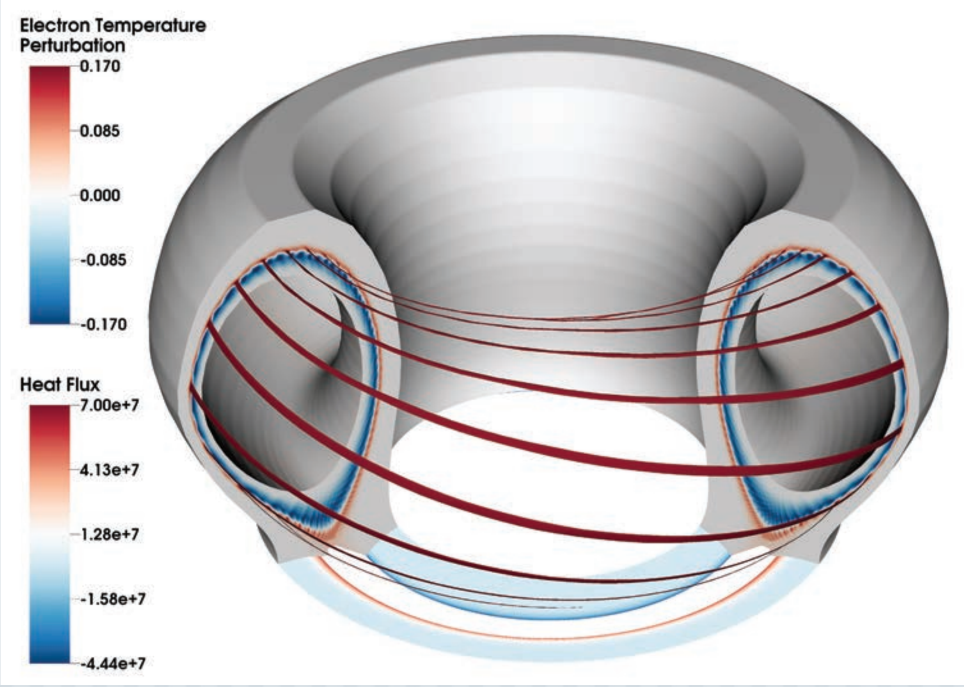
If successful, the edge-plasma model created and validated in this project will generate a theoretical and simulation capability far beyond the present state of the art. An accurate gyro-Landau fluid simulation model may offer orders-of-magnitude savings in computational resources compared to a gyro-kinetic 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 other 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 national security missions in the focus areas of fusion energy, high-energy-density matter, and high-performance computing, and it offers an excellent opportunity to forge alliances with emerging Asian fusion programs.

FY13 Accomplishments and Results

In FY13 we (1) determined requirements for gyro-Landau fluid models to perform well at large perturbations; (2) began deriving second-order-accurate closures; (3) implemented electrostatic and electromagnetic 3 + 1 gyro-Landau fluid models; (4) implemented realistic magnetic x-point geometry; (5) executed linear and nonlinear benchmarks for eigenfunctions, growth rates, turbulence, and equilibria and obtained excellent agreement with gyro-kinetic calculations for ion-temperature-gradient instability growth rates; (6) continued development of radio-frequency sheath models; (7) implemented and verified nonlocal parallel transport operators in toroidal geometry and added Landau heat-flux closures to an electromagnetic, two-fluid, six-field Braginskii model; (8) continued comparisons of simulations with turbulence data from KSTAR and DIII-D—synthetic images from simulations are in qualitative agreement with two-dimensional electron cyclotron emission imaging on the EAST tokamak; (9) continued comparisons of electromagnetic simulations to magneto-hydrodynamic data from DIII-D and EAST; and (10) predicted edge-localized mode filament structure is in qualitative agreement with EAST fast visible camera observations.



A new supercomputer simulation reveals the dynamics of filamentary eruptions of hot plasma from the edge of a tokamak fusion experiment. These relatively hot and dense bursts of plasma, called edge-localized modes, are generated by magneto-hydrodynamic instabilities just inside the plasma edge. During an edge-localized mode crash, a hot filament is rapidly ejected outward, heats the scrape-off layer, and cools the interior (as illustrated by the change in electron temperature at the top of the figure). The magnitude of the power deposition onto the divertor crucially depends on the parallel physics model targets (as illustrated at the bottom on the divertor floor). Accurate predictions of thermal conduction need to incorporate kinetic physics within the model.

Proposed Work for FY14

In FY14 we will (1) analyze the accuracy of nonlinear wave–particle and wave–wave coupling; (2) investigate nonlinear kinetic effects at large perturbation; (3) identify computational bottlenecks and numerical strategies that efficiently project to a massively parallel computational environment; (4) implement a 3 + 1 electromagnetic model; (5) implement 4 + 2 models as needed; (6) start the implementation of second-order closures; (7) investigate advanced numerical techniques such as a generalized three-dimensional Poisson solver; (8) extend the 3 + 0 electromagnetic gyro-Landau fluid model from core to edge; (9) implement a radio-frequency sheath physics model; and (10) validate turbulence, edge-localized mode, and the impact of radio-frequency on plasma turbulence with DIII-D, EAST, and KSTAR experimental data.

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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 missions in stockpile stewardship, energy security, and high-energy-density science.

FY13 Accomplishments and Results

In FY13 we (1) created and validated a two-dimensional, finite-element thermal model that describes the observed seed location in fusion targets; (2) used analysis of x-ray images of the ignition fuel layer evolution in conjunction with a geometric growth model to show that the volume solidified is linear with time; (3) performed molecular dynamic simulations of crystallization on templates using reduced super-cooling, which suggest that specific template orientations are more robust with respect to defects than others; (4) performed initial experiments with candidate template materials—however, nucleation on the container walls prevented good experimental data; and (5) made several modifications to the electro-freezing experiment that was unable to achieve nucleation.

Proposed Work for FY14

In FY14 we will (1) study nucleation on selected template substrates under several super-cooling conditions and compare to simulations, (2) test electro-freezing using a wire inserted into a fusion capsule to achieve higher super-cooling, (3) study layer quality by confining seed location to the bottom of the shell using a thermal gradient, (4) study the effect of surface roughness on seeding using molecular dynamics simulations, (5) continue to develop a layering simulator, (6) develop and test process strategies to improve reproducibility of in situ single-seed isolation, and (7) use model and experiment to understand the driving force responsible for linear mass uptake.

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 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 mono-energetic 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 thrust area in nuclear photo-science 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. It will improve the quality of desired fundamental nuclear physics measurements, as well as expand the envelope of feasible nuclear photonics applications for national and international security.

FY13 Accomplishments and Results

In FY13 we (1) successfully assembled, operated, and characterized the modulator and klystron that will power the accelerator; (2) installed the power distribution and precision accelerator hardware, along with the magnets, diagnostics, control systems, and support services to run the X-band accelerator; (3) received approval to begin implementing our commissioning procedure and powered up the accelerator to condition the hardware; (4) transported the laser beam that will generate the electrons to the location of the accelerator; and (5) demonstrated a gigahertz-rate drive laser with the necessary energy output in the infrared (5 mJ/pulse for 500 pulses and a 28-kHz repetition rate) to meet the requirements for gigahertz electron-beam generation.

Proposed Work for FY14

In FY14 we will (1) operate and characterize the performance of the X-band accelerator at a variety of settings to compare performance with our models and optimize the beam emittance and energy spread; (2) design, build, and install an interaction region to allow generation of x-ray radiation via Compton scattering off the electron beam; and (3) complete the characterization of the gigahertz laser technology, including designing and installing the frequency conversion system to produce the ultraviolet light needed for electron generation.

Publications and Presentations

Gibson, D. J., et al., 2012. *A drive laser for multi-bunch photocathode operation.* 2012 IEEE Particle Accelerator Conf. (IPAC), New Orleans, LA, May 20–25, 2012. LLNL-PROC-557582.

Marsh, R. A., 2013. *Machining precision and small gap effects in high gradient structures.* North American Particle Accelerator Conf. (NA-PAC13), Pasadena, CA, Sept. 29–Oct. 4, 2013. LLNL-ABS-608612.

Marsh, R. A., S. G. Anderson, and J. P. Armstrong, 2012. *Precision magnet measurements for X-band accelerator quadrupole triplets.* 2012 IEEE Particle Accelerator Conf. (IPAC), New Orleans, LA, May 20–25, 2012. LLNL-PROC-557665.

Marsh, R. A., et al., 2013. *LLNL X-band RF system and commissioning.* North American Particle Accelerator Conf. (NA-PAC13), Pasadena, CA, Sept. 29–Oct. 4, 2013. LLNL-CONF-636275.

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.

Marsh, R. A., et al., 2012. *Modeling multi-bunch x-band photoinjector challenges.* 2012 IEEE Particle Accelerator Conf. (IPAC), New Orleans, LA, May 20–25, 2012. LLNL-PROC-557580.

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.

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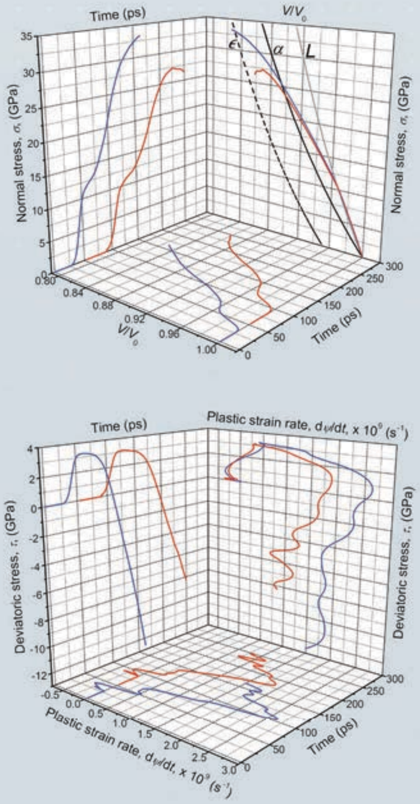
Strength in Metals at Ultrahigh Strain Rates

Jonathan Crowhurst (12-ERD-042)



Results from our ultrafast compression experiments on iron. Normal stress and relative volume versus time is shown at top. The blue and red lines correspond to two different sample thicknesses. The paths the material follows are seen to leave the longitudinal elastic compression curve around 10 GPa and pass through the alpha-phase isentrope at around 25 GPa. The data in the form of deviatoric stress and the plastic strain rate versus time is shown below. The material is seen to sustain a very large deviatoric stress before the majority transforms to the epsilon phase. During the phase transition, the deviatoric stress apparently becomes negative, but this is an artifact of the analysis, which assumes plastic deformation only.

