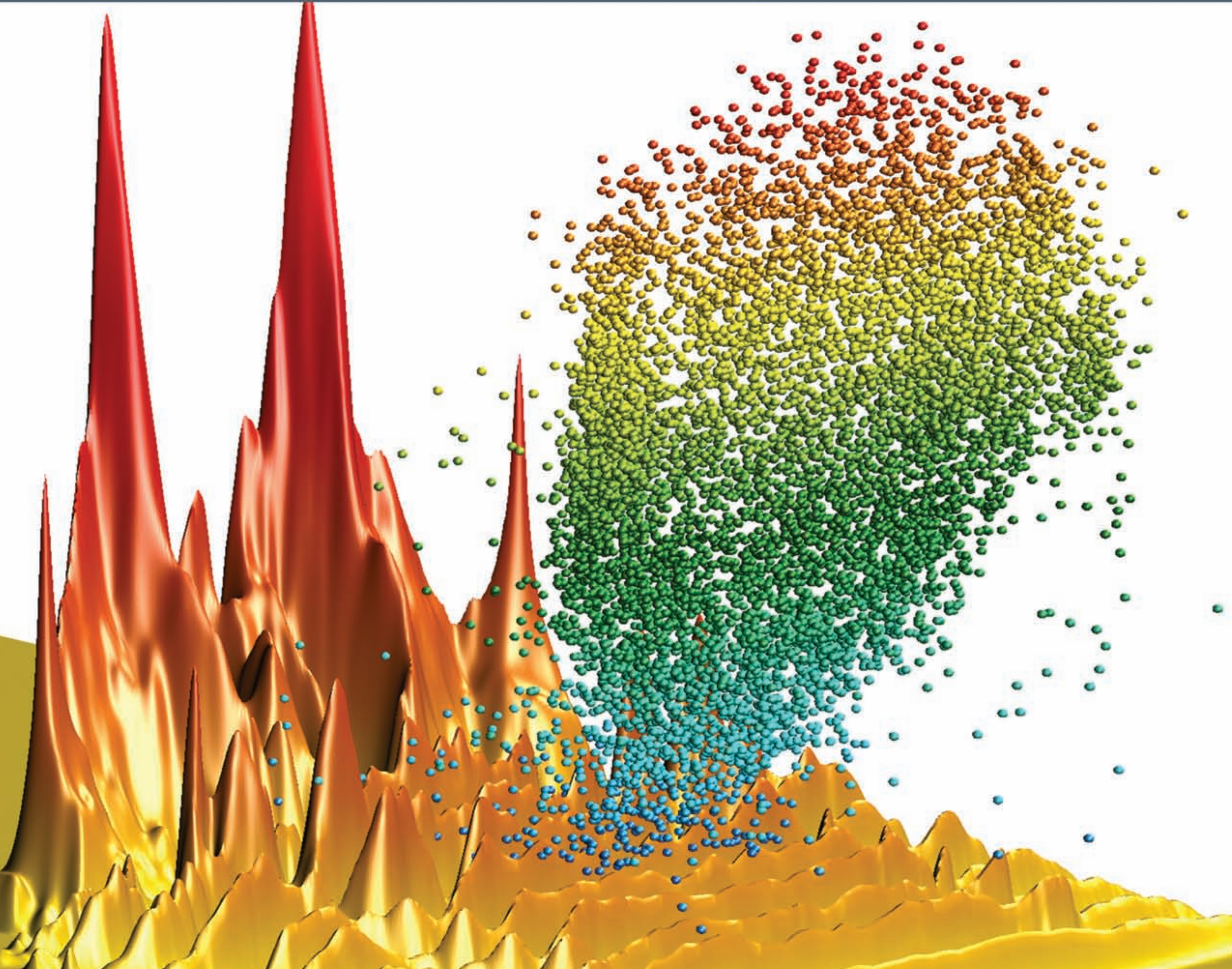
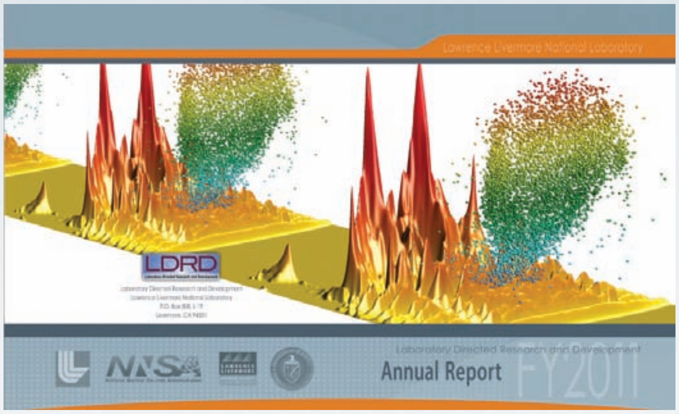
Lawrence Livermore National Laboratory



Laboratory Directed Research and Development

Annual Report FY'2011



About the Cover:

The 2011 Laboratory Directed Research and Development project depicted on the cover, "An Intense Laser-Based Positron Source" (10-ERD-044), seeks to further develop the world's most advanced direct hot-electron diagnostic for high-energy-density physics research. Principal investigator Scott Wilks is laying the science foundation for a new and potentially powerful source of positrons (antimatter counterparts of the electron), which may lead to new approaches for diagnosing experiments in plasma and atomic physics, fusion science, high-energy-density physics, accelerator and particle-beam science, nondestructive interrogation of materials, and astrophysics. These experiments are foundational to the nation's Stockpile Stewardship Program, which is responsible for maintaining confidence in the country's nuclear deterrent in the absence of nuclear testing. The figure is part of an integrated computer simulation that shows, for the first time, all the physical effects believed to occur during the production of electrons and positrons using ultra-intense laser beams. Research to date has led to a provisional patent on a novel positron source for conventional and plasma accelerators.

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Acknowledgments

The Laboratory Directed Research and Development Program extends its sincere appreciation to the principal investigators of fiscal year 2011 projects for providing the content of the annual report. The program also thanks the following for their many contributions to this publication: Barbara Jackson, administrator; Steve McNamara, computer specialist; Iris Lam, resource manager; and Kristen Croteau, business manager. We extend particular thanks to Kenneth Jackson, who successfully directed the Laboratory Directed Research and Development Program as acting director for much of fiscal year 2011.

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Director's Statement

The Laboratory Directed Research and Development (LDRD) Program is the largest single source of internal investment in our future and continues to be critically important for the health of our Laboratory. The LDRD budget of $96.6 million for fiscal year 2011 supported 142 projects. These projects were selected through an extensive peer-review process to ensure the highest scientific quality and mission relevance.

The LDRD projects are aligned with the Laboratory's strategic roadmap 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. Some of these core competency areas—consistent with the science, technology, and engineering pillars as defined in the five-year strategic roadmap—include, but are not limited to:
  - High-energy-density science
  - High-performance computing and simulation
  - Biological science and biotechnology
  - Laser science and optical materials
  - Material properties, theory, and design
  - Radiochemistry and nuclear science
  - Information and network science
  - Extreme measurements
- Developing programs in strategic focus areas, guided by the strategic roadmap, where Laboratory innovation can make an important difference.
- Looking beyond the immediate challenges to future opportunities.

The LDRD Program was conceived as a way for our Laboratory to take on bold initiatives. Scientific and technical risks are essential attributes of a portfolio that expands the 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. Many Laboratory 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.

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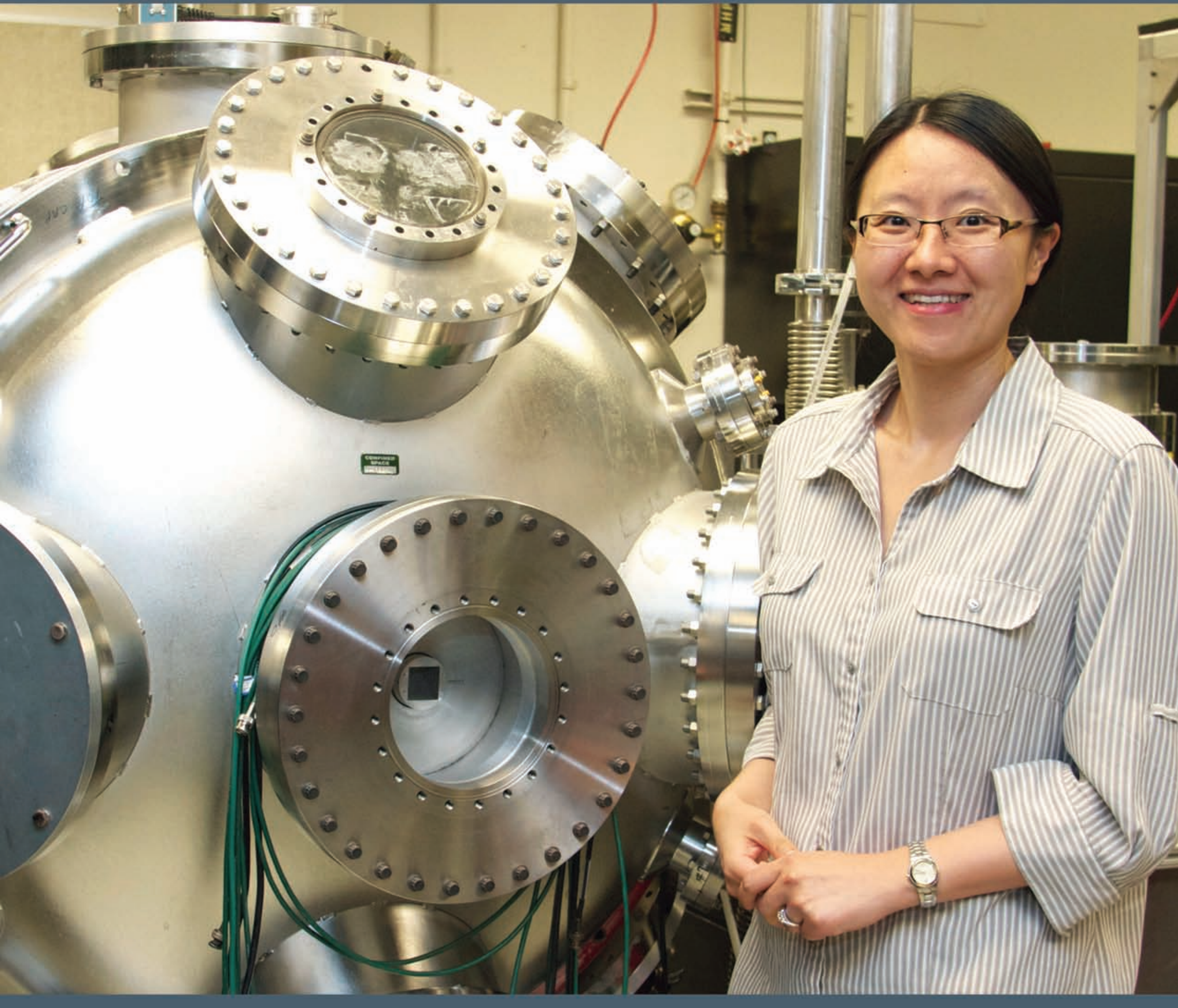


Laboratory Directed Research and Development

FY2011

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 national 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 *FY2011 Laboratory Directed Research and Development Annual Report*

The LDRD annual report for fiscal year 2011 (FY11) 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:

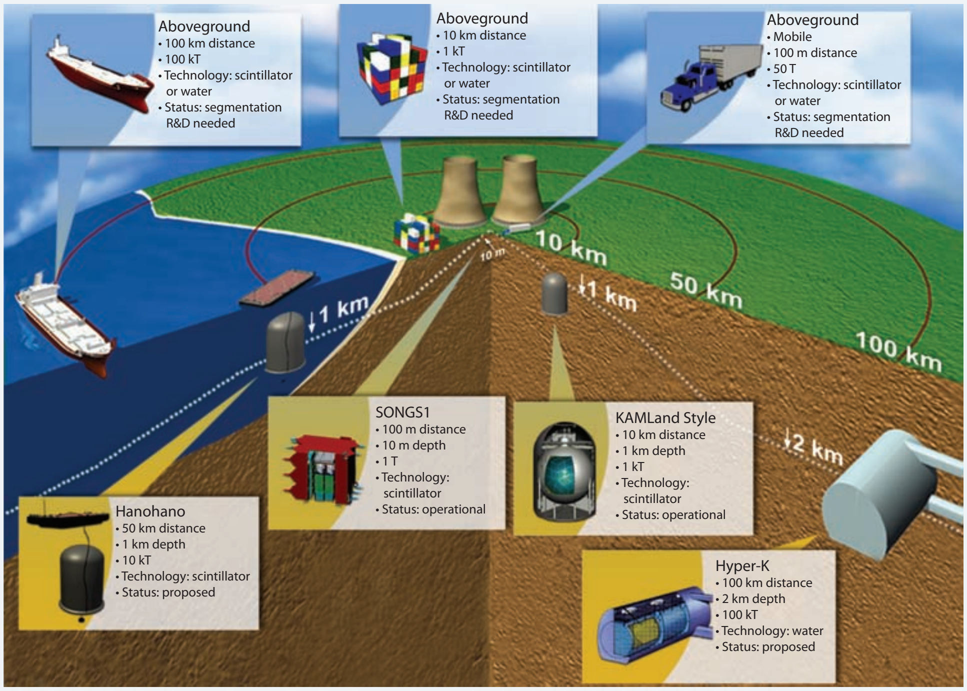
A summary of each project, submitted by the principal investigator. Project summaries include the scope, motivation, goals, relevance to DOE/NNSA and LLNL mission areas, the technical progress achieved in FY11, and a list of publications that resulted from the research.

Highlights of Accomplishments for the Fiscal Year

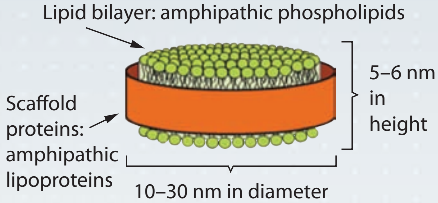
In FY11, the LDRD Program at LLNL continued to be extremely successful in supporting research at the forefront of science, providing new concepts for core missions, and creating an exciting research environment that attracts outstanding young talent to the Laboratory. Wide-ranging projects for this fiscal year exemplify LDRD's noteworthy research in support of the Laboratory's five-year strategic *Roadmap to the Future*, as well as for critical national needs.

Nuclear Threat Reduction

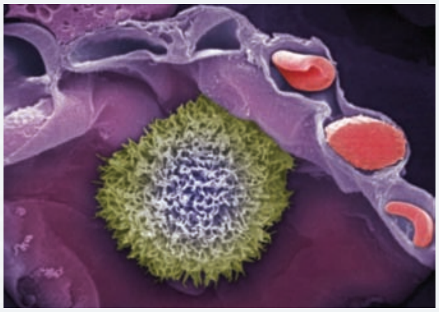
Remote nuclear reactor monitoring using antineutrino detectors could revolutionize global nonproliferation efforts by providing continuous and near-real-time monitoring of plutonium production at its source. An LDRD project is laying the foundation for a program to develop ultrasensitive particle detectors for global nuclear security (10-SI-015). This project integrates the high-profile fundamental science of dark matter and neutrino physics with the very practical research required to achieve breakthroughs in reducing the global nuclear threat. Investigators are collaborating on five international, next-generation physics experiments to enable discovery of operating reactors across borders, as well as provide reactor safeguards and detection of noncritical special nuclear materials that could be used in terrorist threats. To date the team has completed aboveground deployment of the world's largest xenon detector and achieved the first-ever operation of a 1-kg argon dual-phase detector to ultimately detect coherent particle scatter using a reactor neutrino source.



Biosecurity



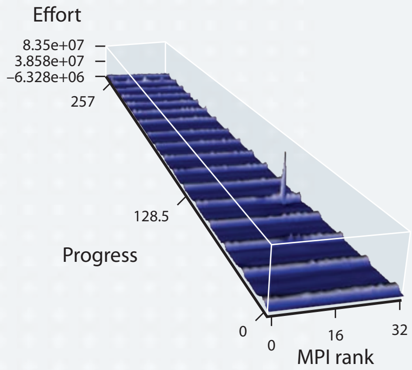
The LDRD Program early on recognized the need for research into countering biosecurity threats, and has supported numerous projects to this end. In FY11, researchers investigated host immunomodulation to rapidly enhance host immune responses to effectively prevent or clear pathogen infection (11-ERD-016). They are developing a novel delivery platform using nanometer-scale lipoprotein technology. 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. The team has successfully demonstrated that the new platform, in conjunction with immune agents, exhibits greater immune-stimulation responses than the agents alone.



By developing a framework for understanding and ultimately predicting natural emerging infectious diseases and differentiating these from man-made pathogens, an LDRD project will lay the foundation for future studies of virus evolution and forensic identification of bioterrorist pathogen agents (10-LW-020). Researchers are developing an understanding of the role of virus evolution in transmissions between animals and humans to ultimately develop models that predict the likelihood of pathogen emergence. This research is also important to healthcare because

- Three quarters of the recently discovered disease agent pathogens are viral.
- One to four new human pathogenic viruses are discovered each year.
- Most of these are animal viruses that infect humans.

High-Performance Computing and Simulation

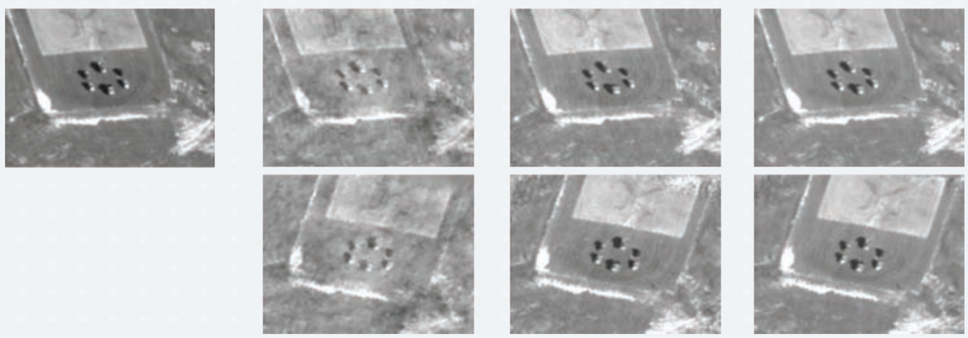


Anticipating the large-scale supercomputer systems of the future, LDRD researchers are developing ExaCT (exascale computing technologies) that will produce tools and strategies to diagnose and overcome difficulties arising in large-scale supercomputer systems (10-SI-014). This will herald a new era of predictive simulation by dramatically improving the scalability and performance of Laboratory applications through innovative algorithms and automated adaptation to systems with huge numbers of computing nodes. Simulation applications include

- Materials modeling
- Fusion energy science
- Physics relevant to stockpile stewardship

Intelligence

An LDRD team is hoping to provide, for the first time, a practical solution to the problem of "data explosion" faced by the defense and intelligence communities as larger surveillance arrays are being readied for deployment (11-ERD-022). 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. The team has determined that compressive sensing recovery of unmanned aerial vehicle video is possible at compression ratios of more than 160:1.

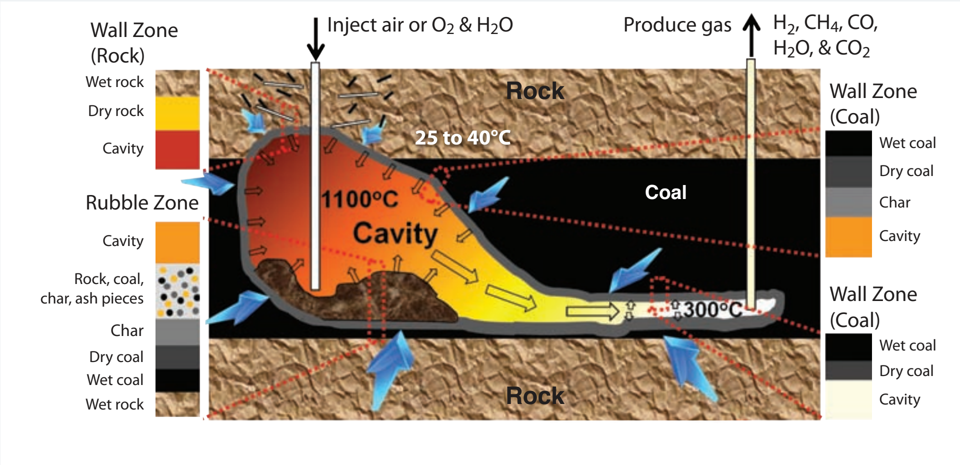


Cybersecurity

Investigators are developing the first simulation platform (NetWarp) to enable realistic studies of cyber attack and defense scenarios that are scalable and accurate in both behavior and performance (10-ERD-025). This LDRD project will enable ensemble studies to optimize cyber defense systems against an array of attack codes and do so without prior understanding of how the attack codes work. In addition, they are creating an automatic rollback code generator (BackStroke) that will restore a database to its previous consistent state, greatly decrease simulation development and maintenance work, and reduce opportunities for extremely difficult computer bugs.

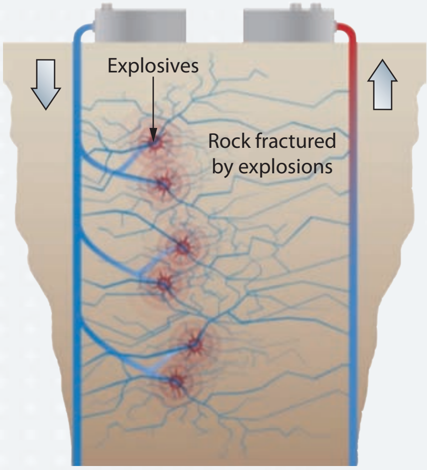
Energy Security

In FY11, LDRD contributions to national energy security included a project to develop the world's most complete and accurate computer model of the underground coal gasification process (10-ERD-055). It will address the many scientific and technical challenges that impede this energy resource, which will also help to mitigate greenhouse gas emissions. The team's approach combines computer simulation with geophysics monitoring, and will allow improved siting, design, permitting, operation, monitoring, and environmental performance of pilot and commercial underground coal gasification projects. In addition, their research will accelerate technology improvements and commercial deployment and serve as a resource in addressing emerging regulations.

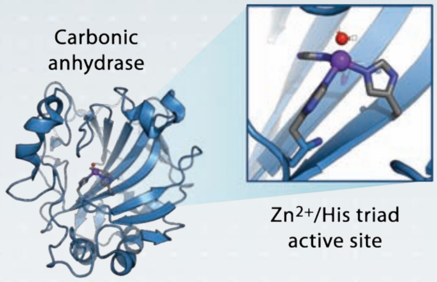


Creating optimal fracture networks for energy extraction (11-SI-006) is the goal of an LDRD project that will use high-performance computing simulations to guide optimization of fracture networks, predict permeability enhancement, and assess associated seismicity in complex, uncertain geologic media. Applications include

- Shale-gas recovery
- Enhanced geothermal systems
- Carbon dioxide sequestration
- Arms control treaty verification



Carbon Capture



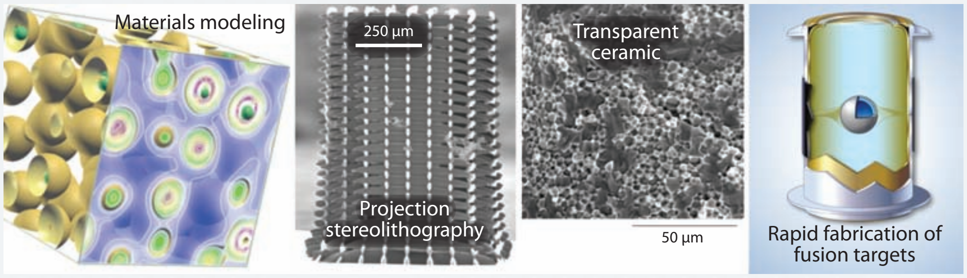
An LDRD project is seeking to develop catalysts to dramatically increase the rate of carbon dioxide separation and thereby reduce the size and cost of industrial processes for keeping carbon dioxide from being emitted to the atmosphere (10-ERD-035). The new technology, which would mimic natural enzymes such as carbonic anhydrase that catalyze carbon capture in animals and plants, could enable the direct capture of carbon dioxide from the atmosphere at an increased rate of up to a factor of 1,000 over conventional systems.

Material Properties, Theory, and Design

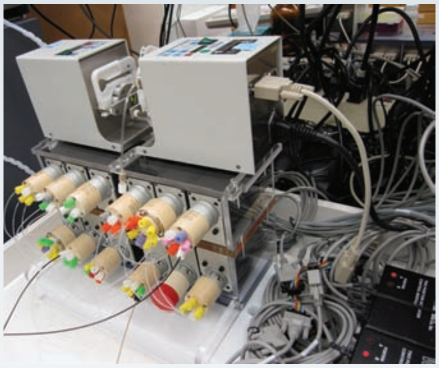
Predictive design and direct digital manufacturing of materials examined by LDRD investigators (11-SI-005) will enable high-volume, low-cost manufacturing of functional parts with novel properties and performance with applications in

- Materials modeling
- Projection stereolithography
- Transparent ceramics
- Rapid fabrication of fusion targets

A three-dimensional manufacturing process combined with multiple materials would provide for new design concepts that have not been possible and offers cost reduction for advanced fusion-class laser system targets and rapid turnaround for new target designs in support of Livermore's mission in stockpile science.



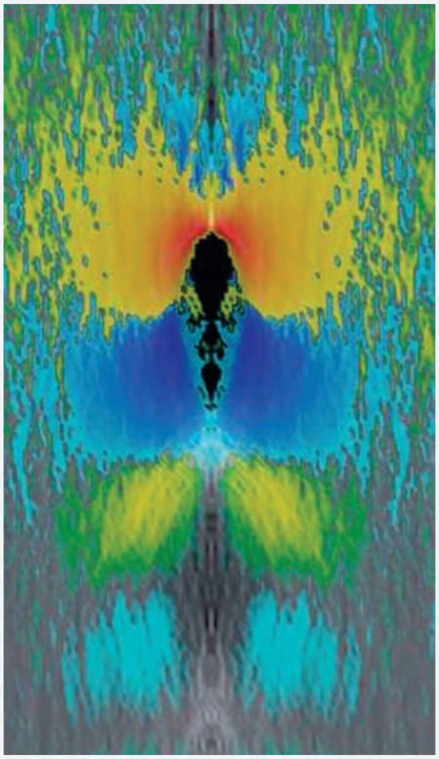
Radiochemistry



The study of heavy elements is a key component of fundamental research in nuclear chemistry and radiochemistry, which are core LLNL competencies. LDRD investigators have created a superheavy element liquid automation system designed to isolate heavy elements for determining their chemical properties (11-ERD-011). The system can be adapted for a variety of chemical systems and can perform a separation in approximately one minute. This research advances the Laboratory's strategic mission of nuclear threat elimination by addressing the challenge of developing autonomous, real-time, forensics methods designed for field deployment. This project will also serve as the foundation for future opportunities in a variety of other areas, such as

- Medical isotope production
- Chemical purification
- Inertial-confinement fusion diagnostics
- Isotope harvesting

High-Energy-Density Science

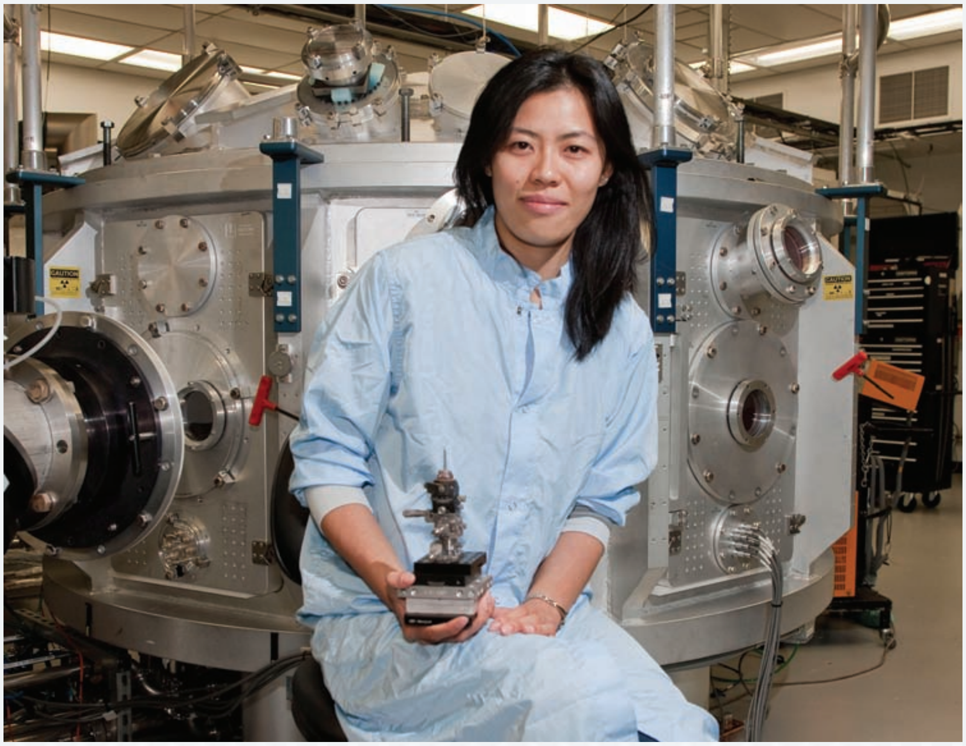


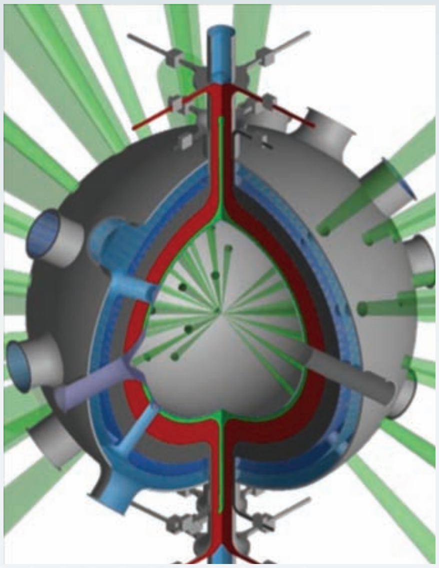
In FY11, LDRD continued to play a central role in building a world-leading capability for high-pressure science on high-power lasers. One project is producing the first precise, detailed, self-consistent description of radiation and matter in a hot, dense burning plasma (09-SI-011). Researchers are building a world-class, massively parallel, many-body simulation capability in conjunction with experimental validation experiments to determine whether or not the current physics used to describe these plasmas is correct. The team is working to establish LLNL as a world center for high-energy-density-physics, and the successful conclusion of the project resulted in a molecular dynamics simulation capability for gaining insights into the behavior of hot dense plasmas such as obtaining new results for electron–ion coupling and charged particle stopping.

The dynamics of ultrafast heated matter (11-ERD-050) is using x-ray Thomson particle scattering techniques to measure the kinetics of novel materials created under high-energy-density conditions, and has enabled research opportunities in astrophysics and high-energy-density science for numerous postdoctoral researchers and students. This newly proposed application of x-ray scattering will provide data important to several Laboratory missions including

- Stockpile stewardship
- Climate and energy challenges
- Laser ignition fusion energy

The LDRD researchers are characterizing heating under ultra-short conditions for direct measurements of temperatures, densities, and collective collisional effects with x-ray Thomson scattering of electrons from laser light interaction with matter. The measurements will determine equation-of-state and collision properties, which are important for successful modeling of experiments on the National Ignition Facility.





Laser Inertial-Fusion Energy (LIFE)

Forwarding national efforts to achieve practical, cost-effective laser inertial-fusion energy plants, an LDRD project is conducting simulations and experiments on laser beam propagation and target chamber clearing (10-SI-009).

- Radiation-hydrodynamics modeling will predict the chamber-gas state after a laser shot and support scaled experiment design.
- Computational fluid dynamics will be used to explore the clearing of hot gas and debris from the chamber.
- Scaled experiments using existing kilojoule-class lasers will be used to observe chamber clearing and beam propagation.

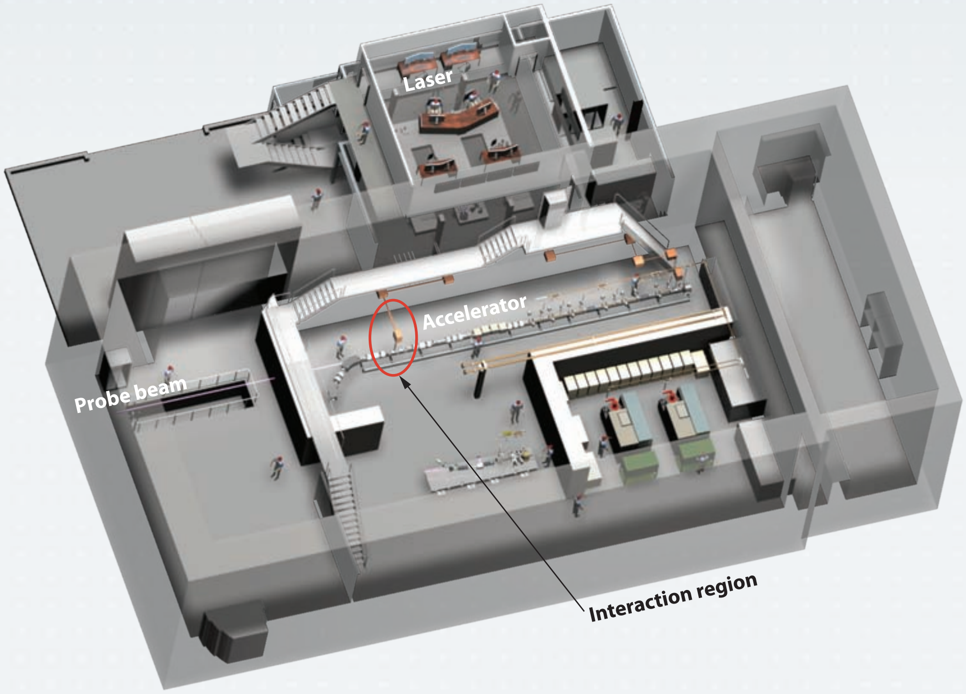
This research is a key component of the Laboratory's strategic roadmap in energy security. Resolving key issues for practical, cost-effective laser inertial fusion energy plants also supports the Laboratory mission in environmental security by enabling a source of abundant, clean power without nuclear waste disposal, safety, carbon sequestration, or proliferation issues.

Advanced Laser Optical Systems and Applications

An LDRD project is developing a concept for a precision mono-energetic gamma-ray (MEGa-ray) capability at Lawrence Livermore for isotope-specific imaging and assaying of highly enriched uranium, as well as for applications in stockpile stewardship and legacy nuclear waste (09-SI-004). A primary accomplishment was the invention of an entirely new configuration for Compton scattering of photons in laser–matter interactions. This approach, for which a patent application was filed,

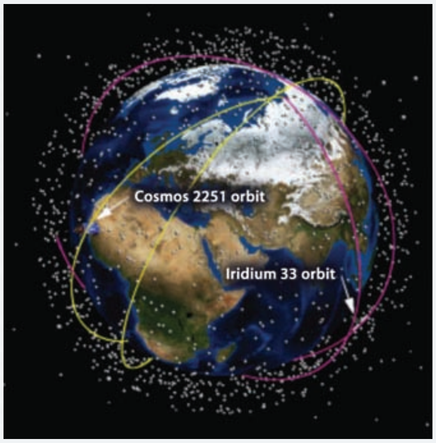
- Operates at a higher effective repetition rate
- Increases the flux of the MEGa-ray source
- Decreases the bandwidth
- Potentially simplifies much of the underlying accelerator and laser subsystems

The successful completion of the project in FY11 resulted in discussions with industry regarding a Cooperative Research and Development Agreement for a MEGa-ray material assay, as well as over 40 conference proceedings articles and papers in peer-reviewed journals such as *Physical Review Letters* and the *Physics of Plasmas*.



Space Security

Because safety in space operations is crucial to U.S. interests, an LDRD team in collaboration with two other national laboratories and the Air Force Research Laboratory is working to improve the nation's capabilities for detecting and monitoring threats to U.S. space operations (10-SI-007). The team is leveraging three relevant key capabilities

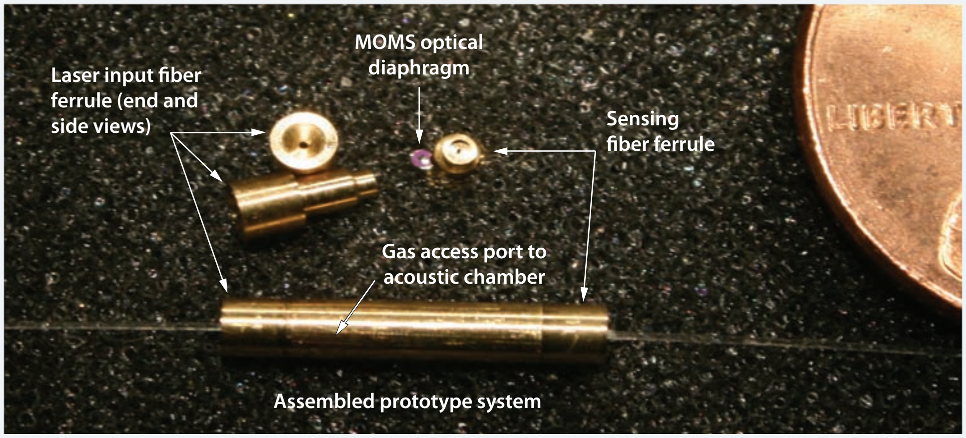


- Extensive experience with numerous operational sensor systems
- Sophisticated analysis tools for interpreting data from multiple sensor systems
- Unparalleled expertise in simulation and modeling of complex systems using the world's largest computers

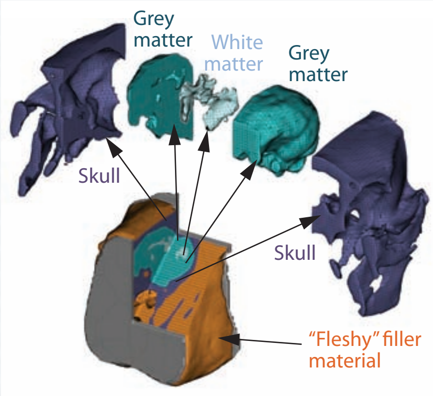
to enable a more accurate assessment of whether or not any orbiting objects pose a threat to active satellites. In addition, techniques from this project would be directly applicable to other national problems, such as nuclear proliferation and climate monitoring.

Stockpile Stewardship Science

Several new sensing capabilities are being considered for nuclear weapons stockpile surveillance in an FY11 LDRD project (10-ERD-043). Broad-spectrum sensors most efficiently address the surveillance challenges and deliver the greatest overall impact. For this reason, this effort pursues the broad-sensing technology of gas sensing. Specifically, optic-fiber-based surface-enhanced Raman scattering, photo-acoustic spectroscopy, and ionization techniques are being studied for detection of unknown gas mixtures. Researchers are exploring novel materials, fabrication processes, and designs for their applicability to the difficult constraints of in situ state-of-health stockpile monitoring.

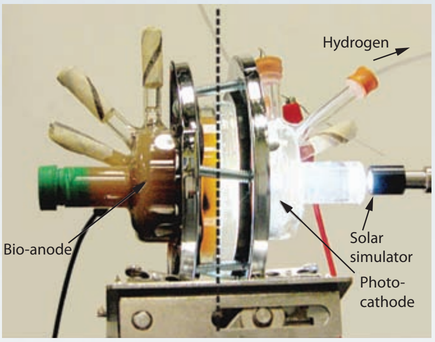


National Security



An FY11 LDRD project is devoted to computational studies of blast-induced traumatic brain injury using high-fidelity models (11-LW-009), a problem relevant to urgent efforts to minimize injuries and fatalities from improvised explosive devices encountered in Iraq and Afghanistan. The investigators are developing high-fidelity models of the head from x-ray computed tomography data, and then leveraging Livermore's computational capabilities and blast expertise to conduct detailed simulations of impacts and blasts on a head model. They will then work with medical researchers to correlate the simulation predictions with actual trauma data to identify the pathways by which blasts cause damaging loads in the brain. Identifying these mechanisms will enable development of more effective protective equipment for reducing traumatic brain injury and saving lives. Understanding damage mechanisms also has implications for rapid diagnosis in the field and could lead to better treatment strategies.

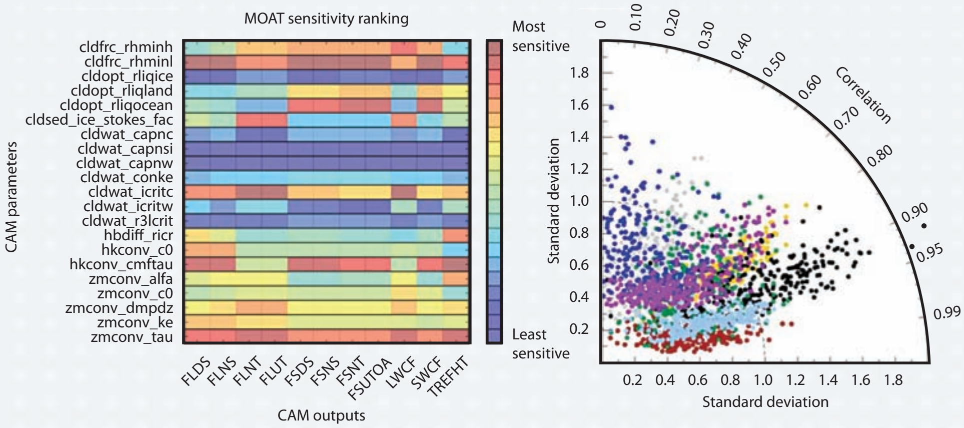
Alternative Energy



Researchers are implementing a straightforward strategy for hydrogen production by photosynthetic microorganisms using sunlight, sulfur- or iron-based inorganic substrates, and carbon dioxide as the feedstock (11-LW-019). The LDRD team hopes to demonstrate, for the first time ever, the ability to produce hydrogen using inorganic substrates coupled with carbon dioxide fixation by photosynthesis. This achievement 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. The 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.

Climate Change

Already a world leader in climate-change detection and attribution, Laboratory efforts in this field are supported by an LDRD project that is applying uncertainty quantification to climate science (10-SI-013). The research team is comprised of both weapons and climate scientists working to understand the fidelity of climate model simulations and diagnose human signatures in climate change. Their research will also have a large impact in stockpile stewardship by providing methods that will enable increasingly precise uncertainty bounds to be placed on the performance of nuclear weapons.



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 technical staff to enhance their skills and expertise. Laboratory LDRD principal investigators and research teams receive numerous prestigious honors, awards, and recognition for LDRD-funded work. These recent honors attest to the exceptional capabilities, talents, and performances of these researchers, while simultaneously highlighting the success and vitality of the LDRD Program at Livermore.

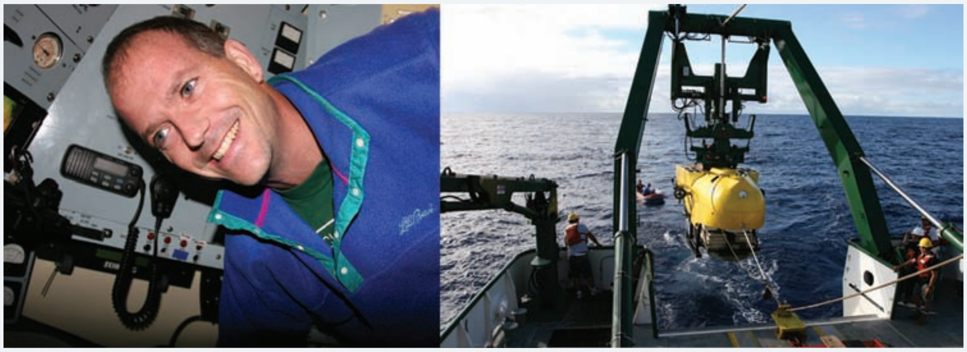
National Academy of Sciences


Benjamin Santer

Physicist, atmospheric scientist, and LDRD researcher Benjamin Santer was elected in 2011 to the National Academy of Sciences for his research in human-induced climate change. The National Academy of Sciences is an honor society of distinguished scholars engaged in scientific and engineering research, dedicated to the furtherance of science and technology and to their use for the general welfare. Santer has participated in two LDRD projects devoted to climate change research (10-ERD-060 and 99-ERD-056). In addition to his recent election to the National Academy of Sciences, Santer is the recipient of the MacArthur "genius" grant; an E. O. Lawrence Award; a Department of Energy Office of Biological and Environmental Research Distinguished Scientist Fellowship; contributor to all four assessment reports of the Intergovernmental Panel on Climate Change, an organization that shared the 2007 Nobel Peace Prize with former Vice President Al Gore; and an American Geophysical Union fellowship.

E. O. Lawrence Award

The award, established by President Dwight Eisenhower in 1959, is given to scientists and engineers for their exceptional contributions to the development, use, or control of nuclear energy (broadly defined to include the science and technology of nuclear, atomic, molecular, and particle interactions, including their effects). The E. O. Lawrence Award is given for relatively recent work rather than for lifetime achievement. Thomas Guilderson received the award in 2011 in biological and environmental sciences for his ground-breaking radiocarbon measurements of corals, advancements in understanding the paleo-history of ocean currents and ocean processes revealing past climate variability, and the elucidation of how physical and biogeochemical oceanic processes affect the global carbon cycle. Guilderson is currently the co-investigator for an LDRD project on land-use impacts on belowground carbon turnover and an ecosystem carbon dioxide source attribution using radiocarbon (11-ERD-053) and the use of late-Holocene lake sediments for mapping patterns of past drought in California (09-ERI-003). He previously was the principal investigator for a fossil fuel emission verification capability (07-ERD-064), atmospheric 14-carbon dioxide constraints for modeling of net carbon fluxes (06-ERD-031), and carbon sequestration and transport in natural environments and the role of organic carbon and microbial processes in the ocean (04-ERD-060), among others.



Thomas Guilderson

Edward Teller Medal

Bruce Remington (left) receives the Edward Teller medal from Bob Kauffman of the National Ignition Facility.

The Edward Teller Medal, presented by the American Nuclear Society, 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. An LDRD researcher since 1996, Bruce Remington was so honored in 2011. His most current LDRD project concerns probing extreme high-energy-density states of matter with x-rays for applications from super-Earths to nucleosynthesis (09-SI-010), but he has also served on various projects related to astrophysical collision-less shock generation by laser-driven laboratory experiments (11-ERD-054), the study of Kelvin–Helmholtz instability in high-energy-density hydrodynamic processes (08-ERD-069), high-energy backlighting for an ignition diagnostic for high-power lasers (07-ERD-004), and ultrafast lattice response of shocked solids (06-SI-004), among others.

Presidential Early Career Award for Scientists and Engineers

This award is the highest honor bestowed by the U.S. government on outstanding scientists and engineers beginning their independent careers, and is intended to recognize and nurture young scientists and engineers who show exceptional potential for leadership at the frontiers of scientific knowledge. In 2010, LDRD established support for Gianluca Iaccarino (Stanford University), who was cited for his extensive and deep scientific contributions in the areas of turbulent flow and uncertainty quantifications and quantified margins of uncertainty (10-SI-013). In 2011, LDRD supported three winners: Greg Bronevetsky (LLNL) who was cited for innovative, cutting-edge research using statistical models to predict the effects of system faults leading to the development of new software tools and more reliable applications and supercomputer systems (10-SI-014); Tina Chow (UC Berkeley) for fundamental research on the simulation of atmospheric turbulence, which has wide-ranging applications that include the dispersal of plumes resulting from atmospheric releases of radioactive or toxic material and the accurate simulation of wind fields for weather prediction (12-ERD-043); and Gang Logan Liu (University of Illinois, Urbana-Champaign) for collaborative development of surface-enhanced Raman spectroscopy techniques for a variety of national security applications ranging from measuring the long-term health of the U.S. nuclear stockpile to bio-detection (12-ERD-065).

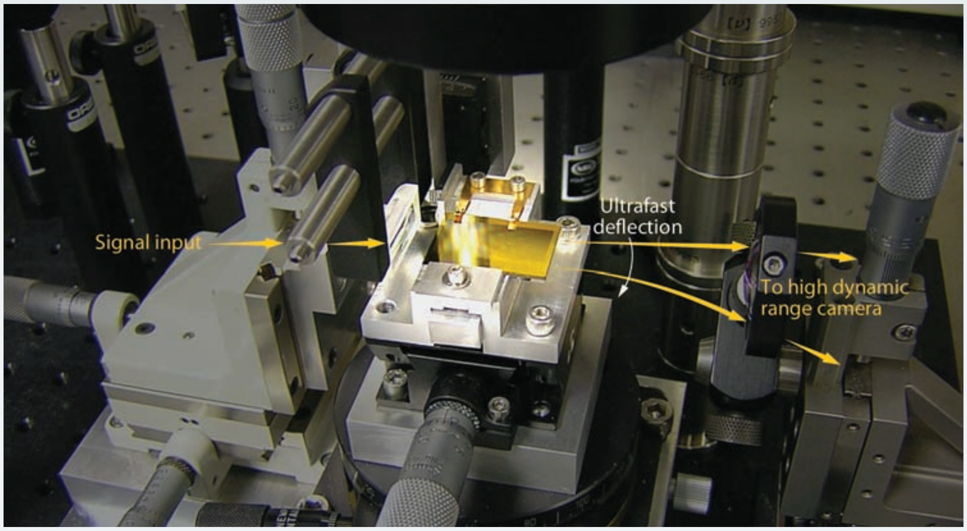
The Presidential early career awards embody the high priority the Obama Administration places on producing outstanding scientists and engineers to advance the Nation's goals, tackle grand challenges, and contribute to the American economy. In the last several years, several LDRD-supported researchers have been named as recipients.

R&D 100 Awards

In 2011, LDRD-supported technologies garnered both of the R&D 100 Awards presented to the Laboratory by *R&D Magazine*.

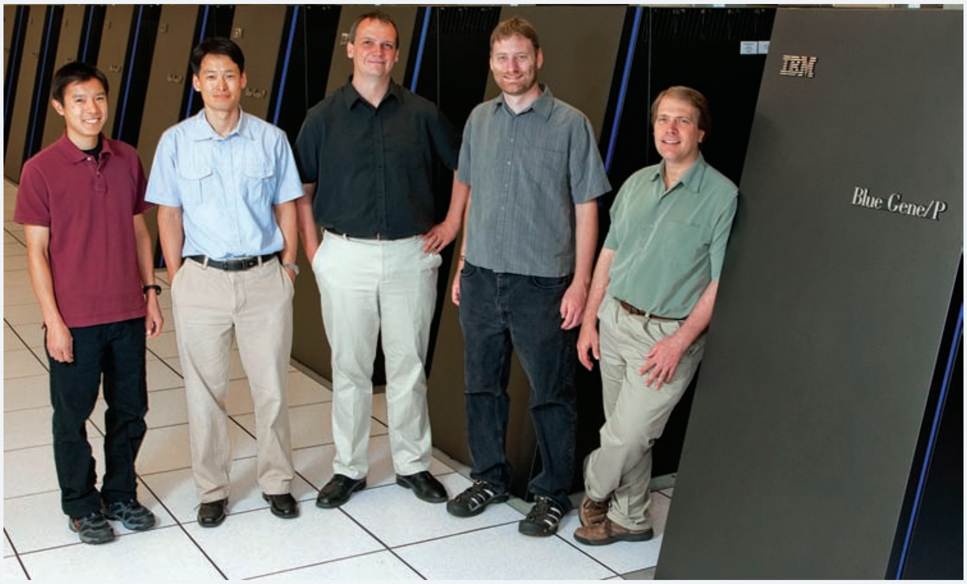
- **World's Fastest Light Deflector.** An LDRD project on serrated light illumination for deflection-encoded recording (07-ERD-017) led by John Heebner has led to an R&D 100 Award for a novel solid-state optical device called SLIDER. When combined with a high dynamic range camera, SLIDER can maintain high temporal resolution and a high dynamic range—two performance parameters that are difficult to meet simultaneously. This unique combination of high resolution and dynamic range will be crucial for better understanding reactions that occur under the extreme conditions—such as temperatures of more than

100 million degrees Celsius—needed for the tritium–deuterium fuel to "ignite" in a National Ignition Facility target and undergo thermonuclear burn.



Signals to be recorded propagate from left to right in a thin waveguide layer at the top of the SLIDER (serrated light illumination for deflection-encoded recording system) deflector. The pump beam illuminates the top of the device where the serrated gold mask defining the prisms resides. Because the pattern pitch is 60 µm, it is too fine to be resolved by the camera and hence the patterned array of gold prisms is discernible only as a gold gradient. The deflected beam emerging from the device is collected and focused by a lens onto a camera for recording.

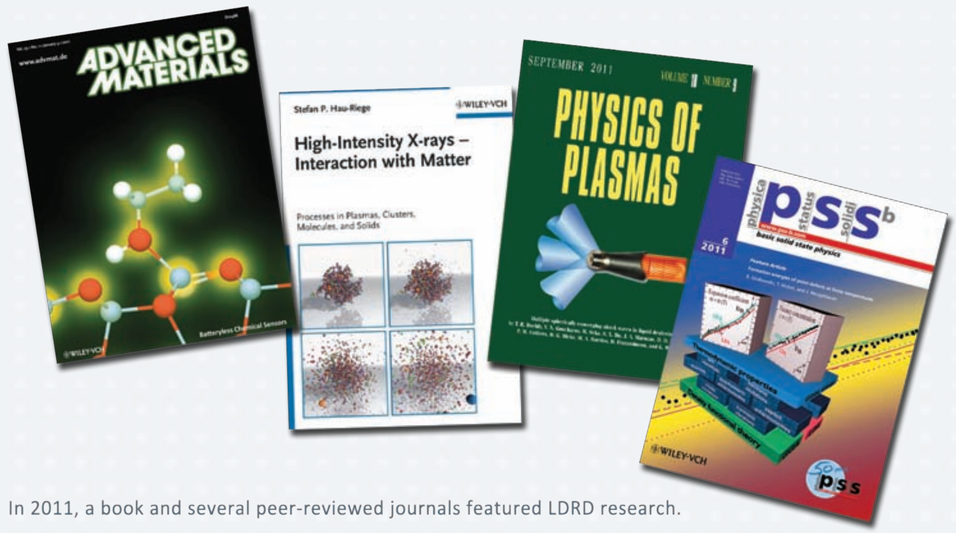
- **Tool Identifies Supercomputers' Errors.** A project on exascale computing technologies (10-SI-014) resulted in a unique R&D 100 Award–winning solution to bugs in supercomputer codes called the stack trace analysis tool (STAT). The tool can identify errors in code running on today's largest machines. It will also work on the even larger machines expected to roll out over the next several years. STAT works by detecting and grouping similar processes at suspicious points in an application's execution. It quickly and automatically identifies anomalies and outliers—processes that cannot be grouped or whose behavior is substantially different—because they often indicate flawed execution.



The Livermore development team for the stack trace analysis tool (STAT): Greg Lee, Dong Ahn, Martin Schulz, Matthew LeGendre, and Bronis de Supinski (left to right).

Book and Journal Covers

In 2011, several LDRD-supported projects were featured on the covers of peer-reviewed journals, and one in a new book on high-intensity x rays. Stefan Hau-Riege, an LDRD researcher since 2002, is currently probing atomic-scale transient phenomena using high-intensity x-rays (12-ERD-021) and recently published a book that comprehensively covers the physics of interactions between x ray and matter in all forms, from condensed phases to plasmas *(High-Intensity X-rays—Interaction with Matter)*. Hau-Riege has led or served on myriad LDRD projects related to the research frontiers made accessible by powerful new x-ray sources, such as x-ray free-electron lasers (notably, 09-ERD-023, 09-LW-044, 09-SI-010, 05-SI-003, and 02-ERD-047). The cover article for the September 2011 issue of *Physics of Plasmas* is a paper by LDRD researchers that reports on the first experiments to time multiple, spherically convergent shocks in cryogenically cooled liquid deuterium. The research was supported, in part, by an LDRD project on the physics and chemistry of the interiors of large planets using a new generation of condensed matter (09-SI-005). The cover of the June issue of *Physica Status Solidi B* highlights, as a featured article, a paper by Blazej Grabowski, an LDRD researcher currently part of a team examining temperature-dependent lattice dynamics and stabilization of high-temperature phases from first-principles theory (11-ERD-033). The journal article concerns the formation energies of point defects at finite temperatures. In January, the cover of *Advanced Materials* featured an article on battery-less chemical detection with semiconductor nanowires, and is the basis for LDRD researcher's Yinmin Wang's current LDRD project related to versatile energy harvesting and battery-less molecule sensors (12-LW-010).



In 2011, a book and several peer-reviewed journals featured LDRD research.

Alameda County Women's Hall of Fame

Dawn Shaughnessy was recently elected to the Alameda County Women's Hall of Fame in the science category for her experimental work in nuclear chemistry and radiochemistry, following in the footsteps of numerous other LDRD researchers so honored. Her most recent project is exploring the fundamental chemical behavior of superheavy elements through applications of online isotope production and automated chemical systems (11-ERD-011). She has also served as a principal and co-investigator for a variety of projects related to collection of refractory debris from the National Ignition Facility for stewardship-relevant measurements (09-ERD-026), nuclear astrophysics studies at the National Ignition Facility (08-ERD-066), rapid radiochemical separations for investigating chemistry of the heaviest elements (08-ERD-030), and fragment separation technology for superheavy element research (04-ERD-085).

Dawn Shaughnessy

American Geophysical Union Fellow

Benjamin Santer was elected in 2011 as an American Geophysical Union fellow for "his insightful and rigorous contributions to climate change detection and attribution research, and tireless communication and public outreach." He most recently served as a co-investigator for an LDRD project to enhance climate model diagnosis and intercomparison (10-ERD-060), and was the principal investigator for research on multivariate climate change detection (99-ERD-056).

American Physical Society Fellow

Michael J. Edwards was nominated by the Division of Plasma Physics and awarded fellow status for "fundamental contributions to hydrodynamics in high energy density physics, and for his leadership in the National Ignition Campaign on the National Ignition Facility." He was the principal investigator for an LDRD project devoted to gaseous laser targets and optical diagnostics for studying compressible turbulent hydrodynamics (02-ERD-023). Most recently, Edwards was co-investigator for the development of scaled astrophysical experiments for current and future lasers (07-ERD-038), which followed his contributions to studies of anisotropic shock propagation (02-ERD-024), a novel multi-layer mix experiment (01-ERD-077), and a double-shell laser target design related to non-cryogenic ignition and nonlinear mix studies for stockpile stewardship (01-ERD-033).

Department of Energy Early Career Research Program

The Early Career Research Program, now in its third year, supports the development of individual research programs of outstanding scientists early in their careers and stimulates research careers in the disciplines supported by the DOE Office of Science. In 2011, LDRD researcher Sofia Quaglioni was selected by the Office of Nuclear Physics for her project on "Solving the Long-Standing Problem of Low-Energy Nuclear Reactions at the Highest Microscopic Level." Quaglioni is currently on the LDRD team exploring nuclear plasma physics (11-ERD-069) and was the principal investigator for studies on carbon and oxygen production in stars using an ab initio approach to nuclear reactions (09-ERD-020).

Sofia Quaglioni

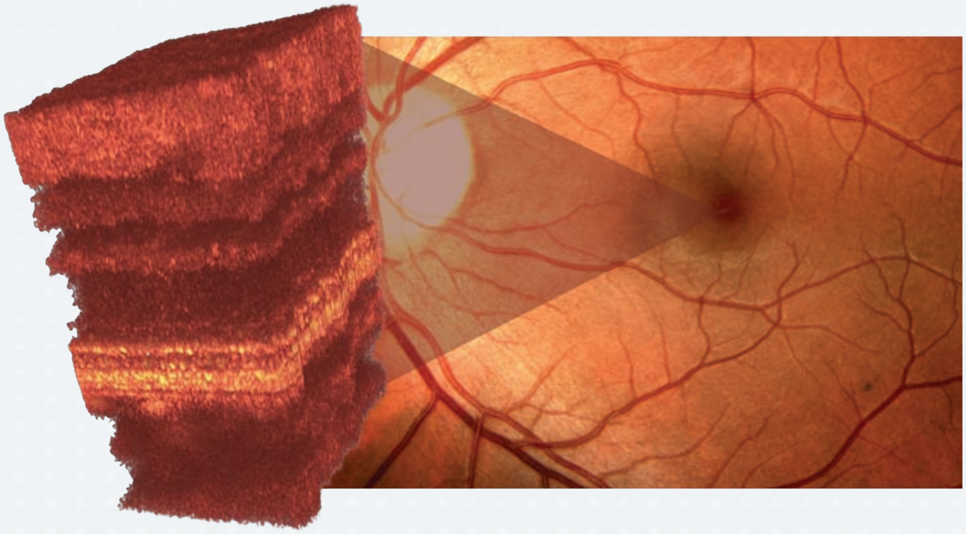
Federal Laboratory Consortium Award for Excellence in Technology Transfer

Since 1974, the Federal Laboratory Consortium Award for Excellence in Technology Transfer has recognized scientists and engineers at federal government and research centers for their "uncommon creativity and initiative in conveying innovations from their facilities to industry and local government." Scientists and engineers from more than 650 federal government laboratories and research centers compete for the 30 awards presented each year. Lawrence Livermore received two awards in 2011, including the intercranial hematoma detector for John Chang and Genaro Mempin, with roots from LDRD work on impulse radar application to cardiac monitoring of hemodialysis patients. The second award was for an environmental sample processor for John Dzenitis, Vincent Riot, Bill Benett, Dean Hadley, and Catherine Elizondo, with early support provided by an LDRD project to replace burdensome, manual techniques with new automated technologies for sample handling and preparation for rapid defense against the next-generation biothreat (05-ERD-084).

LDRD researcher John Chang tests a prototype intracranial hematoma detector on a human skull. Work on this project won an award for excellence in technology transfer from the Federal Laboratory Consortium.

Federal Laboratory Consortium's Far West Region Awards

The Federal Laboratory Consortium "is the nationwide network of federal laboratories that provides the forum to develop strategies and opportunities for linking laboratory mission technologies and expertise with the marketplace." Their primary goal is "to promote and strengthen technology transfer nationwide" for the more than 250 federal members of the organization. Livermore technologies have resulted in several consortium awards in the Western Region competition for 2011, including "MEMs-Based Adaptive Optics Optical Coherence Tomography" (Outstanding Technology Development), a clinical instrument that provides noninvasive, ultra-high resolution, three-dimensional volumetric retinal images for eye doctors to view retinal structures at the cellular level. The device has LDRD roots in several projects including correction of distributed optical aberrations (03-ERD-006), diffraction-limited adaptive optics and human visual acuity (01-LW-036), and advanced wave-front control (98-ERD-061), all with Scot Olivier as the principal investigator.



Micro-electromechanical systems and adaptive optics enable a three-dimensional image of the cellular layers within the retina. An image of the photoreceptor layers within the eye allows physicians to diagnose sight-threatening diseases, such as macular degeneration, in their earliest stages (inset).

SPIE Fellow

Chris Barty

Each year, SPIE (the international society for optics and photonics) promotes members as new fellows of the society. Fellows are members of distinction who have made significant scientific and technical contributions in the multidisciplinary fields of optics, photonics, and imaging. LDRD researcher Christopher Barty, chief technology officer for LLNL's National Ignition Facility and Photon Science Directorate, was recently named as a fellow of SPIE. Barty was recognized for his achievements in a leadership role in the advancement and development of new laser technology. His technical interests include development of new optical capabilities for fusion energy drivers, directed energy systems, nuclear photo-science, high-energy-density science, fast ignition, and laser-based x-ray applications of relevance to national security-related missions, programs, and projects. He has served on numerous LDRD projects since 2002, including research into precision mono-energetic gamma-ray science for NNSA missions (09-SI-004). He is currently a co-investigator for projects related to the next generation of gamma-ray sources (12-ERD-060), Compton-scattering optimization for ultra-narrowband nuclear photonics applications (12-ERD-057), novel multi-gigahertz electron beams for advanced x-ray and gamma-ray light sources (12-ERD-040), and the science and technology of unconventional fiber waveguides for emerging laser missions (10-SI-006).

Graph 500 List

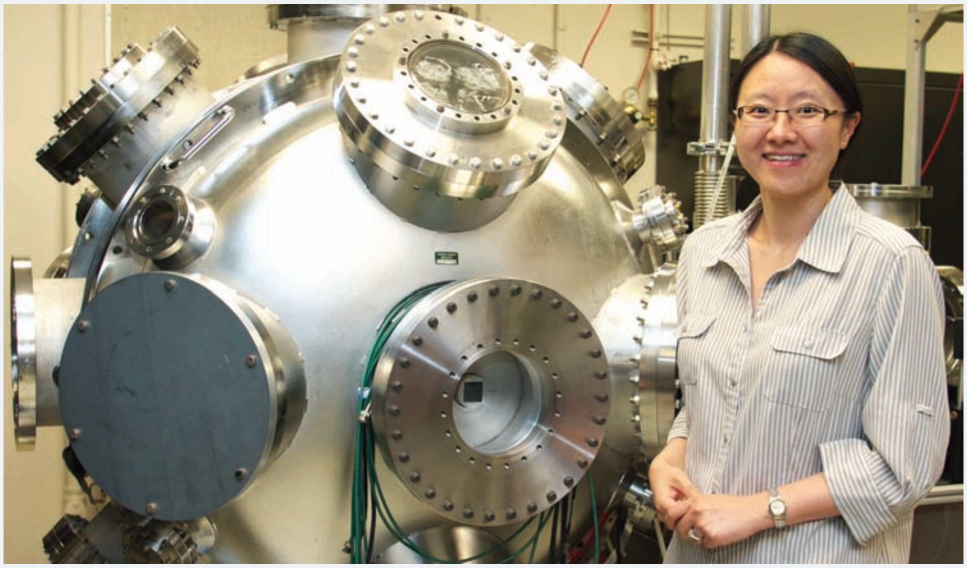
Backed by a steering committee of over 50 international high-performance computing experts from academia, industry, and national laboratories, Graph 500 is establishing a set of large-scale benchmarks for data-intensive supercomputer applications. An LDRD project on unifying memory and storage with persistent random-access hardware (11-ERD-008) was selected as such a benchmark. The team's graph traversal algorithm using a Flash storage array and one compute node ranked 7th place in the international Graph 500 competition in June 2011, in contrast to all other entries that used traditional supercomputers with thousands of nodes.

GRAPH 500

A new set of benchmarks is being established to guide the design of hardware architectures and software systems intended to support supercomputer applications and to help procurements.

Katherine Weimer Award

In 2001, the American Physical Society Division of Plasma Physics Executive Committee established the Katherine E. Weimer award to "recognize and encourage outstanding achievement in plasma science research by a woman physicist in the early years of her career." Researcher Yuan Ping was named the 2011 recipient based on her "pioneering experiments to explore the interaction of high-intensity laser light with matter." Ping is a co-investigator for an LDRD project on the physics and chemistry of the interiors of large planets with a new generation of condensed matter (09-SI-005).



Yuan Ping stands next to the target chamber in the Europa laser bay, part of the Jupiter Laser Facility at Lawrence Livermore.

IEEE Pacific Visualization Conference Best Paper Award

At the IEEE Pacific Visualization Symposium held in Hong Kong, China in March 2011, LDRD research into quantitative analysis of vector field topology (09-ERD-014) received a best paper award.

IEEE VisWeek Best Paper Award

A conference paper submitted as part of an LDRD research project on uncertainty visualization (10-ERD-040) was cited with a best paper award at the 2011 conferences held in Providence, Rhode Island in October.

Program Metrics

Intellectual Property

Projects sponsored by LDRD consistently account for a large percentage of the patents issued for LLNL research, especially considering that the program represents a small portion of the Laboratory's total budget. In FY11, LDRD costs at LLNL were $96.6M, which is 6.03% of total Laboratory costs. The number of patents resulting from LDRD-funded research since FY07 and the percentage of total patents that were derived from LDRD work 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 FY11, LDRD projects generated 53% of Livermore's total patents, even though the LDRD program constitutes only about 6% of the Laboratory's budget.

Patents	FY07	FY08	FY09	FY10	FY11
All LLNL patents	62	57	46	54	60
LDRD patents	32	23	20	27	32
LDRD patents as percentage of total	52%	40%	43%	50%	53%

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

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 FY07 to FY11 account for almost 40% of the 741 total. In FY11, there were 164 records submitted at Livermore, with 59 (36%) of those attributable to LDRD-supported projects.

Record of Invention	FY07	FY08	FY09	FY10	FY11
All LLNL records	162	110	145	160	164
LDRD records	69	44	56	66	59
LDRD records as percentage of total	43%	40%	39%	41%	36%

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

Finally, LDRD plays a role in producing Laboratory copyrighted material. From FY07 to FY11, LDRD-supported projects accounted for over 22% of the 241 Livermore copyrights. In FY11, there were 64 LLNL copyrights, with 16 (25%) 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 FY11 there were 994 such articles, of which at least 207 (21%) resulted from LDRD projects. The downward trend in total Laboratory publications in recent years is in part related to a decrease in the size of the scientific and engineering workforce at LLNL. During FY08 this population at Lawrence Livermore decreased by 15% (from 3,412 to 2,891), and the number of postdoctoral researchers declined by 19% (from 147 to 119). Since that year, the total population of scientists and engineers at LLNL has remained relatively constant at the lower number (~2,900), and the postdoctoral population began to recover in FY10. Because most of LLNL's postdoctoral researchers are sponsored by LDRD and represent a substantial component of the Laboratory's publishing scientists and engineers, the number of postdoctoral researchers is an important contributing factor for the lower number of total LLNL publications from FY07 through FY11. However, over the last several years, the percentage of LDRD-supported articles has remained relatively consistent, with a five-year average of nearly 20% of total Laboratory publications. The following table shows the number of journal articles per fiscal year resulting from LDRD-funded research since FY07 and the percentage of total articles that were derived from LDRD research and development.

Journal Articles	FY07	FY08	FY09	FY10	FY11
All LLNL articles	1,342	1,214	1,311	996	994
LDRD articles	267	233	225	227	207
LDRD articles as percentage of total	20%	19%	17%	23%	21%

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 FY11 portfolio included 75 formal LDRD-funded collaborations involving 51 LDRD projects (68% of the total projects funded). Collaborating institutions included the University of California (15% of total collaborators), other academic institutions (67%), DOE sites (4%), and other collaborators (e.g., other government agencies and industry, 15%). These statistics do not include the numerous informal collaborations that investigators 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 FY11, the LDRD Program supported 68% of the Laboratory postdoctoral researchers—there was an average of 179 postdoctoral researchers at the Laboratory in FY11, of which 122 were supported in some way by LDRD projects. The Laboratory continues significant recruitment efforts to increase the total number of postdoctoral researchers.

Program Mission

To fulfill its missions, LLNL must continually invest in the science and technology that form the foundation of its signature capabilities. The LDRD Program, which was established by Congress at all DOE national laboratories in 1991, is LLNL's most important single resource for fostering excellent 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 basic 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 2010 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,

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[1] *U.S. Department of Energy Order 413.2B, Laboratory Directed Research and Development* <http://doe.test.doxcelerate.com/directives/archive-directives/413.2-BOrder-b/view> (Retrieved March 5, 2012).

[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 15, 2012).

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]

At LLNL in 2011, Laboratory Director George Miller and the Deputy Director for Science and Technology Tómas Díaz de la Rubia were responsible for the LDRD Program. Execution of the program was delegated to the director of the LDRD Program, Kenneth Jackson (acting) and then William Craig (appointed August 2011). 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 the *Roadmap to the Future*, 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) or institute (ERI) 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.

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[3] *FY2010 Laboratory Directed Research and Development at the DOE National Laboratories Report to Congress* <https://ldrdrpt.doe.gov/PUBLICdocument/congress.pdf> (retrieved March 5, 2012).

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.

Project Competency Areas

Although LDRD projects often address more than one scientific discipline, each project is classified into 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 FY11 Portfolio

The FY11 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 *Roadmap to the Future* was developed by multidisciplinary teams under the guidance of the deputy director for science and technology. This document set the strategic context for the LDRD competition for five years, starting in 2009. As a living document, it will be updated periodically to respond to our ever-changing 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 America's future. In FY11, 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 *Roadmap to the Future* guides the LDRD portfolio planning process. This five-year strategic roadmap describes institutional strategic goals and science and technology needs in selected mission focus areas and in critical science, technology, and engineering pillars:

Mission Focus Areas

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

Science, Technology, and Engineering Pillars

- Controlling Fusion and High-Energy-Density Matter
- High-Performance Computing and Simulation
- Materials on Demand
- Measurement Science and Technology
- Energy Manipulation
- Information and Network Systems
- Earth and Environmental Sciences
- Bioscience and Biotechnology

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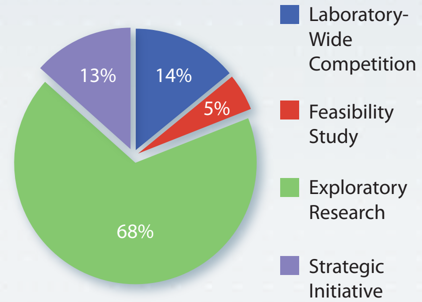
4 *U.S. Department of Energy Strategic Plan, May 2011* <http://energy.gov/sites/prod/files/DOE_2011-Strategic-Plan_Medium-Resolution_Print-Quality.pdf> (retrieved March 5, 2012).

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 5, 2012).

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 FY11 Portfolio

The FY11 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-reviewed selection process and received ongoing management oversight.



Distribution of funding among the LDRD project categories. Total funding for FY11 was $96.6M.

In FY11 the LDRD Program funded 142 projects with a total budget of $96.6M. The distribution of funding among the LDRD project categories is shown in the pie chart to the right.

Strategic Initiative

In FY11, the LDRD Program funded 19 SI projects. Although the SI category represented a little over 13% of the total number of LDRD projects for FY11, it accounted for nearly 45% of the budget. The SI projects ranged in funding from $1.3 to $5.4M.

Exploratory Research

The LDRD Program funded 96 ER projects for FY11. The largest project category, ERs accounted for over 67% of the number of LDRD projects and over 49% of the budget for the fiscal year. Projects in this year's ER category ranged in budget from $70K to $2.1M.

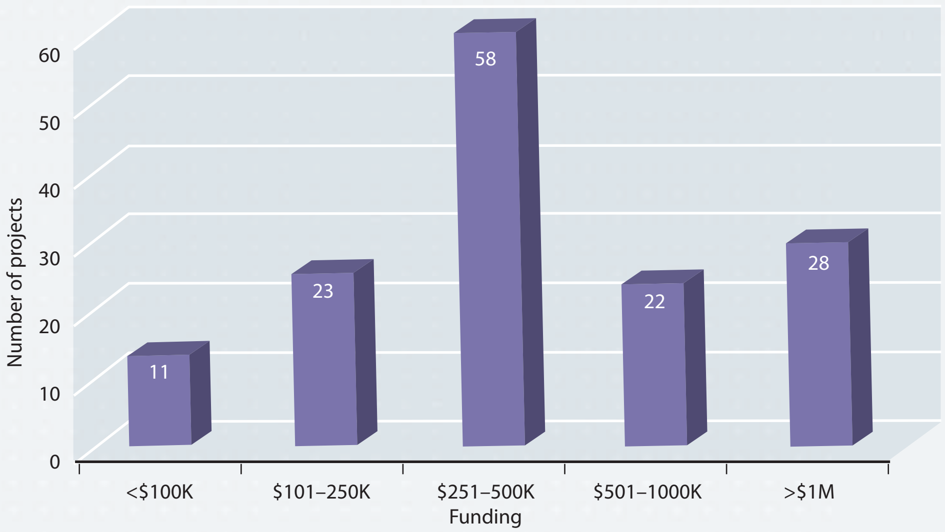
Laboratory-Wide Competition

In FY11, 20 LW projects were funded, which represented about 14% of the LDRD projects for the year and slightly over 5% of the budget. The LW projects for FY11 ranged in funding from $70 to $338K.

Feasibility Study

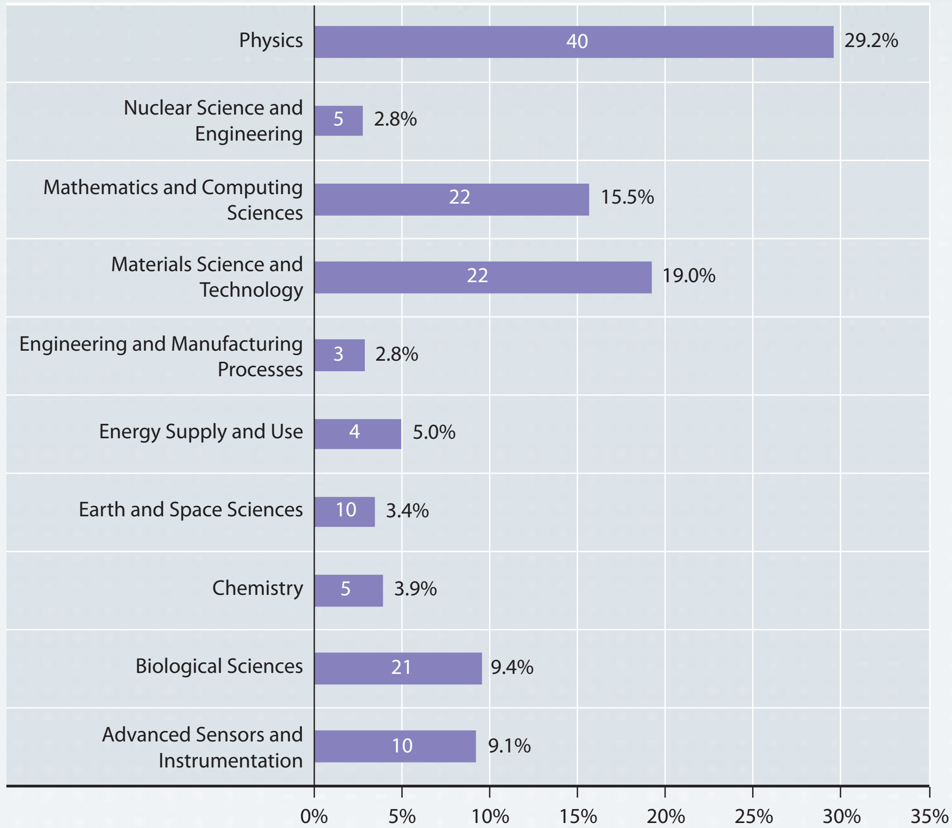
The LDRD Program funded just 7 FS projects in FY11, which represented about 5% of the LDRD projects for the year and 0.3% of the budget. The FS projects for FY11 ranged in funding from $50 to $125K. Project details for some of the FS projects for FY11 have not been included in this year's report because contracts with collaborating institutions have not been finalized and no work was performed during the fiscal year.

The following bar chart shows the funding distribution by dollar amount for the 142 FY11 projects—57% of the projects were in the $101 to $500K range, with around 8% falling below $100K. Projects in the $501K to $1M funding range accounted for over 15% of the total, and almost 20% of the projects received more than $1M. The average funding level for the 142 projects was about $607K.



Number of projects and levels of funding. The average funding level for an LDRD project in FY11 was about $607K.

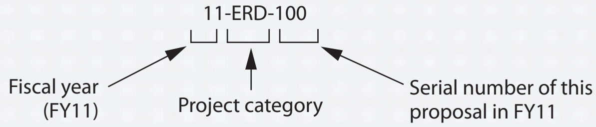
The percentage of LDRD funding and number of projects in each research category for FY11 are shown in the following chart. Nearly two-thirds of the LDRD projects fall under the physics, materials science and technology, and mathematics and computing sciences categories.



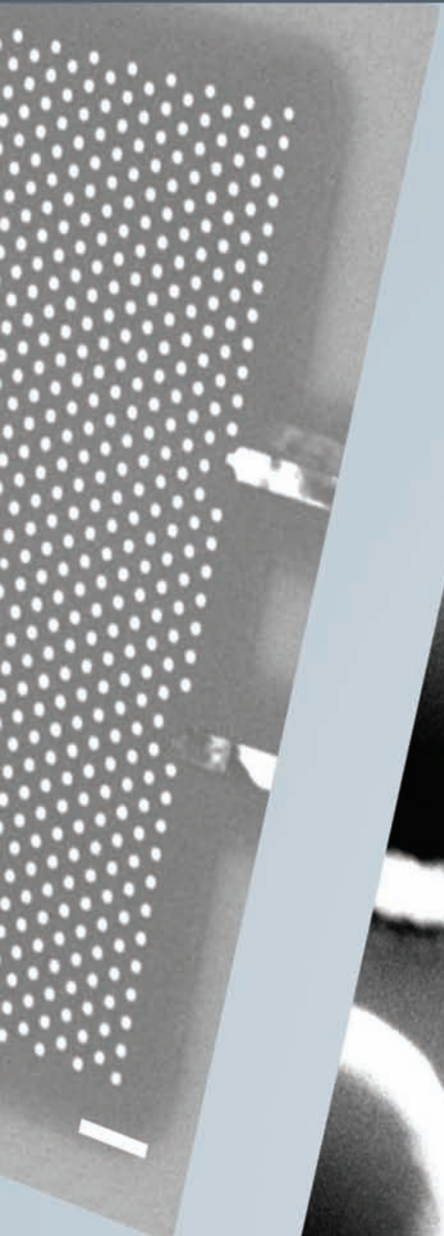
Percentage of LDRD funding and number of projects in each research category in FY11.