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 of, 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 areas of stockpile stewardship science and advanced laser and optical systems. Obtaining a more detailed understanding of the detailed physics associated with high-strain-rate dynamic compression is also important for the foundational science of materials and chemistry at the extremes as well as for fundamental research in a range of disciplines.

FY13 Accomplishments and Results

In FY13 we (1) observed the alpha-to-epsilon transition in iron on an ultrafast timescale at strain rates of 10^9/s, and found the transition began very soon (within ~100 ps) after the arrival of a large elastic precursor and was essentially complete within a similar time period thereafter; (2) determined that the deviatoric stress before the transition begins can exceed 3 GPa; (3) found the majority of the material transformed at stresses around 25 GPa, or nearly twice as large as inferred in longer-timescale experiments; (4) made measurements of free surface velocities in pure body-centered-cubic tantalum in the thickness range of 1 to 2 μm, from which we expect to extract elastic precursor amplitudes, deviatoric stress, relative volume, and normal stress; and (5) completed a new shock platform that will accommodate an evacuated cryostat, which will not only control the initial temperature, but will allow us to deliver much higher energies (up to 100 times) to our metal samples without interference from plasma formation in air.

Proposed Work for FY14

In FY14 we will (1) study single-crystal iron to determine how its transition depends on grain structure by comparing results to our polycrystalline data, (2) conduct further measurements on tantalum in the stress range above 1 MBar to obtain important strength information, (3) continue to study pure aluminum to further refine our technique and test analysis approaches, and (4) begin to investigate the combination of our small-scale shock wave experiment with dynamic electron diffraction, thus taking advantage of the inherent advantages of both, to probe the microstructure of dynamically compressed materials at ultrahigh strain rates with high fidelity and rapid throughput.

Publications and Presentations

Crowhurst, J. C., 2012. *Aspects of the behavior of aluminum and iron at ultrahigh strain rates.* Int. Workshop Strain Measurement in Extreme Environments, Glasgow, Scotland, Aug. 28, 2012. LLNL-PRES-563697.

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 the 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 thrust area and supports the national security mission.

FY13 Accomplishments and Results

In FY13 we implemented the random partially resolved transition-array model algorithm into the OPAL and TOPAZ opacity codes. We then fully tested the new model, demonstrating both its accuracy and speed. For example, the model was tested on the historically difficult to calculate iron opacity in stellar envelopes and obtained 5% agreement in the Rosseland mean opacity with the full detailed-line calculation (well within the expected error of the full calculation), with the new model operating at a factor of 20 faster.

Proposed Work for FY14

In FY14 we will (1) extend our successful partially resolved transition-array concept to the super-transition-array method, which is typically used in many of the Laboratory opacity calculations—however, the latter method contains an approximation because of the grouping of myriad electronic configurations into a single super configuration; (2) use the partially resolved transition-array concept to identify and reduce the uncertainty associated with configurations that least satisfy the super-transition-array approximation; and (3) implement a partially resolved super-transition-array algorithm into Livermore's VISTA relativistic, local thermodynamic equilibrium opacity code, which will be checked against more complete calculations such as those from Livermore's TOPAZ opacity code.

Publications and Presentations

Iglesias, C., 2012. "Statistical line-by-line model for atomic spectra in intermediate coupling." *High Energ. Density Phys.* **8**, 253. LLNL-JRNL-530533.

Iglesias, C., and V. Sonnad, 2012. "Partially resolved transition array model for atomic spectra." *High Energ. Density Phys.* **8**, 154. LLNL-JRNL-520413.

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 the 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 the 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 the 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 strategic science foundation in measurement science and technology by developing diagnostics able to view complex energetic dynamic processes.

FY13 Accomplishments and Results

In FY13 we (1) completed analysis of a relevant subset of data for production of W boson pairs using leptons, (2) began design of a reference clock system with auto-synchronization to the LHC proton beam, (3) led the U.S. collaboration for the new proton-tagging detector upgrade of the Compact Muon Solenoid LHC experiment, and (4) led an international team of about 30 physicists, resulting in a favorable Compact Muon Solenoid management review of the upgrade project.

Proposed Work for FY14

In FY14 we will (1) complete the design of our reference clock system incorporating features of LHC beam bunch signals, (2) continue to lead the U.S. collaboration for the proton-tagging upgrade, and (3) implement recommendations from the experiment management review.

Publications and Presentations

Wright, D. M., et al., 2012. *Exclusive gamma gamma to mu mu production in proton-proton collisions at 7 TeV*. LLNL-JRNL-604993.

Wright, D. M., et al., 2013. "Study of exclusive two-photon production of W+W– in pp collisions at √s = 7 TeV and constraints on anomalous quartic gauge couplings." *J. High Energ. Phys.* **1307**(2013), 116. LLNL-JRNL-644972.

Compton-Scattering Optimization for Ultra-Narrowband Nuclear Photonics Applications

Frederic Hartemann (12-ERD-057)

Abstract

Our main goal with this project is to optimize Livermore's Compton-scattering gamma-ray source technology for a potential 100,000-fold bandwidth reduction by combining gigahertz gamma-ray pulse trains with ultrahigh-resolution stabilized-crystal spectrometers. Gamma-ray lens technology also will be utilized for fractional bandwidths of 100 parts per million. This bandwidth reduction will enable direct fluorescence isotope-specific detection (with nuclear resonance), assaying, and imaging of special nuclear materials and other materials of interest. Key deliverables include detailed computer models of gamma-ray production, tracking, and interaction with gamma-ray optics, along with optimization algorithms. In addition, we will perform experimental characterization of gamma-ray optics.

If successful, this project will bring the emerging new field of nuclear photonics closer to maturity by providing tunable, mega-electronvolt photons within sub-electronvolt bandwidths at high average flux. Our proposed approach is two-pronged. We expect that our intensive theory and modeling component will combine advanced three-dimensional electron-beam dynamics codes with LLNL's Compton-scattering models, laser design tools, gamma-ray particle tracing and interaction codes, and sophisticated gamma-ray crystal diffraction models. Secondly, we will characterize a precision gamma-ray crystal spectrometer on loan from the Institut Laue-Langevin in France, and we may integrate the device with Livermore's Compton gamma-ray source, along with performing characterization of gamma-ray optics.

Mission Relevance

Our project is expected to have maximum impact for the Laboratory's strategic mission in advanced laser optical systems and applications by developing a Compton source with higher flux and brightness than the current single-bunch system to provide improved quality of desired nuclear physics measurements and expanded research possibilities. By enabling imaging of special nuclear materials, this research also directly supports LLNL's national security mission with applications to nuclear threat reduction.

FY13 Accomplishments and Results

In FY13 we (1) studied solid-angle effects such as collimation and detector size on LLNL's Compton gamma-ray source, (2) performed a systematic optimization of the nonlinear brightness of the gamma-ray photon beam in the single-bunch regime, and (3) performed preliminary work on multiple-bunch beam-line operation, as well as optimization for different metrics, including peak brightness and peak brightness divided by the frequency-integrated collimated spectrum, which is important for signal-to-noise consideration when the source is used for nuclear resonance fluorescence experiments.

Project Summary

During the course of this project, we studied solid-angle effects such as collimation and detector size on LLNL's Compton gamma-ray source, performed a systematic optimization of the nonlinear brightness of the gamma-ray photon beam in the single-bunch regime, and published our results in *Physical Review Letters*. In addition, we performed seed work on multiple-bunch beam-line operation, as well as optimization for different metrics, including peak brightness and peak brightness divided by the frequency-integrated collimated spectrum, which is important for signal-to-noise consideration when the source is used for nuclear resonance fluorescence experiments. This work is expected to have programmatic implications, because it provides a path to differential hard x-ray imaging of ion charge states in high-energy-density and inertial-confinement fusion plasmas.

Publications and Presentations

Albert, F., et al., 2012. *Development of Compton scattering gamma ray sources at LLNL*. 15th Advanced Accelerator Concepts Workshop (AAC 2012), Austin, TX, June 10–15, 2012. LLNL-ABS-557757.

Albert, F., et al., 2012. *NRF-based isotope material detection experiments with the LLNL T-REX MEGa-ray machine.* 53rd Ann. Mtg. Institute of Nuclear Materials Management, Orlando, FL, July 15–19, 2012. LLNL-ABS-468934.

Anderson, S. G., et al., 2012. *Compact linear accelerator sources for mono-energetic gamma ray (MEGa-ray) generation and NRF-based nuclear materials management*. 53rd Ann. Mtg. Institute of Nuclear Materials Management, Orlando, FL, July 15–19, 2012. LLNL-ABS-468571.

Hartemann, F. V., and S. S. Q. Wu, 2013. "Nonlinear brightness optimization in Compton scattering." *Phys. Rev. Lett.* **111**, 044801. LLNL-JRNL-622841.

Wu, S. S. Q., and F. V. Hartemann, 2012. *Transient evolution of a photon gas in the nonlinear QED vacuum*. LLNL-TR-503123.

Wu, S. S., F. V. Hartemann, and C. P. J. Barty, 2012. *Ultracompact accelerator technology for a next-generation gamma-ray source*. Intl. Particle Acccelerator Conf. 2012 (IPAC 2012), New Orleans, LA, May 20–25, 2012. LLNL-PROC-557854.