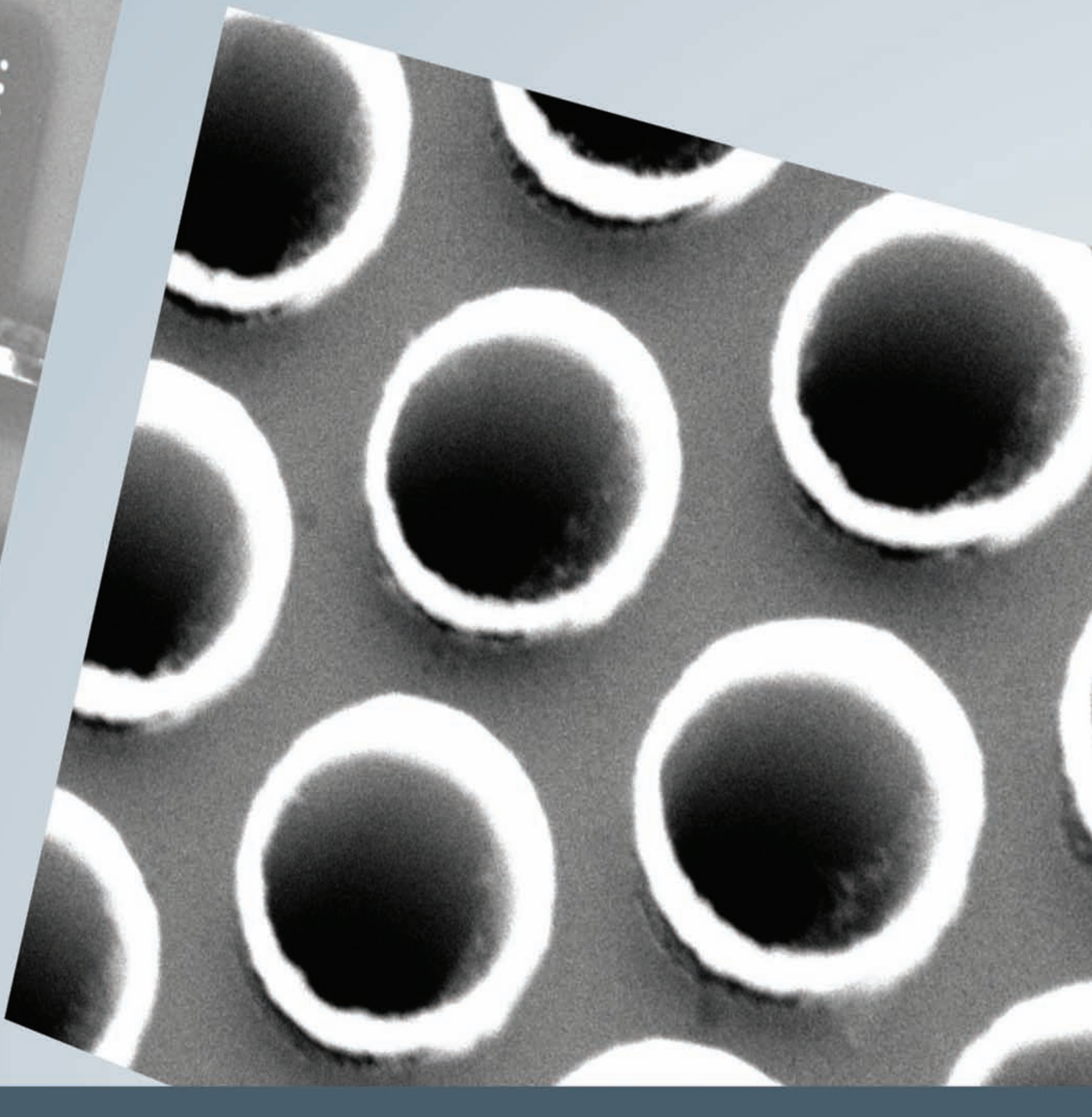
Organization of FY11 Project Summaries

Project summaries for the LDRD FY11 annual report are organized in sections by research category (in alphabetical order), and 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:



Advanced Sensors and Instrumentation





Laboratory Directed Research and Development

FY2011

Hybridization, Regeneration, and Selective Release of DNA Microarrays

Elizabeth Wheeler (08-ERD-064)

Abstract

The DNA microarrays for genetic testing identify hybridization patterns and signatures ideal for environmental and clinical monitoring, but a critical need exists for methodologies enabling rapid and selective analysis of these signatures. Analysis of DNA sequences from selective spots on an array could quickly yield vital information. This is especially important for countering rapidly mutating and emerging pathogens. We will develop a method for selective spot release and analysis. Because microarrays also suffer from long hybridization times (4 to 16 hours) and high chip-replacement costs, we will also study hybridization kinetics and mass transfer to enable chip reuse and faster analysis. This work leverages LLNL expertise in optics, microfluidics, and bioinformatics.

This effort will increase by an order of magnitude the information provided from microarrays. We will do this by investigating the (1) chemical and physical changes that occur during the processes of in vitro DNA hybridization; (2) stabilities of different chemical couplings between DNA molecules and surfaces; (3) controlling mechanisms between DNA in solution and DNA affixed to a solid support in hybridization experiments; (4) influence of optical, thermal, and fluidic effects on the intrinsic binding or stringency for DNA targets on probes; and (5) analysis of individual spots to eliminate the complex background signal.

Mission Relevance

This work supports the national security mission areas of nonproliferation and homeland security by enabling the fast and specific detection of, and response to, biological weapons of mass destruction. The capabilities developed will enable efficient, cost-effective, and highly sensitive and specific pathogen detection. With these improvements, microarray technology can be applied to distributed sensors and systems, enabling nonproliferation, counterterrorism, and force protection efforts.

FY11 Accomplishments and Results

In FY11 we tested our approach for release of selective DNA. As a result, we successfully laser-eluted bound DNA from the surface of a microarray and then recovered the DNA using microfluidics for analysis with polymerase chain reaction technology. Having successfully demonstrated not only selective feature release but also DNA recovery, we are now well positioned to enable even more widespread use of microarrays. Selectively releasing a feature for downstream sequencing or analysis will potentially enable microarray use in personalized medicine, cancer diagnostics, drug discovery, and bio-surveillance.

Publications

Beer, N. R., et al., 2011. *Hybridization and selective release of DNA microarrays.* LLNL-TR-519311.

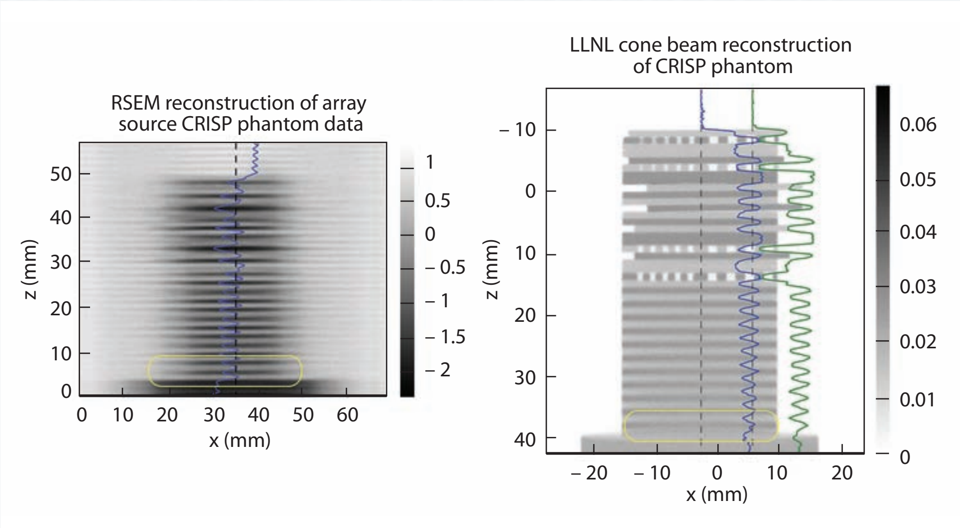
Optimized Volumetric Scanning for X-Ray Area Sources

Angela Foudray (09-ERD-045)

Abstract

Our goal is to perform a systematic study to determine optimal scanning geometries for x-ray area sources and provide a roadmap for area-source imaging in the field. Area sources have a much greater spatial coverage than the traditionally used point sources, enabling a more accurate volumetric scanning of an object. This capability is of high importance in many key efforts at Lawrence Livermore. While area sources are a very promising and critical technology, their full potential cannot yet be quantified or exploited because no study has been performed to optimize the scanning geometry for area sources. We will carry out this optimization by comparing performances of the possible scanning geometries using simulated and real data.

We expect to develop optimal scanning geometries for x-ray area sources, as well as a complete method to perform optimal scanning in the field. Specifically, we will study the feasible scanning geometries and determine which are optimal. A complete methodology for implementing the optimal scan in the field will be developed. Producing such a methodology will greatly advance the use of x-ray area sources, enabling the scientific community to reap full benefits of its greater spatial capabilities than conventional point sources. This is highly significant because the ability to



A reconstruction of array-source data collected with the CRISP (contrast and resolution interleaved stacked plate) phantom (left) is compared to a reconstruction of a single-source cone beam data (right) also collected with CRISP. Although the array-source data lacked sufficient photon count to achieve the contrast and resolution of the single-source data, cone beam artifacts are reduced in the array source data, as indicated by the yellow contours.

perform imaging with area sources will create a paradigm shift in x-ray imaging practice.

Mission Relevance

This project supports Laboratory efforts in national and homeland security as well as energy security by optimizing scanning geometries for x-ray area sources used in detectors and diagnostic instruments. Enabling improved x-ray diagnostics for components vital to advanced energy generation systems benefits the Laboratory's mission in pursuit of future clean energy sources. Improved x-ray sources for detecting explosives in luggage at airports is a key counterterrorism tool of interest to the Department of Homeland Security, and greater efficiency in weapons inspection will benefit the nation's nuclear weapons and complex integration program.

FY11 Accomplishments and Results

We collected single-source and array-source data using the CRISP (contrast and resolution interleaved stacked plate) phantom, then reconstructed both data sets using a randomized subset expectation-maximization algorithm. These reconstructions were compared to single-source cone beam data collected with the same phantom and similarly reconstructed. Although the array-source data lacked the photon count necessary for a fair reconstruction comparison with single-source data, the array-source reconstruction did exhibit fewer off-angle blurring artifacts. The next step will be to strive for higher photon counts and energy levels to verify that our approach outperforms the current single-source systems.

Publications

Foudray, A. M., and S. K. Lehman, 2011. *The contrast and resolution interleaved stacked plate (CRISP) phantom*. LLNL-TR-489551.

Lehman, S. K., and A. M. Foudray, 2011. *X-ray array sources*. LLNL-TR-503894.

Superimposed Plasmonic and Photonic Detection Platform

Sarah Baker (09-LW-003)

Abstract

Current protocols for the detection of pathogenic biological organisms in the environment are time consuming and unreliable, hampering response efforts. Our goal is to design, model, and fabricate a flow-through biosensing platform that will enable the collection, concentration, detection, and identification of low concentrations of pathogens using superimposed surface-enhanced Raman

spectroscopy and photonic crystal-based transduction methods. This radically new biosensing platform would trap organisms in flow-through pores, detect them via optical measurements of photonic bandgap across the membrane, and identify them at the surface of the membrane.

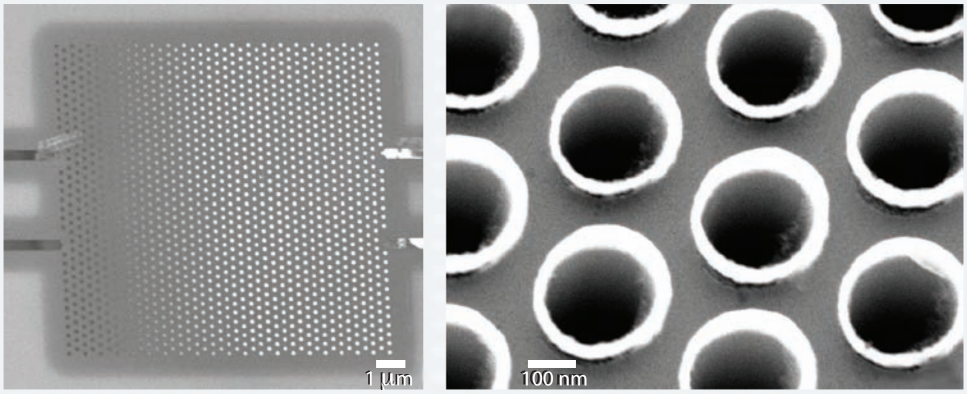
In addition to enabling rapid and sensitive biological organism detection, this work will provide the first proof of concept for the integration of photonic and plasmonic crystal-based signal transduction and separation. This new class of extremely compact and sensitive flow-through sensors will enable real-time detection of known organisms for counterterrorism, environmental, and medical applications, and enable future work focused on generating fingerprints for detecting unknown organisms. Novel design and nanofabrication approaches for integrating photonics and spectroscopy transduction will also be developed, enabling next-generation multifunctional biosensing platforms and paving the way for new membrane-based devices.

Mission Relevance

This project directly supports Livermore's efforts in the rapid detection of threats in support of the biosecurity mission by developing a novel class of platforms for identifying biological particles. Current techniques are based on polymerase chain reaction, which is not easily carried out in the field. The project takes an innovative approach to nanofabrication challenges encountered when creating multifunctional platforms. It will position LLNL as a leader in functional nanostructures, signal transduction, and organism detection and will be of interest to federal security and health agencies.

FY11 Accomplishments and Results

We (1) fabricated gold nanometer-scale rings on a photonic crystal as a test integration of plasmonic elements (the nanorings) and photonic elements into a single flow-through membrane, but found that the nanorings prevented transmission through the crystal and decided to develop and pursue other approaches; (2) used the multiphysics code COMSOL to model gold nanoring resonances and determine



A scanning electron microscopy image of a flow-through photonic crystal (left). Holes through silicon membrane appear as bright dots because of electron reflection from underneath sample. Gold nanorings fabricated on a photonic crystal membrane by templated self-assembly (right). These rings could provide a method to identify particles trapped in the membrane.

the potential for integrating the nanorings as plasmonic elements with standard flow-through membranes, finding geometries suitable for surface-enhanced Raman spectroscopy; (3) fabricated and tested the designed structures; (4) developed a method for fabricating our flow-through photonic crystal membranes; (5) determined that the refractive index sensitivity of these functional membranes was 100 nm/refractive index unit—comparable to other state-of-the-art photonic devices; and (6) used an applied electric field to verify that we could flow bio-organism simulants through the photonic crystal membranes. In summary, this project developed and demonstrated a method for fabricating multifunctional, multi-transduction sensor membranes. The prototype devices will be used as a proof of concept in exploring applications relevant to the Defense Threat Reduction Agency.

Publications

Baker, S. E., et al., 2010. "Detection of bio-organism simulants using random binding on a defect-free photonic crystal." *Appl. Phys. Lett.* **97**, 113701. LLNL-JRNL-432541.

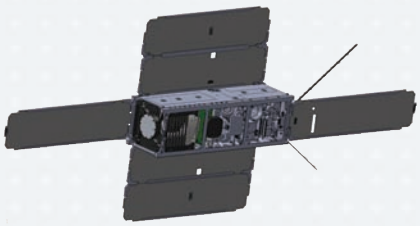
Baker, S., et al., 2009. *Silicon filtration membranes for organism capture and identification.* Molecular Foundry Users Mtg., Berkeley, CA, Oct. 15–16, 2009. LLNL-ABS-416212.

Baker, S. E., et al., 2010. *Silicon filtration membranes for organism capture, detection, and identification.* 2010 Materials Research Society Spring Mtg., San Francisco, CA, Apr. 5–9, 2010. LLNL-POST-426522.

Baker, S. E., et al., 2010. *Superimposed photonic and plasmonic flow-through nanostructures for biodetection applications.* 2010 Materials Research Society Spring Mtg., San Francisco, CA, Apr. 5–9, 2010. LLNL-PROP-464676-DRAFT.

Real-Time Space Situational Awareness

Scot Olivier (10-SI-007)



The space-based telescopes for actionable refinement of ephemeris (STARE) Pathfinder satellite being built by LLNL, Boeing, the Naval Postgraduate School, and Texas A&M University. This miniaturized satellite, scheduled for launch in 2012, will demonstrate a Livermore Laboratory concept to help prevent future collisions in space that could destroy valuable satellite systems.

Abstract

More than 80 countries have joined the space community, making Earth orbit an increasingly congested piece of aerial real estate. Hundreds of active satellites as well as thousands of pieces of space debris orbit Earth. We propose to develop and demonstrate advanced capabilities for "space situational awareness" that will result in enhanced safety for space operations. We will leverage three relevant key capabilities: extensive experience with numerous operational sensor systems, sophisticated analysis tools for interpreting data from multiple sensor systems, and unparalleled expertise in simulation and modeling of complex systems using the world's largest computers.

This project will help create a new paradigm for real-time space situational awareness. Specific outcomes will include new operational methodologies, advanced techniques and technologies, and new analysis capabilities. These new capabilities can be provided to the government as Livermore resources or transferred to other government agencies for operation. Simulation and modeling capabilities we develop can continue to be used by the government to provide an ongoing basis for selecting different technical options in this area. In addition, some of the techniques from this project would be directly applicable to other national problems, such as nuclear proliferation and climate monitoring.

Mission Relevance

Freedom of operation in a crowded space environment is crucial to U.S. interests, and maintaining space flight safety is a key component of this freedom. Utilizing unique Laboratory technical resources to help address this issue directly supports core LLNL missions in national and global security. Through this project, Livermore has an opportunity to establish a major new business area, to enhance core competencies important for basic scientific research, and to provide leadership for the U.S. in an area of intense national interest and enduring importance.

FY11 Accomplishments and Results

In FY11 we (1) developed additional advanced techniques for modeling and simulating threats to space operations by adding more detailed physical phenomena to the models of radar and optical sensors and by developing a method for modeling debris generation from satellite collisions in minutes while maintaining high accuracy, a capability for modeling satellite maneuvers, and a new capability for orbit determination from sparse data; (2) incorporated these advanced simulation and modeling techniques into a comprehensive space situational awareness framework, including upgrading sensor and analysis modules to provide feedback, which resulted in a significant improvement in our ability to quantitatively evaluate satellite threat scenarios; (3) demonstrated the use of real data for refining the orbital trajectories of space objects; (4) developed a prototype sensor design that successfully passed a series of formal engineering reviews; and (5) developed and tested multiple sensor components based on our design.

Proposed Work for FY12

In FY12 we will (1) validate techniques, developed in FY11, for modeling and simulating threats to space operations by comparing model predictions with relevant data from sensors and independent models; (2) use our integrated, comprehensive space situational awareness framework to quantify the benefits of new sensor systems; (3) demonstrate the synthesis of space-based data for refining orbital trajectories of space objects; (4) demonstrate the cueing of space-based sensors; and (5) complete our improved sensor system by integrating, testing, launching, and operating actual sensors based on the prototype we demonstrated in FY11.

Publications

de Vries, W. L., 2011. *Maneuver optimization through simulated annealing.* 12th Ann. Advanced Maui Optical and Space Surveillance Technologies Conf., Maui, HI, Sept. 13–16, 2011. LLNL-CONF-497728.

Horsley, M., 2011. *An investigation into using differential drag for controlling a formation of CubeSats.* 12th Ann. Advanced Maui Optical and Space Surveillance Technologies Conf., Maui, HI, Sept. 13–16, 2011. LLNL-CONF-498275.

Jiang, M., et al., 2011. *Computing and visualizing—reachable volumes for maneuvering satellites.* 12th Ann. Advanced Maui Optical and Space Surveillance Technologies Conf., Maui, HI, Sept. 13–16, 2011. LLNL-CONF-499168.

Nikolaev, S., et al., 2011. *Analysis of Galaxy 15 satellite images from a small-aperture telescope.* 12th Ann. Advanced Maui Optical and Space Surveillance Technologies Conf., Maui, HI, Sept. 13–16, 2011. LLNL-CONF-498573-DRAFT.

Nikolaev, S., et al., in press. "Utility of ground- and space-based sensors in assessing collision probability in near-Earth orbits." *Acta Astronaut.* LLNL-JRNL-485520-DRAFT.

Schneider, M. D., 2011. "Bayesian linking of geosynchronous orbital debris tracks as seen by the Large Synoptic Survey Telescope." *Adv. Space Res.* **49**(4), 655. LLNL-JRNL-481055.

Simms, L., et al., 2011. *Optical payload for the STARE mission.* SPIE Defense, Security, and Sensing 2011, Orlando, FL, Apr. 25–29, 2011. LLNL-CONF-474234.

Simms, L., et al., in press. "The space-based telescopes for actionable refinement of ephemeris (STARE) Pathfinder mission." *Opt. Eng.* LLNL-JRNL-485519.

Compact, Efficient Lasers for Inertial Fusion–Fission Energy

Robert Deri (10-SI-010)

Abstract

To mitigate the challenges of nuclear energy and to advance the timescale for availability of fusion sources, the Laboratory envisions a novel once-through, closed fusion–fission nuclear fuel cycle based upon the Laser Inertial Fusion Energy (LIFE) concept. We propose to develop a compact, economically viable laser system that can drive the LIFE power plant. Current laser designs required for LIFE are large and expensive. Eliminating these impediments is critically important to enabling practical fusion energy, providing abundant clean power without nuclear waste disposal, safety,

or proliferation issues. We will develop key, enabling optical technologies and use them to design a laser with performance, footprint, and costs suitable for LIFE, leading to a laser architecture and design that will guide further development. We will test key technology elements of this laser to validate our approach.

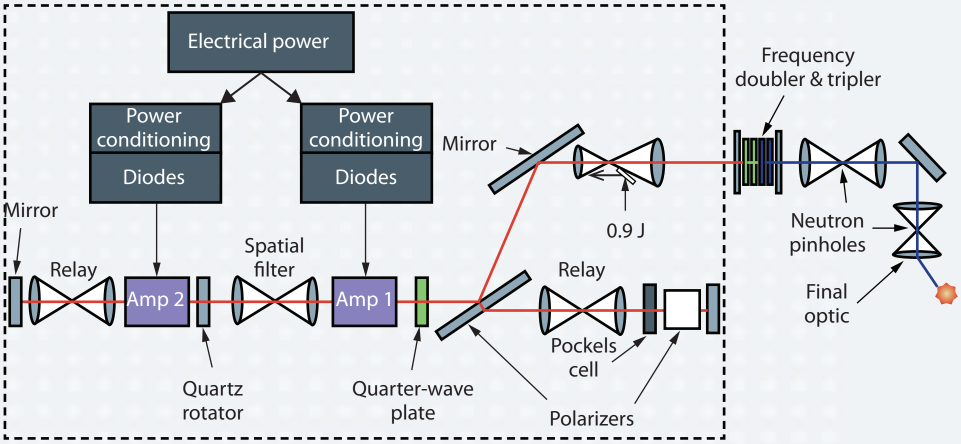
We expect to deliver an advanced laser system architecture and baseline design for LIFE that is compact and cost effective, accompanied by experimental results for key technology elements that validate our approach. Relative to today's baseline, the design could reduce the length of the optical beam path by more than fivefold, leading to a similar reduction in the building size required to house a LIFE power plant. We will develop system concepts and technology that enable a significant improvement in system costs required for the diode laser pumps. Because these components account for over 90% of LIFE laser system cost, as calculated with current technologies, this represents a significant reduction in the overall cost. These results will define a clear path forward for realizing the laser system required.

Mission Relevance

This project supports LLNL's mission of enhancing energy security for the nation and builds directly upon the Laboratory's world-class capabilities in inertial-confinement fusion and laser technologies. Resolving key issues for practical, cost-effective laser inertial fusion energy plants also supports the Laboratory mission in environmental security by eliminating nuclear waste disposal. In addition, high-power laser advances achieved by this work will enable widespread deployment of laser-driven mono-energetic gamma-ray sources, which provide unique advantages for detecting nuclear devices and reducing the threat posed by nuclear terrorism, in support of LLNL's national and homeland security missions.

FY11 Accomplishments and Results

In FY11 we (1) modified our FY10 laser design to improve efficiency by a factor of 1.5, and began detailed performance validation using propagation modeling—models for the infrared portion of the beam line are essentially complete; (2) completed a detailed comparison of three potential gain media for the laser; (3) built a subscale prototype of



Schematic view of laser architecture for a high-repetition rate, high-power and high-energy laser.

a novel spatial filter—testing of this device was postponed because of limited access to the test laser, but is scheduled for completion the first quarter of FY12; (4) fabricated Pockels cell electrodes and demonstrated that they could withstand fluences up to 2 J/cm^2, which is sufficient for our laser architecture; and (5) began to explore bonding technologies for diode arrays, and demonstrated a bonding technique to enable the scaling of a key polarization component in our architecture.

Proposed Work for FY12

In FY12 we will (1) continue to develop our laser design for improved performance and robustness, and to develop our propagation model to enhance its fidelity, particularly in the ultraviolet; (2) develop thermally robust harmonic converter designs and refine our propagation models to include cylindrical filters and improved pump delivery optics; (3) develop a prototype of a Pockels cell that eliminates the need for plasmas, based on the electrodes being demonstrated in FY11; and (4) develop a design for the final laser focusing element based on a diffractive optic.

Publications

Bayramian, A. J., et al., 2010. "A laser technology test facility for laser inertial fusion energy." *J. Phys. Conf.* **244**, 032016. LLNL-PRES-416506.

Bayramian, A., et al., 2011. *Beam in a box: A compact line replaceable unit for LIFE.* 7th Intl. Conf. Inertial Fusion Sciences and Applications (IFSA 2011), Bordeaux-Lac, France, Sept. 12–16, 2011. LLNL-PRES-496497.

Bayramian, A., et al., 2011. "Compact, efficient laser systems required for laser inertial fusion energy." *Fusion Sci. Tech.* **60**, 28. LLNL-JRNL-462671.

Bayramian, A., et al., 2010. *Compact, efficient laser systems required to harness the power of the sun.* 19th Topical Mtg. Technology of Fusion Energy, Las Vegas, NV, Nov. 7–11, 2010. LLNL-PRES-462119.

Bayramian, A., et al., 2011. "Compact, efficient, low-cost diode power conditioning for laser inertial fusion energy." *Proc SPIE* **7916**, 79160B. LLNL-PROC-464105.

Bayramian, A. J., et al., 2010. *Fusion energy: Laser systems required to harness the power of the sun.* 2010 Conf. Lasers and Electro-Optics/Quantum Electronics and Laser Science Conf. (CLEO/QELS), San Jose, CA, May 16–21, 2010. LLNL-PRES-432679.

Bayramian, A. J., et al., 2010. *Heat capacity laser.* HEC DPSSL 6th Workshop High Energy Class Diode Pumped Solid State Lasers, Versailles, France, Sept. 8–10, 2010. LLNL-PRES-452893.

Bayramian, A. J., et al., 2010. *LIFE laser system update*. HEC DPSSL 6th Workshop High Energy Class Diode Pumped Solid State Lasers, Versailles, France, Sept. 8–10, 2010. LLNL-PRES-452916.

Bayramian, A. J., et al., 2010. *LLNL DPSSL activities*. HEC DPSSL 6th Workshop High Energy Class Diode Pumped Solid State Lasers, Versailles, France, Sept. 8–10, 2010. LLNL-PRES-452915.

Bayramian, A. J., et al., 2010. *NIF update*. HEC DPSSL 6th Workshop High Energy Class Diode Pumped Solid State Lasers, Versailles, France, Sept. 8–10, 2010. LLNL-PRES-452971.

Bullington, A., et al., 2011. "Thermal birefringence and depolarization compensation in glass-based high-average-power laser systems." *Proc SPIE* **7916**, 79160V. LLNL-CONF-461272.

Caird, J. A., et al., 2009. *DPSSL design for LIFE*. 6th Intl. Conf. Inertial Fusion Sciences and Applications (IFSA 2009), San Francisco, CA, Sept. 6–11, 2009. LLNL-PRES-416598.

Deri, R., et al., 2010. *Semiconductor laser diode pumps for inertial fusion energy.* LLNL-TR-465931.

Deri, R., et al., 2010. *The LIFE laser design in context: A comparison to the state-of-the-art.* LLNL-TR-47795.

Erlandson, A., et al., 2011. *Efficiency modeling for laser inertial fusion energy drivers.* 7th Intl. Conf. Inertial Fusion Sciences and Applications (IFSA 2011), Bordeaux-Lac, France, Sept. 12–16, 2011. LLNL-ABS-480402.

Rubenchik, A. M., et al., 2010. "Modulation instability in high power amplifiers." *Optic. Express* **18**(2), 1380. LLNL-JRNL-431613.

Schaffers, K., et al., 2011. *Advanced materials for inertial fusion energy lasers.* 18th American Conf. Crystal Growth and Epitaxy, Monterey, CA, July 31–Aug. 5, 2011. LLNL-ABS-481338.

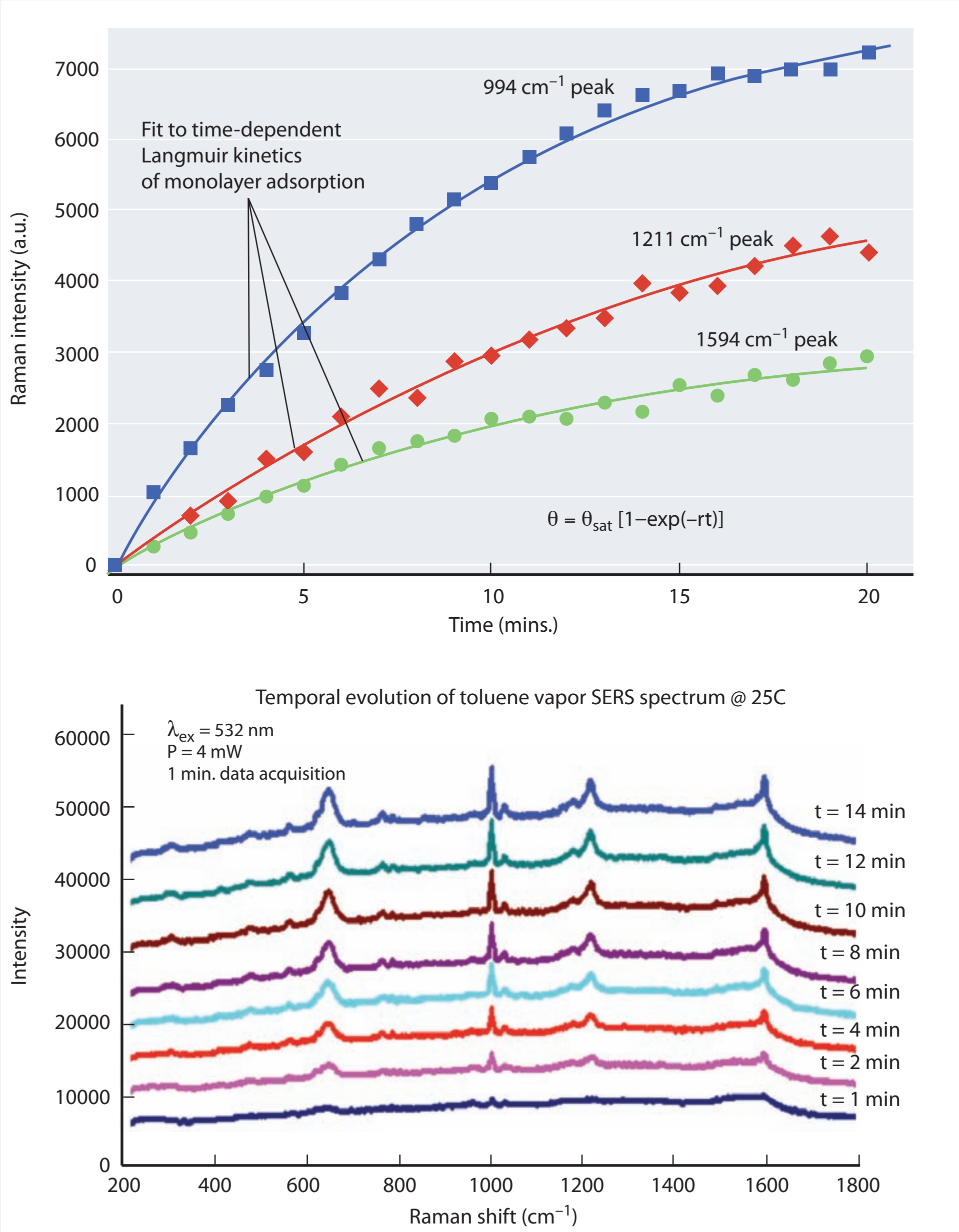
Traggis, N., et al., 2011. "Improved characterization of transmitted wavefront error on CADB epoxy-free bonded solid state laser materials." *Proc SPIE* **7912**, 79121K. LLNL-PROC-464931.

Embedded Sensors for Monitoring Complex Systems

Jack Kotovsky (10-ERD-043)

Abstract

As the nation's nuclear stockpile is reduced in size, stockpile surveillance approaches must change significantly to increase safety, security, and cost effectiveness. We will explore broad-spectrum sensing technologies for this purpose. Several embedded sensing methods that have not been previously explored will be pursued. Broad-



The kinetics of surface-enhanced Raman scattering (SERS) as indicated by peak-to-background measurements (top). Langmuir kinetics behavior is indicated by substrate exposure time to toluene vapor as expected with a mean time constant of 12 minutes (bottom).

spectrum sensors most efficiently address the surveillance challenges and deliver the greatest overall impact. For this reason, this effort pursues the broad-sensing technology of gas sensing. Techniques for assessing noble and non-noble species will be considered using methods that are compatible with actual stockpile applications.

If successful, several new sensing capabilities will be produced for stockpile surveillance. Specifically, optic-fiber-based surface-enhanced Raman scattering (SERS), photo-acoustic spectroscopy (PAS), and ionization techniques will be considered for detection of unknown gas mixtures. Novel materials, fabrication processes, and designs will be explored for their applicability to the difficult constraints of in situ state-of-health stockpile monitoring.

Mission Relevance

If successful, this project will lay the groundwork for a game-changing comprehensive sensor suite that will dramatically enhance stockpile surveillance and significantly advance the entire nuclear weapons complex, in support of the Laboratory's national security and stockpile stewardship missions.

FY11 Accomplishments and Results

We (1) tested PAS spectrometer designs with the laser system we designed in FY10; (2) used a glove-box test environment with CO_2 monitoring to quantify the performance of the PAS designs; (3) conducted extensive modeling with the COMSOL multi-physics code to further improve the PAS designs, exploring system sensitivities to acoustic resonator shape and size, laser stimulus, and detector geometries and thereby producing designs that anticipate enhanced detection sensitivity and include several features to speed experimental development of the system; (4) began fabricating the modified PAS system that we designed; (5) used a laser vibrometer system with a custom fiber assembly to continue miniaturization of the overall detection system; and (6) continued using a system based on 532-nm fibers to study toluene vapor as a representative of volatile organic compounds, including time and temperature studies of the vapor SERS signal that provided new insights on adsorption kinetics—observing, for instance, that the peak intensity of the three main modes saturates with time in agreement with a Langmuir trend.

Proposed Work for FY12

In FY12 we will (1) achieve an order-of-magnitude improvement in SERS sensitivity by upgrading our current regulator-based pressure-control system to a concentration-control scheme that uses our newly built delivery system with mass-flow controllers; (2) continue theoretical and experimental studies of adsorption and desorption kinetics for SERS; (3) provide the first fiber-based measurements using photonic crystal fibers coated with metal nanoparticles; (4) adapt vibrometer hardware to achieve a system-relevant fiber-based detection system for use with PAS, reserving a Fabry–Perot readout scheme as an alternative approach if the vibrometer hardware turns out not to perform as well as we hope; (5) continue refined modeling of latest-generation prototypes to enhance PAS detection limits; and (6) test and improve the PAS noise-cancellation system.

Publications

Bond, T. C., et al., 2011. *Broadly tunable plasmonic nanogap resonators or black plasmons.* 4th EOS Topical Mtg. Optical Microsystems, Capri, Italy, Sept. 26–28, 2011. LLNL-PRES-502853.

Bond, T. C., et al., 2011. *Nanostructures for surface-enhanced spectroscopy.* 2011 CMOS Emerging Technologies Workshop, Whistler, Canada, June 15–17, 2011. LLNL-PRES-487811.

Bond, T. C., et al., 2010. *Large-area nanopillars SERS arrays.* 38th Ann. Conf. Federation of Analytical Chemistry and Spectroscopy Societies (FACSS 2010), Raleigh, NC, Oct. 17–21, 2010. LLNL-POST-433531.

Bora, M., et al., 2010. "Plasmon-resonant cavities in vertical nanowire arrays." *Nano Lett.* **10**, 2832. LLNL-JRNL-425128.

Bora, M., et al., 2010. *Vertical pillar arrays as plasmon nanocavities.* 2010 Solid-State Sensors, Actuators, and Microsystems Workshop, Hilton Head, SC, June 6–10, 2010. LLNL-ABS-420921.

Chang, A. S., et al., 2011. "Nanopillar array for surface enhanced Raman scattering." *Proc. 2011 SPIE Defense, Security and Sensing.* LLNL-PROC-480794.

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 to quantify carbon isotope ratios from atmospheric CO_2 at concentrations as low as one ^{14}C atom per 10^{13} atoms of carbon

using coupled rotational–vibrational excitation lines. We will use a prototype cavity ring-down spectroscopy system to measure $^{14}CO_2$, and develop a novel approach to measure ^{14}CO derived from $^{14}CO_2$. 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 ^{14}C 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 ^{14}C measurement methods and maintain LLNL as the leader in the biomedical and environmental uses of radiocarbon by enabling a new, transformative ^{14}C 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 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.

FY11 Accomplishments and Results

In FY11 we (1) established a three-party Commercial Research and Development Agreement with our industry partner and university collaborator, (2) began fabricating our cavity ring-down spectroscopy instrument and hired a graduate student to assist with this device, (3) designed and began construction of a CO_2-to-CO conversion cell as part of our novel optical spectrometry approach, and (4) modeled the spectra of CO_2 and CO, demonstrating that we should be able to reduce the interfering isotopomers for the CO_2 spectrum, but finding that there may be issues with the CO spectrum.

Proposed Work for FY12

In FY12 we will (1) begin testing our prototype cavity ring-down spectroscopy system with atmospheric gases, (2) test system response with elevated levels of $^{14}CO_2$, (3) begin benchmark testing by comparing the results from this system with results from accelerator mass spectrometry, (4) continue testing the CO_2–CO conversion cell, and (5) finish evaluating the overall approach based on CO_2–CO conversion and decide whether or not to move forward with it.

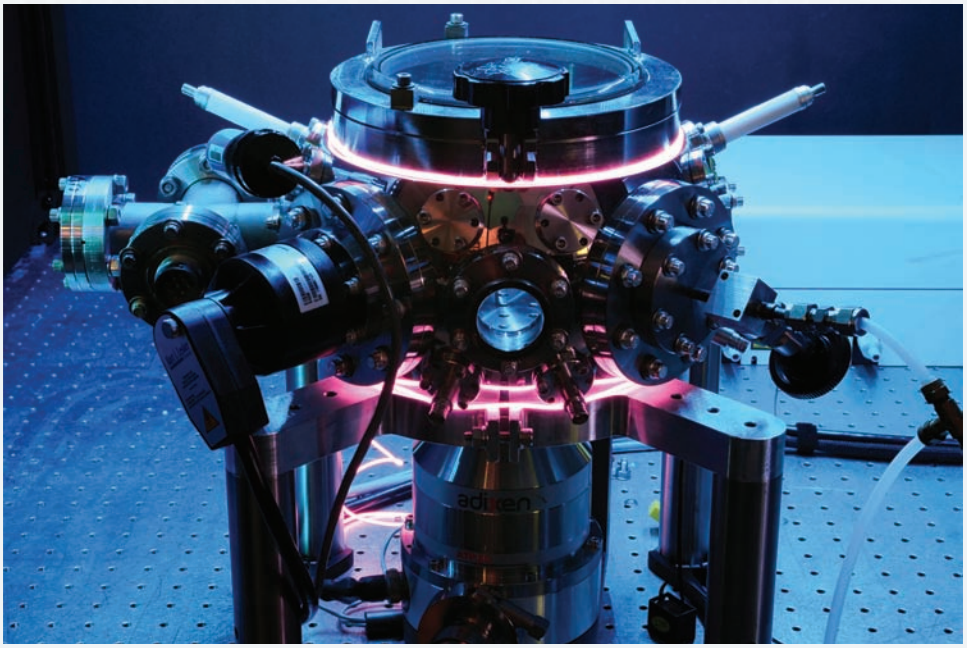
Ultrafast, Sensitive Optical Radiation Gamma, Neutron, and Proton Detector Development

Stephen Vernon (11-ERD-052)

Abstract

The National Ignition Facility will enable the weapons physics and high-energy-density scientific communities to investigate previously inaccessible plasma physics, particularly phenomena associated with the deuterium–tritium fusion burn process 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 very short temporal durations 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 the fusion gammas, neutrons, protons, x rays (in some cases), and relatively weak signals expected from fusion-class experiments, we propose to develop a very-high-speed detection capability with extremely good particle-measurement quantum efficiencies 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



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

signals (usually electrons), (2) develop electron optics techniques for concentrating 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.

FY11 Accomplishments and Results

Major accomplishments for FY11 include significant progress in modeling and experimental development of the sensor systems. Specifically, theoretical accomplishments included completion of model-based designs for both 900-nm and 1550-nm electron sensors and modeling analysis of the conversion efficiency and temporal response of gamma and neutron converters using the Monte Carlo radiation transport code MCNPX—initial MCNPX simulations results are extremely encouraging and indicate that appropriately designed gamma and neutron converters can produce secondary electrons with approximate picosecond temporal response. We also made significant progress toward developing a model analysis of the Cherenkov process in the gamma converter using the GEANT geometry and tracking code, as well as developing preliminary designs for the electron optics system using the SIMION electric field and ion trajectory code. Experimental accomplishments included the design and implementation of an experimental system to enable rapid prototyping and characterization of candidate electron optics systems, beginning the multistep process to fabricate both 900-nm and 1550-nm Fabry–Perot electron sensors. We also performed characterization and optimization of an optical streak camera system that is a potential recorder for 900-nm implementation.

Proposed Work for FY12

In FY12 we will (1) fabricate prototype neutron and gamma converters and characterize them using designs guided by, and developed in conjunction with, the simulation results obtained in FY11; (2) design, fabricate, and test prototype electron optics systems; (3) perform tests by integrating the electron optics systems with prototype electron sensors demonstrated in FY11, using short-pulse (266-nm) ultraviolet laser radiation to generate picosecond electron pulses with a back-side-illuminated gold photocathode maintained in high vacuum; and (4) demonstrate an integrated detector consisting of an optimized electron demagnification system for electron flux concentration and an optimized radoptic electron sensor.

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

Jerry Carter (11-ERD-061)

Abstract

A recognized need exists 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 lack of size constraints, a rugged device for operation in a wide range of environments, high sensitivity with low sample volume, and immunity to environmental noise.

We expect our research will significantly improve the sensitivity of PARS and provide a viable alternative to traditional optical-signal-based measurement techniques that suffer a number of limitations for multiple applications. For many applications, 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.

FY11 Accomplishments and Results

In FY11 we (1) designed a multi-pass Raman converter cell, two acoustic resonator cells, and a microphone-based acoustic detector for proof-of-concept measurements using hydrogen–nitrogen gas mixtures; (2) completed our tuning-fork PARS experimental setup, which combines the 266-nm pulsed excitation of a neodymium-doped yttrium aluminum garnet laser with our multi-pass Raman converter; and (3) integrated our achievements into a new analytical technique for the non-optical, in situ remote gas and condensed-phase material characterization and mapping of dynamic processes, in either imaging or non-imaging modalities, and submitted a record of invention for this technique. We also presented results on our dynamic model, experimental platform design, and performance analysis for the pulsed and resonant PARS analysis of gases at an Acoustic Society of America conference—a presentation that was also published in the Journal of the Acoustical Society of America.

Proposed Work for FY12

In FY12 we propose to complete the resonantly detected PARS analysis of gases and direct our attention toward proof-of-concept demonstrations using solid-phase analytes. Specifically, we will (1) extend the theoretical system performance model developed in FY11 to include solid-phase materials; (2) construct proof-of-principle experiments for generating a PARS signal of model solid materials using our tuning fork and acoustic resonator setup; (3) configure an experimental setup including the laser system, a high-quality tuning-fork resonator and associated electronics, and acoustic resonator cell; and (4) perform preliminary experimentation and analysis of single-crystal and amorphous solid samples to demonstrate the feasibility of our approach.

Publications

Chambers, D. H., and J. C. Carter, 2011. "Modeling of photoacoustic Raman spectroscopy." *J. Acoust. Soc. Am.* **128**(4), 2412. LLNL-PRES-484444.

Non-Acoustic Secure Speaker Verification in High-Noise Environments

John Chang (11-LW-042)

Abstract

We will experimentally demonstrate a capability for noninvasive, real-time voice authentication and alteration using non-acoustic electromagnetic voice sensing. We will first build a dual-transducer sensor system using existing LLNL micro-power radar technology and a traditional microphone, and then evaluate the system on a statistically relevant sample of subjects. We will also develop a new near-field, low-profile antenna that provides efficient sensing of the vocal cord region of the speaker. Building upon prior work on single-parameter detection algorithm concepts, we will also develop the multiparameter detection algorithms and architecture for high-confidence, real-time speaker validation, verification, and alteration.

We will demonstrate the noninvasive in situ electromagnetic sensing of the sound-producing tissues of the human voice box, which will realize new capabilities in biometrics, de-noising, and secured communications. This capability could be used, for instance, to authenticate an individual based upon prior baseline information and to detect intrinsic levels of behavioral stress not detectable by traditional acoustic means. By integrating real-time processing and filtering with a new "cone of silence" algorithm

we develop, we can realize secured peer-to-peer communication in the form of a miniaturized radar device.

Mission Relevance

This project supports the Laboratory's national security mission by developing a new capability with applications in secured communications, cybersecurity, and biosecurity.

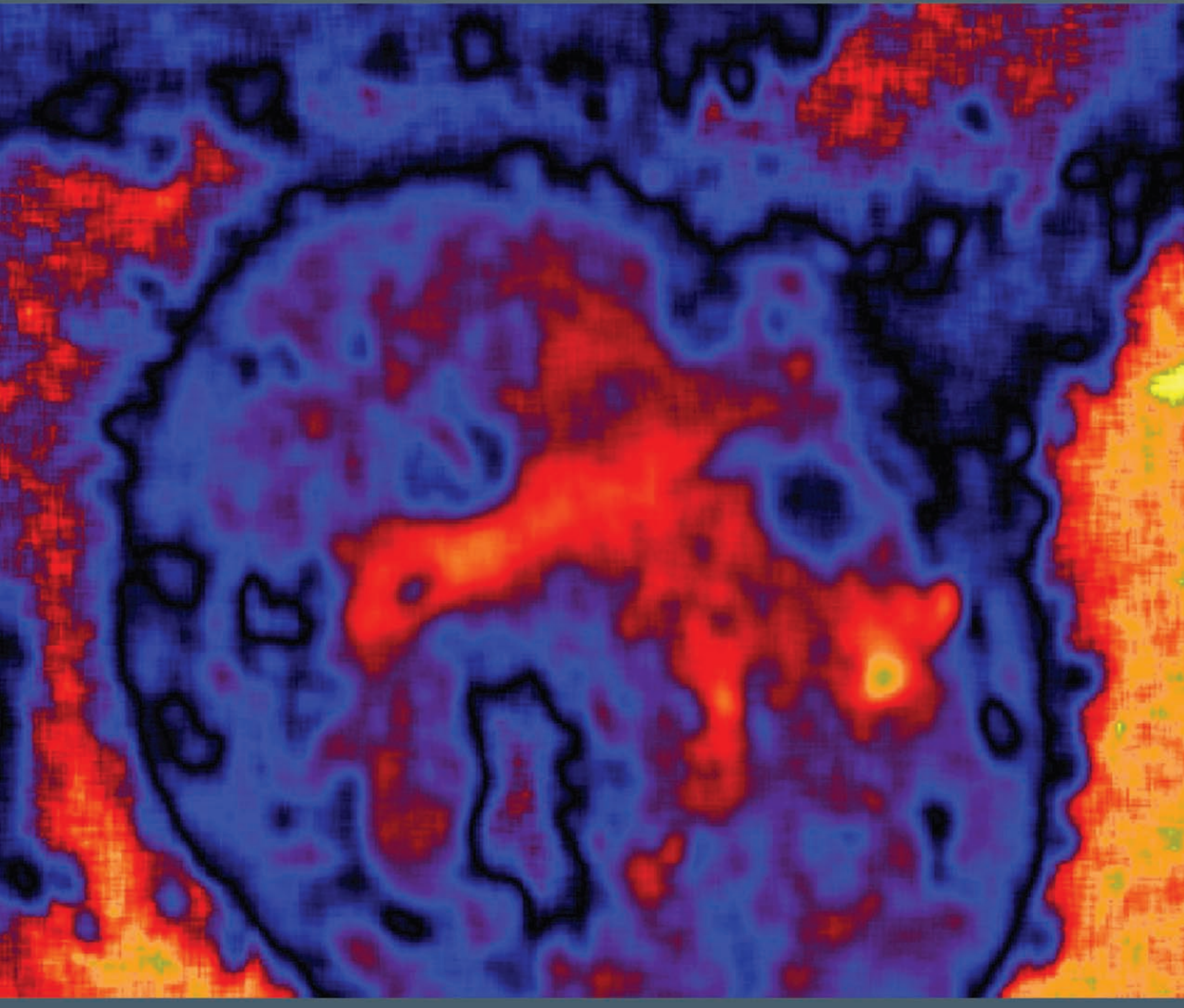
FY11 Accomplishments and Results

In FY11 we (1) built a preliminary version of a dual-transducer sensor system using noninvasive micro-power radar combined with traditional microphones as the baseline sensor suite, achieving a functional system; (2) began evaluating different near-field antenna designs that enable efficient sensing of the vocal cord region, including the prototype antennae suggested by initial results; (3) used a conventional signal-processing algorithm and existing data to identify signal features that could enable anticipatory voice-activation capabilities; and (4) made preparations for human subjects research in FY12.

Proposed Work for FY12

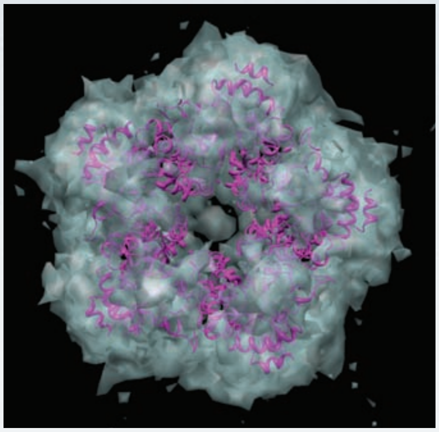
In FY12 we will (1) continue with human subject data acquisition, data analysis, and documentation; (2) demonstrate and document a background noise-removal algorithm; (3) demonstrate and document a speech-silencing algorithm; and (4) demonstrate a real-time voice signature-alteration algorithm.

Biological Sciences



Laboratory Directed Research and Development

FY2011



An electron microscopy image of the protein LsrF, with the ribbon representation of the protein model overlaid (purple).

Coupling Advanced Cryo-Electron Microscopy with High-Performance Computing to Resolve Biomolecular Function

Felice Lightstone (09-ERD-009)

Abstract

Our objective is to combine advancements in aberration-corrected cryo-electron microscopy with high-performance computing to rapidly determine biomolecular structures and their function. We plan to be the first to use aberration-corrected electron microscopy to image a protein at atomic resolution while avoiding the current limiting factor of protein crystallization. Specifically, we will (1) adapt ion mobility to improve sample preparation and deposit more homogeneous populations of proteins on a clean substrate, (2) obtain aberration-corrected electron microscopy data on protein, (3) develop image-processing techniques to reconstruct the three-dimensional image and model, and (4) apply high-performance computing to simulate the protein to predict its function.

If successful, we will be the first to use aberration-corrected electron microscopy to image a protein complex at atomic resolution, which will revolutionize the biological field by eliminating the need to crystallize samples prior to structural determination. Ultimately we will develop a single-molecule method for determining biomolecular structures at resolutions conducive to rational drug design. To achieve this, we will make advancements in each step necessary for three-dimensional reconstructions of proteins, including sample preparation, high-resolution imaging, and algorithms for image analysis. By utilizing constraints revealed from the solved structure with high-performance-computing simulations, we will also begin to probe the predicted function of the protein.

Mission Relevance

The national and homeland security missions of Lawrence Livermore include developing new countermeasures to combat chemical and biological warfare threats. Our success will enable a new capability to determine structures and functions of unknown proteins in host–pathogen pathways, which will provide rational approaches that can be applied to the development of new therapeutics and pre-treatments and can be applied to all fields of biology.

FY11 Accomplishments and Results

In FY11 we (1) optimized our sample preparation techniques and applied them to inhomogeneous protein complexes; (2) collected aberration-corrected cryo-electron microscopy data on the LsrF and LsrF–LsrG protein complexes; (3) implemented a new, fully two-dimensional noise-canceling algorithm and tested it on simulated and acquired data sets, finished implementing the performance-measurement algorithm, computed its quantitative performance, and implemented and tested iterative image-restoration algorithms to mitigate image distortion; and (4) simulated the LsrF and LsrF–LsrG proteins using our models on a high-performance computing platform. In summary, this project has enabled the development of high-quality, conformational separated biological sample preparation, developed new algorithms for data classification, and improved signal-to-noise ratios and iterative image restoration. Our sample-preparation and image-processing techniques were adopted by LLNL researchers in developing a free-electron laser process for imaging biological samples at the Linac Coherent Light Source, with the ultimate goal of advancing biological structure determination to the level of single molecules.

Publications

Benner, W. H., et al., in press. "Re-electrospraying splash landed ions and nano-particles." *Anal. Chem.* LLNL-JRNL-516331-DRAFT.

Flexible and Rapid Therapeutic Countermeasures for Global Biosecurity

David Rakestraw (09-ERD-054)

Abstract

Emerging and engineered infectious diseases are a threat to political, social, and economic stability. A robust global biodefense strategy requires anticipation, detection, and rapid response. Currently, biodefense is guided by knowledge of state-sponsored bioweapon programs and a list of biological threat agents. This strategy is poorly suited to address the rapidly evolving nature of biological threats. Our goal is to create a science and technology base to significantly reduce development time for antimicrobial drugs. The foundation we lay will help reduce drug development times to months rather than years. This project integrates advanced scientific computing, microfluidics, accelerator mass spectrometry, and select-agent science to create a unique approach for rapid development of new therapeutics.

We expect to develop automated, high-throughput extraction and culturing techniques for processing existing libraries of marine natural products and chemical genomics, which will make it possible to create a huge new library of drug candidates from these compounds. We will develop techniques for rapidly identifying drug candidates using host gene expression, gene knockout libraries, and metallome analyses as well as employing model systems to illuminate these pathways and identify factors involved in the early immune response to pathogen infection. In addition, we will accelerate the process for predicting the effectiveness of drugs using ultrasensitive accelerator mass spectrometry.

Mission Relevance

This project supports LLNL's mission to reduce or counter threats to national security by helping enable a flexible biodefense capability to detect and characterize unknown and engineered pathogens and rapidly develop new medical countermeasures.

FY11 Accomplishments and Results

In FY11 we (1) completed a drug interaction study between caffeine and ciprofloxacin in rats to demonstrate linearity of absorption, distribution, metabolism, excretion, and toxicity properties over a wide dose range—experiments confirmed that the pharmacokinetics of ciprofloxacin in rats is linear, therefore the micro-dose is predictive of the pharmacokinetic at the macro-dose; (2) identified a gap in the flavin biosynthesis annotation in the *Yersinia pestis* plague bacterium by genome-wide computational analysis—several putative phosphatases were proposed to fill the gap, models of their three-dimensional structures were built, and interactions with the model enzymes' known substrates were examined; (3) completed a lead-compound screening for an inhibitor to the pathogenic bacterium *Francisella tularensis* REP24 using the library of all commercially available drug-like compounds (approximately one million); (4) identified two compounds from a library of drugs approved by the U.S. Food and Drug Administration that exhibit activity against the multidrug-resistant *Acinetobacter baumannii* pathogenic bacterium and select-agent surrogate pathogens; (5) completed a multi-variable array, bacterial infection, transcriptomics experiment of *Y. pestis* gene expression during human cell infection; and (6) determined the crystal structure of an inhibited co-complex of a *F. tularensis* rapid-encystment protein. The successful completion of this project helped to identify new potential targets for countermeasures, new lead compounds for countermeasures, and further established micro-dosing as an effective method for studying pharmacokinetics of drug compounds. Data, methods, and conclusions drawn from this project helped to provide preliminary results, credentials, and capabilities that helped to generate new extramural funding from the National Institutes of Health and the Defense Threat Reduction Agency, and provides the basis for a number of other planned research proposals.

Publications

Cadag, E., et al., 2012. "Computational analysis of pathogen-borne metallo β-lactamases reveals discriminating structural features between B1 types." *BMC Res. Notes* **5**, 96. LLNL-JRNL-476671.

Segelke, B. W., et al., 2010. *Structure-function of* Y. pestis *LsrG, a protein involved in AI-2 quorum sensing.* LLNL-JRNL-427462.

Wheeler, K. E., et al., 2010. "Functional insights from computational modeling of orphan proteins expressed in a microbial community." *J. Proteomics Bioinform.* **3**, 266. LLNL-JRNL-421048.

Zemla, A., and B. W. Segelke, 2008. *Practical applications of structural genomics technologies for mutagen research.* 39th Ann. Mtg. Environmental Mutagen Society, Rio Grande, PR, Oct. 18–22, 2008. LLNL-PRES-407861.

Zemla, A., et al., 2011. "StralSV: Assessment of sequence variability within similar 3D structures and application to polio RNA-dependent RNA polymerase." *BMC Bioinformatics* **12**, 226. LLNL-JRNL-463671.

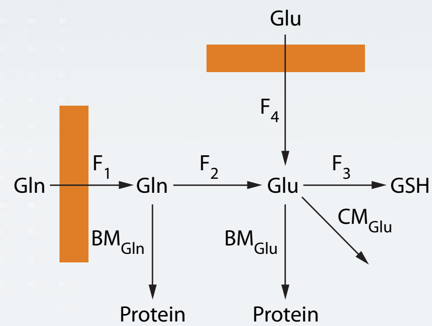
Biological Testing of Systems Biology: Validation of Flux-Balance Analysis Predictions

Benjamin Stewart (09-ERI-002)

Abstract

Flux-balance analysis can determine metabolic capabilities of cellular systems and growth rates of organisms, but current techniques for measuring metabolic fluxes lack sensitivity and rely only on concentration measurements from which flux ratios are calculated. The objective of this project is to develop a protocol for measuring metabolite fluxes to test, validate, and further constrain flux-balance models. We will use accelerator mass spectrometry to measure the rate of metabolite formation using a tracer labeled with carbon-14 and use these results to improve flux-balance simulations for the yeast *Saccharomyces cerevisiae*. We will also develop a protocol using accelerator mass spectrometry to measure fluxes for amino acid biosynthesis metabolites as a proof of concept to constrain and validate flux-balance models.

Using whole-cell labeling techniques, we will determine the rate of incorporation of glucose metabolism products into cellular pools of DNA, protein, lipids, and small molecules in yeast cells over time. We will also develop an extraction method and high-performance liquid chromatography protocol for separating and quantifying several metabolites and free amino acids along phenylalanine and tyrosine biosynthesis pathways. The extraction and purification protocols will be used to measure the fluxes of metabolite intermediates chorismate, prephenate, and arogenate and the free amino acids phenylalanine and tyrosine. Experimentally derived fluxes will be compared to fluxes predicted by the existing flux model for yeast and used to experimentally constrain and validate the model.



External and internal metabolic fluxes culminating in the production of the antioxidant glutathione were measured in the yeast *Saccharomyces cerevisiae* using accelerator mass spectrometry and other analytical methods. These experimental results were then used to significantly improve a mathematical model of yeast metabolism, enhancing the model's ability to identify target pathways for bioengineering applications such as drug development and biofuel production.

Mission Relevance

By developing advanced flux-modeling technology with biodefense and bioenergy applications, this project supports LLNL's missions in national and energy security by assisting efforts in the rapid detection and characterization of emerging and unknown biothreats as well as efforts to deliver clean energy systems.

FY11 Accomplishments and Results

We successfully measured extracellular and intracellular amino acid fluxes in the yeast *S. cerevisiae* using a combination of analytical methods including accelerator mass spectrometry. We then used our experimental data to constrain the iDN750 computational model of yeast metabolism. The applied experimental constraints resulted in a nearly 25% reduction in model uncertainty. Our results demonstrated that internal metabolic flux measurements provided more information than external flux measurements and were more useful in constraining the computational model. We performed additional flux experiments to identify amino acid metabolic pathways affected by oxidative stress mediators and to determine the relationship between glucose flux and generation of the toxic metabolite methylglyoxal. These results demonstrated that experimental data can significantly reduce uncertainty in mathematical models of cell metabolism and improve model predictions. We intend to seek support from the National Institutes of Health to investigate the contribution of methylglyoxal flux to the pathology of type 2 diabetes mellitus.

Publications

Sporty, J., et al., 2009. "Quantiation of NAD+ bioynthesis from the salvage pathway in *Saccharomyces cerevisiae*." *Yeast* 26, 363. LLNL-JRNL-410719.

Stewart, B. J, et al., 2011. *Glutathione and amino acid flux measurement under normal and oxidative stress conditions using HPLC and accelerator mass spectrometry*. Society of Toxicology National Mtg., Washington, D.C., Mar. 6–10, 2011. LLNL-POST-471528.

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Stewart, B. J., et al., 2010. *Yeast metabolic flux measurement by accelerator mass spectrometry*. American Chemical Society National Mtg., San Francisco, CA, Mar. 23, 2010. LLNL-POST-425375.

The Role of Dendritic Cells in Tularemia Pathogenesis

Sahar El-Etr (09-LW-036)

Abstract

Our objective is to understand the pathogenesis of *Francisella tularensis*, with the ultimate goal of helping to develop countermeasures for tularemia (rabbit fever). Hypothesizing that *F. tularensis* disrupts host cellular trafficking pathways upon entry into immune cells, we will identify and localize the stage at which this disruption occurs and identify the bacterial and host genes involved in the process. Using immunofluorescent microscopy and biochemical analysis and a panel of pathogenic and nonpathogenic strains, we will compare the localization of known protein markers along the trafficking pathway of human dendritic cells and identify the stage where

F. tularensis causes aberrant host-protein localization. We will also identify bacterial proteins involved in the process.

We expect this study to result in the identification of host proteins that are directly disrupted by *F. tularensis*. These findings will have a broad impact on development of potential therapeutics for tularemia and possibly other intracellular pathogens that interfere with host immune responses in similar ways. In addition, identifying bacterial proteins involved in the disruption of host cellular functions will provide potential bacterial candidates for vaccine development. Our study will be the first ever to compare the behavior of multiple pathogenic and nonpathogenic strains in human host cells and will have a broad impact on the scientific community, as well as provide critical information on the pathogenesis of tularemia.

Mission Relevance

Understanding the mechanisms of pathogenesis of *F. tularensis* and identifying bacterial and host genes involved in the virulence process will contribute significantly to development of medical countermeasures against tularemia, a Class A biothreat agent. This project thus supports LLNL's national and homeland security mission areas of countering the use of weapons of mass destruction and strengthening homeland security.

FY11 Accomplishments and Results

In FY11 we completed the analysis of endosome pathway disruption by *F. tularensis* strains and identified steps that were unique to the pathogenic strains. Although isolation of the phagosomes and proteins from infected cells was not possible because of equipment capabilities in the biosafety laboratory, we were able to identify host genes that are involved in the immune response to pathogenic *F. tularensis* 2, 6, 24, and 48 hours after infection, and identified differences between host proteins responding to infection by pathogenic and nonpathogenic strains. These host proteins identified are likely to be involved in pathogenesis and would represent targets for developing countermeasures against tularemia. In this study we have shown that pathogenic *F. tularensis* strains escape the endosome pathway in dendritic cells faster and in greater numbers than nonpathogenic strains. This suggests that pathogenic *F. tularensis* actively secrete effectors that modulate their escape from the endosome pathway and underscores important differences between animal and human innate immune response to *F. tularensis* infection. We also observed production of chemokines that suggest that pathogenic *F. tularensis* strains may actively inhibit the human inflammatory response post-infection and subsequent antigen process, a strategy that is likely employed by a number of relevant biowarfare agents. Future research will focus on identification of bacterial effector proteins involved in this process and the host proteins that they interact with. We expect these findings to have broad implications on the identification of potential drug or vaccine targets for tularemia. We are hopeful that the Defense Threat Reduction Agency will sponsor further research to examine the antigen process by dendritic cells in response to *F. tularensis* infection.

Publications

El-Etr, S. H., 2011. *The role of dendritic cells in Tularemia pathogenesis.* LLNL-TR-520212.

El-Etr, S. H., L. Vergez, and A. Rasley, 2011. *Differential intracellular trafficking of* Francisella tularensis *strains in human dendritic cells.* IMMUNOLOGY 2011: 98th Ann. Mtg. American Association of Immunologists, San Francisco, CA, May 13–17, 2011. LLNL-POST-483430.

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Verges, L., A. Rasley, and S. H. El-Etr, 2010. *Differential endosomal trafficking of pathogenic and nonpathogenic* Francisella tularensis *strains in human dendritic cells.* American Society of Microbiology 110th General Mtg., San Diego, CA, May 23–27, 2010. LLNL-POST-432756.

Verges, L., A. Rasley, and S. H. El-Etr, 2011. Francisella tularensis *Type A strains modulate cytokine production in human dendritic cells post infection.* ASM2011: General Mtg. American Society for Microbiology, New Orleans, LA, May 21–24, 2011. LLNL-POST-484452.

Verges, L., et al., 2010. *Pathogenic* Francisella tularensis *exhibit differential trafficking in human dendritic cells: Addressing the therapeutic potential for dendritic cell targeting drugs in human tularemia.* Chemical and Biological Defense Science and Technology Conf., Orlando, FL, Nov. 15–19, 2010. LLNL-POST-458478.

Versatile Delivery and Immune-Stimulatory Platform for Just-in-Time Vaccine Development

Craig Blanchette (09-LW-077)

Abstract

The goal of this project is to develop new vaccines in which a versatile and rapidly manufactured platform can be used for efficiently delivering pathogenic antigens. Although traditional use of whole- or killed-cell pathogens has proven successful in the past, this method is plagued with technical difficulties. Currently, researchers are attempting to use subunit vaccines as a substitute but have had little success in getting efficient delivery of the recombinant protein to immunogenic cells. We propose to use nickel–nanolipoprotein (NLP) particles as an antigen-delivery vehicle. This method will allow us to combine the delivery vehicle, immune stimulation, and

pathogen antigen into a single entity, thus eliminating problems currently facing researchers in the field.

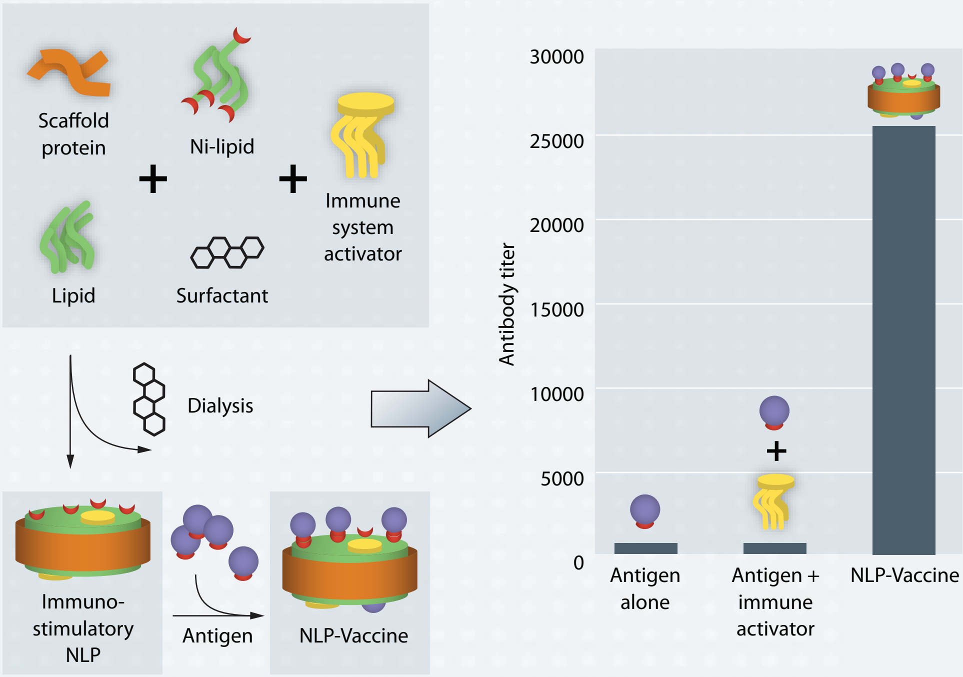
The expected result of our proposed research is just-in-time vaccine production in which a robust and highly effective antigen-platform vaccine is manufactured from a gene of interest and ready for use in a matter of hours. This technology will also greatly enhance the field of disease prevention by helping to develop vaccines against hundreds if not thousands of pathogens. Furthermore, development of this vaccine technology will launch a whole new and unexplored field of vaccine development.

Mission Relevance

Our proposed work is crucial to developing potent vaccines that can be applicable to both disease prevention and increased biodefense capabilities, in support of Laboratory missions in homeland and national security.

FY11 Accomplishments and Results

In FY11 we (1) successfully cloned, expressed, and purified polyhistidine-tagged *Francisella tularensis* (tularemia) and *Yersinia pestis* (anthrax) antigens in *Escherichia coli* bacterial expression systems; (2) evaluated the immune-stimulating properties of nickel–NLPs containing adjuvant lipid A and the cholesterol cytosine–phosphate–guanine sequence in vivo using the antigens; and (3) tested the efficacy of these NLP-based vaccines in live pathogen-challenge experiments at Livermore's the biosafety level 3 laboratory. This project successfully demonstrated the potential efficacy of using the nickel–NLP platform as an immunostimulatory delivery vehicle for vaccine applications. The data generated in this project attracted interest from the



Nanolipoproteins assembled with immune-stimulating compounds and antigens significantly enhance antibody production. This technology forms a vaccine-delivery vehicle being investigated for use against pathogens such as those that cause anthrax and tularemia.

National Institutes of Health and the Department of Defense to continue developing this platform as a vaccine-delivery vehicle against *B. anthracis*, *F. tularensis*, and *Burkholderia* bacteria species.

Publications

Blanchette, C. D., et al., 2010. "Kinetic analysis of His-tagged protein binding to nickel-chelating nanolipoprotein particles." *Bioconjugate Chem*. **21**, 1321. LLNL-JRNL-425115.

Fischer, N. O., et al., 2010. "Conjugation to nickel-chelating nanolipoprotein particles increases the potency and efficacy of subunit vaccines to prevent West Nile encephalitis." *Bioconjugate Chem*. **21**, 1018. LLNL-JRNL-420436.

Fischer, N. O., et al., 2010. "Isolation, characterization, and stability of discretely-sized nanolipoprotein particles assembled with apolipophorin-III." *PLoSONE* **5**, e11643. LLNL-JRNL-427664.

Genomics of Cell to Cell Communication: Identification of DNA Sensors in Humans

Gabriela Loots (10-ERD-020)

Abstract

Communication between individual cells is an essential process in all living organisms. Although several signal transduction pathways have been identified, we do not yet know how a signal from the cell surface is interpreted at the genomic level to control transcription. Furthermore, tumor formation is a direct result of the failure to carry out normal intercellular communication. We propose to undertake a genomic approach to identify noncoding regions in the human genome that are activated in a receiver cell by signals from a cancer cell. We will use the metastasis of prostate cancer to bone as a model for testing our hypothesis and for identifying genomic signals that are activated in response to prostate cancer cells.

Our experiments will address the consequences of intracellular communication and identify changes in gene expression that are induced either in osteoblasts—cells responsible for the synthesis and mineralization of bone—or prostate cancer cells that are grown in mixed co-cultures. We will elucidate gene expression changes in prostate cells in response to osteoblasts, as well as changes in osteoblasts resulting from cancer response. The goal of these experiments is to gain insight into how osteoblasts respond to the presence of cancer cells in an in vitro, co-culture system as a first approximation of changes in gene expression that may be involved in early stages of bone metastasis. This study will be the first global genomic survey of gene expression in response to intracellular communication in the context of bone metastasis.

Mission Relevance

We will use cancer metastasis as a model of intracellular communication to develop novel genomic tools and methods that will help us understand what regulatory elements are critical for sensing tumors, which would comprise a significant advancement in basic biology in support of LLNL's mission in the basic sciences. These tools can then be applied to understanding how cells recognize pathogens, including potential bioterror pathogens, in support of the Laboratory's national security mission area of countering bioterrorism.

FY11 Accomplishments and Results

In FY11 we (1) isolated RNA from osteoblasts and conducted control microarray experiments; (2) isolated RNA from prostate cancer cells and conducted control microarray studies on two cell lines, each with different metastatic potential, and a control healthy prostate cell line; (3) optimized our enhancer co-culture assay in osteoblasts; (4) isolated high-molecular-weight human DNA using standard methods, and initiated DNA engineering to create transgenes for analysis in enhancer assays; and (5) performed co-cultures of cancer and osteoblast cells and carried out microarray experiments at three time points—preliminary results have identified over 1,000 genes that are differentially expressed as a function of cancer–bone co-culture, and some of these genes were further validated using cell proliferation, migration, and invasion assays.

Proposed Work for F12

In FY12 we will (1) analyze microarray expression data from prostate cancer cell–osteoblast co-culture experiments; (2) identify differentially expressed genes likely to contribute to prostate cancer metastasis to bone; (3) screen 1,000 or more clones for human enhancers activated in osteoblasts only when co-cultured with prostate cancer cells; (4) use microarray expression data to select 10 loci, then clone and test evolutionary conserved elements for enhancer functions in osteoblasts; and (5) validate 10 enhancer genes by sequencing, reverse-transcription polymerase chain reaction, and in situ hybridization.

Understanding the Role of Virus Evolution in Interspecies Transmission

Monica Borucki (10-LW-020)

Abstract

We propose to investigate the role of genetic mutation in the generation of bovine coronavirus variants that acquire the ability to adapt to human laboratory cell lines. This will allow us to assess the likelihood of the occurrence of a host-jumping event.

Data obtained from this work will be analyzed using advanced pattern recognition and other bioinformatics techniques to identify genomic markers applicable to future studies of adaptation of a zoonotic virus to human cells. This research will lay the foundation for virus evolutionary and forensic studies that further the development of a framework for understanding and ultimately predicting natural emerging infectious diseases and for differentiating these from man-made pathogens.

This project will advance our understanding of host–pathogen interactions, predictive biology, and forensic microbiology. The results of these experiments will provide the first systematic, controlled data set that can be used to develop models that predict the likelihood of pathogen emergence. In addition, these experiments will provide information that may be valuable for forensic identification of intentional pathogen introductions, because they will allow us to test the hypothesis that viral genomes undergo predictable changes when transferred in cell cultures, and these differences can be used to distinguish naturally occurring strains from strains propagated via passage in cell culture.

Mission Relevance

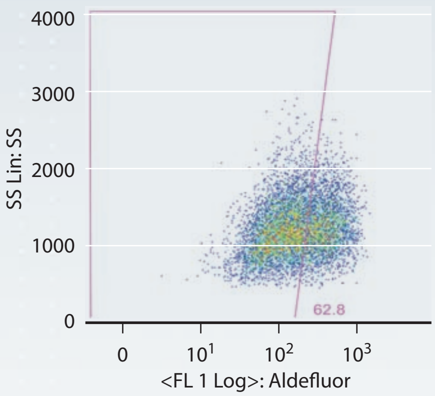
By developing a framework for understanding and ultimately predicting natural emerging infectious diseases and differentiating these from man-made pathogens, this research will lay the foundation for future studies of virus evolution and forensic work in support of Livermore's missions in national and homeland security.

FY11 Accomplishments and Results

In FY11 we determined that mutational dynamics of bovine virus evolution during passage in bovine and human cell lines appears to be driven by positive selection of a rare variant that is present in the natural samples and that may broaden host range. The rare variant selected for during passage has genetic characteristics that are used by other closely related viruses to broaden host range—phenotypic testing indicated that passage of the virus in human macrophage rapidly increased growth rate of the virus in this cell type. The virus did not replicate well in lung cells, which limited analysis. Computational analysis, however, of quasi-species data resulted in the identification of 42 markers of passage in cell culture and indicated that two genotypes circulate in natural and passaged samples with one genotype being rapidly selected for during passage. We could not recombine this virus with human coronavirus because of problems with both viruses consistently forming plaques in the same cell line. Overall, this study resulted in identification of a genetic insert in the viral genome that is present at low numbers within the viral population and is likely to impact host range. This suggests that deep sequencing analysis of viral populations may be necessary to fully understand evolutionary mechanisms and interspecies transmission events. We have obtained short-term support from the Defense Threat Reduction Agency to continue this work and are currently pursuing longer-term funding.

Establishing Cancer Stem Cell Longevity and Metastatic Potential

Bruce Buchholz (10-LW-033)



Aldefluor assay results for a bladder cancer cell line with resistance to the chemotherapy agent Cisplatin. A specific inhibitor of aldehyde dehydrogenase was used to set the gate for sorting the cells. Of the cells, 37.2% fall outside of this gate—that is, to the right of the pink line—and are therefore high in aldehyde dehydrogenase and exhibit stem-cell-like properties.

Abstract

We propose to demonstrate that cancer stem cells in human cancers are long-lived, resistant to conventional therapies, and are prime suspects in metastasis and relapse. Our goal is to advance cancer treatment and to initiate transformation in cancer research by targeting these stem cells. We intend to use the spike in carbon-14 that occurred as a result of atmospheric nuclear testing to date stem cells isolated from actual tumors and produce highly labeled cells as a tool for research. Techniques we developed for the DNA dating of healthy tissue using the carbon-14 spike will be used for cancer stem cells, and we will grow cancer cell lines on highly labeled DNA precursors to produce labeled cells. The stem cells will then be isolated from other cancer cells for use as indicators of metastatic potential.

We expect our DNA dating of cancer stem cells to clearly establish that they are long-lived cells that exist well before tumors occur. Producing highly labeled cancer stem cells in cell culture will provide a tool for measuring the metastatic potential of any cell line and will potentially lead to a clinical tool to assess biopsied tissue. We believe our results will provide clear evidence that cancer stem cells are cancer initiators and need to be the targets of all cancer therapies. Because most cancer deaths are associated with metastasis, changing the targets of therapy to cancer stem cells will revolutionize the treatment of cancer and potentially reduce cancer deaths.

Mission Relevance

This project supports the Laboratory's efforts in exploring the forefronts of science and technology, as well as bringing specialized expertise and Livermore's unique measurement capabilities to bear on problems of national and international interest. If successful, measuring the longevity of cancer stem cells and their metastatic potential will focus future cancer research to target these cells for eradication of metastatic cancer.

FY11 Accomplishments and Results

In FY11 we focused on identifying a marker for the proliferative and invasive behavior of cancer stem cells rather than growing labeled stem cells. Specifically, we (1) identified a cancer stem cell marker from bladder cancer cell lines as a potential marker in actual tumors, (2) completed initial measurements of the metastatic potential of these cancer stem cells, and (3) performed scoping studies, based on ratios of carbon-14 to carbon-12, for measuring natural DNA using the new moving-wire interface for the gas ion source at Livermore's Center for Accelerator Mass Spectrometry.