Early-Phase Hydrodynamic Instability Development in National Ignition Facility Capsules

Daniel Clark (12-ERD-058)

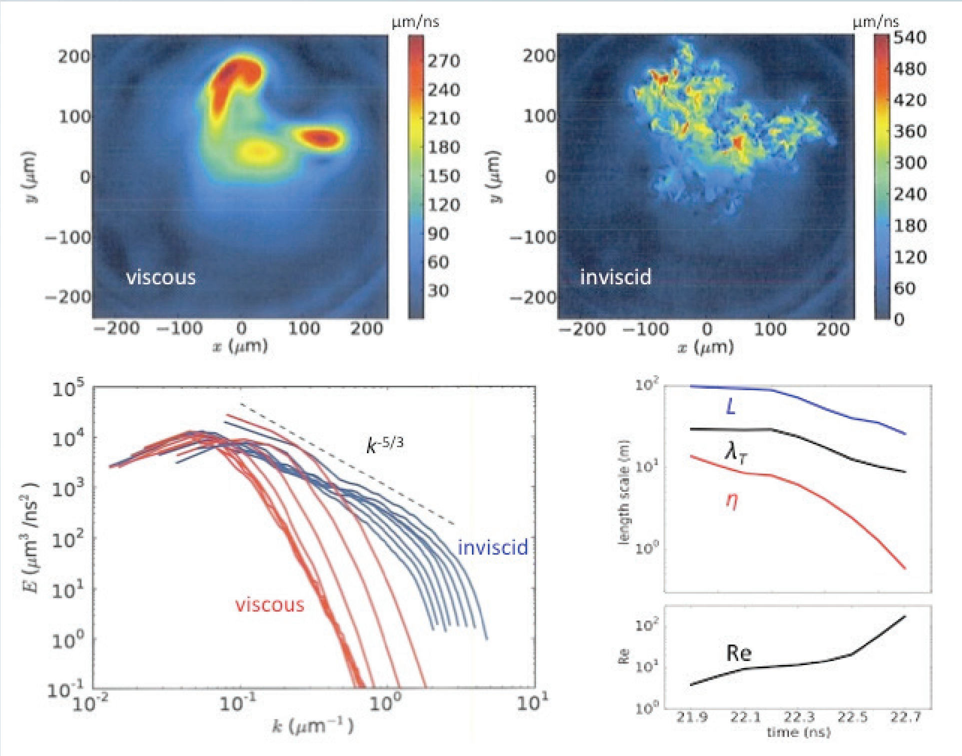
Abstract

Recently, the importance of the effect of early-phase Richtmyer–Meshkov instability growth on later Rayleigh–Taylor instability has been appreciated in determining the pulse shape needed to achieve fusion ignition in National Ignition Facility (NIF) experiments. 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 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 those from mixed models 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 the details of instability development more thoroughly and by precisely determining acceptable stability boundaries, this project will also help improve ignition-capsule



Comparison of viscous and inviscid three-dimensional Miranda simulations of an experimental laser implosion shot showing late-time instability development in the hot spot. The upper panels show the fluctuating velocity fields near peak compression and quasi-turbulent behavior in the inviscid case that is heavily damped in the viscous case. The lower panels show the development of an approximate inertial range of turbulent fluctuations in the inviscid case.

hydrodynamic stability and therefore enable the high-gain target designs required for inertial fusion energy.

FY13 Accomplishments and Results

In FY13 we (1) refined our semi-analytic model for the evolution of growth factor phase in NIF implosions by elaborating our understanding of existing ablative Richtmyer–Meshkov theory and showing good agreement with HYDRA simulations, with a crucial ingredient to this evolution being the impact of returning rarefaction on setting the final perturbation phase; (2) found a prescription for capturing the perturbation phase evolution during this crucial rarefaction interaction and began work to develop a more rigorous understanding of this; and (3) worked on Miranda (a hydrodynamics code for simulating Rayleigh–Taylor and Richtmyer–Meshkov instability growth) simulations of three-dimensional instability evolution in NIF implosions, which showed very good agreement with companion three-dimensional HYDRA simulations—an important verification of the modeling fidelity of both codes—and also showed the surprisingly large impact of hot spot viscosity on late-phase instability evolution.

Proposed Work for FY14

In FY14 we will (1) complete our phenomenological, simulation-based study of Richtmyer–Meshkov and Rayleigh–Taylor phase effects and the dependence of these effects on the strength of the onset of the radiation drive pulse; (2) leverage these results to investigate the sensitivity of Richtmyer–Meshkov and Rayleigh–Taylor instability growth to equation of state, opacity, and other factors; and (3) continue our Miranda–HYDRA comparisons; and (4) continue our Miranda simulations of early- and late-time instability development in NIF implosions, which will enable very-high-resolution modeling of vorticity deposition and evolution that is crucial to instability growth.

Publications and Presentations

Peterson, J. L., et al., 2012. *Altering the hydrodynamic mixing of NIF capsules with shocks and rarefactions.* 54th Ann Mtg. APS Division of Plasma Physics, Providence, RI, Oct. 29–Nov. 2, 2012. LLNL-ABS-563711.

Peterson, J. L., et al., 2012. *The phase of hydrodynamic growth in inertial confinement fusion capsules.* 42nd Ann. Anomalous Absorption Conf., Key West, FL, June 25–29, 2012. LLNL-ABS-552711.

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. Created by Compton scattering of short-duration laser pulses interacting with relativistic electrons, MEGa-rays are a new class of light source with extraordinary qualities. 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 under development. 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 strategic goal for advanced laser optical systems of 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, narrowbandwidth 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.

FY13 Accomplishments and Results

In FY13 we (1) used the existing code base for the Monte Carlo radiation transport code COG to model and analyze the potential of a narrowband gamma-ray source for detection and assay of special nuclear materials; (2) modeled and examined multiple scenarios of interest to the nuclear resonance fluorescence characterization community (fuel rod assay, soil contamination, and illicit material transport) to optimize the anticipated configuration of a potential DINO detection system (shielding, Compton liner, foil, and scintillator); (3) examined the conditions under which successful assay of special nuclear materials in fuel rod assemblies, as one example, could be determined; (4) successfully fielded an experiment at the High Intensity Gamma-Ray Source at Duke University with a unique gamma-ray detector arrangement; and (5) observed polarized interaction and discrimination during the Duke experiment, as predicted by modeling, using an isotopic-enriched surrogate material.

Proposed Work for FY14

In FY14 we will (1) use experimental data from the High-Intensity Gamma-Ray Source at a 3.4% bandwidth to enable a comparison of the capabilities of a narrowband source; (2) use COG coupled with experimentally obtained data to optimize design and model the construction and layout of a DINO detection system (shielding, Compton liner, foil, and scintillator); (3) add additional nuclear resonance fluorescence lines and capability to COG in collaboration with Texas A&M University; and (4) demonstrate, in combination with a novel narrowband gamma-ray spectrometer, the capabilities of a DINO-type detector in the presence of a low-, medium-, and narrowbandwidth gamma-ray source at the Institut Laue-Langevin in Grenoble, France.

Publications and Presentations

Brantley, P. S., et al., 2013. R*ecent advances in the mercury Monte Carlo particle transport code.* Int. Conf. Mathematics and Computational Methods Applied to Nuclear Science and Engineering (M&C 2013), Sun Valley, ID, May 5–9, 2013. LLNL-PROC-60997.

Johnson, M. S., 2013. *Photonuclear physics research at LLNL in collaboration with SJSU.* LLNL-PRES-622692-DRAFT.

Johnson, M. S., J. M. Hall, and D. P. McNabb, 2013. *Detector array performance estimates for nuclear resonance fluorescence applications.* DNP 2012 Fall Mtg. Division of Nuclear

Physics of the American Physical Society, Newport Beach, CA, Oct. 24–27, 2012. LLNL-PRES-594633.

Johnson, M. S., et al., 2012. *Detector array performance estimates for nuclear resonance fluorescence applications.* DNP 2012 Fall Mtg. Division of Nuclear Physics of the American Physical Society, Newport Beach, CA, Oct. 24–27, 2012. LLNL-ABS-563014.

Johnson, M. S., et al., 2012. *Preliminary study of the efficacy of using nuclear resonance fluorescence with quasi-monoenergetic gamma-ray sources for nuclear safeguards assay.* LLNL-TR-483851.

Johnson, M.S., et. al., 2012. "Searching for illicit materials using nuclear resonance fluorescence stimulated by narrow-band photon sources." *Nucl. Instrum. Meth. Phys. Res.* **285**, 72. LLNL-JRNL-422898.

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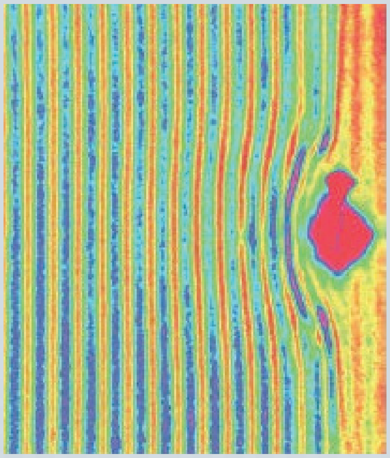
Pair-Plasma Creation Using the National Ignition Facility

Hui Chen (12-ERD-062)



Interferogram measured when the Livermore Titan laser interacts with a target in pair-plasma experiments. The laser irradiates the target from left to the right. The bright red area is the self-emission of the target, and plasma density is inferred from this measurement.

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 Radiographic 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 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.

FY13 Accomplishments and Results

In FY13 we (1) performed five experiments optimizing pair-plasma density and temperature on Livermore's Titan laser and the OMEGA Extended Performance laser at the University of Rochester, (2) tested a prototype electron and positron spectrometer on the Titan laser in preparation for shots at the National Ignition Facility, and (3) designed an astrophysics-relevant experiment on relativistic pair-plasma jets that could be conducted on the Titan, OMEGA Extended Performance, or Advanced Radiographic Capability laser.

Proposed Work for FY14

In FY14 we will (1) conduct our first experiments on focusing pair jets with externally applied magnetic fields, (2) begin the first pair jet–plasma interaction experiments using our FY13 simulation results, (3) perform our final pair-scaling experiments, (4) analyze data produced by our experiments, and (5) design the initial pair-plasma experiment to be conducted on the Advanced Radiographic Capability laser using up-to-date parameters for that laser system.

Publications and Presentations

Chen, H., 2013. "New insights into the laser produced electron-positron pairs." *New J. Phys.* **15**, 065010. LLNL-JRNL-591692.

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Kerr, S., et. al., 2013. *Characterization of laser-accelerated protons from the front surface of metal foils using the Titan Laser.* NIF and Jupiter User Group Mtg., Livermore, CA, Feb. 10–13, 2013. LLNL-POST-618097.

Park, J., et al., 2012. *Experimental study of preformed plasmas*. California–Nevada APS 2012, San Luis Obispo, CA, Nov. 2–3, 2012. LLNL-ABS-586772.

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Williams, G. J., et al., 2012. *Measuring gamma ray production from laser-produced pair plasmas*. California–Nevada APS 2012, San Luis Obispo, CA, Nov. 2–3, 2012. LLNL-ABS-588298.

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Volume Collapse: Finger on the Pulse of f-Type Electrons

Magnus Lipp (12-LW-014)

Abstract

We propose to examine a very important correlated-electron phenomenon: electron-driven volume collapse. Volume collapse is related to one of the most fascinating and important properties of f-orbital electrons, which is their ability to transition between localized-like and delocalized-like behavior. This transition is a key element in determining important material properties, including the temperature phase diagram of plutonium, the great power of rare earth magnets, and exotic, condensed-matter effects like heavy fermions, valence-mediated superconductivity, and quantum criticality. We will employ new experimental probes—pressure-dependent advanced x-ray spectroscopy techniques—to distinguish between competing theoretical descriptions of the f-electron delocalization and volume collapse. Direct examination of the microscopic physics of f-electrons promises to create a fertile feedback loop with theory, in direct contrast with the standard paradigm, where theory is often expected to derive f-electron physics from crystallographic structure alone.

If successful, we will provide a systematic description of the delocalization process of f-electrons, in order of increasing pressure, for classes of rare earth materials. Such a description would be an immediate benchmark for the theory of correlated f-electron systems, which is still under development. The project will also offer the x-ray community a new and powerful methodology for addressing questions of electronic structure under pressure. Furthermore, LLNL will have developed the infrastructure

and expertise to examine and explain the electronic structure of more complex rare earth materials, which have important applications that depend on the subtle details of their microscopic electronic states.

Mission Relevance

Because of the strategic importance of fundamental research on rare earth metal systems, this project has relevance to LLNL's nuclear and energy security missions. In the future, this work can be extended to the actinides, where chemical and equation-of-state modeling both depend strongly on understanding intimate details of the microscopic physics of f-electrons.

FY13 Accomplishments and Results

In FY13 we completed, using a newly developed poly-capillary x-ray spectroscopy setup, our non-resonant x-ray emission experiments of all the light lanthanides under pressure from 0 to beyond 50 GPa, starting with lanthanum, praseodymium, neodymium, samarium, and europium—cerium had been investigated previously and all isotopes of promethium are radioactive. In addition, we completed our resonant x-ray emission investigation by studying neodymium and now have a complete set of the first three lanthanides (cerium, prometheum, and neodymium).

Project Summary

After only two years, in collaboration with the University of Washington, we have developed two new kinds of experimental equipment that allow a much faster collection of signals for studying atomic electronic structure under pressure. The poly-capillary-based x-ray spectrometer boasts a twentyfold improvement in signal-to-noise ratio and was mainly used for the normal x-ray emission spectroscopy experiments. The miniature x-ray emission spectrometer, with a factor of six improvement in collection angle, served as the workhorse for the resonant x-ray emission spectroscopy studies. The development of these two techniques has enabled the complete normal x-ray emission spectroscopy study of all light lanthanides under pressure to almost 1 MBar, and the resonant x-ray emission spectroscopic pressure study of the elements prometheum and neodymium. Theoretical calculations modeling these data have definitely ruled out the previously advocated Hubbard–Mott volume collapse mechanism for cerium and established the Kondo volume collapse as the correct theory. While this was expected by dynamical mean field combined with density-functional theory calculations, our experimental data show that the electron occupation in the f orbital and total angular momentum actually track in an opposite fashion to dynamical mean field theory, which needs to be modified significantly. The current efforts are being incorporated into programmatic studies of actinides (5f metals) under conditions of high pressure and temperature, and will provide information about the multi-configurational nature of the f orbitals and the total angular momentum. Both observables are of crucial importance to understanding macroscopic materials properties and will guide and constrain theoretical investigations.

Publications and Presentations

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Lipp, M. J., 2013. *Cerium under high pressure (and temperature): f-electron correlation physics.* LLNL-PRES-635641.

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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 relativistic quantum-field theory of electrodynamics. 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—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 will also mean that the interpretation of the 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 alter our understanding of the fundamental forces of physics. It may thus affect the underpinnings of the physics of many LLNL missions and scientific thrusts, including atomic physics in stockpile stewardship, high-energy chemical compounds with national security relevance, energy manipulation, and materials on demand.

FY13 Accomplishments and Results

In FY13 we (1) successfully optimized the production and trapping of large numbers of lead ions in SuperEBIT, with a newly installed lead sublimation injection system to top off the number of ions in the trap, simultaneous use of two high-resolution spectrometers, and the operation of the SuperEBIT facility at 140-keV energy (an increase of 40 keV); (2) developed improved analysis software to process the spectral data by optimizing the signal-to-noise discriminator across the two-dimensional spectral image; and (3) made accurate calculations for comparison with the spectral data.

Proposed Work for FY14

A five-month delay in optimizing machine alignment meant that the goal of collecting enough photon counts at the level needed to make a definitive measurement was not achieved in FY13. Therefore, we will collect more data in FY14, with the goal of collecting enough photon counts for deriving the QED contribution for determining the two-loop term for behavior of subatomic particles at a tenfold increase in accuracy over what has been previously achieved.

Publications and Presentations

Trabert, E., et al., 2012. *Hyperfine splitting of the* $2s_{1/2}$ *and* $2p_{1/2}$ *levels in Li- and Be-like ions of the open-nuclear shell isotope* ^{141}Pr. LLNL-JRNL-594313.

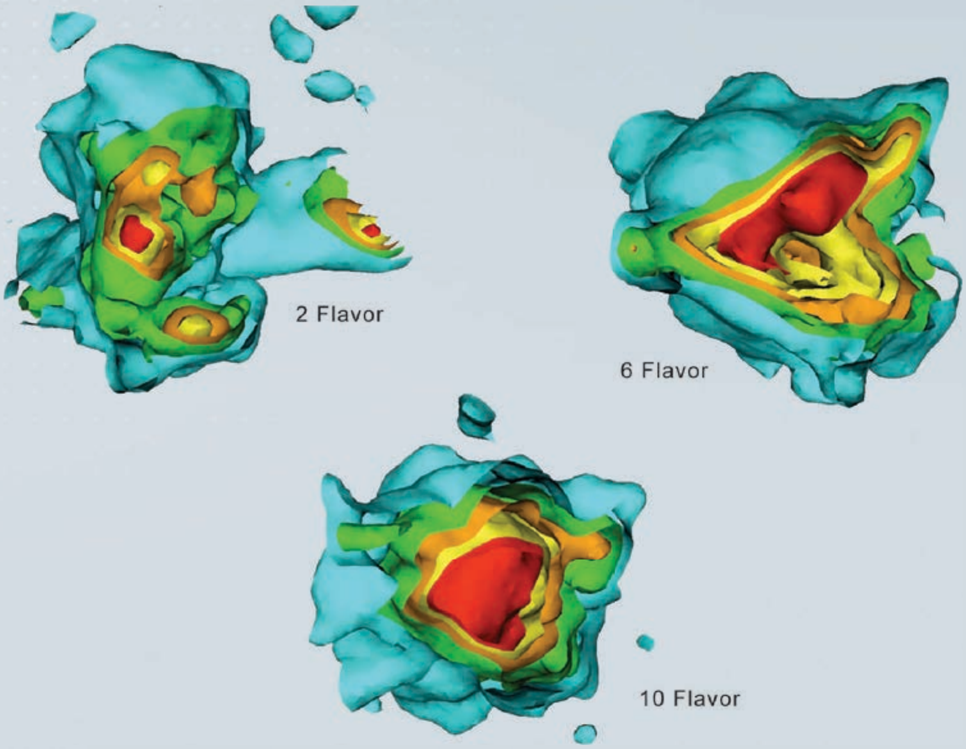
Illuminating the Dark Universe with the Sequoia Supercomputer

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



The neutron electric charge density in our world (2 flavors) as compared to what it would look like in a world with 6 and 10 flavors with a similar but new strong nuclear force from lattice numerical simulations on the BlueGene supercomputer. The neutrons in these simulated worlds may in fact exist as the dark matter that makes up more than 80% of all matter in the Universe. Blue indicates low density, going through green, orange, yellow, and then red for higher densities.

moments, as well as 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.

FY13 Accomplishments and Results

In FY13 we (1) finalized our findings on strong-force dynamics 3-color gauge theory on existing lattice configurations relevant to charge radius and anomalous magnetic moment that can play a significant role in direct detection of composite dark matter; (2) begun studying the 2-color gauge theory that has a candidate Higgs elementary particle (theorized to be the building block of the Universe); (3) generated quantum chromodynamics lattices with various 4-dimensional lattices and heavy 300-MeV pions relevant to nuclear states studies; (4) simulated the 3-color quantum chromodynamics thermodynamic transition on 32-lattice systems with physical 140-MeV pion masses, which provided us with an estimate of the critical temperature and an indication of the transition order; and (5) were selected as 1 of 6 finalists for the Gordon Bell competition on high-performance computing for our topic on the origin of mass, which describes how our work was done using algorithms and code implementation on Livermore's BlueGene/Q supercomputer.

Proposed Work for FY14

In FY14 we will (1) study the 4-color gauge theory on quenched 32-lattice linear sites, calculate the particle spectrum of the theory, identify the dark matter candidate particle, and begin to calculate its properties; (2) simulate the 2-color gauge theory and determine if the light-boson state present is consistent with our current understanding of the Higgs particle; (3) begin simulations of zero-temperature quantum chromodynamics by generating lattices with large linear size of 64 sites and light pions; (4) begin the cross-section calculation of the 4-color theory with current dark matter detectors; and (5) simulate the 3-color thermal transition on very large lattices (64 in the linear dimension).

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 to develop and retain a workforce of highly qualified scientists and engineers, and position the Laboratory for future astrophysics and x-ray optics projects.

Mission Relevance

Participation in space science is a crucial element in LLNL's cyber and space security and intelligence strategic mission thrust. 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 the Laboratory's foundational science in high-energy-density matter.