Proposed Work for FY12

In FY12 we propose to date DNA from 20 to 25 tumor samples using our established carbon-14 techniques to determine the longevity of cancer stem cells. In addition, we will use the bladder cancer stem cell marker we identified in FY11 to target specific cells and investigate our marker's potential for identifying the stem cells of other cancers.

Publications

Sarachine, M. J., and B. A. Buchholz, in press. "Bomb pulse biology." *Nucl. Instrum. Meth. B.* LLNL-JRNL-485203-DRAFT.

Sarachine, M. J., B. A. Buchholz, and R. W. de Vere White, 2011. *An investigation of cancer stem cells in three bladder cancer cell lines with differential sensitivity to Cisplatin.* Keystone Symposium: Cancer, Stem Cells and Metastasis, Keystone, CO, Mar. 6–10, 2011. LLNL-POST-471696.

Sarachine, M. J., B. A. Buchholz, and R. W. de Vere White, 2010. *An investigation of stem-like cancer cells in three bladder cancer cell lines.* 15th Ann. Cancer Research Symposium, Sacramento, CA, Oct. 28, 2010. LLNL-POST-461177.

Sarachine, M. J., et al., 2010. *Establishing cancer stem cell longevity in bladder cancer.* LLNL-POST-433311.

Radiation Biodosimetry Using Loop-Mediated Isothermal Amplification

Lawrence Dugan (10-LW-041)

Abstract

We propose to develop rapid radiation biodosimetry assays using loop-mediated isothermal amplification (LAMP)—a novel DNA amplification technology—to detect abnormal blood cells with chromosomal translocations induced by exposure to ionizing radiation and other agents. As proof of principle, we will use LAMP to amplify the messenger RNA (mRNA) transcripts resulting from the Philadelphia chromosome translocation breakpoint in chronic myelogenous leukemia cells. We will then focus on a genomic "hot spot" on a chromosome associated with radiotherapy-induced acute myelogenous leukemia. Identification of low-frequency precancerous cells provides

an opportunity for early therapeutic intervention and medical triage following an unanticipated radiological event.

We expect to develop LAMP assays to detect the primary mRNA transcripts resulting from Philadelphia chromosome translocation using chronic myelogenous leukemia cell lines, which could yield a patentable, clinically deployable laboratory-based assay for detecting and staging these leukemia patients. We will optimize LAMP assays to detect normal cells carrying radiation-induced mRNA transcripts and chromosome translocations in acute myelogenous leukemia cells to determine the assay's sensitivity, dynamic range, and radiation dose response for applications in medical triage of a radiological event as well as in radiotherapy clinics. We also propose to integrate the LAMP assay into hand-held point-of-care devices in development at LLNL for microbial pathogen detection.

Mission Relevance

Our LAMP assay supports the Laboratory mission to ensure national security by aiding medical efforts in the event of a radiological attack. Further development of LAMP-based DNA detection assays at Livermore will benefit current efforts to develop assays to detect other chemical and biological threats and may also provide valuable patient information for clinical oncologists to individualize therapy regimens and identify at-risk individuals earlier, when treatment may be far more effective for controlling disease progression.

FY11 Accomplishments and Results

We (1) designed LAMP primer signatures for detecting mRNA representing the wild-type breakpoint cluster region (bcr) and fusions of bcr and the Abelson gene (abl)—these fusions represent over 90% of all bcr–abl fusions in human chronic myelogenous leukemia; (2) tested three to six sets of primers for each target, consisting of a bcr exon paired with an abl exon; (3) synthesized a control plasmid containing a sequence covering the entire sequence of primers; (4) used the plasmid to generate control mRNA to validate the primer signatures, then down-selected the signatures to a manageable number for studies on cell-line-derived mRNA; and (5) developed protocols for studying in situ LAMP. In summary, we successfully developed LAMP signatures targeting chromosomal rearrangements found in human chronic myelogenous leukemia. The next step would be to validate the signatures in human cell lines and clinical samples, which would establish the signatures as a diagnostic tool for cancer research and treatment.

Publications

Dugan, L. C., et al., 2011. *Detection of BCR–ABL fusion mRNA using reverse transcriptase loop-mediated isothermal amplification*. LLNL-TR-520096.

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 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 the biological half-life of the target toxin and increase clearance and 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. For instance, this approach could also be used to treat 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.

FY11 Accomplishments and Results

The FY11 efforts were devoted to purifying NLPs and creating a functioning complex. The greatest challenge thus far has been to obtain a sufficient expression of the CYP450 into the NLP. Specific work included (1) incorporating the specific protein CYP450 responsible for metabolizing T-2 into NLPs, (2) co-expressing both the NLP-associated apolipoprotein and the target CYP450 protein with phospholipids as well as with added surfactants and detergents, and (3) optimizing the reaction for increased efficiency and yield to test functionality of the protein complex using a colorimetric assay. Once we have proven the CYP450–NLP complex is functional, we will test the ability of CYP450 to metabolize a surrogate compound in vitro.

Proposed Work for FY12

In FY12 we will (1) test the stability and metabolic functionality of the CYP450–NLP complex in vitro and assess the ability of the CYP4501A2 protein containing NLPs to metabolize caffeine (a surrogate) and T-2 toxin in a cell-free system, (2) administer the CYP450–NLP metabolizing enzymes to mice in an effort to enhance their capability to detoxify T-2 in vivo, and (3) determine the differences in the T-2 toxin's pharmacokinetic profiles and detoxification rates between mice treated with the CYP450–NLP complex and untreated controls. We will quantify these parameters by accelerator mass spectrometry.

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 cost 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.

FY11 Accomplishments and Results

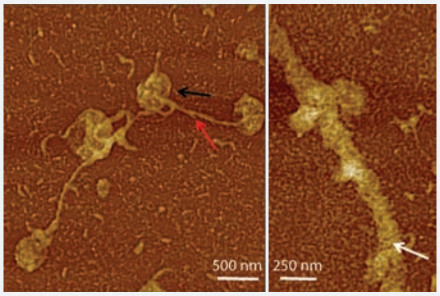
In FY11 we (1) conjugated three innate immune agonists (a chemical that binds to a cellular receptor to induce a biochemical response) into a nanometer-scale lipoprotein particle (NLP) platform—a fourth agonist, LL-37, was incompatible with the NLP platform and could not be conjugated; (2) assessed cellular activation in response to these NLP–agonist constructs in vitro, using a combination of multiplex cytokine arrays, enzyme-linked immune-sorbent assays, Greiss assays, and NanoString transcipt analyses; (3) assessed both cellular and subcellular localization of NLP constructs in vitro; (4) quantified the ability of NLP constructs to prevent the entry and survival of fully virulent *Francisella tularensis* (the bacteria that causes tularemia) in mouse macrophages; (5) obtained a semi-quantitative biodistribution profile of the NLP constructs in vivo after intraperitoneal injection; and (6) demonstrated that NLP–agonist constructs exhibit significantly more immuno-stimulation responses compared with free agonists in vivo.

Proposed Work for FY12

In FY12 we will (1) continue characterizing the in vivo distribution and pharmacokinetic profile of the NLP constructs after inoculation by intraneoplastic, intraperitoneal, intravenous, and subcutaneous injection; (2) assess inflammation in vivo using nuclear factor kappa B transgenic mice; and (3) use accelerator mass spectrometry to obtain quantitative and robust measurements of NLP half-life, biodistribution, and immunogenicity in vivo.

Unraveling of Assembly and Structure–Function Relationships of Poxviruses

Alexander Malkin (11-ERD-027)



Atomic force microscopy images of the unraveling of vaccinia virus architecture and assembly. A combination of chemical and enzymatic treatments of intact vaccinia virion results in the extrusion of a tubular nucleoprotein complex (red arrow) attached to the sacculus-type remnants of the viral core wall (black arrow), shown on the left. A high-resolution structure of the nucleoprotein complex with DNA strands clearly seen (white arrow) is shown on the right.

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 architecture and assembly pathways for the vaccinia virus, a cowpox virus that is used to vaccinate against smallpox. We will unravel roles played by individual viral genes in virion assembly and host–pathogen interactions and determine forensic signatures using a multidisciplinary approach uniquely combining nanometer-scale physico-chemical techniques and genetic and biochemical analysis. This improved knowledge will be pivotal for elucidating mechanisms of pathogenesis and development of countermeasures against viral agents.

We will identify and map target protein locations in vaccinia virion, and establish the relationships and links between viral assembly, chemistry, proteomic structure, and replication cycle, and 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. We will link viral molecular-scale structural and elemental attributes to production conditions. Our research will provide important insights into the viral replication cycle and physico-chemical properties, and will serve as an enabling platform to identify protein targets for the 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 structural and elemental attributes for pathogen forensics and attribution contributes to more efficacious preventive and therapeutic measures for emerging diseases and biodefense-related agents.

FY11 Accomplishments and Results

In FY11 we (1) developed experimental approaches for atomic force microscopy and nanometer-scale secondary-ion mass spectrometry (nanoSIMS) characterization of single viruses and subviral structures; (2) developed chemical and enzymatic methods for selectively degrading the vaccinia virion; (3) began experimental analysis of wild-type intact virions and selected subviral structures; (4) developed novel isotopic labeling procedures for localizing lipids in viral samples and for performing elemental analysis of viral and subviral structures, and conducted the corresponding nanoSIMS analysis; and (5) recruited a graduate student from Stanford University to work on the project.

Proposed Work for FY12

In FY 12 we will (1) complete atomic force microscopy and initiate nanoSIMS characterization of subviral structures of the wild-type vaccinia virus, (2) begin the same experimental characterization of the structure of the virion and subviral structures of selected mutants representing phenotypic classes with distinctive defective replication properties, and (3) finish characterizing the formulation attributes of a vaccinia virus manufactured using different processing procedures.

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, a specific need exists for robust antibodies for multiple research applications as well as therapeutic use. This is especially true for pathogen detection, biodosimetry, and cancer detection. 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

This 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.

FY11 Accomplishments and Results

We (1) developed and validated fluorescence correlation spectroscopy instrumentation for detecting the topology and diameters of protein complexes, including an excitation setup combining three laser beams—with the frequencies 487, 561, and 635 nm, an improvement from our previous two-color detection capability—into a single-mode fiber capable of delivering the light to the detection setup, which will allow us to characterize antibodies and study protein–protein interactions in FY12; (2) engineered and expressed recombinant proteins for 1 bacterial muramidase, 3 secretion III proteins, and 2 human apolipoproteins, achieving better than 95% homogeneity after affinity purification; (3) collaborated with the University of Illinois at Chicago to generate antibodies; and (4) identified and characterized the antibodies produced, including 200 positive single-chain antibodies and antibodies unique to the secretion III protein LcrV, and performed downstream characterization to select 10 antibodies with the necessary affinity and sensitivity for use in FY12.

Proposed Work for FY12

We will further develop recombinant antibody interaction assays, using fluorescent correlation spectroscopy to characterize complexes that form interactions between host or pathogen membranes, and use cell-free expression to engineer antibodies in a high-throughput format. Specifically, we will (1) characterize protein interactions for complexes generated in FY11, (2) engineer and analyze recombinant antibodies, and (3) perform single-molecule characterization of interactions using labeled affinity reagents and biophysical methods such as fluorescent correlation spectroscopy.

Publications

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-459318l.

Targeted Drug Delivery for Treating Traumatic Bone Injury

Nicole Collette (11-ERD-060)

Abstract

A significant category of 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 cases. Most existing bone-injury interventions are mechanical, designed to stabilize the break and facilitate natural repair. However, no existing treatment can heal broken bones and build bone de novo. To increase bone formation and reduce healing time, we will develop an approach that couples strontium and anti-sclerostin antibodies to nanolipid particles (NLPs) for delivery to bone-forming cells at the site of injury. In vitro and in vivo studies of effectiveness will be analyzed by histology, mechanical testing, and LLNL's world-leading accelerator mass spectrometry capabilities.

We will deliver a fully functional, tissue-specific drug-delivery system tested in vivo with synergistic bone-healing therapy that is ready for clinical settings. This therapy system will provide much-needed effective, nonsurgical treatments for traumatic bone injuries. In addition to defense applications, the resulting bone therapy system will also have applications in biodefense and public medicine in general.

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 engineering and analytical tools, for future research in the areas of infection treatment and prevention.

FY11 Accomplishments and Results

In FY11 we created, characterized, and optimized strontium NLPs in vitro. Specifically, we (1) demonstrated and quantified binding of strontium to NLPs and varied their presence on these particles in controllable amounts; (2) identified a dose-dependent response to the loading of strontium and other divalent cations on the particle (particle cation concentration) that demonstrates the ability of the particles to bind to the cell surface, be taken up, and promote cell proliferation and mineralization of cells in vitro; and (3) prepared and received approvals for upcoming in vivo experiments and acquired necessary surgical proficiency for the experiments.

Proposed Work for FY12

To follow up our FY11 in vitro work, we will (1) create, characterize, and optimize the strontium and anti-sclerostin antibodies coupled to nanolipid particles in vivo; (2) establish a surgical technique to deliver reproducible and humane fractures in mice; (3) induce fractures and dose animals with optimized NLPs; and (4) use samples from our in vivo studies to examine the efficacy of the therapeutic particles by histology, accelerator mass spectrometry, and molecular biology.

Principal investigator Mayali with the "rolling bottle" laboratory setup where isotope-labeled marine particles are incubated with bacteria under different climate conditions.

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.

We will produce 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.

FY11 Accomplishments and Results

In FY11 we (1) performed initial mesocosm experiments and grew stable-isotope-labeled marine particles; (2) collected samples for chip stable-isotope probing, isotope-ratio mass spectrometry, and 16S RNA sequencing; (3) optimized extraction protocols for proteins, lipids, and intracellular and extracellular polysaccharides; (4) identified stable-isotope-ratio mass spectrometry detection as a cheaper alternative for laboratory studies, compared to radioisotope detection by accelerator mass spectrometry; (5) analyzed 16S sequences from degradation experiments and designed probes for these sequences and printed a RNA microarray to target these organisms; (6) performed chip stable-isotope probing analyses from samples collected during degradation experiments; (7) obtained isotope-ratio mass spectrometry data from collected particles over time; and (8) planned subsequent experiments with climate effects on particle degradation.

Proposed Work for FY12

In FY12 we will (1) continue our mesocosm experiments, which should produce the first results by the end of FY12; (2) begin RNA sequencing using the technique developed in FY11; (3) continue probe design; and (4) begin analysis using both nanometer-scale secondary-ion mass spectrometry and accelerator mass spectrometry.

Publications

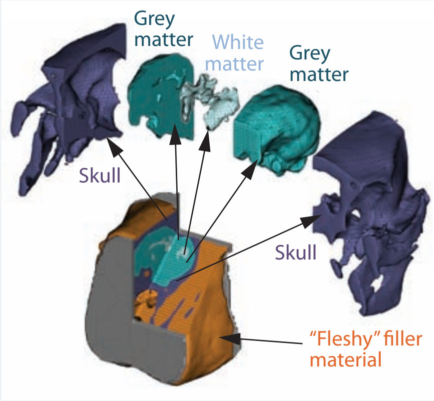
Mayali, X., et al., 2011. "High-throughput isotopic analysis of RNA microarrays to quantify microbial resource use." *ISME J.*, Dec. 8, 2011. LLNL-JRNL-470805.

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.

Computational Studies of Blast-Induced Traumatic Brain Injury Using High-Fidelity Models

Michael King (11-LW-009)



A finite-element model of a pig brain generated with computed tomography images.

Abstract

Our objective is to explore the mechanisms by which explosive blasts cause traumatic brain injury. Unlike civilian injuries, which are usually caused by impacts and have been extensively studied, military traumatic brain injuries are often caused by explosive blasts and are poorly understood. Determining the mechanisms by which blasts damage the brain would enable better protective armor, diagnosis, and treatment. We will develop high-fidelity models of the head from x-ray computed tomography data, and then leverage Livermore's computational capabilities and blast expertise to conduct detailed simulations of impacts and blasts on our head model. We will then work with medical researchers to correlate the simulation predictions with actual trauma data to identify the pathways by which blasts cause damaging loads in the brain.

We expect to identify mechanisms by which explosive blasts damage the brain. This could allow development of more effective protective equipment for reducing traumatic brain injury and saving lives. Understanding damage mechanisms also has implications for rapid diagnosis in the field and could lead to better treatment strategies. Even ruling out potential damage mechanisms would be a significant contribution to the research community seeking to understand this phenomenon.

Success in this project will provide LLNL with a great deal of visibility through our planned publications and conference attendance and provide us with a sophisticated set of tools that will enable further research.

Mission Relevance

The prevalence of traumatic brain injury among our combat forces, accounting for more than 30% of all combat injuries, is both a challenge to our national security and a significant drain on our economy because of the rising cost of veterans care. By marrying LLNL's expertise in blast–structure interactions, multi-scale physics, and high-fidelity simulation with our extensive computational capabilities, we have a unique opportunity to address this challenge in support of the Laboratory's mission to reduce or counter threats to national security.

FY11 Accomplishments and Results

In FY11 we worked to develop an understanding of what kind of simulation-predicted mechanical loads correlate to observed pathologies in the brain. Specifically, we (1) created mesh models of both pig and human heads from x-ray computed tomography data, (2) performed a literature review of biomaterial properties and populated our models with these properties, (3) conducted simulations of animal experiments performed by our collaborators at the University of Pennsylvania to assess how well our models predict brain damage, (4) continued the process of refining these models with more data from our collaborators and examining correlations with increasingly complex damage mechanisms, and (5) developed a software application that generates biological meshes from computed tomography data quickly and easily, including analysis setup and code verification. Our preliminary correlations to actual data imply promising paths towards understanding damage metrics.

Proposed Work for FY12

By the end of FY12 we hope to understand the mechanisms by which blasts can cause damage in a human brain and to correlate simulation predictions to human trauma data. Specifically, we will (1) continue simulations of pig brain injuries with our collaborators, (2) correlate fractional anisotropy data to simulation damage metrics, (3) improve biological material models, (4) perform multi-scale modeling of biological structures as needed, and (5) apply what we have learned from studying pig injuries to create blast simulations of the human head to better understand how blasts cause traumatic brain injury.

In Vivo Modulation of MicroRNA for Cancer Therapy

Nicholas Fischer (11-LW-015)

Abstract

Our goal is to develop cancer therapies based on the targeted modulation of microRNA in vivo. In living systems, microRNA is an important regulatory molecule, and aberrant regulation of cellular microRNA has been linked to cancer. In vitro modulation of microRNA in cancer cells has demonstrated their potential as therapeutic targets. However, targeted in vivo delivery of microRNA modulators is currently a major obstacle to using this modality. We will develop nanometer-scale lipoprotein (nanolipoprotein) particles as universal, in vivo delivery platforms incorporating both microRNA modulators and tumor-targeting moieties. The delivery and efficacy of the microRNA modulators will be assessed using in vitro and in vivo tumor models.

If successful, we will demonstrate the therapeutic efficacy of in vivo delivery of microRNA modulators to specific tissues using nanolipoprotein platforms. The implications for success are twofold—these findings can provide the foundation for a novel targeted cancer therapy and demonstrate the utility of functional nanolipoproteins for a range of in vivo applications. Currently, the promise of microRNA modulation as a possible therapy is precluded by the difficulty of its in vivo delivery. Consequently, the delivery of microRNA modulators using cancer-targeting nanolipoproteins is expected to alleviate this bottleneck, effectively accelerating the clinical application of this therapeutic technology.

Mission Relevance

This approach can have significant implications not only for cancer treatment but also national security—specifically, the Laboratory's mission thrust area of biosecurity, by providing a potential therapeutic modality to address disease and illness caused by natural or engineered pathogens.

FY11 Accomplishments and Results

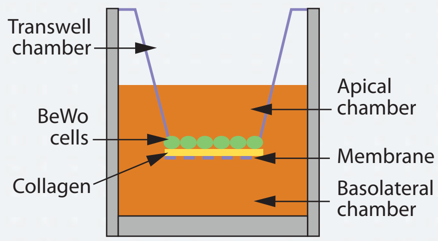
In FY11 we (1) demonstrated incorporation of cancer-targeting moieties into nanolipoprotein particles and developed strategies for conjugating microRNA modulators to the nanoparticles; (2) started to develop novel attachment methodologies for incorporating targeting moieties and microRNA modulators in the nanoparticles, as well as techniques to quantify individual components upon incorporation into the nanolipoprotein particles; (3) established five cancer cell lines and began initial in vitro assessments, including targeting, internalization, stability, and function of modulators; and (4) optimized our protocols and established advanced experimental procedures for in vivo applications.

Proposed Work for FY12

In FY12 we will finalize our in vitro assessment of the targeting, internalization, stability, and function of the microRNA modulator nanolipoproteins and demonstrate the ability of targeted nanolipoproteins to deliver microRNA modulators to tumors in mice, elicit tumor cell death, and reduce tumor burden.

Prenatal Exposure to Endocrine-Disrupting Compounds Found in the Water Supply

Miranda Sarachine (11-LW-018)



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.

Abstract

The growth of the U.S. population is placing an increasing demand on fresh water supplies. However, water quality is being threatened by endocrine-disrupting compounds that are sometimes found at extremely low levels in the water supply. 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. This compound has received much attention lately.

We expect to see the transfer of TCC labeled with carbon-14 through a polarized cell line in assays, 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 also expect to investigate the kinetics of TCC placental transfer and explore the developmental effects of TCC exposure using a mouse model. The method we develop will lay the groundwork for further studies of other endocrine-disrupting compounds that have also been reported in water supplies.

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.

FY11 Accomplishments and Results

In FY11 we (1) obtained viable BeWo b30 placental cells, (2) developed and optimized culturing of these cells and established a frozen stock, (3) developed and optimized a Transwell assay of the cells and validated the technique by measuring the trans-epithelial electrical resistant and transport of fluorescein isothiocyanate and fluorescein isothiocyanate dextrans, and (4) performed treatment of the placental cells in Transwell culture with environmentally relevant levels of carbon-14 labeled TCC. Our initial measurements with atomic mass spectrometry show that approximately 2% of the carbon-14 TCC dose is transferred across the cell barrier.

Proposed Work for FY12

In FY12 we will (1) complete screening of TCC labeled with carbon-14 in the BeWo b30 Transwell assay and complete measurements of concentrations with atomic mass spectroscopy to determine TCC transport across the placenta at environmentally relevant concentrations, and present the results at a professional society meeting and in a peer-reviewed paper; (2) develop a mouse model for TCC drinking water exposure and placental transfer; (3) quantify levels of environmentally relevant concentrations of TCC transferred to the whole mouse fetus and specific tissues using atomic mass spectroscopy and determine the effects of fetal exposure on gestational age, birth weight, and sex organ development; and (4) present the results of exposure to TCC at a professional conference and in a peer-reviewed journal article.

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. Biological hydrogen is one of the most promising of these fuels and is seen as a future energy carrier. 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 of 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 ever, the ability to produce hydrogen using inorganic substrates coupled with CO_2 fixation by photosynthesis. This achievement 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 gather a multidisciplinary group of scientists at LLNL to collaborate in achieving a basic understanding of critical microbial metabolisms 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.

FY11 Accomplishments and Results

In FY11 we (1) developed a purple nonsulfur bacterium, the TIE-1 strain of *Rhodopseudomonas palustris*, as a model system for hydrogen production; (2) created a flux balance model for *R. palustris* and used it to calculate maximum hydrogen production for different substrates and simulate the relationship between bacterial growth, substrate consumption, and hydrogen production; (3) experimentally achieved steady growth of *R. palustris* with thiosulfate as an inorganic electron donor, and compared the growth rate to that possible with organic substrates; (4) used gas chromatography to confirm hydrogen production in our system; and (5) tested the relationship between substrate concentration, bacterial growth, availability of fixed nitrogen source, and hydrogen production.

Proposed Work for FY12

In FY12 we will genetically transform our *R. palustris* strain to overcome the energy barrier and redirect electron-transport pathways to the biosynthesis and liberation of hydrogen. To this end, will use enzyme expression and metabolic modeling to identify the most promising enzyme combination and electron-transfer pathways needed to efficiently produce hydrogen. We will focus on hydrogenases and nitrogenases, with which the bacterium metabolizes hydrogen. We hypothesize that the regulation of these enzymes can influence the internal reduction and oxidation balance and therefore affect the cellular ability to use certain electron donors for hydrogen production. We will grow *R. palustris* cells on reduced inorganic substrates, such as sulfur or ferrous iron, as the electron donor.

Protist Power: Deconstructing Termite Conversion of Wood to Biofuels

Kevin Carpenter (11-LW-039)

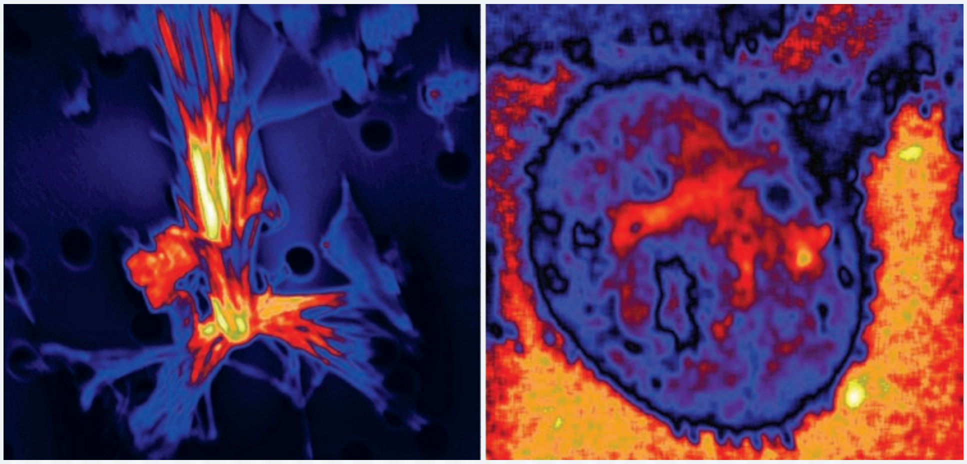
Abstract

We intend to investigate the metabolic pathways and symbiotic interactions of microbes called protists in termite guts, which are responsible for highly efficient conversion of wood to simple carbohydrates and hydrogen gas—both of great interest in biofuels production. This builds on recent advances in microbial ecology using isotopes and advanced mass spectrometry here at LLNL, as well as discoveries about these organisms by our team. We will identify key metabolic pathways and organisms so that their capabilities may be applied to biofuels production. We will accomplish this by introducing isotopically labeled substrates into termite air and food supply and monitoring the flux of metabolites through various microbial species with nanoscale secondary ion mass spectrometry.

We expect our results will form the basis for greatly improving efficiency and reducing cost in converting plant biomass to biofuels, and will reduce dependence on foreign oil, lessen climate impacts, and potentially create new jobs. The potential application of termite microbes to biofuels production is currently a topic of great interest, and we expect results to be disseminated through high-profile publications and presentations at scientific meetings.

Mission Relevance

The Energy Independence and Security Act of 2007 mandates a great increase in U.S. biofuels production capability. The goal of our research is to provide a scientific



Nanometer-scale secondary-ion mass spectroscopy images showing the carbon-13 isotopic enrichment of *Hoplonympha natator* (left) and *Oxymonas dimorpha* (right), two protists that are symbiotic in the hindgut of the termite *Paraneotermes simplicicornis*.

basis for greatly enhancing U.S. biofuels production capability. This research directly addresses two Laboratory missions of enhancing the nation's energy and environmental security and strengthening the nation's economic competitiveness. Furthermore, the techniques we develop will be directly applicable to the study of protist disease agents such as malaria, which supports Livermore's mission to reduce or counter threats to national and global security.

FY11 Accomplishments and Results

In FY11 we completed the first successful in situ, stable isotopic labeling experiments on live termites with subsequent analysis of the hindgut microbial community by nanometer-scale secondary ion mass spectrometry. We (1) found that hindgut microbes exhibit an order-of-magnitude enrichment in carbon-13 (^{13}C) after 6 weeks; (2) conducted a 2-day ^{13}C enrichment study that showed heterogeneous enrichment, suggesting the existence of microbes that degrade lignocellulose and others that are downstream consumers of its breakdown products—we infer that a protist species of unknown function may play a central role in nitrogen metabolism; (3) conducted an analysis of ^{13}C data supporting the genomic hypotheses of protist lignocellulose degradation; (4) determined that symbiotic *Bacteroidales* bacteria living on the surface of *Hoplonympha* protists consistently have higher enrichments of ^{13}C than the host protist—as inferred from analysis of protist flagella—and higher than that of nearly all other microbes surveyed, suggesting an important role in carbon metabolism; and (5) began to sequence the genome of these bacteria with the help of collaborators.

Proposed Work for FY12

In FY12 we will track the flux of energy and nutrients in the termite hindgut microbial system through protists and their bacterial symbionts using isotopically labeled substrates such as ^{13}C cellulose. Specifically, we will (1) complete experiments with different ^{13}C-labeled feeding substrates—cellulose, hemicellulose, and lignocellulose—on at least one termite species to assess niche partitioning among gut microbes with respect to these three substrates; (2) complete a comparative functional survey with ^{13}C and nitrogen-15 labeling on additional termite species to assess the relationship between microbial function and morphology (i.e., phylogenetic affiliation) in the termite gut; and (3) use these results to design novel microbial consortia for the efficient, cost-effective conversion of woody biomass into biofuels.

Publications

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Solar-Powered Microbial Electrolysis Cells for Hydrogen Production

Fang Qian (11-LW-054)

Abstract

Our objective with this project is to develop a self-biased, solar-driven microbial electrolysis cell that uses solar energy to electro-hydrogenate organic waste into hydrogen fuel. Compared to existing fermentation approaches based completely on microbial catalysis, our unique microbial electrolysis cell design will, for the first time, couple a semiconductor photocathode with an electrogenic bacteria-colonizing anode, which functions synergistically for enhanced photon–electron conversion, accelerated waste removal, and increased hydrogen yield. The solar microbial electrolysis cell approach will provide new insights into the fundamentals of microbe–inorganic interfaces and the basis for diverse, self-sustained, and cost-effective energy recovery systems.

We will provide fundamental insights into electrical coupling between electrogenic bacteria and inorganic materials and evaluate a transformative concept for using sunlight to convert organic wastes into renewable fuels. Based on our proof-of-concept experiments, we will provide the first direct evidence for hydrogen production based on complementary functions from a combined microbial–inorganic system. We will characterize how semiconductor material properties, such as band gap, can influence interactions with microbes. The solar microbial electrolysis cell system should readily scale up to generating hydrogen fuel by treating industrial wastewater from food, animal, fermentation, and municipal facilities, and also enable small, self-sustaining power generators.

Mission Relevance

Efficient and economic hydrogen production is a considerable challenge on the road to new, environmentally sound energy systems. By developing a new concept for extracting hydrogen from water without significant power input, this project—which will be synergistic with DOE projects at LLNL in biological hydrogen production—will help provide clean, useful energy. This goal directly supports the energy security mission of reducing dependence on foreign petroleum and energy sources that damage the environment. Determining how microbes and semiconductors can be used for electrolysis also supports the mission of strengthening the nation's economic competitiveness.

FY11 Accomplishments and Results

In FY11 we achieved several key results towards realization of a solar-driven microbial electricity generation system. We successfully prepared a cuprous photocathode with a photocurrent density of 2.1 mA/cm^2 at –0.4 V bias. We also created a small-scale bacteria-colonizing anode that generated a high power density of 62.5 W/m^3 in a microfluidic microbial fuel-cell device. Our work included (1) design and construction of reconfigurable microfluidic platforms for microbial fuel-cell studies, (2) development of microbial fuel-cell assays to test the reduction capability of the *Shewanella* strain ANA-3 and several heme cytochrome mutants, (3) chemical synthesis of cuprous oxide nanowire arrays as the photocathode component of the microbial electrolysis cell, and (4) demonstration of the ability of our prototype solar-driven microbial electrolysis cell to generate current using only light and biodegradable organic matter in a self-sustained manner for several days.

Proposed Work for FY12

In FY12 we will (1) continue developing high-performance photo-electrodes to increase the chemical stability of current cuprous oxide electrodes and use other semiconductor materials, such as ferric oxide and titanium dioxide, as nanometer-scale electrodes within the solar microbial electrolysis cell configuration; (2) test the proposed synergistic effect of current generation in the hybrid microbial–semiconductor system; (3) load platinum nanometer-scale particles on the photo-electrode to convert the generated current on site into hydrogen; (4) isolate electrogenic strains from the environment to enable the solar-driven microbial electrolysis cell to use waste water instead of pure chemicals; and (5) use other catalysts and precursors to explore the potential of solar-driven microbial electrolysis cells for generating other high-value chemicals or fuels besides hydrogen.

Publications

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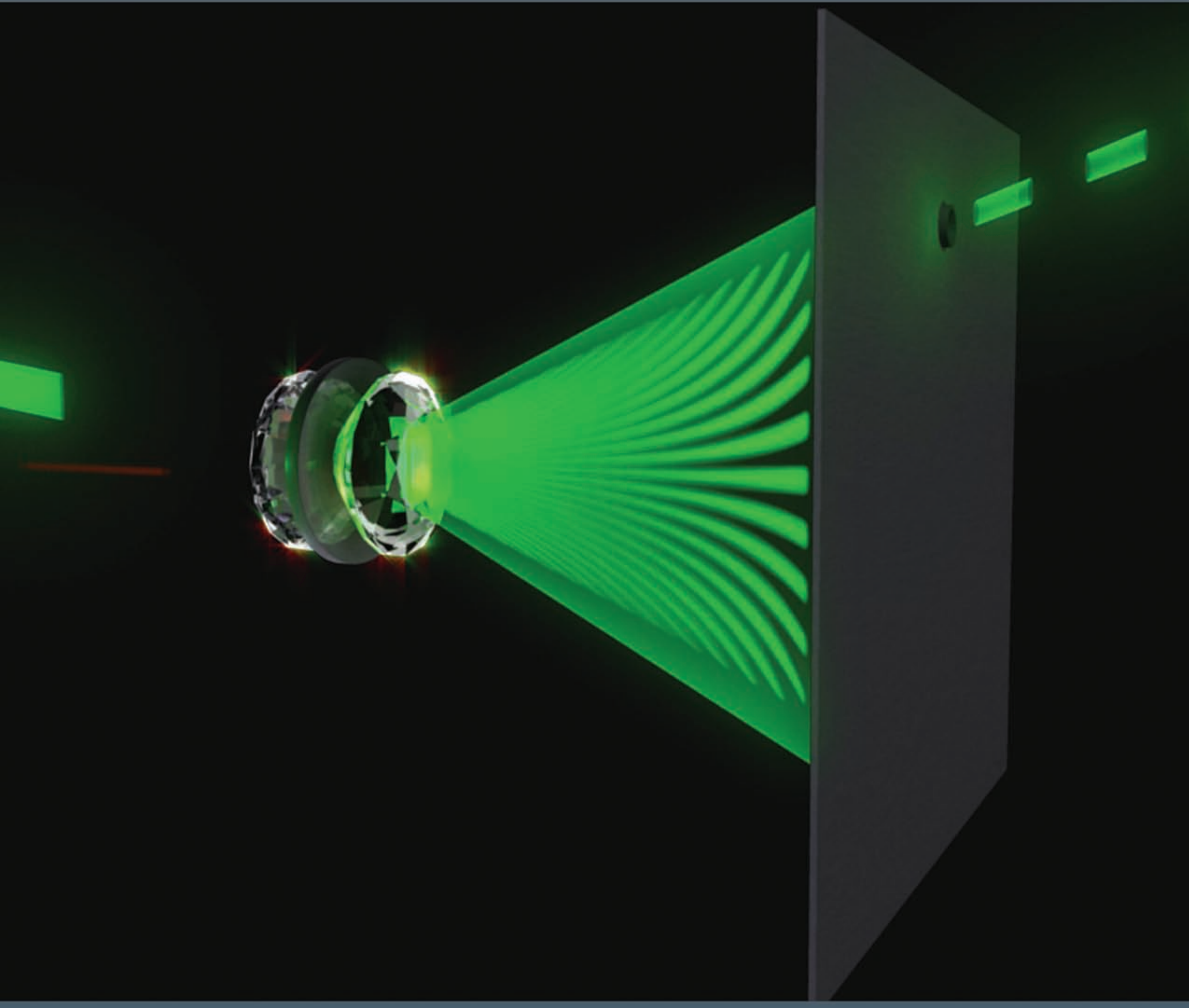
Chemistry



Laboratory Directed Research and Development

FY2011

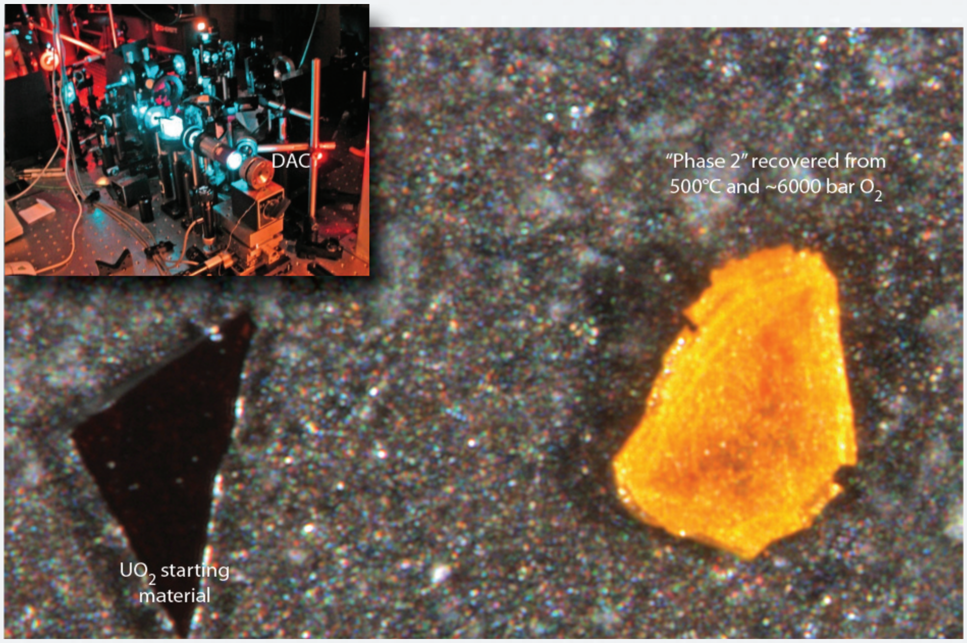
Nuclear Forensics: An Integrated Approach for Rapid Response

Ian Hutcheon (10-SI-016)

Abstract

We propose to develop a robust and responsive nuclear forensics capability to reconstruct a nuclear incident quickly and with high fidelity, even in a crisis situation. We will leverage unique LLNL capabilities in mass spectrometry, actinide and forensic science, laser spectroscopy, weapon physics, and material synthesis to develop scientific and technological breakthroughs critical to nuclear threat elimination. Although focusing initially on the challenges of post-detonation forensics, the expected advances will also be of great utility for assessing pre-detonation events and signatures.

We expect to achieve breakthroughs in experimental science to increase the speed and fidelity of nuclear forensic analysis by meeting the following challenges: (1) development of resonance ionization mass spectrometry to provide isotope-specific, definitive information on fuel composition in post-detonation scenarios within hours, without chemical separation; (2) development of a cavity ion source to increase mass spectrometry sensitivity by a factor of 10 to 25; (3) development of accelerator mass spectrometry to measure activation products at ultralow concentrations; (4) application of in situ Raman and infrared spectrometry to evaluate actinide chemical behavior at high temperature; (5) synthesizing uranium oxides by precipitation, hot pressing, and sintering to interpret material signatures; and (6) characterization of nanometer-scale particles in fallout to provide the "ground truth" to evaluate new experimental techniques.



Optical image showing the dramatic change in color after UO_2 has been heated in pure oxygen above 450°C under pressure. "Phase 2" is almost certainly a form of UO_3, although there is not a definitive match with any of the known polymorphs—phase 2 is most similar to the gamma phase. The inset shows the Raman system with the diamond cell used to generate the sample.

Mission Relevance

This project supports LLNL's national security mission by developing nuclear forensics and attribution capabilities for reducing the threat of the proliferation and use of weapons of mass destruction. This project will also help cultivate the next generation of scientific leaders necessary to ensure the nation's ability to safeguard nuclear weapons and respond to nuclear threats.