FY13 Accomplishments and Results

In FY13 we (1) improved the modeling of the optics response for the NuSTAR telescopes by refining and upgrading ray-trace simulations for the mission—specifically, upgrading the simulated NuSTAR optics response by implementing new optical constants derived from the latest metrology measurements, and comparing the results to existing raw calibration data; (2) processed and analyzed observations of neutron stars acquired by NuSTAR; and (3) developed algorithms for neutron star data analysis, and applied those algorithms to data from observations of magnetars (neutron stars with very high magnetic fields) acquired by NuSTAR.

Proposed Work for FY14

For FY14 we will work towards completing and refining the NuSTAR optics models and finalizing and extending the analysis and interpretation of data for different classes of neutron stars in the light of current and emerging models. Specifically, we will (1) implement upgrades to the simulation of the NuSTAR optics response to fully understand the physics of on-ground and in-orbit calibration data, (2) upgrade and extend the algorithms we developed for analysis of neutron stars data to date, and (3) complete the ongoing data analysis and interpretation of currently available NuSTAR data and prepare for new upcoming observations.

Publications and Presentations

An, H., et al., in press. "NuSTAR observations of magnetar 1E 1841-045." *Astrophys. J.* LLNL-JRNL-642086.

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Optimized Diode-Pumped Alkali Lasers for National Security Applications

Thomas Reese (13-ERD-037)

Abstract

Diode-pumped alkali lasers have many advantages over existing laser technologies for national security applications such as directed-energy weapons. Specifically, these advantages translate into a concept for the first operationally viable laser system that can project energy at distances that address current operational missions, while configured to the size, weight, power, and volume constraints of deployable systems. However, key risk-reduction demonstration experiments are needed to prove that the unique merits of this technology can be transformed into operationally suitable systems. We intend to conduct comprehensive rubidium management testing that enables comparison and selection of optimum materials and coatings for the laser system based on experimental data.

With the successful completion of this project, we expect to characterize rubidium management for laser media at the temperatures and pressures needed with the range of materials required to implement the laser system. We will employ a new approach that minimizes the volume and surface area of the system and is optimized for cleanliness and functionality, as well as establish control and response for the system in terms of density, flow rate, and temperature. We will validate the full-scale kinetics model for the diode-pumped alkali laser and evaluate the parasitic processes

such as energy pooling to ensure the efficient operation of a power-scaled system. In addition, we will identify engineering challenges for scaling that may not have been apparent at lower power levels.

Mission Relevance

This project supports the Laboratory's central mission in national security by focusing on rubidium management for a diode-pumped alkali laser system as well as gaining a better understanding of using this type of laser as a directed energy weapon. Diode-pumped alkali lasers offer a promising approach for high-power lasers in military applications that will not suffer from the long logistical trails of chemical lasers or the thermal management issues of conventional solid-state lasers.

FY13 Accomplishments and Results

In FY13 we developed the infrastructure required in three separate laboratories at LLNL to execute the full range of rubidium management experiments—rubidium exposure, in-situ transmission measurement, surface passivation, surface chemistry characterization, and performance evaluation in a laser environment. This infrastructure enables each of these critical characterization experiments as well as allowing the transfer of substrates between passivation, photoelectron spectroscopy, and laser laboratories in vacuum or in an inert gas environment. This capability did not previously exist at the Laboratory, and it enabled us to perform experiments that revealed complex surface chemistry interactions that we must master to develop an operationally suitable alkali laser. Specifically, we developed an initial operating capability for a rubidium laser-media flow system that ensures cleanliness and rapid control. In addition, we performed management diagnostics and experiments for a rubidium laser medium that lay the foundation for all future alkali laser designs.

Proposed Work for FY14

In FY14 we will characterize the susceptibility of different substrates (e.g., materials, surface defects, and surface preparation techniques) to rubidium adsorption. We also propose to determine the approach needed to maximize our contamination design margin for an operationally suitable system.

Specialty Optical Fibers for Unique National Missions

Michael Messerly (13-ERD-040)

Abstract

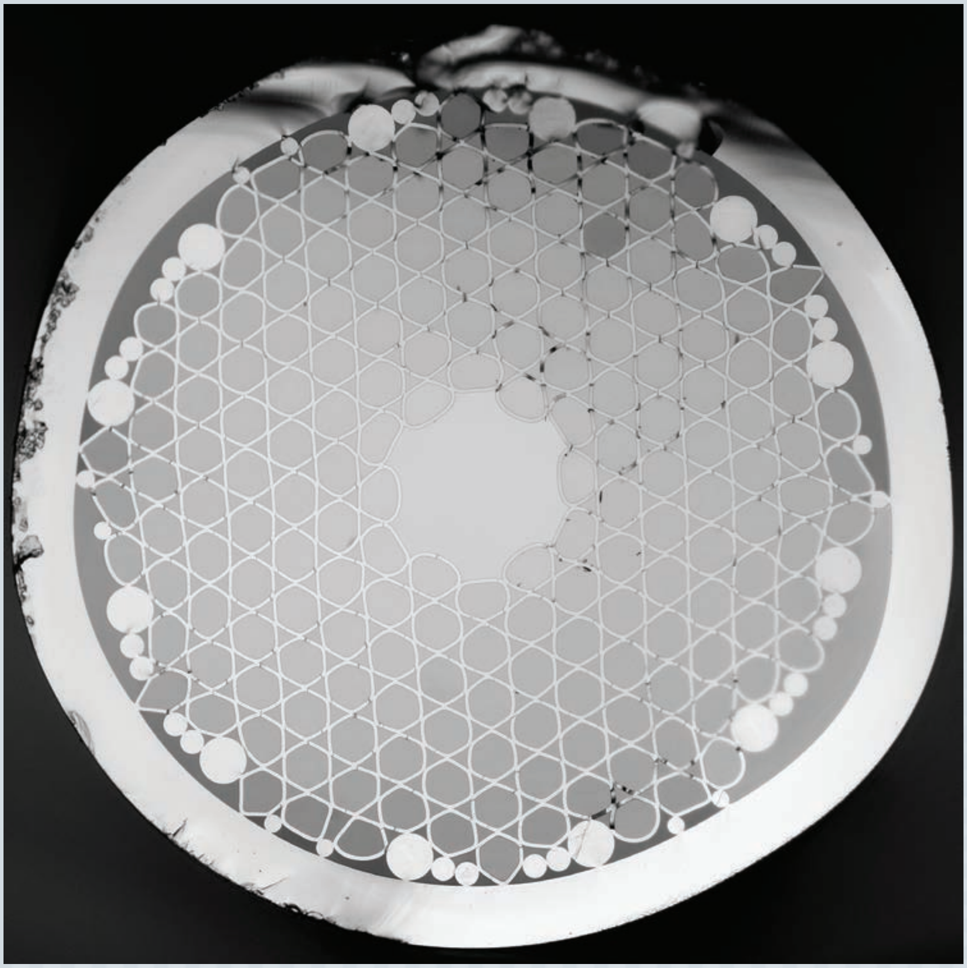
Our objective is to fabricate photonic crystal fibers for mission-relevant applications, including specialty fibers able to transport the beams of extremely high-power lasers (100 kW to 1 MW) and hollow-core fibers to accelerate and guide charged particles for laser-based particle accelerators or to enhance the interaction of photons with accelerated ions. Such fibers, although critical to Laboratory missions, are not attractive to commercial vendors because of difficulty, cost, or time to market. We will

use LLNL's new fiber draw tower to produce fibers to advance development of alkali-vapor lasers, laser-based particle accelerators, x-ray and high-harmonic sources, sensors for national security, and systems for transporting ultraviolet light from test shots to sensors at the National Ignition Facility.

We expect to produce fibers able to transport megawatt-laser power, capable of withstanding neutron and gamma-ray environments, able to detect nuclear species, and capable of advancing the state of the art in laser-based particle acceleration. More specifically, we will produce optical fibers relevant to high-power lasers for missile defense and future laser-based particle accelerators; the radiation-hardened transport of ultraviolet light at the National Ignition Facility; hollow-core fibers for lasers, high harmonic generation, and sensors; and field-flattened fibers for the transport of high-power beams, large pulse energies, and particle acceleration.

Mission Relevance

This project is relevant to Livermore's mission focus area of advanced laser optical systems and applications, in particular to missile defense and nuclear detectors in support of national security, as well as applications supporting the energy security mission, such as fusion energy research.



We fabricated a hollow-core optical fiber prototype that could be used to accelerate and guide charged particles for laser-based particle accelerators or to enhance the interaction of photons with accelerated ions.

FY13 Accomplishments and Results

In one month in FY13 we modeled and fabricated our first hollow-core optical fiber.

Project Summary

For this one-month research effort, we fabricated LLNL's first hollow-core optical fiber, which led to interest from agencies in the Departments of Defense and Energy for transporting high-average-power lasers and high-peak-power pulses, as well as from three private medical companies and two aerospace companies. The follow-on effort will be supported by work-for-others funds from outside interests.

Publications and Presentations

Messerly, M. J., et al., 2013. *Optical fibers having field-flattened, ring-like higher order modes*. LLNL-JRNL-629312.

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 material 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 the 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 interactions with multilayers 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–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 the safety of the country through detection of special nuclear materials.

FY13 Accomplishments and Results

In FY13 we (1) performed experiments at the European Synchrotron Radiation Facility to constrain the physics of photon–multilayer interactions in the 100- to 644-keV energy range, specifically the way that Compton scattering and other processes influence the performance of hard x-ray optics; (2) successfully demonstrated the first-ever multilayer reflectivity above 500 keV, at 508 and 644 keV; (3) performed Monte Carlo simulations of our experimental setup to quantify background contribution at both energies and completed the main data analysis; (4) developed the first predictive model of multilayer reflective properties, taking into account depth-grading and surface uniformity data; (5) identified a type of polycarbonate as a promising substrate and investigated chemical and heat-based surface-smoothing techniques, although surface metrology indicated that the surface characteristics in the mid-spatial frequency range might not be sufficient for use in hard x-ray optics; and (6) focused on demonstrating the feasibility of a new glass slumping process and interferometric measurements to assess figure error.

Proposed Work for FY14

In FY14 our goal is to (1) extrapolate results from prototype optics into full systems with realistic multilayer coatings, (2) compute the expected response of the optics, (3) design instruments and techniques for a selected nuclear security application, (4) compute the notional performance of the system, and (5) determine its utility, which will be measured by its ability to meet requirements and the practical constraints that will affect its use.

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.

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.

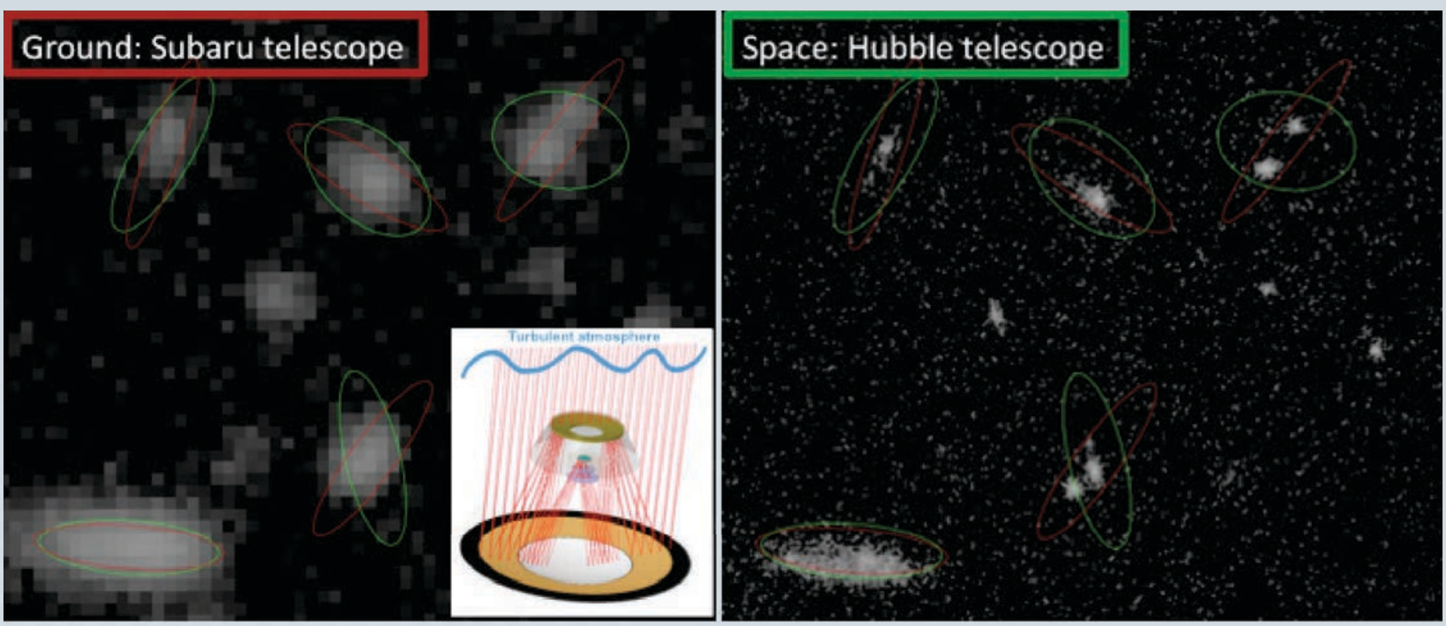
Measuring Dark Energy with the Large Synoptic Survey Telescope

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



The Large Synoptic Survey Telescope is sensitive to the effects of dark energy through the gravitational lensing distortion of galaxy shapes. But the true shapes as seen from space (right) often look different from the ground (left) after they are blurred by atmospheric turbulence and optical distortions in the telescope. The red ellipses show the space-based shapes to compare with the ground-based shapes shown by the green ellipses. Our simulations are allowing us to invert the measurement process to infer the true galaxy shapes from ground-based observations.

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. We expect to establish LLNL as a leader in the 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 high-performance computing and simulation foundation in science, technology, 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.

FY13 Accomplishments and Results

In FY13 we (1) recruited a postdoctoral researcher for the project with experience in image analysis for gravitational lensing measurements; (2) modified and tested image simulation codes for the LSST field of view and local atmospheric conditions, including a comparison of high-fidelity LLNL algorithms with the LSST project image simulator; (3) analyzed the point-spread function size, shape, centroid, and wavelength-dependent variations for the LSST field of view leading to metrics for the successful application of fast numerical approximations; (4) characterized requirements for the active optics wave-front control driven by weak lensing measurements, which could introduce low-order aberrations affecting the interpolation of point-spread function shapes; and (5) planned the statistical framework to incorporate our high-fidelity point-spread function models in galaxy shape estimators, including potential fitting and interpolation errors.

Proposed Work for FY14

In FY14 we will (1) enhance the simulation of active optics wave-front control in LSST; (2) improve algorithms for correcting the perturbed wave front based on measurements from the wave-front sensors; (3) develop new atmosphere simulation capabilities that can generate arbitrarily large phase screens without frozen flow approximations; (4) implement our statistical galaxy shape-estimation methods, incorporating arbitrarily complex point-spread function models relevant to LSST; and (5) compare and validate our galaxy shape estimation with ground- and space-based observations.

Publications and Presentations

Dodelson, S., and M. D. Schneider, 2013. *Simulation covariance error.* LLNL-JRNL-632261.

Morrison, C., and M. D. Schneider, 2013. "On estimating cosmology-dependent covariance matrices." *J. Cosmol. Astropart. Phys.* **2013**(11). LLNL-JRNL-635596-DRAFT.

Schneider, M. D., 2013. *An optimal estimator for cosmological cross-correlation functions.* LLNL-JRNL-644558-DRAFT.

Schneider, M. D., et al., 2013. *Efficient cosmological simulation frameworks for parameter estimation*. American Astronomical Soc. 221st Mtg., Long Beach, CA, Jan. 6–10, 2013. LLNL-POST-609413.

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—that is, the behavior of matter under a given set of physical conditions—at these pressures has never been measured for any material. We propose to measure the equation of state of matter at greater than gigabar pressures for the first time at the 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 National Ignition Campaign 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 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 basic science. The new x-ray scattering capability at NIF that will result from this project can be employed on NIF shots to provide more information on target ablator implosion dynamics, which is important for achieving ignition.

Mission Relevance

Our high-energy-density experiments and development of a new x-ray scattering technique closely aligns with Laboratory missions in stockpile stewardship, energy security, basic science, and understanding giant gas planets and highly evolved stars.

FY13 Accomplishments and Results

In FY13 we (1) completed radiation-hydrodynamic simulations of polystyrene target conditions using the Laboratory’s equation-of-state tables, (2) performed research and development and completed the fabrication of coated diamond spheres to be used in FY14, (3) completed the conceptual design of a multi-angle scattering spectrometer for probing the ionic structure of material at extreme conditions and tested the lithium–

fluoride crystal to be used, (4) developed an analysis technique to extract opacity and density simultaneously from radiographic data and applied it to experimental data, and (5) completed two successful shots on NIF and analyzed the results.

Proposed Work for FY14

In FY14 we will (1) complete radiation-hydrodynamics simulations of solid diamond and carbon–deuterium targets using the revised ignition platform and the indirect-drive exploding pusher platform, (2) fabricate solid carbon–deuterium targets, (3) continue the development effort for coated solid diamond targets, (4) complete design of a modified NIF spectrometer for imaging x-ray Thompson scattering (elastic scattering of electromagnetic radiation by charged particles), and (5) complete analysis for use of radiography marker layers to constrain equation-of-state measurements.

Publications and Presentations

Doeppner, T., 2013. *Developing x-ray Thomson scattering for the NIF*. LLNL-PRES-637813.

Doeppner, T., 2013. *X-ray Thomson scattering for characterization of dense plasma states in high-energy-density physics experiments on large-scale laser facilities.* 2013 Users Meeting—Advanced Photon Source, Center for Nanoscale Materials, Electron Microscopy Center, Chicago, IL, May 6–9, 2013. LLNL-PRES-639680.

Doeppner, T., and R. Falcone, 2013. *EOS experiments at Gbar pressures on the NIF.* LLNL-PRES-609802.

Doeppner, T., and L. Fletcher, 2013. *Tracking megabar shock waves to coalescence with an x-ray laser*. 8th Intl. Conf. Inelastic X-Ray Scattering, Menlo Park, CA, Aug. 11–16, 2013. LLNL-ABS-637079.

Doeppner, T., et al., 2013. *Equation of state measurements of CH plastic at Gbar pressure.* 55th Ann. Mtg. APS Division of Plasma Physics, Denver, CO, Nov. 11–15, 2013. LLNL-ABS-640933.

Doeppner, T., et al., 2013. *X-ray Thomson scattering for characterization of dense plasma states in high-energy-density physics experiments on large-scale laser facilities.* LLNL-PRES-636158.

Hawreliak, J. A., et al., 2013. *Gbar equation of state measurements in convergent shock experiments.* AGU Fall Mtg., San Francisco, CA, Dec. 10–13, 2013. LLNL-ABS-577793.

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Kritcher, A., et al., 2013. *EOS measurements of extreme Gbar pressures at the NIF.* LLNL-PRES-558691.

Kritcher, A., et al., 2013. *Probing matter at extreme Gbar pressures at the NIF.* 4th Intl. Conf. High Energy Density Physics, Saint-Malo, France, June 25–28, 2013. LLNL-JRNL-575292.

Kritcher, A., et al., 2013. *Probing matter at Gbar pressures at the NIF.* 8th Intl. Conf. Inertial Fusion Sciences & Apps. (IFSA 2013), Nara, Japan, Sept. 8–13, 2013. LLNL-PRES-644639.

Why Is Nuclear Matter So Red?

Lee Bernstein (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 to explain the 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, 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 science, technology, and engineering foundation for fusion and high-energy density matter.

FY13 Accomplishments and Results

In FY13 we (1) procured equipment to enable experiments for mapping enhancements in the low-energy part of the radiative strength function of nuclear matter, (2) tested this equipment and shipped it to iThemba Laboratory for Accelerator Based Sciences in South Africa, where it will be used in an approved experiment in early 2014; and (3) recruited a University of California, Berkeley undergraduate to analyze a data set from the University of Oslo to measure the level density and radiative strength function in lanthanide-139.