FY11 Accomplishments and Results

In FY11 we (1) continued development and optimization of methods for rapid analysis of actinide isotope ratios in nuclear materials using resonance ionization mass spectrometry, and successfully analyzed uranium and plutonium at low (micrograms per gram) concentrations in fallout glass from U.S. weapons tests; (2) completed the design of a cavity ion source for a next-generation actinide mass spectrometer; (3) synthesized a suite of uranium oxide compounds using commonly utilized precipitation processes, analyzed the materials for morphology and structure, and compared them to a set of real-world samples with known process histories—results strongly suggest morphology provides a new forensic signature indicative of material processing history, despite otherwise identical chemical properties; (4) examined oxidation of uranium oxide in pure oxygen at pressures up to 0.9 GPa and temperatures of 450°C and used in situ Raman scattering, scanning electron microscopy, transmission electron microscopy, and x-ray photoelectron spectroscopy to demonstrate that the synthesized uranium oxide differs significantly from common U_3O_8, having far fewer crystalline defects and exhibiting a hexagonal rather than orthorhombic form; (5) successfully demonstrated the feasibility of measuring activation products using accelerator mass spectrometry by analyzing samples from the Oak Phoenix nuclear forensics exercise, which contained trace levels of aluminum-26 resulting from activation of the aluminum foil during irradiation—the measured aluminum-26/aluminum-27 ratios agree well with expected values; and (6) analyzed melt glass in fallout debris from a uranium-fueled, low-yield nuclear test and demonstrated highly enriched levels of a number of radioactive isotopes—uranium isotopes were measured without the need for extensive chemical separation, and stable isotope signatures provided insight into construction and sophistication of the test device.

Proposed Work for FY12

We will (1) use Raman and infrared spectroscopy to examine uranium-bearing systems at high temperatures and varying oxygen fugacity to obtain structural and vibrational data, phase boundaries, and thermal-transport properties relevant to uranium volatility; (2) apply resonance ionization mass spectrometry to achieve rapid and accurate analysis of uranium and plutonium isotopes in debris samples; (3) develop a cavity ion source, based on the design developed in FY11, to increase the sensitivity of mass spectrometry of uranium and plutonium by a factor of ten; (4) use the results of nuclear test debris analysis to develop quantitative models of fallout formation and transport; (5) develop thermal-gradient chromatography to speed up chemical purification and analysis of nuclear debris; and (6) synthesize uranium oxide compounds using different chemical and physical treatments to understand forensic signatures.

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Microbes and Minerals: Imaging Carbon Stabilization

Jennifer Pett-Ridge (10-ERD-021)

Abstract

Understanding the mechanisms and persistence of interactions between organic molecules and minerals is crucial to the maintenance of sustainable organic matter levels in soils, especially in systems stressed by climate change and intensive agriculture. We propose to study the interactions between organic molecules, soil minerals, and microbes by marrying high-resolution spectroscopy and imaging mass spectrometry, which will allow us to map organic carbon distribution and image associations of organic material with specific minerals in soil. We will develop new preparation techniques for complex organic and mineral samples, new ways to navigate and relocate analyses, and mechanistic explanations for the carbon saturation behavior of soil fractions crucial to long-term soil carbon stabilization.

We propose to develop advances in sample preparation and imaging analysis by combining scanning transmission x-ray microscopy and nanoscale secondary-ion mass spectrometry to address the stability of relationships between organic molecules and minerals. Specifically, we intend to determine specific preferential sorption relationships, whether sorption processes amplify over time, and the effects of microbes. Our technology will have great potential for applications in industrial chemistry, geochemistry, and cosmochemistry, as well as for terrestrial carbon sequestration.

Mission Relevance

Our approach will allow us to characterize highly complex associations of biological and mineral materials and contribute to an improved understanding of carbon stabilization in soils, which supports the Laboratory's mission of enhancing the nation's environmental security. With our unique combination of high-sensitivity and high-resolution imaging for coupled analyses of organic, mineral, and isotopic distributions, this research advances LLNL measurement science and technology efforts for a strong foundational science and engineering base. Our technology is also relevant to a primary mission area in the DOE's Genomic Science Program.

FY11 Accomplishments and Results

In FY11 we (1) performed several multifactorial soil incubation experiments with "synthetic soil" (mineral mixtures) and isotopically labeled organics (chitin, lignocellulose, and plant cells) both with and without microbial catalysts, finding that a nitrogen-15-labeled fungal cell-wall amide material is preferentially associated with soil iron oxide and hydroxide minerals, and that the microbial transformation and partitioning of amide nitrogen can occur extremely rapidly within weeks; (2) performed analysis by both bulk and micro-characterization techniques—we used scanning transmission x-ray microscopy to elucidate spatial association of the key elements of interest including carbon, nitrogen, oxygen, and transition metals within micro-aggregates, and used nanoscale secondary-ion mass spectrometry to produce high-resolution correlated maps of the distribution of isotopically labeled organic matter relative to microscopy mapping; and (3) prepared a manuscript describing the nature and stability of sorption relationships and the effects of microbial catalysts on mineral–organic association patterns.

Proposed Work for FY12

In FY12 we will continue to improve sample preparation using thin sectioning and cryo-sectioning techniques. We will focus on the role manganese oxides play in decomposition and stabilization of phenolic compounds (lignocellulosic materials). Specific work includes performing multifactorial incubations involving carbon-13-labeled Zinnia elegans plant cells, diffusible manganese(III)–ligand complexes, pure and native soil minerals, and fungal catalysts. We predict that litter-decomposing fungi create diffusible manganese(III)–ligand complexes that accelerate decomposition and have a significant effect on the biogeochemistry and stabilization of phenolic soil organic matter. We also expect to submit at least two papers for publication in the peer-reviewed literature describing our proof-of-concept micro-characterization techniques and the results of an early study mapping associations of amino sugars and iron oxides.

Publications

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

Dawn Shaughnessy (11-ERD-011)

Abstract

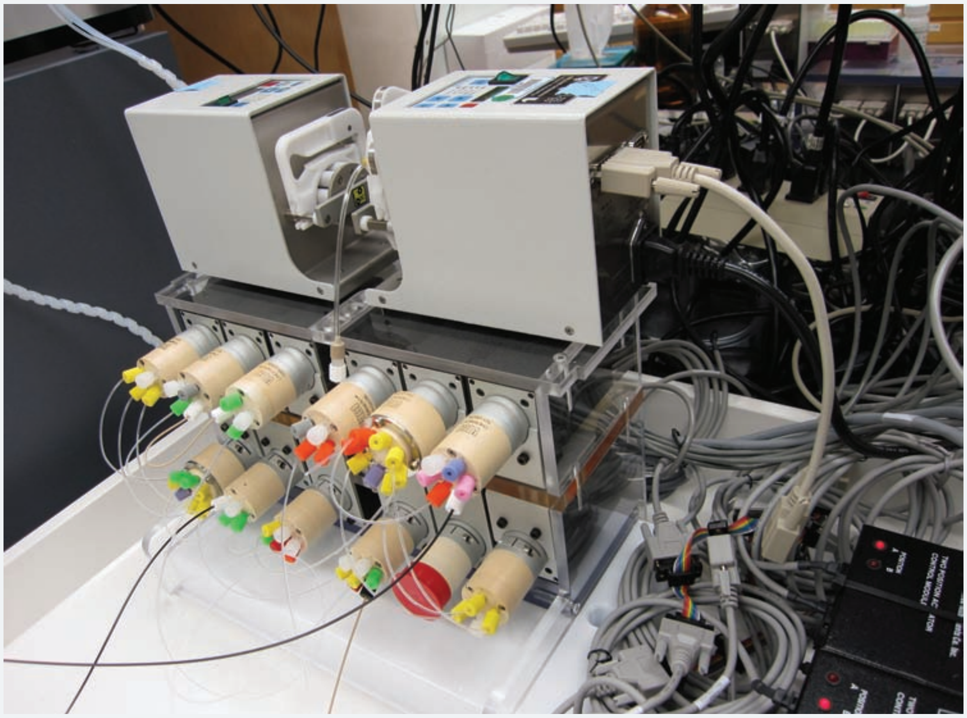
Our goal is to study fundamental chemical properties of the heaviest elements. In particular, we will explore reported modifications caused by relativistic effects of group behavior in the periodic table for certain transactinides (elements 104–106), and observe the unknown behavior of element 114 in aqueous media. We propose to develop two chemical separation methods for heavy elements with atomic numbers of 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 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 behavior down 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 elimination by addressing the challenge of developing autonomous, real-time, forensics methods designed for field deployment. This project will also serve as the foundation for future opportunities in a variety of other areas, such as medical isotope production, chemical purification, inertial-confinement fusion diagnostics, and isotope



The superheavy element liquid automation system designed to isolate heavy elements for determining their chemical properties. The system can be adapted for a variety of chemical systems and can perform a separation in approximately one minute.

harvesting. The study of heavy elements is a key component of fundamental research in nuclear chemistry and radiochemistry, which are core competencies at LLNL.

FY11 Accomplishments and Results

In FY11 we (1) developed chemical separation methods with extraction chromatographic materials for periodic table groups 4 and 5; (2) began our research on the use of crown ether extractants for groups 13 to 15, starting with germanium tracers on Eichrom lead resin; (3) designed an interface for the accelerator mass spectrometry beam line so that short-lived isotopes can be produced without a carrier and transported to the automated chemistry system; (4) began the engineering analysis of the interface design, created a complete list of reactions relevant to heavy-element work at CAMS, and began planning an initial set of tests at CAMS; and (5) modified our existing automated chemistry apparatus to be more flexible and more chemically resistant.

Proposed Work for FY12

In FY12 we propose to (1) develop model chemical systems for group 14 elements, (2) continue to modify an automated chemical system for chemistry at CAMS and also for future chemistry performed behind mass separators at accelerator facilities, (3) design and fabricate a CAMS–chemistry interface chamber, (4) perform transport efficiency tests using our CAMS target and interface chambers, and (5) begin experiments searching for new elements 119 and 120 in collaboration with the GSI Helmholtz Centre and the Flerov Laboratory of Nuclear Reactions.

Publications

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Oganessian, Y. T., et al., 2011. "Eleven new heaviest isotopes of elements Z = 105 to Z = 117 identified among the products of ^{249}Bk + ^{48}Ca reactions." *Phys. Rev. C* **83**, 054315. LLNL-JRNL-512855.

Oganessian, Y. T., et al., 2011. *Synthesis of a new element with atomic number Z = 117.* 241st American Chemical Society Natl. Mtg., Anaheim, CA, Mar. 27–31, 2011. LLNL-PRES-432557.

Oganessian, Y. T., et al., 2010. "Synthesis of a new element with atomic number Z = 117." *Phys. Rev. Lett.* **104**, 142502. LLNL-JRNL-462051.

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Shaughnessy, D. A., 2010. *Current status and future plans for aqueous chemistry of the heavy elements.* Future Directions of Heavy Element Science Workshop, Berkeley, CA, Dec. 10–11, 2010. LLNL-ABS-463763.

Shaughnessy, D. A., K. J. Moody, and R. A. Henderson, 2010. *Synthesis and observation of new chemical element number 117*. APS April Mtg., Washington, DC, Feb. 13–17, 2010. LLNL-ABS-462040.

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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 and 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.

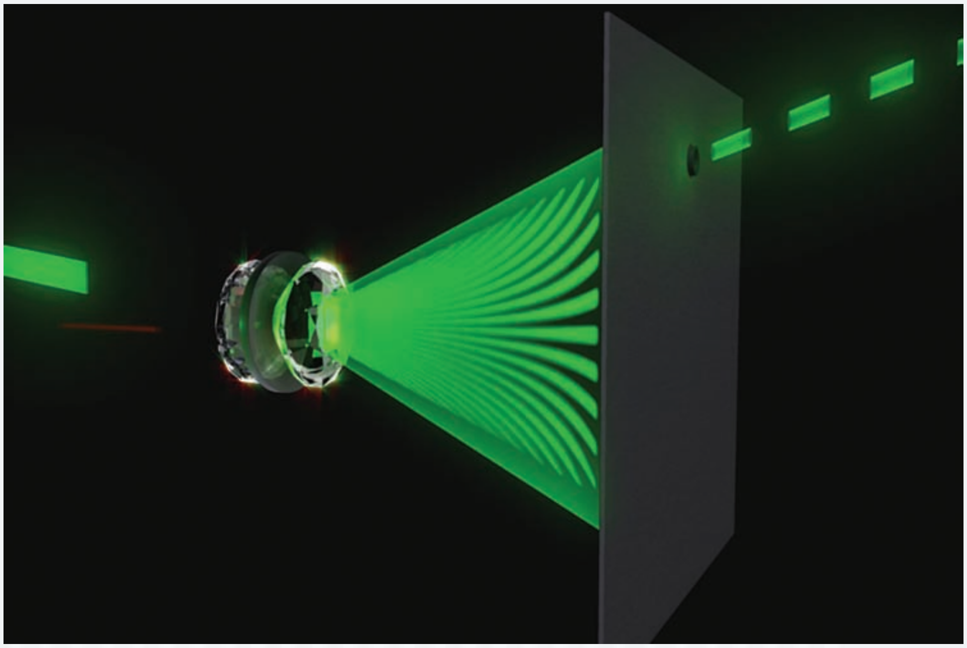
Hydrogen peroxide is a common oxidizer that has been shown to yield powerful energetic formulations when mixed with organic fuels. We expect to provide a thermodynamic and kinetic description of this material and a model for its behavior under detonation conditions. We will perform reactive flow simulations 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 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.

FY11 Accomplishments and Results

In FY11 we (1) performed ultrafast shock measurements on aqueous solutions of hydrogen peroxide, in particular with 90% concentration and at two different laser power settings, which enabled us to observe experimentally, for the first time, the transition between unreactive and reactive shocks in energetic liquids; (2) performed sound speed measurements on unreacted hydrogen peroxide at room temperature and pressures up to 2.5 GPa; (3) performed density functional theory and density



Schematic image of a setup combining a diamond anvil cell and photo-acoustic light scattering to measure sound speeds at high pressures.

functional theory tight-binding simulations of pure hydrogen peroxide at shock speeds from 5 to 10 km/s, which encompass the detonation velocity of this compound, then analyzed the results; (4) developed a decomposition kinetics model for hydrogen peroxide at detonation conditions; (5) used our shock experiment data, sound speed measurements, and simulation results to develop a thermodynamic model for unreacted hydrogen peroxide and used it to perform thermodynamic calculations of unreactive and reactive shocks in aqueous hydrogen peroxide mixtures; and (6) used our hydrogen peroxide thermodynamic model and decomposition kinetics in hydro-kinetics simulations of hydrogen peroxide under detonation conditions.

Proposed Work for FY12

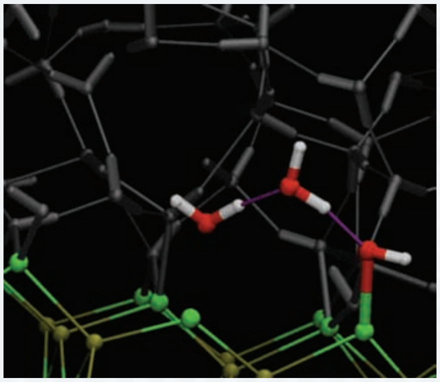
In FY12 we will (1) combine our experimental and simulation results to develop a full detonation kinetics model for hydrogen peroxide aqueous solutions; (2) employ our model in hydrodynamics and kinetics simulations, using the ALE3D hydrodynamic and Cheetah thermochemical codes, to quantify the detonation performance of peroxide solutions at different sample sizes and confinements and to determine the critical charge diameter; (3) develop a detonation kinetics model for hydrogen peroxide and nitromethane solutions (chosen as a representative example of a liquid oxidizer and fuel solution), and use the model in hydrodynamics and kinetics simulations to predict detonation properties of such a mixture; and (4) perform additional high-pressure experiments and density functional theory simulations in support of our objectives.

Publications

Zaug, J. M., et al., 2011. "Ultrafast shock interrogation of hydrogen peroxide–water mixtures: Thermochemical predictions of shock condition chemistry." *Proc. 17th Intl. Conf. APS Topical Group on Shock Compression of Condensed Matter*. LLNL-ABS-471470.

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

Tadashi Ogitsu (11-ERD-073)



A simulation showing the formation of a hydrogen-bonding network at the water–semiconductor interface, along with enhanced proton transfer along the network.

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 the microscopic properties of the surfaces of semiconductor elements in groups III and V of the periodic table as well as general structural and dynamical properties of water–semiconductor interfaces. In addition, we aim to gain understanding of the stabilities of various surface oxides and gas-phase water adsorption on those surfaces, and on 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.

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.

FY11 Accomplishments and Results

In FY11 we (1) studied the morphologies of oxygen- and hydroxyl-rich surfaces of group III through V semiconductors using ab initio simulations; (2) identified common local bonding configurations that showed different chemical affinities for the adsorption of water molecules, along with other topologies that form a hole-trapping state that may be linked to known surface corrosion effects in these materials; (3) successfully applied an analysis scheme based on the local bonding topology to ab initio molecular dynamics of the water–semiconductor interface to explain the observed behaviors; and (4) successfully compared theoretical and experimental x-ray spectroscopic data for the reference group III through V systems.

Proposed Work for FY12

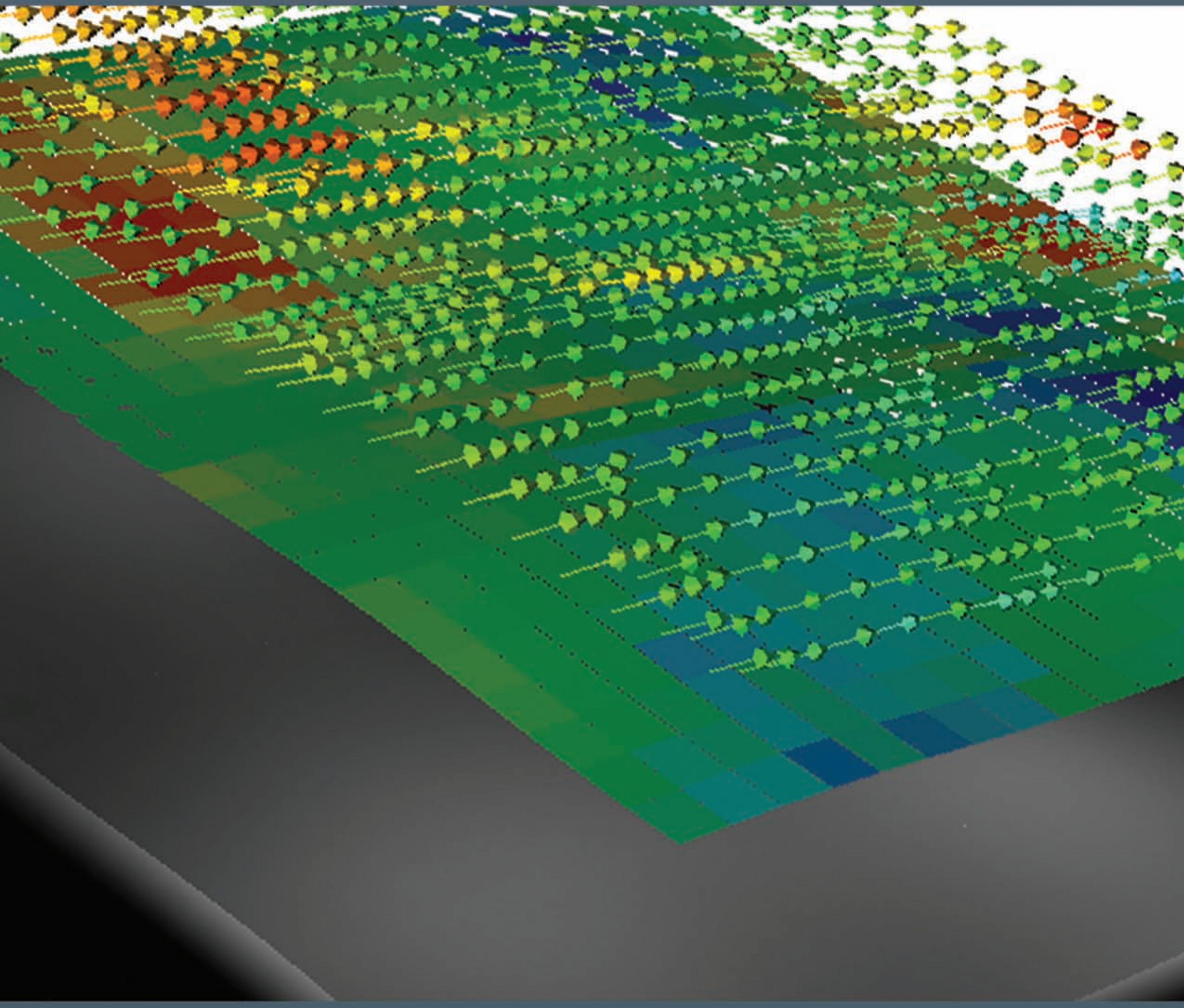
In FY12 we will complete our study of the morphologies of surface oxygen and hydroxyl on indium phosphide and gallium phosphide, following up on evidence found in FY11 that those surface morphologies can affect the hydrogen-bond network properties of interfacial water, which in turn govern proton transport parallel to the surface. Specifically, we will (1) investigate further the effect of surface morphologies on surface proton transfer and how that might lead to water formation, building on knowledge about the role of surface proton transfer in catalytic water formation; (2) examine the potential effects of nitrogen doping on surface hydrogen-bond network formation; and (3) refine our theoretical model of water–electrode interface based on the x-ray spectroscopic data for actual electrode samples collected in collaboration with the University of Nevada, Las Vegas.

Publications

Wood, B., T. Ogitsu, and E. Schwegler, 2011. "Ab initio modeling of water–semiconductor interfaces for direct solar-to-chemical energy conversion." *Proc. SPIE Optics + Photonics 2011*. LLNL-PROC-444431.

Wood, B. C., T. Ogitsu, and E. Schwegler, 2011. "Structure and reactivity of III–V semiconductors for photoelectrochemical hydrogen production." *Abstr. Paper. Am. Chem. Soc.* **241**. LLNL-JRNL-461893.

Earth and Space Sciences



Laboratory Directed Research and Development

FY2011

Coordinated Analysis of Geographic Indicators for Nuclear-Forensic Route Attribution

M. Lee Davisson (08-ERD-065)

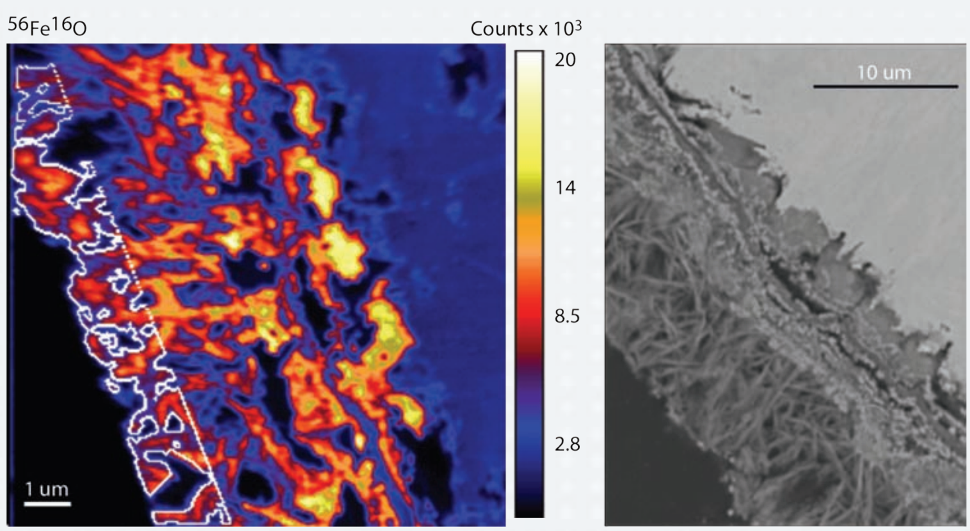
Abstract

We propose to develop a scientific foundation for determining the route of smuggled nuclear materials via nuclear forensics by exploring which types of signatures are viable over what timescales and why. The pathways and means of transport are key to uncovering smuggling routes and potential distribution sites of illicit nuclear material. Environmental and geological particulates and surficial deposits will be analyzed in coordinated studies by x rays, electron and ion microscopy, and gas chromatography–mass spectrometry to extract geographically specific signatures such as molecular and trace-element chemistry and stable isotope abundances. Accuracy and limitations will be analyzed using real-world materials.

This work will improve our understanding of which, why, and how geographically specific clues identify their environment and can be used as measurable, meaningful signatures of the route of smuggled radioactive and associated materials. We anticipate the first-ever demonstration of isotope signatures for route attribution. The analytical methods and technical capabilities we develop will significantly impact the scientific foundations for more efficient and effective route attribution. This project will leverage, enhance, and expand LLNL's capabilities and expertise in nuclear forensics; produce publications; and position Livermore for upcoming work-for-others projects in nonproliferation and nuclear attribution.

Mission Relevance

This project supports the Laboratory's national security mission by furthering nuclear forensic efforts relevant to nonproliferation and homeland security. The microscale and



A scanning electron microscope image of iron bead oxidized in lab experiments (left) is compared to a nanometer-scale secondary-ion mass spectroscopy map of iron oxide (right).

nanoscale characterization capabilities developed will also support the Laboratory's efforts in bioscience and technology by developing new analytical techniques.

FY11 Accomplishments and Results

In FY11 we focused on iron oxidation experiments with water of different compositions of oxygen-stable isotopes. Specifically, we (1) analyzed mineralogy and trace elements with scanning electron microscopy and by measuring oxygen-18/oxygen-16 ratios with nanometer-scale secondary-ion mass spectrometry, (2) collected field samples at different elevations in the Sierra Nevada mountains of California, (3) performed initial bulk stable-isotope measurements of the field samples, and (4) measured the mineralogy and isotope compositions of individual iron-oxide layers from the packaging material of an interdicted nuclear material. In summary, this project has enabled the testing and definition of various potential nuclear-forensic measurements designed specifically to produce information about the route over which a package of illicit nuclear material was transported before interdiction. These initial achievements formed the basis of a new hypothesis that resulted in new external funding from DOE.

Publications

Davisson, M. L., H. A. Ishii, and R. N. Leif, 2010. *Potential route evidence from fugitive chemicals adsorbed to packaging materials and metal oxides*. 51st Mtg. Institute of Nuclear Materials Management, Baltimore, MA, July 11–15, 2010. LLNL-CONF-434293.

Davisson, M. L., R. N. Leif, and H. A. Ishii, 2010. *Potential for detecting fugitive chemicals sorbed to packaging material*. LLNL-JRNL-461054.

Improving Atmospheric Flow Prediction at Intermediate Scales

Jeffrey Mirocha (09-ERD-038)

Abstract

A significant problem in simulating atmospheric flow in the lower atmosphere is how to accurately represent the entire spectrum of flow features, from the large scales of weather to the small scales of turbulence, that influence near-surface flow. To overcome this problem, we must fill an important knowledge gap involving modeling flow at intermediate scales—that is, those between the large- and small-scale endpoints that we understand more fully. In this project, we aim to fill this gap by developing both the physical models and technical expertise required to achieve an accurate multiscale flow simulation and prediction capability. Filling this knowledge gap is critical to many important scientific thrusts, including wind energy, climate modeling, and emergency preparedness and response.

If successful, this project will overcome several key obstacles to multiscale atmosphere flow simulation and prediction, such as parameterizations for modeling sub-filter turbulence at intermediate mesh resolutions, algorithms to represent turbulence interactions across computational meshes of different sizes, and guidance for appropriate computational domain and mesh configurations required for various spatial and temporal scales and flow regimes. These results will yield significant advances in simulating, predicting, and understanding complex flows that are vital components of many important applications across the atmospheric sciences.

Mission Relevance

By extending the ability to simulate and predict complex atmospheric flows, this work directly benefits efforts in risk and response management in support of the national security mission. In addition, wind-resource characterization and dynamic downscaling approaches address regional climate adaptation and mitigation in support of the Laboratory's missions in energy security and regional climate modeling.

FY11 Accomplishments and Results

In FY11 we (1) implemented new algorithms to improve the exchange of flow properties at nest interfaces for nested large-eddy simulations; (2) evaluated these improvements in several idealized simulations; (3) performed additional simulations over complex terrain and under different atmospheric stability categories, which necessitated further modifications to the new algorithms; and (4) validated the new nesting capability in a limited set of applications. This project has provided an advanced tool for conducting high-resolution large-eddy simulations of the atmospheric boundary layer under real-world conditions involving complex terrain and atmospheric buoyancy effects. We intend to seek further support from the DOE Energy Efficiency and Renewable Energy program, which is currently interested in projects that involve application of new methods and models to multiscale atmospheric simulations for wind energy.

Publications

Kirkil, G., and J. Mirocha, 2010. *Nested high-resolution mesoscale/large eddy simulations in WRF: Challenges and opportunities.* 5th Intl. Symp. Computational Wind Engineering (CWE2010), Chapel Hill, NC, May 23–27, 2010. LLNL-PRES-432948.

Kirkil, G., et al., in press. "Implementation and evaluation of dynamic subfilter-scale stress models for large-eddy simulation using WRF." *Mon. Weather Rev.* LLNL-JRNL-459613.

Mirocha, J., et al., 2012. *Transition and equilibration of flow parameters in one-way nested large eddy simulations using the weather research and forecasting model.* LLNL-JRNL-501655.

Mapping Patterns of Past Drought in California: Late-Holocene Lake Sediments as Model Diagnostics

Susan Zimmerman (09-ERI-003)

Abstract

We propose to create high-resolution chronologies for records of drought over the last 2,000 years from California lake sediments. From these, spectral analysis will identify dominant periodicities, and time-slice maps will show spatial patterns, both of which may be linked to forcing mechanisms of climate change. Our results will be used to improve the ability of computer models to predict the likelihood of future droughts, which is critical to helping resource managers plan water projects to meet societal demands. To accomplish this, we will measure radiocarbon dates of macrofossils such as pollen and twigs from outcrop and core samples for which collaborators have climate records under development. The goal is to create high-resolution, high-precision radiocarbon and calendar chronologies for these climate events.

We expect to produce (1) four to six high-resolution records of past droughts in California over at least the last 2,000 years, with high-precision chronologies; (2) spectral analysis of these records; and (3) proof-of-concept time-slice maps at 100-year intervals. The maps will demonstrate the utility of this approach and demonstrate the methodology for more such high-resolution, well-dated records. The individual records will be an important comparison to annual-resolution tree-ring records over the last 1,000 years and extend drought reconstructions 1,000 years farther back in time. We expect to better define the paleontological environmental response to the widespread droughts of 900 to 1300 A.D. and describe earlier changes at high resolution.

Lakes act as climate stations, recording changes such as temperature and precipitation over thousands of years. Through reconnaissance of 14 lakes across the state (yellow stars), we have identified 7 (red stars) that have been exceptional recorders of California climate over the last two millennia, providing insight into the patterns and causes of drought—past, present, and future.

Mission Relevance

Creating drought records for California supports the Laboratory's mission in energy security and climate. We will ultimately provide diagnostic tools in support of LLNL work to modify the Weather Research and Forecasting Model for regional climate prediction, which will enhance collaborations between Lawrence Livermore and outside organizations as well as support programs at DOE and various other federal agencies.

FY11 Accomplishments and Results

In FY11 we (1) completed high-quality age models for Zaca, Big Bear, Fish, and Frog lakes, discovering in the last two lakes intense hydrological variability on sub-decadal timescales; (2) collected 5-m-long core samples in several meadows in the southern Sierra and analyzed them, yielding an estimated recovery of approximately 10,000 years ago; (3) dated a new Mono Lake core sample, finding it to be the only continuous core covering the whole Holocene in existence, which was reported at the

Annual Meeting of the Geological Society of America; (4) recovered and dated very high-quality cores from Fallen Leaf Lake in the Tahoe basin—the first continuous cores from that basin—which showed decadal-scale recovery over the last few thousand years; and (5) tested a new method for concentrating pollen grains for radiocarbon dating using flow cytometry, which is a substantial step forward in dating lake sediments, in which macrofossils are rare and 10,000 to 30,000 grains are required for a single dating. In summary, this project advanced our understanding of the patterns and driving mechanisms of drought in California by dating 13 lakes and producing 6 exceptional age models. This project also contributed to 5 Ph.D. dissertations and resulted in 14 posters and talks presented at national meetings, in addition to a journal paper. Because of these achievements and the collaborations created, this work has already received support from the National Foundation of Science and the Moore Foundation. Next steps include conducting millennial-scale climate model runs to identify the precursors to drought.

Publications

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LeRoy, S. L., et al., 2009. *Radiocarbon constraints on fossil thinolite tufa formation in the Mono Basin, CA.* American Geophysical Union 2009 Fall Mtg., San Francisco, CA, Dec. 14–18, 2009. LLNL-POST-421330.

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Zimmerman, S. R., et al., 2011. *Holocene sedimentation patterns and volcanic history from the BINGO core, Mono Lake, CA*. 2011 GSA Annual Mtg., Minneapolis, MN, Oct. 9–12, 2011. LLNL-PRES-503461.

Zimmerman, S., et al. in press. "Potential for accurate and precise radiocarbon ages in deglacial-age lacustrine carbonates." *Quat. Geochronol.* LLNL-JRNL-501033.

Stardust Science: Nanometer-Scale Analytical Studies of Materials

John Bradley (09-ERI-004)

Abstract

The National Aeronautics and Space Administration's high-profile Stardust mission has brought back sample cometary grains that can provide new insight about the early solar system. We will leverage Lawrence Livermore's analytical instruments to perform research on such natural materials and on cadmium zinc telluride, a strategically important man-made semiconductor material with potential application in room-temperature radiation detectors. All of these materials are heterogeneous at the nanometer scale and we plan to obtain scientific insight about them by interrogation at that scale using LLNL's unique combination of state-of-the-art instruments.

We seek to achieve a fully integrated analytical capability for analyzing a single, expensive nanoscale sample using multiple instruments simultaneously to maximize the science yield. This capability will also be applicable to nuclear forensics samples. We expect to better understand the degree of modification sustained by the comet Wild 2 samples during capture and to gain important new insight about comets, asteroids, other primitive meteoritic materials, and the early solar system. Moreover, our studies of cadmium zinc telluride will provide fundamental scientific insight at the nanoscale about the growth of defects in semiconductors, thus laying the foundation for high-performance, defect-free cadmium zinc telluride for room-temperature radiation-detection devices.

Mission Relevance

This project supports the Laboratory's national security mission by providing insight into an important material for radiation detection. Other aspects of the project support efforts in breakthrough science by addressing fundamental planetary science questions and the properties of these and other materials at atomic or near-atomic level. The proposed research will also attract top young scientists to the Laboratory.

FY11 Accomplishments and Results

We successfully diversified use of the Titan "SuperSTEM" transmission electron microscope at Livermore to launch new projects of strategic interest to the DOE. Specifically, we (1) studied defects in cadmium zinc telluride, a semiconductor suitable for use as a room-temperature gamma-ray detector; (2) examined atomic bonding and functional groups at the atomic scale in natural organic matter (kerogens) and cellulose-based propellants used in U.S. Army munitions; (3) examined the light lanthanide metals cerium, praseodymium, and neodymium with electron energy-loss spectroscopy—the higher energy resolution and high signal-to-noise allowed for measurement of complex and subtle excitation spectra in the lanthanide metals (surrogates for actinides and also integral components in a wide range of technologies such as energy storage and magnets), validating the applicability of the screened trivalent atomic model used for these materials; and (4) extracted and compared momentum-transfer dependence of the x-ray scattering against atomic calculations for the most tightly bound excitonic resonances, which provides a direct test of the predicted atomic radial wave functions. The unprecedented 0.1-eV energy resolution combined with approximately 1-Å spatial resolution of the Titan instrument enables investigation of solid-state bonding and electronic structure of materials at the single-atomic-column scale. In addition to ongoing funding from the National Aeronautics and Space Administration, the unique analytical capabilities of the Titan are now attracting exploratory funding from DOE and the U.S. Army.

Publications

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Chi, M., et al., 2009. "The origin of refractory minerals in comet 81P/Wild 2." *Geochim. Cosmochim. Acta* **73**(23), 7150. LLNL-JRNL-410054.

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Kearsley, A. T., et al., 2011. *Experimental impact features in Stardust aerogel: How track morphology reflects particle structure, composition and density.* LLNL-JRNL-517492.

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Wozniakiewicz, P. J., et al., 2011. *The origin of crystalline residues in Stardust Al foils: Surviving cometary dust or crystallized impact melts?* LLNL-JRNL-488931.

Enhancing Climate Model Diagnosis and Intercomparison

Karl Taylor (10-ERD-060)

Abstract

Current capabilities in global climate modeling and the evaluation of those models lack information about why the models differ in their projections. They also fail to incorporate carbon cycle and atmospheric chemistry information. In addition, it is not understood how uncertainties in cloud feedbacks and other key mechanisms vary over different timescales, and simplistic metrics are used for model performance. We intend to conduct innovative research that would address these issues. We will develop concepts for globally distributed and synthetic data sets, as well as advanced metrics for model simulations of ocean, land-surface, and sea-ice processes. We will also provide new analysis and diagnostic methods for cloud and other feedbacks and new measures of model performance.

This project will result in the most comprehensive and scientifically meaningful set of climate modeling and evaluation tools available to the scientific community to date. Specifically, the enhancements over existing global climate modeling and evaluation approaches are expected to (1) improve the ability to constrain the climate model projection uncertainties related to structural uncertainties in the models themselves (e.g., in the physics and parameterizations); (2) improve the ability to constrain inter-model feedback differences on century timescales, which are of most interest; (3) increase our understanding of model differences and reliability; and (4) maintain and strengthen LLNL's national and international preeminence in climate model diagnosis and intercomparison.

Mission Relevance

This work supports LLNL's mission focus area in energy and climate by furthering regional and global climate predictive capabilities, and it is directly relevant to DOE's mission in understanding and mitigating global climate change. It will address scientific questions that underpin the upcoming Coupled Model Intercomparison Project, a flagship activity of DOE's Climate Change Prediction Program.

FY11 Accomplishments and Results

In FY11 we developed innovative concepts for data management, analysis, and reduction in uncertainty for globally distributed data sets. Specifically, we (1) improved the software library used by modeling groups worldwide to prepare climate model output for analysis, (2) helped develop quality control codes for uncovering discrepancies in the output produced by models, (3) developed a graphical interface to a new search capability for climate data discovery and download, (4) established

Web-based organizational structures to accommodate "sibling" projects to phase 5 of the Coupled Model Intercomparison Project, and (5) followed up on FY10 accomplishments by preparing publications on an innovative data set used to compare models and observations.

Proposed Work for FY12

We will (1) develop and use salinity data sets for diagnosing and evaluating climate models from phase 5 of the Coupled Model Intercomparison Project, (2) continue developing advanced performance metrics for evaluating ocean and sea-ice processes in those models, and (3) continue to develop innovative concepts for data management and analysis for globally distributed data sets.

Publications

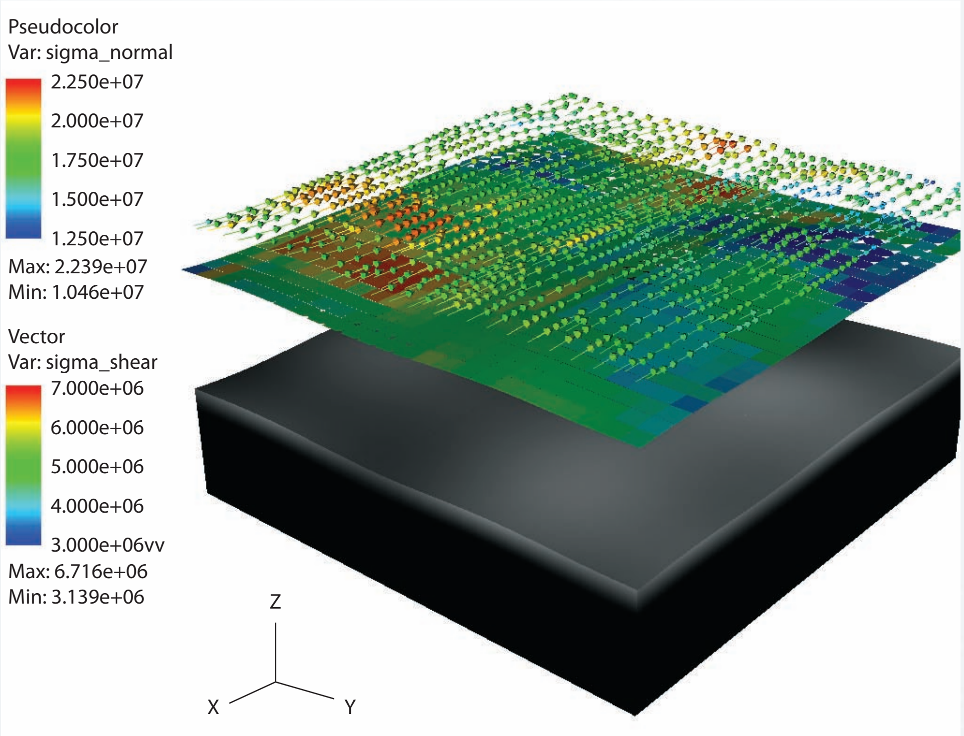
Santer, B. D., et al., 2011. "Separating signal and noise in atmospheric temperature changes: The importance of timescale." *J. Geophys. Res.* **116**, D22105. LLNL-MI-499392.

Creating Optimal Fracture Networks for Energy Extraction

Frederick Ryerson (11-SI-006)

Abstract

The technology required to extract energy resources contained within the Earth's crust is constantly evolving. Resources that were once considered unconventional



Simulation of the distribution of tangential (vectors) and normal (grid) stress along a rough fracture surface. Stress is intensified on the "windward" side of small features on the surface plot.

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 circulation of water for more effective use of hydrothermal energy resources. 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 capability—GPAC (GeoPhysics Analysis Code)—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 produced from shales. This capability will describe the optimal fracture network, how to create this network, how to understand what has been created, and how this perturbed system evolves over time. We will develop a high-fidelity hydro-geomechanics code that describes both hydraulic and explosive fracturing in the subsurface. This code will be linked to a wave propagation code for predicting seismicity associated with the hydraulic fracturing process.

Mission Relevance

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

FY11 Accomplishments and Results

In FY11 we (1) established the basic fracture simulation framework, implementing an explicit mechanics solver and a small strain and linear elastic implicit mechanics solver in parallel, an implicit and explicit parallel-plate flow solver, mesh splitting along element boundaries in serial, common plane contact for rough surfaces in parallel, and a coupled explicit mechanics solver and parallel-plate flow solver in parallel, including node-splitting and element remapping; (2) formulated the seismic source term for a rough, confined fault and began a parameter study to determine the response under different loading conditions—these capabilities allow us to address problems such as coupled flow in evolving fracture networks in three dimensions and fault activation from hydraulic pressure; (3) demonstrated switching between implicit and explicit solvers, allowing transition from static loading (implicit) to dynamic loading (explicit) when fracture occurs; (4) applied the earthquake location program Bayesloc to Salton Sea geothermal field data, and found that location scatter may be because of errors in arrival times that can be improved with a cross-correlation algorithm; (5) analyzed Salton Sea data using adjoint wave tomographic methods and established that earthquake source parameters can be recovered—the workflow developed will be used to improve further crustal models and source parameter accuracy; (6) prepared, with Altarock Energy, seismic stations for deployment at the Newberry Enhanced Geothermal System demonstration site in Oregon to record micro-seismicity and characterize the velocity structure using ambient noise methods; and (7) extended quantification of uncertainties in the geological characterization of fractures to include the effect of mean fracture radius, variance of fracture radius, and mean orientation and densities on the topology of the fracture network, as well as to assess the overall system response.

Proposed Work for FY12

We will (1) develop hydraulic flow simulations by adding fracture elements and hydraulic pressures on fracture surfaces to existing codes; (2) develop a seismic source term within a continuum framework to model the full-range of motion for rock masses using implicit and explicit methods; (3) develop capabilities to accommodate multiple, interacting fractures, including the effects of induced stress fields in the near field of the fracture process zone, as well as the coalescence of fractures; (4) couple adjoint wave tomography with Livermore's existing seismic code WPP (Wave Propagation Program); and (5) use the seismic array to monitor stimulation at the Newberry Caldera.

Publications

Ezzedine, S. M., et al., 2011. *A surrogate hybrid three dimensional model for fluid-induced fracture propagation under conditions of uncertainty.* AGU Fall Meeting 2011, San Francisco, CA, Dec. 5–9, 2011. LLNL-ABS-492063-DRAFT.

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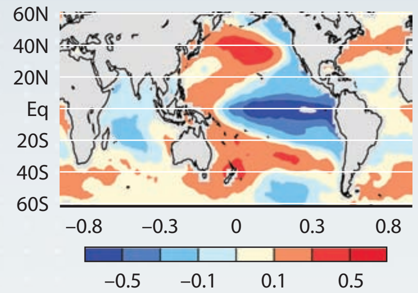
Detection and Attribution of Regional Climate Change and Drought Precursors

Celine Bonfils (11-ERD-006)



Drought-conducive pattern of a La Niña phase related to subtropical droughts inferred from observed sea-surface temperature anomalies. We found that the temporal component associated with this spatial pattern can explain a large fraction of the observed variability of tropical and subtropical droughts.

Abstract

For centuries, droughts have affected human and environmental systems. Understanding the primary causes of droughts has become even more crucial in the context of global warming, with drier conditions predicted to intensify in already arid regions. The detection and attribution techniques typically used to identify human-induced climate change have seen little application in drought research. Sea-surface temperature patterns similar to those under La Niña conditions and the changes in atmospheric circulation are widely recognized as key factors triggering subtropical droughts. We will determine whether an emerging human signal exists in the frequency and spatial structure of drought precursors and how the relative weights of forced and natural contributions to drought evolve over time.

This project will improve our scientific understanding of drought mechanisms. We expect to investigate new avenues in detection and attribution, including new variables and regional-scale tools. Ultimately we will generate knowledge that can inform policy decisions on adapting to drought in a changing climate.

Mission Relevance

This work supports basic research that is relevant to the Laboratory's mission in climate challenges by increasing our understanding of climate impacts and the key climate processes that drive uncertainty in climate prediction. We will deal with climate change's impact on water resources and fresh water availability—critical to society, food production, and regional sustainability.

FY11 Accomplishments and Results

In FY11 we made major progress in clarifying the role of oceans in drought initiation. Specifically, we identified an El Niño–Southern Oscillation spatial sea-surface pattern as a natural trigger of historical droughts. We then applied a detection and attribution technique to assess whether the temporal statistics of this pattern will evolve in the future in interaction with model-projected greenhouse warming. This assessment showed a trend toward less La Niña (more El Niño) conditions that would potentially initiate fewer subtropical (more tropical) droughts. However, we anticipate that any shift in future drought characteristics will instead be driven more by an external climatic forcing such as a slowly evolving greenhouse-warming signal (e.g., as manifested by warmer global oceans or by a related change in large-scale atmospheric circulation features), rather than by a change in the El Niño–Southern Oscillation mode of variability. Finally, we have gathered various metrics on drought-related changes in atmospheric circulation and collaborated with scientists who are investigating drought behavior in various simulations included in Phase 3 of the Coupled Model Intercomparison Project.

Proposed Work for FY12

In FY12 we will (1) continue to characterize drought behavior in observations and various model runs, (2) determine whether including external forcing yields variations in droughts consistent with observations, (3) use simulations included in Phase 5 of the Coupled Model Intercomparison Project (as available) to begin examining whether a human influence can be detected in the recently observed expansion of subtropical droughts, and (4) determine a suitable tropical atmospheric circulation metric for formal detection and attribution of climate change, and compute this metric for both observations and model runs.

Publications

Bonfils, C., et al., 2011. *Investigating the nature and causes of historical changes in droughts: Exploration phase.* LLNL-POST-471695.

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Images and Spectra of Extrasolar Planets from Advanced Adaptive Optics

Bruce Macintosh (11-ERD-048)

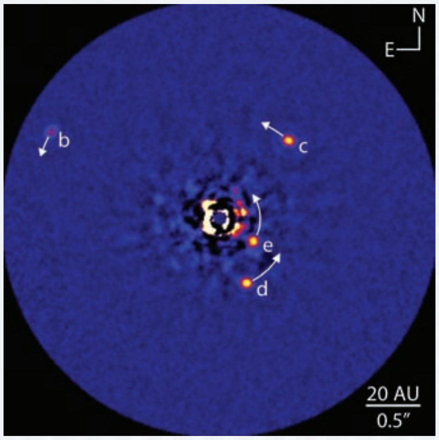

Image of the HR8799 system showing the newly discovered fourth planet HR8799e. Arrows show the direction and speed of orbital motion.

Abstract

The newest frontier in exoplanet studies 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 telescope. Specifically, we will characterize the HR8799 multi-planet system, determining the composition and evolution of the planets. In addition, we will develop tools for the unbiased estimates of planetary spectra and lead a 100-night survey campaign on the new LLNL-built Gemini Planet Imager adaptive optics system. Our new capability will be an order of magnitude more sensitive than any to date and could enable 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 c—making them the lowest-temperature exoplanets ever spectroscopically characterized—and use these results to adapt models of the planets' complex atmospheres. We will use precision adaptive optics astrometry and numerical integration to find orbital parameters that are consistent with observations and with formation and evolution scenarios. Next, through our large-scale Gemini Planet Imager project that will survey 600 stars—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. Remote sensing is also a possible application of the techniques we will develop for extracting faint planetary signals from hyper-spectral data with highly correlated noise.

FY11 Accomplishments and Results

During FY11 we had several significant achievements. Specifically, we (1) used improved image processing to discover a fourth planet in the HR8799 system, which makes HR8799 the most massive planetary system ever discovered and a puzzle for planet-formation theorists; (2) used orbit simulations to study the dynamics of this puzzling system, exploring its orbital stability; (3) published our first spectroscopic observations of the atmosphere of HR8799b, which showed complex, nonequilibrium chemistry and cloud structure; (4) developed new algorithms for analyzing the spectroscopic data and explored new formal algorithms for extracting faint signals from speckle-noise-dominated images; and (5) assembled an international team for a new exoplanet survey campaign using the Gemini Planet Imager, for which LLNL is leading construction. Our research was the only campaign selected by Gemini—we have been allocated 890 hours of telescope time, the largest program ever planned at the observatory, and the most statistically powerful planet-imaging campaign ever conducted.

Proposed Work for FY12

During FY12 we will (1) continue our observations and modeling of the HR8799 system, conducting a Markov-chain Monte Carlo exploration of the enormous space of potential orbital configurations to find stable solutions for the system's evolution; (2) complete our analysis of the HR8799c spectrum; (3) design adaptive estimation of noise correlations for extracting faint signals from speckle noise using spectral information; (4) characterize the Gemini Planet Imager in the laboratory using simulated starlight before it begins science operations and test our observing strategies and algorithms on real data; and (5) carry out "first light" observations of extrasolar planetary systems on the Gemini Planet Imager as soon as it is operational.

Publications

Barman, T. S., et al., 2011. "Clouds and chemistry in the atmosphere of extrasolar planet HR8799b." *Astrophys. J.* **733**, 65. LLNL-JRNL-476433.

Barman, T. S., et al., 2011. "The young planet-mass object 2M1207b: A cool, cloudy, and methane-poor atmosphere." *Astrophys. J.* **735**, L39. LLNL-JRNL-485291.

Galicher, R., et al., 2011. "M-band imaging of the HR 8799 planetary system using an innovative LOCI-based background subtraction technique." *Astrophys. J.* **739**, 41. LLNL-JRNL-490367.

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

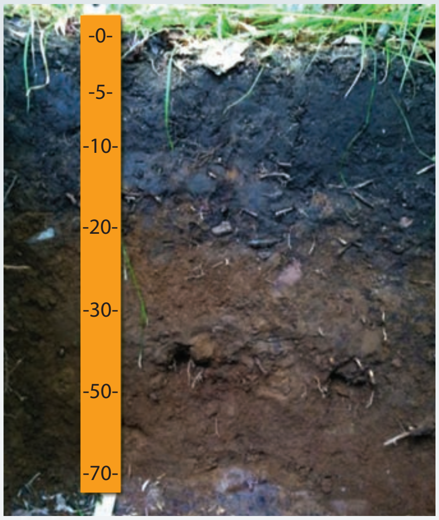
Karis Mcfarlane (11-ERD-053)

Abstract

Our goals are to determine the short-term, inter-annual 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 ecosystem and soil respiration, and specifically that disturbances will cause a shift in carbon sources used by soil microbes from primarily labile 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 ecosystem and regional scales, altering the biosphere signal for CO_2 partial pressure. We expect our work will demonstrate the utility of carbon-14 for partitioning ecosystem and soil respiration fluxes, quantifying shifts in microbial carbon sources, and the sensitivity of regional carbon fluxes and carbon-14 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.



A soil profile at the Willow Creek site, located in a northern hardwood forest in Wisconsin. The yellow ruler shows depth in centimeters.

Mission Relevance

This work addresses a central Laboratory mission in energy and climate. Specifically, this research will make valuable contributions to the improved 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.

FY11 Accomplishments and Results

We began data collection for the pre-harvest treatment period at two sites in Wisconsin, conducting $^{14}CO_2$ sampling at the WLEF-TV tower—in collaboration with the National Oceanic and Atmospheric Administration—and initial sampling at Willow Creek. Specifically, we (1) installed instrumentation at the Willow Creek site, including flask samplers on the flux tower, transects for surveying soil CO_2 flux across the tower footprint, and four intensive plots; (2) instrumented the intensive plots for continuous measurements of surface CO_2 flux, moisture, and temperature; (3) installed, at each plot, soil gas wells at six different depths for determining CO_2 flux and levels of ^{13}C and ^{14}C along the soil profile; (4) sampled plots for soil carbon characterization, microbial biomass, soil physical characteristics, and incubations; (5) began sampling canopy air, including intensive sampling to establish the ^{14}C end member for ecosystem respiration and a time series to continue throughout the project; and (6) made surface soil CO_2 flux measurements, including six measurements of soil profiles and collection of some soil gas samples from the profiles for ^{14}C and ^{13}C determination. We also hired a new postdoctoral researcher to perform the measuring and modeling of soil respiration.

Proposed Work for FY12

In FY12 we will (1) continue $^{14}CO_2$ measurements from air sampled at the Willow Creek and WLEF-TV tower sites, (2) continue CO_2 purification at the University of Colorado for air from the WLEF-TV tower, (3) perform ^{14}C measurements of the purified CO_2 at Livermore's Center for Accelerator Mass Spectrometry, and (4) continue soil respiration flux measurements and sampling for $^{14}CO_2$.

Using Aerosols to Discriminate and Quantify Greenhouse Gas Emissions by Source

Eric Gard (11-FS-010)

Abstract

We propose to assess the feasibility of using aerosol (particulate) composition, concentration, and size distribution information to improve the discrimination and quantification of greenhouse gas emissions, and assess how these analyses might help determine the emission source. An independent verification system is a critical element for any future regional, national, or international greenhouse gas agreement, because it allows all parties to trust that their counterparts are adhering to the terms of their agreement. It is hoped that this effort will aid in the conceptual development of a future greenhouse gas treaty verification system and ultimately will contribute to its implementation. To this end, we will explore the best ways that aerosol information can be used in conjunction with gas phase data to demonstrate that a verification system will perform at the level required to instill confidence that agreements are being upheld.

It is clear from decades of environmental and pollution research that both natural and anthropogenic activities are correlated with patterns in gas phase and aerosol

emissions. The extent to which quantitative assessments of aerosol emissions can be used to improve our discrimination of both natural and anthropogenic greenhouse gas sources has not been well explored. This project will result in a detailed study of the existing aerosol analysis techniques and how they can be used to better understand and quantify the anthropogenic greenhouse gas emissions at the regional and country level. In addition, both the advantages and difficulties in using this aerosol information to infer greenhouse gas emissions will be detailed. Finally, the extent to which aerosol information can be used as an independent diagnostic for atmospheric transport and inversion models will be investigated. These results will be detailed in several peer-reviewed articles.

Mission Relevance

This project supports Laboratory and DOE missions in assessing and potentially mitigating the effects of climate change and helps quantify the regional consequences of choices about fossil fuel use.

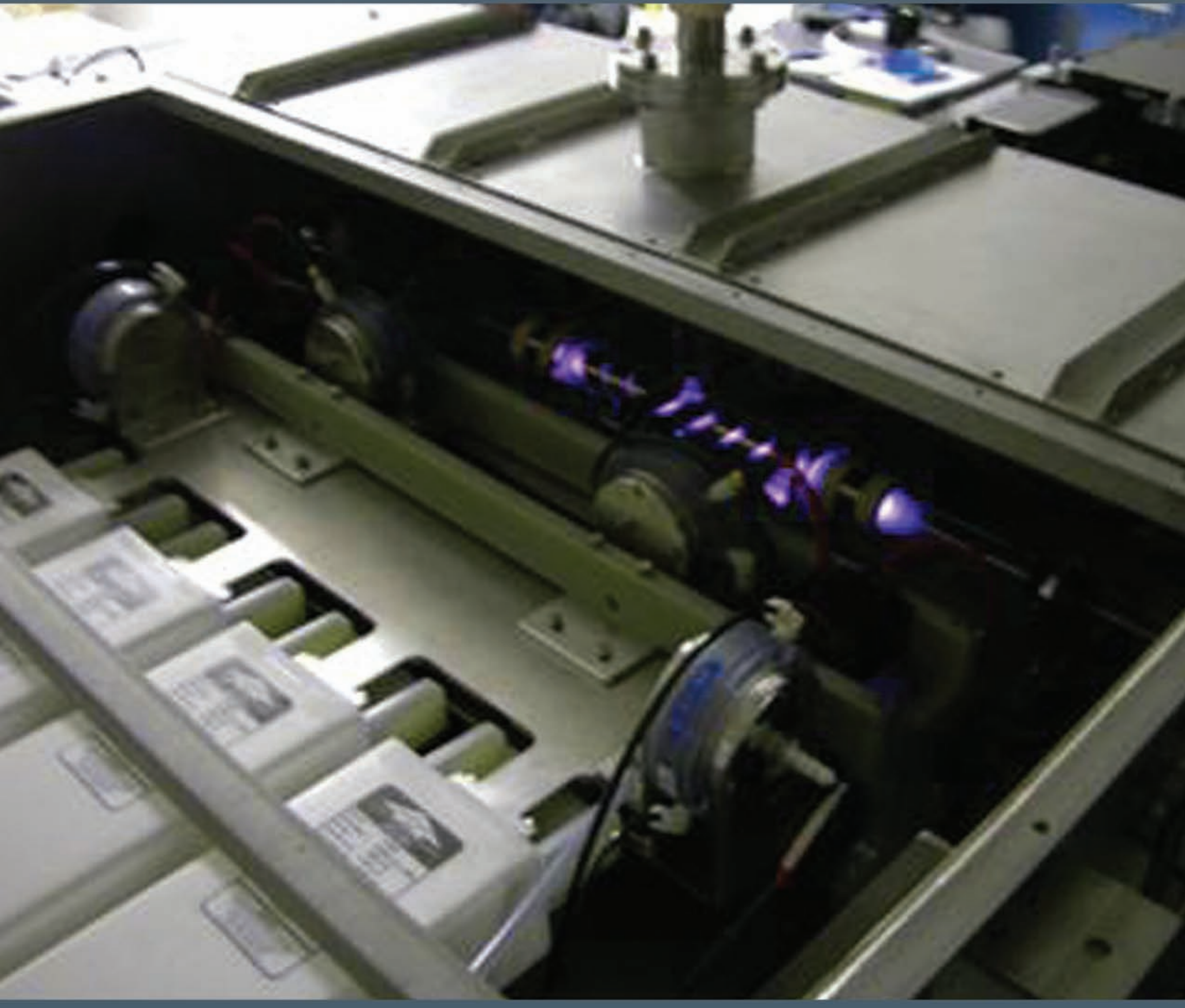
FY11 Accomplishments and Results

After a late FY11 start, we (1) began a detailed study of existing aerosol techniques to document the current state of aerosol analysis technology, (2) initiated formulation of several analysis schemes with varying information and cost trade-offs, and (3) worked to develop an overall feasibility of coupling these new and older aerosol analytical methods with gas-phase methods for anthropogenic greenhouse gas quantification.

Proposed Work for FY12

In FY12 we will complete the detailed study of existing aerosol analysis techniques and documentation of both advantages and difficulties in using this aerosol information to infer greenhouse gas emissions. We will determine how aerosol information can be used as an independent diagnostic technique for guiding atmospheric transport and inversion models.

Energy Supply and Use



Laboratory Directed Research and Development

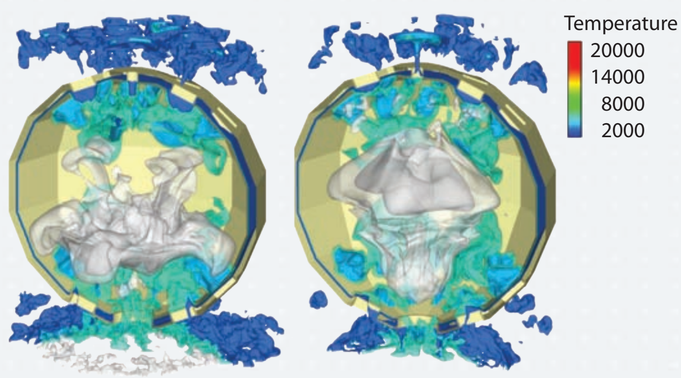
FY2011

Dynamic Chamber Processes for LIFE: Simulations and Experiments on Beam Propagation and Chamber Clearing

Jeffery Latkowski (10-SI-009)

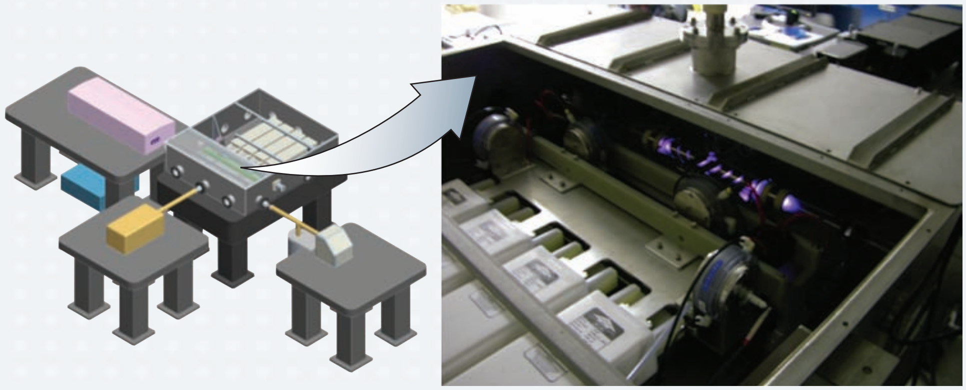
Abstract

To mitigate the challenges of nuclear energy and to advance the timescale for availability of fusion sources, the Laboratory envisions a novel once-through, closed nuclear-fuel cycle based upon the Laser Inertial Fusion Energy (LIFE) concept. We propose to couple modeling and experiments to explore and resolve fundamental issues for the LIFE chamber, including gas clearing between rapid (10-Hz) shots and laser beam propagation. Radiation-hydrodynamics modeling will predict the chamber-gas state after a LIFE shot and support scaled experiment design, while computational fluid dynamics will be used to explore the clearing of hot gas and debris from the chamber. Scaled experiments using existing kilojoule-class lasers will be used to observe chamber clearing and beam propagation.



0.5 Mj blast wave, 100 cm debris source region, after 3 shots

1 Mj blast wave, 200 cm debris source region, after 3 shots



Computational fluid-dynamics modeling used to examine the impact of source energy and size on debris dispersion in the Laser Inertial Fusion Energy (LIFE) chamber over many shots (top). The gray iso-surface bounds the region where lead makes up more than 1% of the local density. The theta-pinch facility (bottom) is generating plasmas with an electron density greater than 10^{16} atoms/cm^3 and temperatures in excess of 1.6 eV to improve our understanding of cooling times and charge states for low-temperature plasma regimes of interest to LIFE.

We expect to achieve an integrated, validated assessment of the behavior of chamber fill gases within the LIFE chamber including (1) a consistent model of post-shot plasma conditions; (2) characterization of beam propagation, including gas ionization, debris, density gradients, and turbulence; (3) assessment of chamber gas dynamics, including aerosol formation; (4) an optimized vent and fill protocol; and (5) demonstration of plasma cooling, clearing, and beam propagation in a scaled chamber. In addition, we will develop plans for a high-energy experimental campaign.

Mission Relevance

This project supports LLNL's mission of enhancing energy security for the nation and builds directly upon the Laboratory's world-class capabilities in inertial-confinement fusion and laser technologies. Our research is a key component of the Laboratory's strategic roadmap in energy security. Resolving key issues for practical, cost-effective laser inertial fusion energy plants also supports the Laboratory mission in environmental security by enabling a source of abundant, clean power without nuclear waste disposal, safety, carbon sequestration, or proliferation issues. A demonstration of chamber clearing and beam propagation at high repetition rates will address key technical issues about the feasibility of the LIFE concept while producing important scientific results.

FY11 Accomplishments and Results

We (1) completed activation of the theta-pinch facility, including installation of laser tables, cable trays, electric power, and cooling water as well as the fabrication, assembly, and operation of the plasma source and diagnostics systems; (2) routinely formed a xenon plasma (with an electron density of 3×10^{16} cm^{-3} and electronic energy of 1.6 eV) while operating the source at low energy (115 J); (3) activated diagnostic systems that included voltage and current monitors, a spectrometer system with photomultiplier and gated intensified charge-coupled device camera detectors, single and double Langmuir probes, and a Thomson scattering system; (4) began developing spectroscopic methods to measure temperature and density by identifying strong emission lines of xenon ions; (5) conducted Hydra code simulations to guide theta-pinch design, examining both coil stress and plasma conditions as a function of drive current; (6) conducted computational fluid dynamics studies of the fusion chamber using the Caltech Virtual Test Facility to explore the impact of source strength and size on debris fate and to quantify turbulence and post-shot density gradients; (7) studied the atomic physics of xenon at low-charge state and the cooling times of a xenon–debris mixture to improve model predictions of chamber response; (8) developed 1,000-shot blast-wave models of the chamber system to explore gas exchange between the fusion and vacuum chambers—in the models, the chamber gas density was sustained, neither voiding nor growing unacceptably; (9) performed radiation-hydrodynamics simulations of the chamber system with different first-wall materials and molten lead at the first wall; (10) identified a window for successful laser propagation through lead vapor that suggests aggressive chamber clearing is not required; and (11) identified regions in which temperatures and debris concentrations could favor aerosol development.

Proposed Work for FY12

In FY12 we will (1) study the radiative cooling of xenon and demonstrate laser beam propagation through hot xenon, using the theta-pinch facility; (2) optimize the vent and fill protocol using many-shot simulations in two- and three-dimensional integrated MIRANDA and HYDRA calculations to estimate quasi-steady state densities and temperatures; and (3) improve our understanding of the early-time gas response to target explosion using various integrated radiative-hydrodynamic simulations and datasets to explore hydrodynamic motions under the influence of a blast wave induced by target explosion, including the effects of radiation, turbulence, and shock propagation in the fusion chamber.

Publications

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Kane, J. O., et al., 2011. "Modeling of the LIFE minichamber Xe theta-pinch experiment." *Proc. SPIE* **7916**, 791605. LLNL-CONF-461704.

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Design of Novel Catalysts to Capture Carbon Dioxide

Roger Aines (10-ERD-035)

Abstract

We propose to develop new, robust, small-molecule catalysts that mimic the behavior of the natural enzyme carbonic anhydrase. Such catalysts can dramatically increase the rate of carbon dioxide (CO_2) separation and thereby reduce the size and cost of industrial processes that seek to keep CO_2 from being emitted to the atmosphere. This separation cost is the primary barrier to worldwide carbon capture and storage necessary to control climate change. We will use quantum mechanical predictions of the catalytic behavior of small molecular systems that mimic the active centers in natural proteins that catalyze this reaction in animals and plants. The best-prospect molecules will be synthesized and tested for both catalysis and robustness to environmental interference.

Comparison to natural systems suggests that it may be possible to increase the chemical capture rate of CO_2 separation systems by up to a factor of 1,000 using small, industrially robust catalysts that mimic the behavior of protein systems without their frailty. This would dramatically decrease the cost of carbon capture from point sources. More importantly, it could enable the direct capture of CO_2 from the atmosphere. This would allow us to manage diffuse sources of CO_2 emissions from airplanes and home heating, for example, via centralized air-capture facilities.

Mission Relevance

As part of the Laboratory's mission to enhance the environmental and energy security of the nation, LLNL is committed to developing innovative technologies to reduce atmospheric CO_2. For feasible application, the chemistry of the capture process must be more efficient than current methods to keep the capture device size manageable (which also controls capital expenditure). If successful, our project will provide the necessary increase in capture rate, which can make air capture a key new technology for the Laboratory.

FY11 Accomplishments and Results

In FY11 we focused on zinc–nitrogen capture systems with similar configurations to known catalysts using screening that examines the spatial configuration of essential features that enable a molecular complex to interact with a specific target receptor. Specifically, we (1) compared bond lengths and angles from our energy maps to determine likely catalyst candidates for measurement or modification, which allowed us to screen a great number of compounds without concern for whether they can be synthesized; (2) considered molecule fragments that hold potential for development into complete catalysts and that we can evaluate like building blocks; (3) considered, from previous experience, new molecules that appear to have the desired properties;

and (4) produced designs for 21 catalysts, synthesizing 16 of these and testing 5 with reaction rates faster than our target catalysts.

Proposed Work for FY12

In FY12 we will (1) continue the task of identifying and developing catalysts with an emphasis on designs that mimic the hydrogen bonding, important for product release, seen in carbonic anhydrase; (2) continue evaluating which designs have resulted in the fastest catalysts; and (3) incorporate results from environmental testing of catalysts to develop longer-lived versions for industrial applications.

Publications

Aines, R., et al., 2011. "Modeling, synthesis, and characterization of zinc-containing carbonic anhydrase active site mimics." *Energ. Proceedia* **4**, 2090. LLNL-PROC-451972.

Kulik, H., 2010. *Computational design of carbonic anhydrase mimics for carbon dioxide capture.* LLNL-ABS-426922.

Wong, S. E., et al., 2011. "Designing small-molecule catalysts for CO_2 capture." *Energ. Proceedia* **4**, 817. LLNL-PROC-451972.

Prediction of Underground Coal Gasification Cavity Growth, Coal Conversion, and Geophysical Signatures

David Camp (10-ERD-055)

Abstract

Underground coal gasification (UCG) is an emergent technology critical for boosting U.S. energy security while mitigating greenhouse gas emissions. However, scientific questions and technical challenges impede sustainable UCG production. Industry is seeking improved simulation and monitoring capabilities to predict performance and minimize environmental impact as a function of site, design, and operational characteristics. These challenges demand an accurate, coupled-process simulator. We propose to develop the world's most complete and accurate UCG simulator and resolve outstanding process questions. We will identify gaps in prior methods and develop a new simulator, leveraging Livermore investments in UCG operations and modeling and in computational geosciences.

We expect to provide a far stronger understanding and predictive capability for UCG operations. Our coupled simulation and geophysics monitoring approach will allow improved siting, design, permitting, operation, monitoring, and environmental performance of pilot and commercial projects. In addition, our research will accelerate technology improvements and commercial deployment and serve as a resource in addressing emerging regulations. Development of detailed sub-models will require and allow focused inquiry into complex transport, reaction, and mechanical phenomena, and the integrated model will enable a new level of scientific and technical interrogation of complex UCG behavior.

Mission Relevance

This project is highly aligned with the energy and environmental security mission of the Laboratory and will help spur the deployment of UCG to reduce the cost and accelerate delivery of potentially low-carbon, secure energy to the nation.

FY11 Accomplishments and Results

In FY11 we (1) created an initial rubble-zone model; (2) created a one-dimensional (1D) cavity-gas and heat-transfer model that provides much better spatial resolution than the previous best model and made substantial progress towards an even better 3D cavity-gas model; (3) added a simple continuous representation of spalling to the wall-zone model, which we then validated against laboratory experiments; (4) completed the second spiral of integrated model development by dynamic linking, through a sub-model manager, our improved 3D cavity-boundary tracking, wall-zone, rubble-zone, cavity-gas, heat-transfer, and hydrology models; (5) statically linked our geomechanics model for a field test, discovering a numerically stable method for interfacing thee sub-models; and (6) calculated surrounding temperature fields for use as a geophysical monitoring signature.

Proposed Work for FY12

In FY12 we will (1) finish developing and validating our wall-zone model, 3D boundary tracker, cavity-gas model, and rubble-zone model; (2) test the geomechanics and hydrology models over a range of conditions; (3) interface all models to provide an integrated simulation capability; (4) compare simulation results against available field test data; and (5) publish our results in a peer-reviewed journal.

Publications

Nitao, J. J., et al., 2010. *An integrated 3-D UCG model for predicting cavity growth, product gas, and interactions with the host environment*. 27th Ann. Intl. Pittsburg Coal Conf., Istanbul, Turkey, Oct. 11–14, 2010. LLNL-CONF-459611-DRAFT.

Nitao, J. J, et al., 2011. *Progress on a new integrated 3-D UCG simulator and its initial application.* 28th Intl. Pittsburgh Coal Conf. Pittsburgh, PA, Sept. 12–15, 2011. LLNL-CONF-500551.

Test apparatus for measuring crystallization temperature and vapor pressure of hydrogen isotopes in nanometer-scale porous foams.

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

Robin Miles (11-SI-004)

Project Description

This project addresses 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 integration of 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 the 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.

Mission Relevance

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

FY11 Accomplishments and Results

In FY11 we (1) designed and built a system to measure the range of temperatures over which hydrogen isotopes freeze in nanometer-scale porous foams to qualify the use of liquid DT-foam layers for LIFE targets; (2) performed initial experiments that verified a depressed freezing point of about 1 K for deuterium isotopes in an approximately 360-mg/cm^3 silica aerogel foam, and began vapor pressure measurements of the hydrogen isotopes in foams; (3) began to verify that a liquid-filled foam will not distort appreciably under acceleration loads such as those experienced during injection into the fusion chamber—initial centrifugal experiments indicated no delamination of foams lining cylindrical tubes and no discernable compression of low-density toluene-filled foams under a hydrostatic loading to an equivalent acceleration of about 850 g for a DT system; (4) performed structural modeling that confirmed that the response of the capsule support membrane is critical for proper functioning of the inertial-confinement fusion target when exposed to acceleration forces; (5) tested a number of potential membrane materials at room temperature and at cryogenic conditions; (6) conducted modeling that indicated polyimide capsule-support membranes would deflect about 400 µm, making them less attractive than stiffer membranes—many carbon-based membranes such as chemical vapor deposition diamond do not exhibit the desired strength and some materials such as diamond-like-carbon exhibit a large amount of film pre-stress associated with fabrication; (7) fabricated nanometer-scale composite films and tested them as possible membrane materials—a dynamic mechanical analysis instrument was acquired to measure the effect of dynamic forces on the membranes, (8) performed thermal modeling that indicated that the temperature of the laser entrance hole window could reach very high temperature (1200 K), and investigated carbon-based window materials that would survive this temperature; and (9) began measurements of the infrared emissions of thin films at these temperatures.

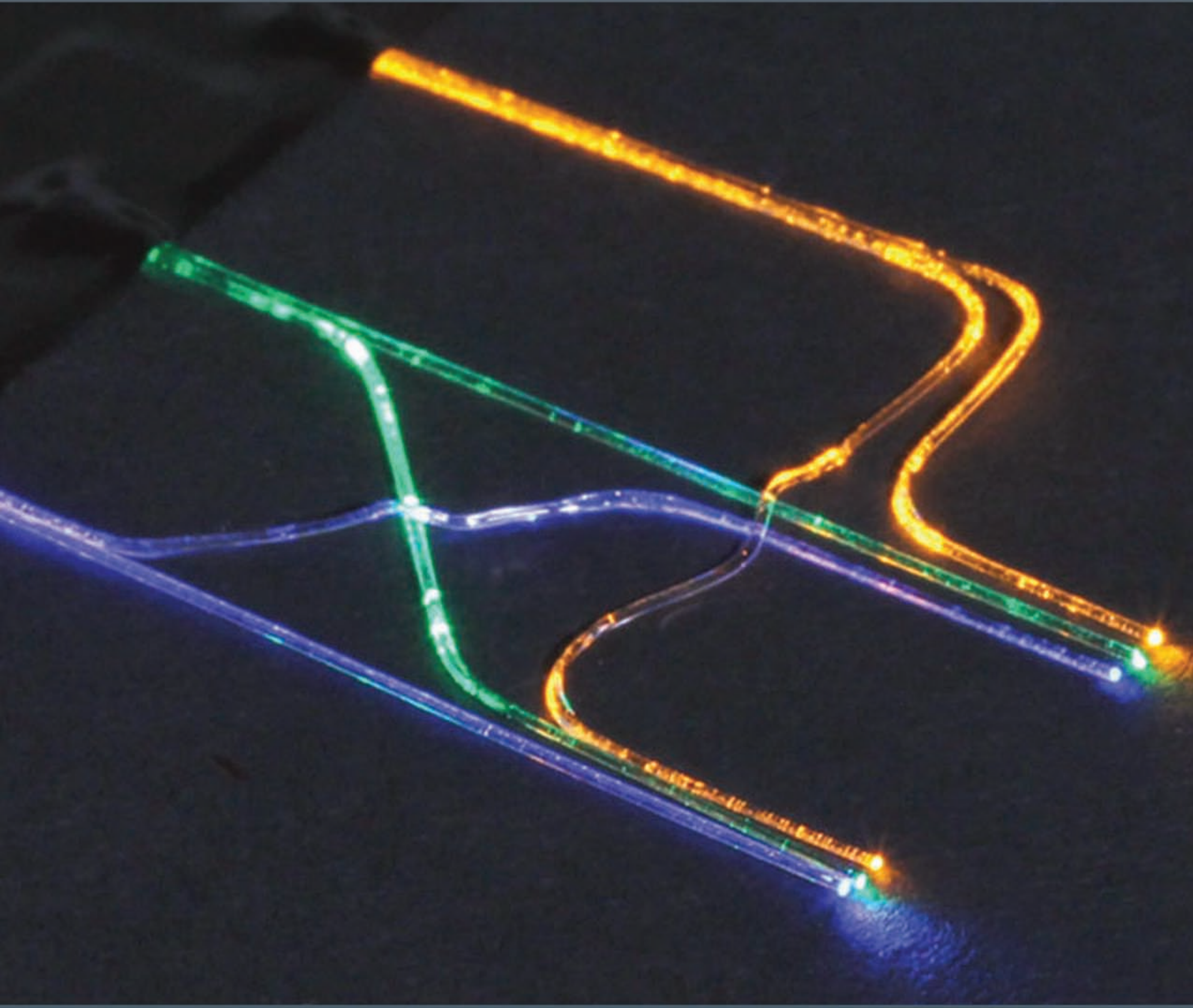
Proposed Work for FY12

In FY12 we will (1) complete experiments to test the viability of wetted foam as a target material, which includes building a specialized cryogenic centrifuge apparatus for initial acceleration tests; (2) design and build an integrated test apparatus for solid DT measurements to determine the mechanical properties of DT layers; (3) complete static measurements and begin dynamic loading of cryogenic thin-film materials to determine whether they can withstand sustained high acceleration forces; (4) develop methods and models to study the absorption, transmission, and reflectivity of ultrathin films relevant to LIFE targets; and (5) obtain target component samples made using relevant manufacturing processes to test material properties and heating effects.

Publications

Miles, R., et al., 2011. "Challenges surrounding the injection and arrival of targets at LIFE fusion chamber center." *Fusion Sci. Tech.* **60**(1), 61. LLNL-JRNL-464793.