Proposed Work for FY14

In FY14 we will participate in a follow-on experiment studying the radiative strength function at collaborators' laboratories that will be analyzed by graduate students at those facilities. In addition, we will procure a high-resolution gamma-ray detector for use at both collaborating institutions.

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.

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 the structures of the ordered phases have been obtained experimentally.

Over the last decade, first-principles computer simulations have been crucial in correctly interpreting 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 the molecules in the 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 LLNL missions in stockpile stewardship and high-energy-density foundational science. 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.

FY13 Accomplishments and Results

In FY13 we (1) used a combination of quantum Monte Carlo methods with advanced density-functional theory and path-integral methods to calculate the free energy of the most important structures of both solid molecular and atomic hydrogen, at a single temperature of 200 K, allowing us to determine the correct structure of the solid as a function of pressure; (2) calculated the melting line of the atomic solid at pressures beyond those needed to achieve the dissociation transition, finding that nuclear quantum effects are critical in this transition and leading to a reduction in the melting line from about 300 to 325 K for classical particles to temperatures below 200 K at pressures above 600 GPa; and (3) calculated the optical properties of solid hydrogen.

Proposed Work for FY14

In FY14 we will (1) use the structural information obtained in FY13 to calculate solid–solid and solid–liquid phase transitions as a function of pressure and temperature in the pressure range of 300 to 750 GPa and in the temperature range of 0 to 600 K, (2) explore the regime of the phase diagram where the metal–insulator transition in the liquid meets the melting line of the solid and the liquid–liquid first-order transition, and (3) use semi-classical quantum dynamical methods based on path integrals to calculate vibrational spectra (infrared and Raman) for quantum ions and compare to experimental results.

Publications and Presentations

McMahon, J. M., et al., 2013. *Atomic hydrogen: A ground-state fluid or a metallic solid?* LLNL-JRNL-641058.

Morales, M. A., et al., 2013. "Toward a predictive first-principles description of solid molecular hydrogen with density-functional theory." *Phys. Rev. B* **87**, 184107. LLNL-JRNL-612081.

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, we can manipulate the rate of spontaneous emission. Our main goal 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 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.

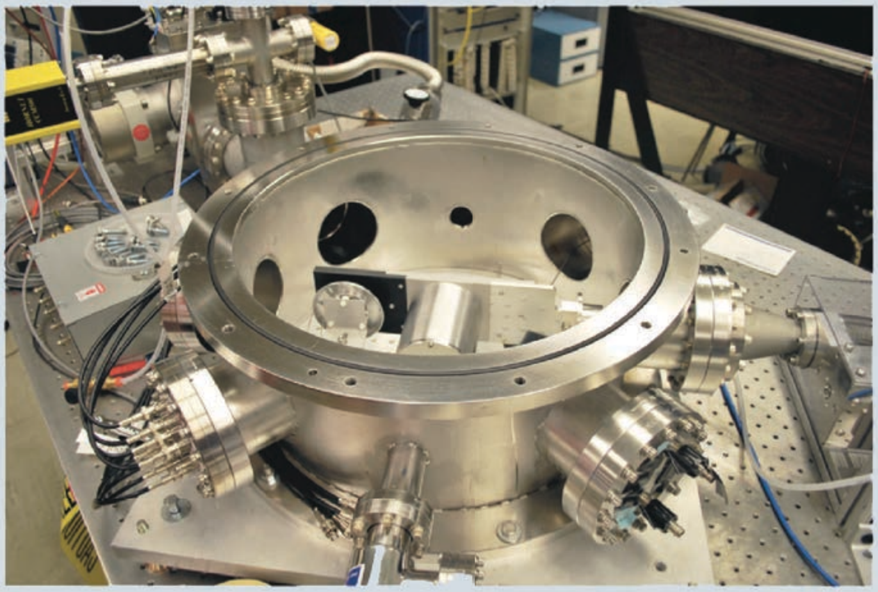
We expect to modify the decay rate of the uranium-235 isomer by up to 60%, extending the 26-minute half-life of the isomer to over an hour. This will represent a modification over ten times larger than previous experiments attempting to alter nuclear decay rates. A successful measurement will open the 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 will 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.

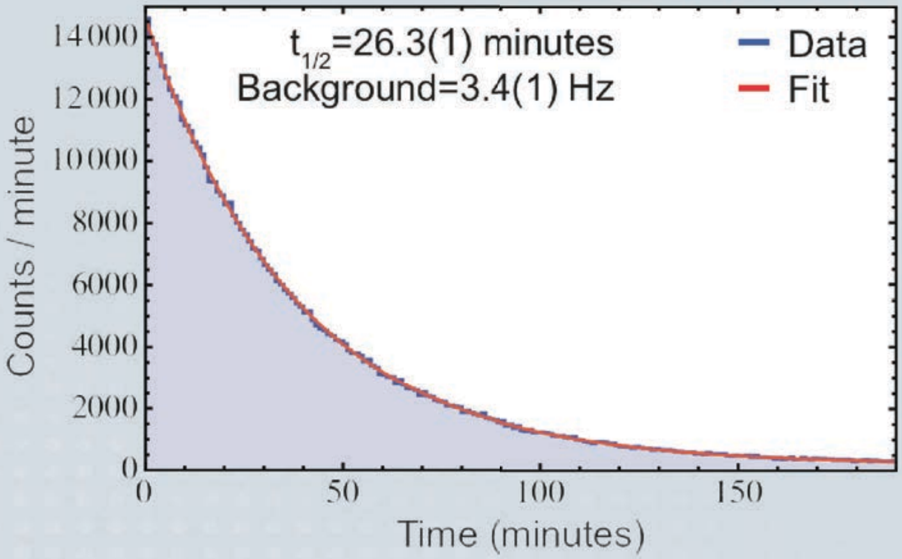
Mission Relevance

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.

FY13 Accomplishments and Results

In FY13 we (1) hired a postdoctoral researcher; (2) constructed the experimental setup for measuring bulk dielectric suppression of the uranium-235 isomer, including a vacuum chamber with vacuum pumps, a micro-channel plate for electron detection, a vacuum translation stage, and a pressure controller for maintaining a specific pressure of argon; (3) determined that pressing wafers together would not be adequate for simulating a bulk material because of dust particles that exist even in clean environments; (4) chose liquid mercury to replace one of the wafers because it should conform to the surface of the solid wafer—although isomer losses during mercury exposure complicate the half-life analysis, decay measurements of the isomer yielded promising results; (5) produced





Vacuum chamber, where the uranium-235 isomer measurements take place (top). The long paddle-shaped object in the center of the chamber is attached to the translation arm, which transports the platinum-coated wafer between the plutonium-239 source (long aluminum cylinder) and the micro-channel plate electron detector (aluminum disk). A uranium-235 isomer decay curve (below), measured after electrostatically drifting plutonium-239 alpha decay recoils through argon, into the platinum-coated wafer. The data is binned in one-minute intervals and fit with an exponential decay curve and a flat background.

uranium-235 isomers from alpha decay of a plutonium-239 source that was created by the LLNL radiochemistry group; and (6) used silicon wafers coated with a thin film of platinum to collect the uranium-235 isomer—platinum was used rather than molybdenum to prevent amalgamation during contact with mercury.

Proposed Work for FY14

In FY14 we will (1) develop and create photonic stop bands (a spectral range of large reflectivity), by pressing together ultraflat molybdenum and silicon multilayer mirrors under vacuum; (2) tune these mirrors to the specific energy of the uranium-235 isomer; (3) adjust the decay rate by varying the thickness of the silicon layer between the two mirrors; (4) use multilayer mirrors created by LLNL's x-ray science and technology group to create a dielectric micro-cavity structure; (5) use the vacuum system that we developed in FY13 to take necessary measurements; and (6) measure bulk suppression to more precisely determine the modified spontaneous emission rates caused by different materials.

A Compact, Femtosecond Hard X-Ray Source for Materials Characterization and High-Energy-Density Science

Felicie Albert (13-LW-076)

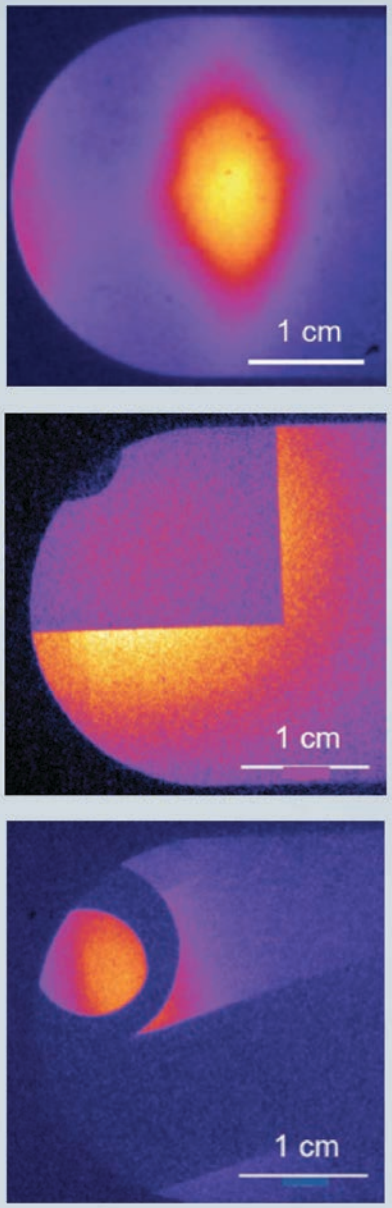
Abstract

New x-ray techniques are a critical tool for investigating materials related to energy conversion, high-energy-density science, and manufacturing. With a new generation of ultrafast x-ray sources, partnered with novel pump and probe tools and capabilities, we can gain insight into material 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 sources' potential as a new x-ray capability.

We expect to create a new ultrafast x-ray source capability at Livermore that is broadband, well collimated, and has femtosecond pulse widths. 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 µm. 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 mission to create an integrated center for the application of advanced lasers to solve 21st-century national security challenges. It leverages Livermore's unique expertise in accelerator, laser, and x-ray sciences and will strengthen our 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.



Betatron x-ray radiographs obtained at the Jupiter Laser Facility in Livermore reveal images of a beam profile (top), silicon wafer (center), and gold ignition hohlraum target capsule (bottom).

FY13 Accomplishments and Results

In FY13 we (1) developed the experimental x-ray source using the Callisto laser; (2) performed two experiments to measure such properties as broadband spectrum up to 80 keV with a sub-5-µm source size and about 10^8 photons per pulse; (3) developed a code calculating electron trajectories and their betatron radiation in the plasma, which agrees with experimental results; (4) observed betatron radiation up to 80 keV with image plates and spectrometers; (5) developed a code (Fresnel diffraction formalism) to analyze phase contrast images obtained with the source; (6) radiographed materials including silicon wafer and tantalum thin-film targets at energies of more than 10 keV; and (7) secured beam time at the Jupiter Laser Facility at LLNL and Astra-Gemini at Rutherford Appleton Laboratory in the United Kingdom to successfully accomplish our FY14 tasks.

Proposed Work for FY14

In FY14 we will (1) begin using the new x-ray source for high-energy-density science applications; (2) perform an experiment at the Jupiter Laser Facility to characterize the source using a longer-pulse and higher-energy laser and to probe a laser-driven shock using the x rays; (3) explore the possibility of future betatron x-ray source development on large facilities, including the OMEGA laser in Rochester, New York, and the Advanced Radiography Capability at Livermore; (4) collaborate with the SLAC National Accelerator Laboratory in Menlo Park to develop pump and probe extended x-ray absorption fine-structure experiments using our x-ray source; and (5) collaborate with Rutherford Appleton Laboratory in the United Kingdom to perform x-ray phase-contrast imaging experiments using our source at the Livermore Gemini laser.

Publications and Presentations

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Albert, F., et al., 2013. "Angular dependence of betatron x-ray spectra from a laser wake." *Phys. Rev. Lett.* **111**, 235004. LLNL-JRNL-642092.

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Albert, F., et al., 2012. *High energy betatron x-rays in the ionization-induced trapping regime.* 54th Ann. Mtg. APS Division of Plasma Physics, Providence, RI, Oct. 29–Nov. 2, 2012. LLNL-PRES-599061.

Albert, F., et al., 2013. *Imaging electron trajectories in a laser-wakefield accelerator by measuring the betatron x-ray spectrum angular dependence.* 55th Ann. Mtg. APS Division of Plasma Physics, Denver, CO, Nov. 11–15, 2013. LLNL-ABS-641274.

Albert, F., et al., 2013. *Multi-keV betatron x-ray source development at the Jupiter Laser Facility.* LLNL-POST-617452.

Albert, F., et al., 2013. *Tomographic reconstruction of electron trajectories in a laser–plasma accelerator using betatron x-ray radiation.* LLNL-POST-644261.

Enhancing Laser-Driven Ion Beams by Self-Guiding of Intense and Ultrashort Laser Pulses in Plasma

Yu-Hsin Chen (13-FS-006)

Abstract
Generating high-energy, high-quality proton and ion beams is important to LLNL'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 a 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 of the laser system. It can be leveraged towards the development of highest-energy proton and ion sources on future large-scale laser facilities at LLNL.

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 advanced accelerator technologies for lasers and the study of properties of matter under extreme conditions of temperature and pressure in high-energy-density matter science.

FY13 Accomplishments and Results

In FY13 we (1) 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; (2) conducted an experiment at Livermore's Jupiter Laser Facility using the Callisto laser; (3) set up diagnostics for measuring target normal sheath acceleration proton-beam characteristics, including radiochromic film stacks, a Thomson parabola ion spectrometer, and imaging plates; (4) estimated the self-guiding length in the gas cell for optimized contrast enhancement to be about 4 mm and compared it to the experiment—beam emission characteristics were improved using our new design; and (5) performed preliminary data analysis with a detailed comparison to simulations.

Proposed Work for FY14

Considering the promising results from preliminary analysis of FY13 experimental data, in FY14 we propose to (1) complete our data analysis, (2) extend the scaling of proton energy to foil thickness to the entire proton spectrum, (3) determine why proton-beam energy is independent of foil thickness for guided laser pulses, and (4) determine if this method is a feasible approach for contrast enhancement in relativistic laser interactions with matter, and publish the results in a peer-reviewed journal.

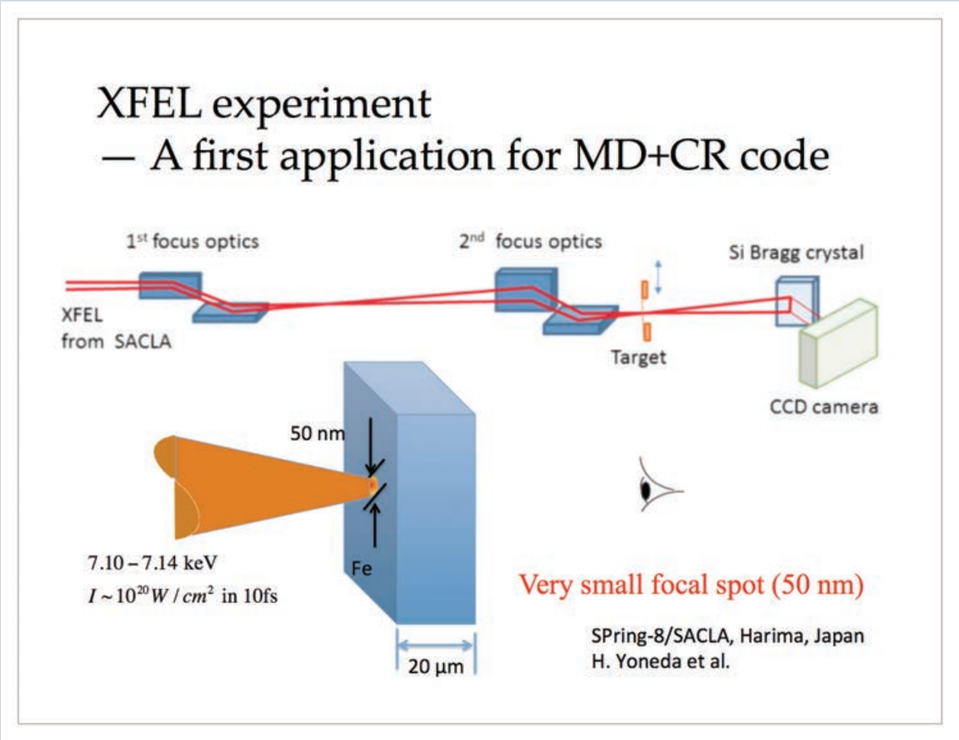
Coupled Computer Code for Modeling Ultrahigh-Intensity X-Ray Interactions with Matter

John Barnard (13-FS-011)

Abstract

We propose to investigate the feasibility of combining molecular dynamics particle simulation with an atomic kinetics model based on radiation transport and collisional processes to explain a variety of new high-intensity x-ray laser and matter interaction experiments. The SPring-8 electron synchrotron in Harima, Japan, is currently being used for experiments that produce powerful beams of kilovolt x rays with a thousandfold-higher intensity than the nearest competitor in a short-pulse, high-repetition-rate facility. Currently, there is no model that can accurately represent these experiments. The need for modeling will grow rapidly as other laboratories continue improving their x-ray sources. A new level of control over x rays will open up a world of possible applications. For example, the development of visible lasers led to new ways to probe materials, to make high-precision measurements, to heat and process materials, to read and record stored information, and even to fundamentally change manufacturing processes. We propose to adapt our existing coupled molecular dynamics and atomic kinetics code based on collisional radiative processes to treat iron inner-shell excited states and compare our results to experiments at SPring-8.

We expect our calculations will provide the first theoretical model for the frontier science experiment at the SPring-8 free-electron laser facility in which the x rays burn through thin iron foils and produce a remarkable "hollow atom" state where most



Particle simulations of high-intensity free-electron laser interaction with an iron target were performed using 20,000 iron atoms, with 48 inner-shell excited states for each atom. The simulations predict nonlinear transmission ("bleaching") and inversion and gain for the iron K-alpha emission at 6.4 keV.

atoms have one electron removed from the innermost K shell. Recently, modeling efforts have focused on combining molecular dynamics simulations with atomic kinetic models of the sort originally used for x-ray laser experiments, which pose various difficulties and have not produced a satisfactory computer code that can accurately represent the SPring-8 experiments. Our work will form the basis for long-term collaboration with future x-ray laser experiments underway around the world and reclaim an LLNL leadership role in x-ray laser science. Applications for high-intensity x rays include research into dense plasmas not transparent to visible radiation, as well as x-ray microscopy, medical imaging, material surface research, and weaponry.

Mission Relevance

Modeling of ultrahigh-intensity x-ray interaction with matter is central to understanding physical processes related to astrophysics and high-energy-density science. This, in turn, supports Laboratory strategic focus areas in stockpile stewardship science and the core competency in high-energy-density science.

FY13 Accomplishments and Results

In FY13 we (1) wrote several new computer codes that combine molecular dynamics particle simulation with nonequilibrium atomic kinetics, (2) applied these codes to successfully match ultrahigh-intensity x-ray laser interaction experiments performed at the SPring-8 free-electron laser x-ray light source, and (3) found that the new phenomena observed in our simulations (and in the experiments) are nonlinear transmission (bleaching) and x-ray laser amplification by iron or copper foil targets.

Project Summary

New free-electron laser x-ray sources have opened the door to a new regime of ultrahigh-intensity (10^{20} W/cm^2) x-ray interactions with solid and plasma targets. Our project was designed to demonstrate the feasibility of particle simulation with atomic kinetics on the moving atoms to model this new type of experiment. Having a validated modeling tool would enable LLNL scientists to develop an applied-science research program using high-power short-pulse multiple-kiloelectronvolt x rays. A logical next step involves assembling a team from among the many LLNL scientists with x-ray laser expertise to design and prepare experiments using the new x-ray sources, especially at the Linac Coherent Light Source in Menlo Park. Our computer codes would be important tools for designing credible experiments.

For a PDF of the *FY 2013 Laboratory Directed Research and Development Annual Report,* scan the QR code below.

or go to the Tri-Lab LDRD Website by scanning the QR code below.

LDRD

Laboratory Directed Research and Development

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Lawrence Livermore National Security LLC