Engineering and Manufacturing Processes



Laboratory Directed Research and Development

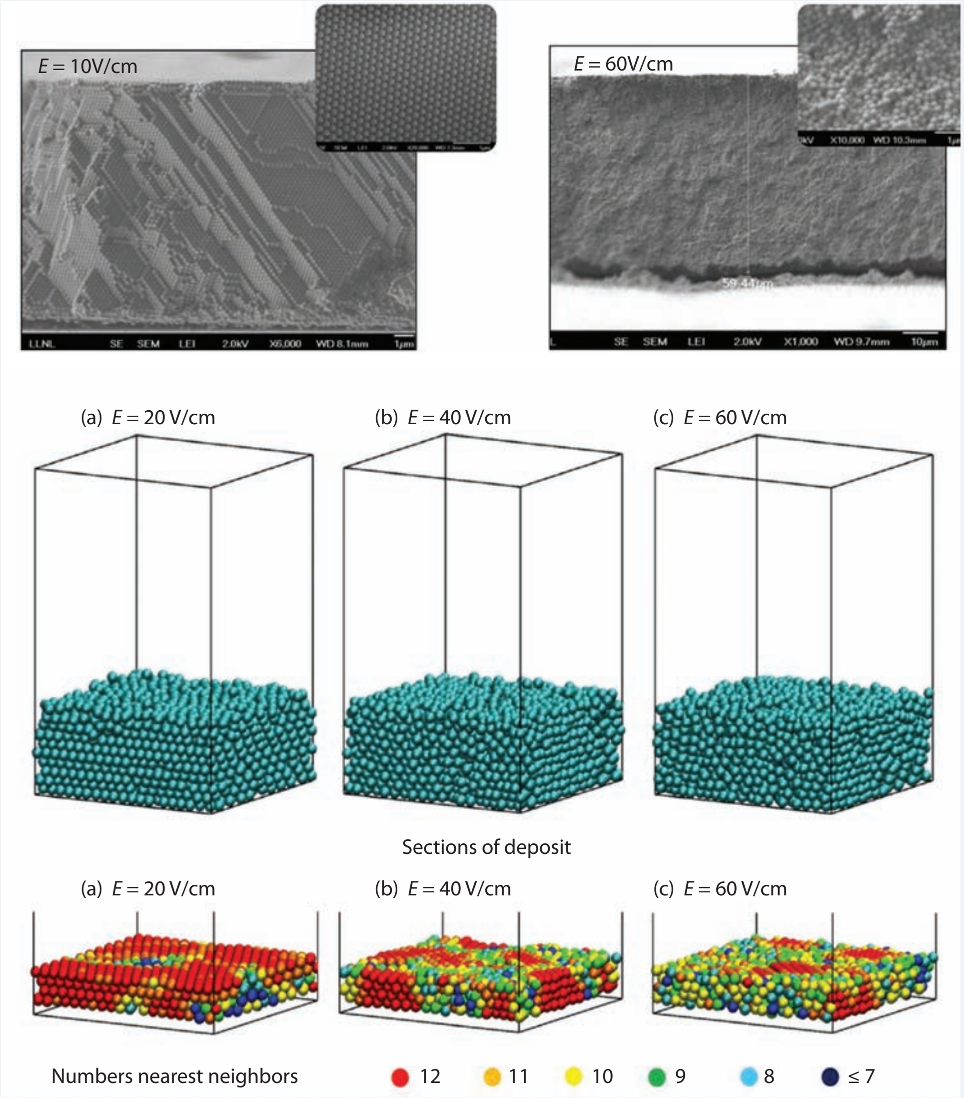
FY2011

Enabling Transparent Ceramics Optics and Advanced Armor with Nanostructured Materials Tailored in Three Dimensions

Joshua Kuntz (09-ERD-029)

Abstract

Our objective is to develop three new techniques using electrophoretic deposition to fabricate novel nanostructured materials. Functionally graded materials fabricated with gradients in composition, microstructure, or density produce enhanced bulk properties, but current techniques are limited to gradients of composition along a



Experimental (top) and modeled (bottom) results demonstrating the transition from ordered to disordered packing in electrophoretic deposition. Higher electric field strength (*E*) results in more-disordered deposits.

single axis. Using our new techniques, we plan to demonstrate new nanostructured materials including transparent ceramic optics with three-dimensional tailored doping profiles and non-cubic transparent ceramics fabricated from aligned nanometer-scale rods. To achieve this, we will develop the necessary particle chemistry, instrumentation, and protocols optimized through experiments and modeling.

Overall, we expect to develop a new nanofabrication system with three-dimensional composition control and controlled orientation of precursor material. Success in reaching these milestones will (1) dramatically expand the design of ceramic optics and immediately impact high-average-power laser design, (2) potentially create a paradigm shift in the field of optical materials by producing transparent ceramics from non-cubic materials, (3) provide an empirically validated deposition model for future designs and applications, and (4) produce high-impact publications on the fabrication technique as well as the new materials and structures from each application.

Mission Relevance

This project will provide a new nanofabrication capability that supports the Laboratory's national and energy security missions. This new technology will enable novel composites and structures using readily available precursor materials, which will enhance ongoing Laboratory efforts in ceramic optics, provide future capability in fabricating inertial-fusion energy targets and enhanced ceramic armor, and potentially enable super-lattice substrates from novel materials for radiation detection and other national security applications.

FY11 Accomplishments and Results

In FY11 we (1) successfully deposited a two-dimensional extruded pattern onto a photolithographically patterned metal electrode with a resolution lower than 7.5 µm, with the sharp edges of the deposit indicating that a much finer resolution is possible; (2) developed and implemented a Brownian dynamics model for electrophoretic deposition; (3) validated the model through correlation with experimentation on the transition of monodisperse spheres from ordered to disordered; (4) developed multiple synthesis routes to near-monodisperse fluorapatite nanorods and demonstrated alignment of the rods in a 300-V/cm electric field; and (5) demonstrated our electrophoretic deposition technique by fabricating functionally graded transparent ceramic structures with dopant concentrations varied as a function of height. This project has significantly advanced the state of the art in electrophoretic deposition, enabling the technique's use in fabricating tailored transparent ceramics and bringing us closer to the goal of transparent ceramics made from non-cubic materials. Further development and implementation of this technique is being supported by the Defense Advanced Research Projects Agency and Air Force Special Operations Command.

Publications

Kuntz, J. D., et al., 2011. *Colloidal crystal assembly by electrophoretic deposition—experiments and modeling.* Technologies for Future Micro–Nano Manufacturing, Napa, CA, Aug. 8–10, 2011. LLNL-POST-491953.

Pascall, A. J., et al., 2011. *Electrophoretic deposition for the fabrication of materials with designer microstructure via dynamic electrodes and electric field sculpting.* 2011 MRS Fall Mtg. and Exhibit, Boston, MA, Nov. 28–Dec. 3, 2011. LLNL-PRES-515011.

Pascall, A. J., et al., 2011. *Functionally graded, nanostructured materials via patterned electrophoretic deposition.* 4th Intl. Conf. Electrophoretic Deposition (EPD 2011), Puerto Vallarta, Mexico, Oct. 2–7, 2011. LLNL-PRES-500471.

Worsley, M. A., et al., 2011. *Patterned electrophoretic deposition: A bottom-up approach to functionally graded materials.* 2011 MRS Fall Mtg., Boston, MA, Nov. 28–Dec. 2, 2011. LLNL-POST-516711.

Maskless, Low-Cost, High-Performance Polymer Waveguides

Eric Duoss (09-ERD-057)

Abstract

Our proposed study will explore a new maskless material fabrication approach being developed at the University of Illinois that has multiple technological applications in composites, microfluidics, and photonics. This technique, direct ink writing, will enable new capabilities in the synthesis of complex structures from a variety of materials. As an initial proof-of-concept demonstration, we will use direct ink writing to fabricate a new generation of high-performance polymer optical waveguides with high optical transmission. We will collaborate directly with the development group at Illinois to develop and optimize the fabrication technique for our desired materials, surface roughness, and geometry specifications.

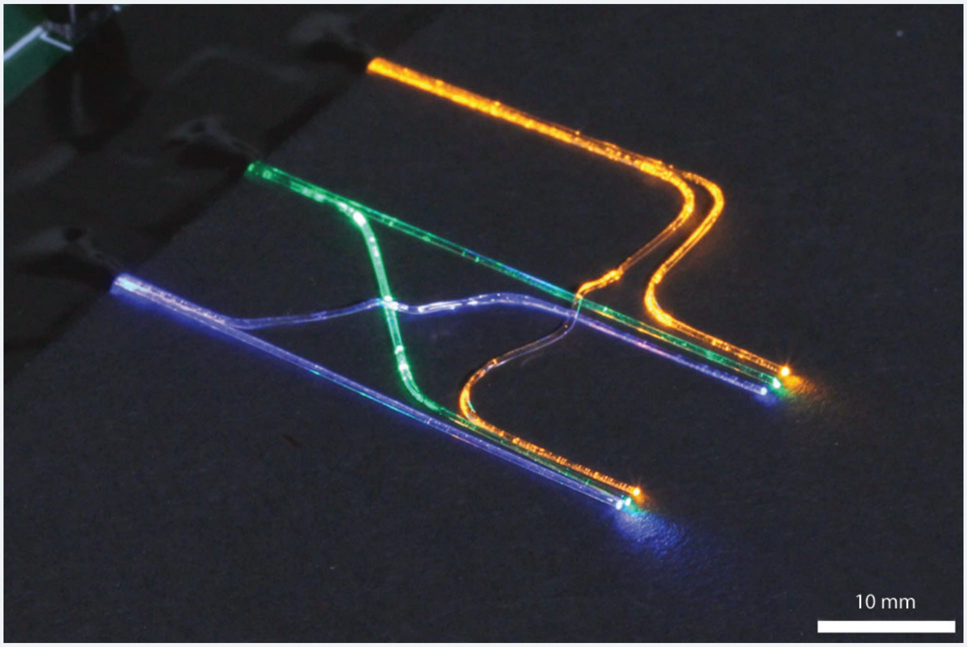
Throughout this study we will work with our collaborators at the University of Illinois to develop the necessary protocols to fabricate polymeric waveguides to achieve optical transmission losses below 0.1 dB/cm. These losses are strongly correlated with the material, surface roughness, and geometry of the waveguide. By optimizing direct ink-writing fabrication for a selected set of polymers, we expect to achieve this low-loss level. We also expect to demonstrate this with multilayer waveguides patterned into compact three-dimensional structures. This optimization process will enable future applications requiring complex three-dimensional structures and materials not compatible with standard microfabrication techniques.

Mission Relevance

The broad range of structures and filament cross sections achievable with direct ink writing and the diverse materials compatible with the process will enable many applications of interest to LLNL in support of several missions, including national and homeland security as well as energy security. Examples of potential applications include self-healing materials, inertial-fusion energy targets, three-dimensional microfluidics, and photonic crystals. Direct ink writing may be ideal for fabricating low-cost polymer waveguides with low optical losses, which are also of interest to numerous Livermore photon science efforts in defense, energy, and basic science.

FY11 Accomplishments and Results

We demonstrated the patterning of hybrid organic–inorganic optical waveguides in straight, curved, and out-of-plane configurations with the direct-writing assembly of photo-curable liquid core and viscoelastic fugitive shell inks. The hybrid organic–inorganic core fluid was encapsulated within a viscoelastic shell composed of an aqueous triblock copolymer solution. The fugitive shell served as a sacrificial support for the core fluid before curing with ultraviolet light. These materials were co-extruded in the desired core–shell configuration using a custom print head consisting of two cylindrical nozzles aligned coaxially. The core fluid and viscoelastic fugitive ink shell were loaded into separate reservoirs and printed simultaneously by applying air pressure to each reservoir. The printed waveguides exhibited nearly cylindrical morphology and low optical loss throughout the visible spectrum—the lowest optical loss observed was approximately 0.1 dB/cm. This successful project has thus produced a new approach that enables printing of optical waveguide networks coupled to



An optical waveguide network composed of six printed waveguides coupled with three light-emitting diodes that distribute colored light with minimal crosstalk.

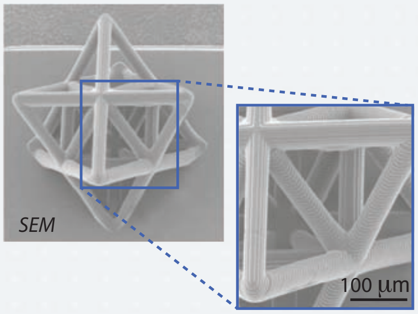
multiple light-emitting diodes, with gaps at the crossover points between waveguides to reduce crosstalk. This innovative fabrication approach can be used to create waveguide networks for integration with high-bandwidth, next-generation optical systems and optical sensor arrays. This new capability is already being used in work supporting LLNL missions in defense, energy, and basic science.

Publications

Lorang, D. J., et al., in press. "Photocurable liquid core–fugitive shell printing of optical waveguides." *Adv. Mater.* LLNL-JRNL-488631.

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

Christopher Spadaccini (11-SI-005)



The smallest known fabrication of the "octet truss," an ultrahigh-stiffness, low-weight structure fabricated in this project using projection micro-stereolithography.

Abstract

Our goal is to fundamentally understand and develop, from the ground up, new micro-manufacturing techniques applicable to a variety of materials such as polymers, metals, and ceramics. These techniques would enable the production of three-dimensional (3D) mesoscale geometries with micrometer-scale precision and scalable 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 and wasteful in energy and material. In addition, grain size becomes problematic and material handling is difficult. This project will revolutionize the way we manufacture parts for advanced fusion-class laser system targets as well as for myriad other devices.

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 3D mesoscale structures with microscale architectures and sub-micrometer precision. These processes will be revolutionary to manufacturing and have a broad impact because of compatibility with a wide range of materials, rapid translation from computer model to fabricated component, and scalability 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 of those attainable with bulk materials processed via traditional synthesis methods. Success in meeting our proposed objectives will both illustrate capabilities of the process and demonstrate novel materials and structures 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 3D manufacturing process combined with multiple materials enables new design concepts that have not been possible and offers cost reduction for advanced fusion-class laser system targets and rapid turnaround for new target designs in support of Livermore missions in stockpile science, as well as reducing or countering threats to national and global security.

FY11 Accomplishments and Results

We (1) used projection micro-stereolithography to fabricate components with nanometer-scale particles embedded within a polymer matrix, thus expanding the technique's usable material set to include metals, ceramics, and semiconductors; (2) used the technique to also fabricate heterogeneous components with two different polymers in the same structure by integrating microfluidics into the system; (3) added light-scattering physics to our micro-stereolithography process model to predict system performance with nanoparticles suspended in the photosensitive liquid; (4) fabricated the smallest known "octet truss"—a high-stiffness, low-density structure; (5) improved our electrophoretic deposition technique to fabricate multi-scale components with features as small as approximately 7 µm, and demonstrated the first-ever light-directed electrophoretic deposition of particles—a key capability for generating 3D structures with this technique; (6) used electrophoretic deposition to co-deposit thermite materials, achieving radically improved power density upon deflagration; (7) developed several new printable materials for direct ink writing such as slioxane, aluminum nanoparticle, and copper oxide inks, and fabricated structures with direct ink writing at multiple length scales; (8) combined direct ink writing with electrophoretic deposition to generate unique structures; and (9) demonstrated the ability to print energetic thermite materials.

Proposed Work for FY12

In FY12 we will (1) continue to advance our fabrication capabilities by further broadening the usable material set, improving resolution, reducing feature size, and increasing fabrication speeds; (2) focus on fabricating unique structures and materials such as high-stiffness, low-density microstructures, binary thermites with improved and controlled energy propagation via microstructural design, and 3D mesoscale components from single materials, in preparation for fabricating these same shapes with multiple materials in FY13; and (3) combine fabrication techniques to enable new structures, among other objectives.

Publications

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Kuntz, J. D., 2011. *Colloidal crystal assembly by electrophoretic deposition—experiments and modeling.* LLNL-POST-491953.

Pascall, A. J., et al., 2011. *Electrophoretic deposition for the fabrication of materials with designer microstructure via dynamic electrodes and electric field sculpting.* 2011 MRS Fall Mtg. and Exhibit, Boston, MA, Nov. 28–Dec. 2, 2011. LLNL-ABS-489152.

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Worsley, M. A., et al., 2011. *Patterned electrophoretic deposition: A bottom-up approach to functionally graded materials.* 2011 MRS Fall Mtg. and Exhibit, Boston, MA, Nov. 28–Dec. 2, 2011. LLNL-ABS-487832.

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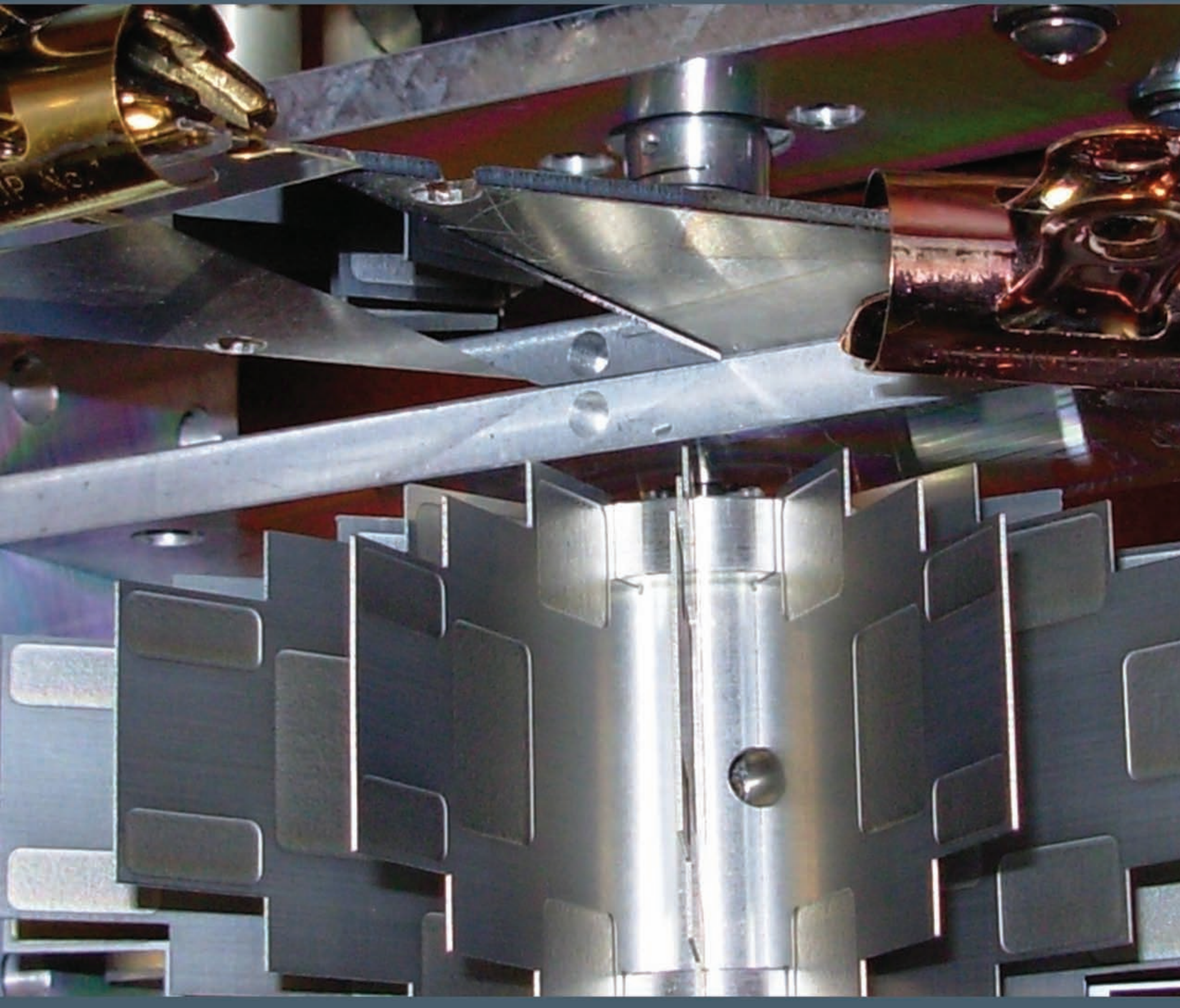
Materials Science and Technology



Laboratory Directed Research and Development

FY2011

Radiation-Tolerant Materials

Michael Fluss (09-SI-003)

Abstract

One challenge specific to sustainable nuclear energy is developing radiation-tolerant materials for encapsulating reactor fuel as well as structural components of nuclear energy systems that can maintain required properties despite neutron-induced changes. Materials with nanometer-scale dispersed particles appear to be good candidates for radiation tolerance, but significant gaps exist in our understanding of the properties of these metals. Our goal is to understand the scientific principles that govern the behavior of this class of materials, to explore the limits of their radiation tolerance, and to discover means by which this tolerance can be extended through materials refinement or development of new material systems.

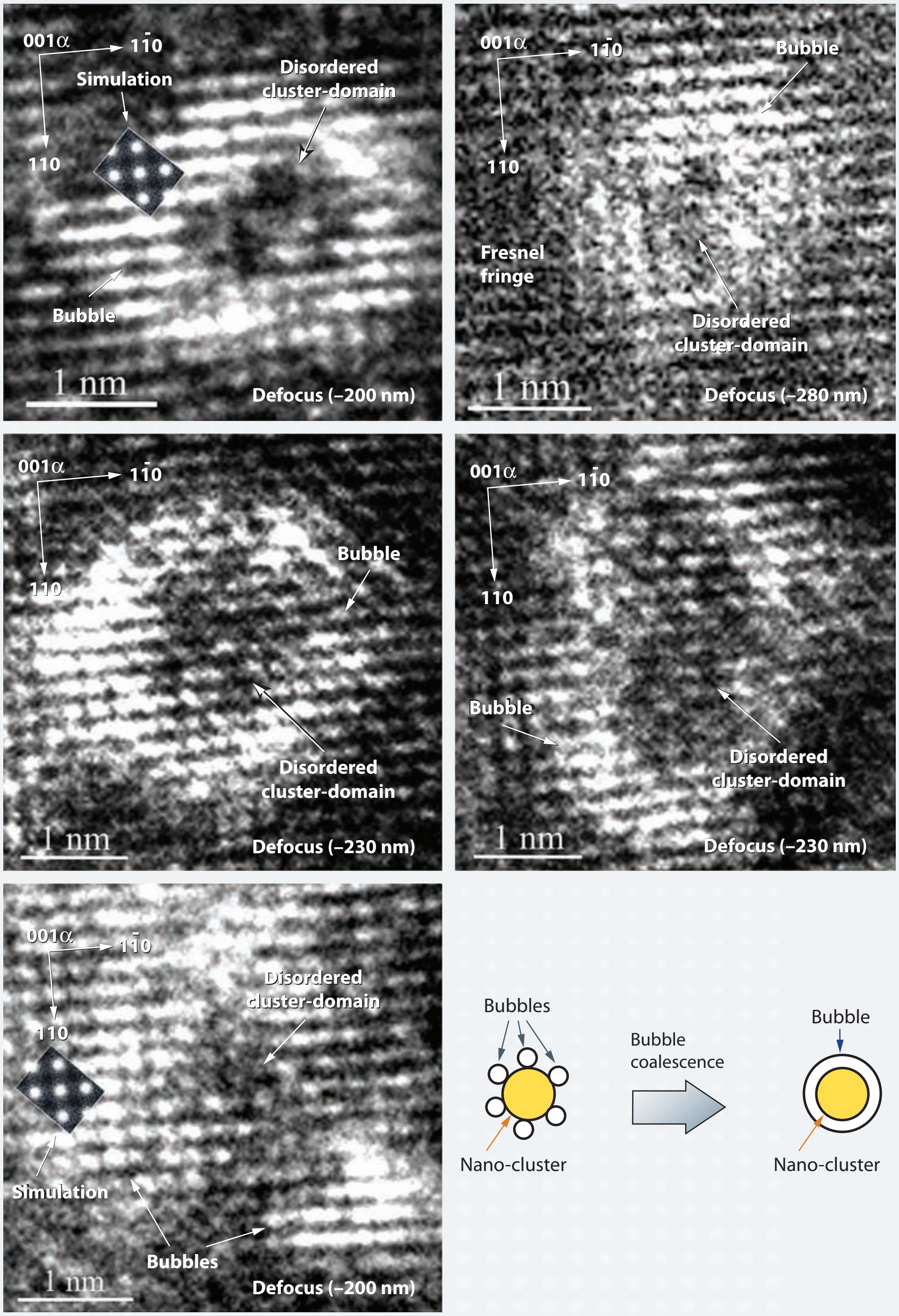
We hope to discover the fundamental materials science necessary to realize radiation-tolerant materials that can maintain required operational specifications despite neutron-induced changes. The behavior of nanoscale-structured materials through modeling validated by sequential and simultaneous ion-beam experiments with heavy ions, helium, and hydrogen will bridge length scales from microscopic to macroscopic and timescales from days to decades in describing advanced radiation tolerance. Key deliverables are both experimental radiation tools for the discovery of advanced radiation-resistant materials and simulation and modeling tools for the design, development, and deployment of advanced nuclear energy systems for fission or fusion.

Mission Relevance

Our proposed research supports Laboratory missions in both energy and national security. Elimination of reprocessing in the fuel cycle would reduce cost and increase energy security. Materials for the ultra-deep burn up of fission fuel are key to using all the potential energy in uranium. This simple idea is the path to a sustainable nuclear fuel cycle that lowers the barriers of waste disposition, decreases the threat of proliferation, incorporates safeguards by design, and enhances economic competitiveness through sustainability and energy independence.

FY11 Accomplishments and Results

We continued our analysis and: (1) developed new techniques to simulate the radiation damage of nuclear energy materials from neutrons by using multiple ion beams to produce displacement damage simultaneously with the injection of helium or hydrogen; (2) determined that the consolidation and heat treatment of mechanically alloyed Fe(Cr)–YO_3 powders lead to a crystallization of the oxide through an amorphous precursor; (3) determined the critical cavity diameter for void growth in an iron–chromium alloy to be 2.5 nm—important data for modeling purposes; (4) discovered that below a critical size of 1 nm, the recrystallization of dispersed oxide in oxide-dispersion-strengthened steels is not energetically favored—instead, the resulting molecular clusters are efficient sequesters of helium and play a major role in helium management by inhibiting the growth of large voids; and (5) confirmed the accelerated growth of cavities in iron–chromium alloys when hydrogen is implanted simultaneously with displacement damage induced by helium and iron ions. This project has provided the scientific basis for a research and development plan for the experimental determination of advanced structural steels to be used for the first wall and blanket of an inertial fusion energy engine.



High-resolution transmission electron microscopy images of helium bubbles associated with cluster domains of various shape. In each under-focused image, helium bubbles appear as white contrast surrounded by a dark Fresnel fringe (top and middle rows). The image in the bottom row shows the trapping of several individual bubbles at a disordered cluster domain, suggesting that the appearance of the cluster core and bubble shell is the result of the coalescence of small bubbles, as conceptualized in the illustration.

Publications

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Nanosecond Characterization of Dynamic Void Evolution in Porous Materials

Anthony van Buuren (09-ERD-002)

Abstract

Very little is currently understood about how the morphology of nanostructured foams responds to shock conditions. Existing models of porous materials are believed to be well grounded experimentally at very low and very high levels of porosity—however, the models make inconsistent assumptions and none have been verified in situ. We propose to characterize the morphology of nanometer-scale foams under shocked conditions and use these results to validate models of void evolution in the materials. We will quantify dynamic structural changes with a state-of-the-art, ultrafast,

small-angle x-ray scattering system. Ultimately, we hope to obtain a predictive capability for these materials under extreme conditions as a function of initial structure and compression dynamics.

We will leverage LLNL's recent successful capability of measuring small-angle x-ray scattering using a single x-ray pulse, which enables the use of pump–probe experiments to measure structural changes in situ during a shock. We will apply these methods to nanofoams for the first time and expect to answer a major question: How reliable are existing theoretical models used to describe the compression of foams?

Mission Relevance

Key deliverables of this project include the ability to characterize the evolution of foam microstructures under shock conditions and the validation of theoretical models, both of which are critical for future laser targets for stockpile stewardship and inertial-confinement fusion experiments, which support LLNL's national and energy security missions.

FY11 Accomplishments and Results

In FY11 we measured dynamic x-ray scattering on carbon aerogels as a function of density as planned, and also explored the effects of drive pressure and various pore structures. Using a drive laser, we produced compressive loading in aerogel samples by direct ablation using an aluminum ablator (lower drive pressure) on the carbon aerogel and by tamped ablation (higher pressure) using a quartz tamper placed on top of the aluminum. We found that the small-angle x-ray scattering signal at early time of less than 50 ns appeared surprisingly unchanged, which is consistent with propagation of a shockwave that is short relative to the sample thickness and released to a structure similar to the original aerogel. At later times, the total small-angle x-ray scattering intensity dropped, consistent with a decrease in the number of scatters and possibly indicative of break-up of the pore structure late in release. The fact that the larger-length-scale feature decreases at early time and disappears at late time greater than 80 ns is consistent with pore structure collapse. Tamped and directly ablated foams behave nominally the same; however, the tamped samples results occur at earlier time. Decreasing density of the carbon aerogel caused the change in small-angle x-ray scattering to be shifted to a much later time, perhaps because of weaker coupling of the laser drive. We are discussing Laboratory programmatic use of ultrafast small-angle x-ray scattering to quantify dynamic changes in carbon condensation during high-explosive detonation and to determine reliability of existing theoretical models used to describe detonation of explosives, in particular the carbon cluster condensation.

Publications

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Understanding the Surface Properties that Lead to Optical Degradation in High-Fluence, High-Average-Power Optical Materials

Jeffrey Bude (09-ERD-003)

Abstract

Laser-induced surface damage to optical materials limits the maximum operating fluence of fusion-class laser systems. Moreover, surface properties that limit optical performance are not well understood. We will develop a fundamental understanding of these properties in high-fluence, high-average-power optical materials. A suite of sensitive, high-resolution optical techniques and advanced computational modeling will be employed to study the electronic and optical properties of dielectric surfaces at the nanometer scale. We intend to clarify the links between surface properties, laser-induced surface damage, and the effects of long-term, high-fluence optical fluxes on damage resistance in optical materials.

We expect to (1) determine the physical origin of ultrafast photoluminescence from defects associated with damage in silica and determine whether fast photoluminescence is a predictor of damage in other optical materials using confocal, time-resolved photoluminescence imaging; (2) determine the effects of long-term, high-fluence optical exposure on surface properties and damage resistance of optical materials; and (3) clarify the links between material properties, surface nanostructure, electronic structure, optical absorption, and high-fluence optical damage in optical materials. This work will guide the development of damage-resistant optics and help to understand the limits of optical materials operated under high fluence, as well as understand high-fluence, high-average-power conditions.

Mission Relevance

This research directly addresses stockpile stewardship and fusion energy challenges by optimizing large advanced laser systems. It will serve to establish science-based rules for optics reliability prediction, improve damage diagnostics, and suggest pathways to increase damage resistance in optical materials. More broadly, understanding defect-assisted absorption and material modification is a frontier problem in condensed-matter physics.

FY11 Accomplishments and Results

In FY11 we (1) completed analysis of optical stress effects on silica, potassium dihydrogen phosphate, and calcium fluoride surfaces, including tests of up to 40,000 subdamage fluence shots, finding small changes in surface roughness, defect density, and transmission but no reduction in damage thresholds—these effects were found to occur at high total fluence even at low per-pulse energy; (2) built and tested a system to measure high-fluence damage precursors up to 150 J/cm^2 and found that for good silica optics, precursors were isolated and distributed up to 30 µm apart, indicating that the intrinsic surface threshold is beyond 150 J/cm^2;

(3) determined that infrared laser heating can reduce precursors, but its effects are eliminated by typical surface processing, including acid etching; (4) experimented with antireflective coatings and found them to reduce damage resistance above 30 J/cm^2—most significantly for etched scratches—but to have little effect at lower fluences; (5) correlated quasi-continuum photoluminescence to defect density and built models for this correlation; and (6) confirmed our model for absorption fronts in silica up to four orders of magnitude in intensity and experimentally estimated bulk absorptivity in silica up to 6000 K. In summary, this project helped develop an advanced mitigation process for silica optics that has dramatically reduced laser damage in lenses at the National Ignition Facility and has added to our understanding of both laser–matter interactions, materials under extreme conditions, and the optical properties of defects in transparent solids. Future research efforts would include pushing the optics to higher per-pulse fluences and investigating the response of optical materials under the multipulse environments of laser-ignition fusion energy.

Publications

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Multi-Resolution Adaptive Monte Carlo for Microstructure Simulations

Vasily Bulatov (09-ERD-005)

Abstract

The objective of our project is to extend the time and length scales of material microstructure simulations to those relevant for engineering applications. Our development will focus on atomistic (lattice) Monte Carlo as a general and accurate framework and will rely on iron–copper and iron–chromium binary alloys as test-bed material systems. Of principal interest is the kinetics of diffusive-phase transformations, including nucleation and growth of precipitates, ordering, segregation, and the effects of continuous and pulsed irradiation drive on these phenomena. Several novel methods will be developed that, in combination, will significantly enhance the computational efficiency of microstructure simulations while maintaining or improving their accuracy.

We expect to develop a novel computational approach of multi-resolution adaptive Monte Carlo that will extend the time and length scales for accurate simulations of the kinetics of microstructure evolution in binary alloys. Eventually, we will extend this approach to more complex materials such as those used in nuclear reactors. Unlike existing phenomenological methods, our multi-resolution approach is entirely self-consistent—on its fully refined level it reduces to a detailed atomistic representation. At the same time, our simulation approach links the fundamental mechanisms of atomic diffusion directly and seamlessly to alloy microstructure evolution on scales relevant for current and future engineering applications.

Mission Relevance

Our new simulation capability can find multiple applications in several existing and emerging mission areas in which behavior of materials away from equilibrium is important, including national and energy security. For example, our new efficient methods and computer codes can be employed for computational extrapolation of material damage observed in an accelerated radiation test to the expected radiation resistance of the same material over its work life in nuclear reactors.

FY11 Accomplishments and Results

We (1) completed an exact, accelerated kinetic Monte Carlo algorithm practically applicable to Markov models with a sparsity of connections (transitions) from individual Markov states; (2) attempted two different implementations based on the incremental expansion of a trapping subset of Markov states—specifically, the numerical solution of a projected master equation and the incremental graph reduction followed by randomization—finding that the latter implementation allows us to overcome, for the first time ever, trapping basins spanning several million Markov states; and (3) used the new method to simulate anomalous diffusion on a two-dimensional substrate and the kinetics of diffusive first-order phase transformations in binary alloys. We verified that, depending on temperature and alloy super-saturation conditions, speedups of up to several orders of magnitude are possible without compromising simulation accuracy. This successful project has paved the way to a new class of Monte Carlo algorithms in which random walks in the space of Markov states are performed whenever the walks are expansive (non-returning) and in which trapping, when detected, is overcome with a solution of the master equation on the trapping basin. This unique capability could have applicability to a number of LLNL missions.

Publications

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Collection of Refractory Debris from the National Ignition Facility for Stewardship-Relevant Measurements

Dawn Shaughnessy (09-ERD-026)

Abstract

Solid debris collected from National Ignition Facility (NIF) shots will be used to measure cross sections of radiochemical detectors for nuclear testing. Such measurements will significantly reduce uncertainties in interpreting radiochemical data. We propose to develop a system for collecting solid debris samples at fusion-class lasers for radiochemical analysis and measurement of stockpile stewardship cross sections. Distribution of the debris will first be determined through simulation and experiment. Next, several designs will be tested, and a priority list of stewardship-relevant measurements will be determined. The methodology will be benchmarked

through measurement of yttrium cross sections, but ultimately a variety of radiochemical detectors will be evaluated.

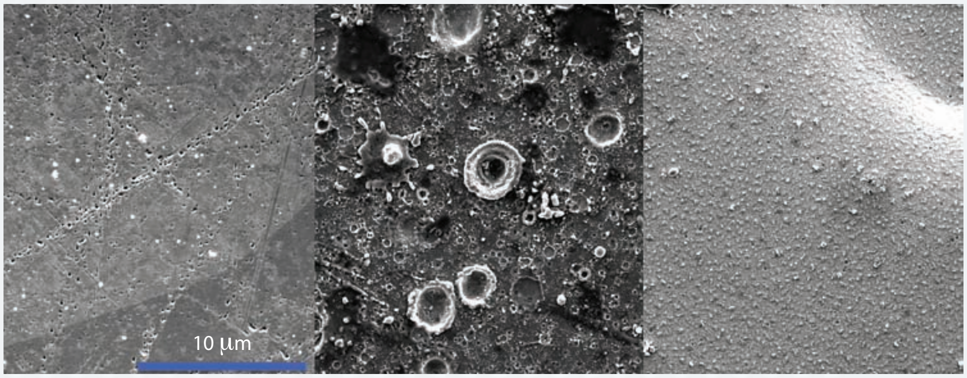
If successful, this project will yield a prototype solid collector that can be fielded during early NIF shots for testing. If, as we expect, this new device significantly improves the acquisition and understanding of radiochemical data, it would then be a candidate for future program support and deployed in the NIF chamber for routine radiochemical measurements. Initially, we will consider several collector designs, then select the most promising ones and conduct testing at NIF to evaluate collection efficiency and ease of insertion and extraction from this particular chamber. Once a prototype has been fully developed, measurements of the yttrium cross section will begin as ride-along experiments on other NIF shots.

Mission Relevance

This project supports LLNL's national security mission by reducing thermonuclear device uncertainties—a major goal of stockpile stewardship—by enabling solid collection to reliably measure cross sections, validate code calculations, and lower uncertainties in the interpretation of radiochemical data. In addition, the measurements on NIF that this project enables will help recruit staff in the core competency area of radiochemistry.

FY11 Accomplishments and Results

In FY11 we (1) concluded that passive collectors work for solid collection but were unable to complete our design of a larger area, ride-along collector mounted on the diagnostic instrument manipulator because of insufficient engineering support; (2) designed experiments for ignition and non-ignition shots for measuring yttrium cross sections; (3) investigated the feasibility of various collector insertion and extraction options and concluded that collectors mounted on the diagnostic instrument manipulator are the most straightforward; (4) began analysis of material test and ablation test samples—results indicated that aerosol-assisted ablation was not required for effective debris collection and initial analysis of material test samples show that tantalum and vanadium make the best collectors; (5) completed sensitivity analysis and determined that first-order (n,2n) reactions on yttrium can be measured to 5% while second-order reactions can be measured to about 15%; and (6) designed



Pre-shot Post-shot (0-0, Polar) Post-shot (90-78, Equator)

Comparison of niobium metal pieces fielded during National Ignition Facility shots: pre-shot piece (left), post-shot fielded on the pole of the diagnostic instrument manipulator (middle), and post-shot fielded on the equator of the manipulator (right). There are obvious differences between the polar and equatorial samples in terms of type of debris collected and amount of shrapnel encountered.

a tritium removal system using glow discharge. The results from this project showed that a ride-along, passive collector can collect solid debris from a NIF shot with a reasonably high efficiency to perform cross-section measurements with an uncertainty rivaling those from calculated cross-section sets. The NIF is now supporting solid debris collection, which means that experiments designed to diagnose capsule performance will begin in the following year with cross-section measurements to be performed after the ignition campaign.

Publications

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Magnetorheological Finishing for Large-Aperture High-Fluence Optical Applications

Joseph Menapace (09-ERD-049)

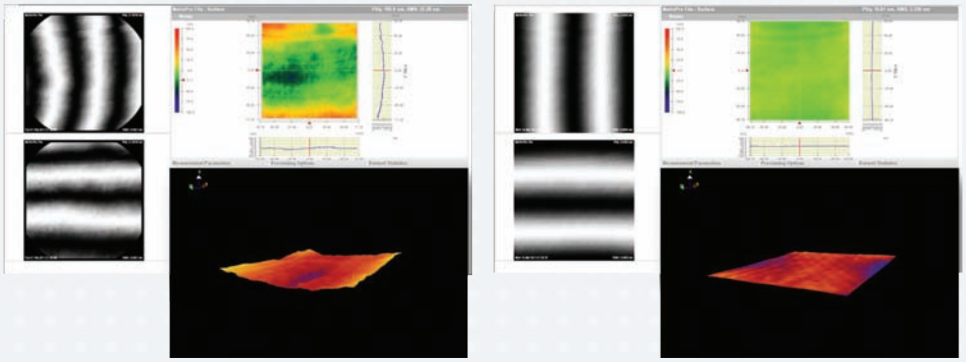
Abstract

Our goal is to understand the magnetorheological finishing (MRF) processes governing the deterministic removal of material from, and achieving figure control on, optical surfaces and how the processes affect the manufacture of precise, high-fluence optics for inertial-confinement fusion and other high-power laser systems. Specifically, we will (1) develop an understanding of the details of MRF material removal on free-form optics so that state-of-the-art protocols can be developed for meter-size optics, (2) develop new polishing slurries and protocols to polish novel crystalline optical materials that are important for inertial-confinement fusion laser systems, and (3) investigate MRF to precisely polish out or mitigate flaws and damage on high-fluence optical materials.

This study will greatly enhance the scientific knowledge of MRF polishing, which has applications in precision optics, space science, and semiconductors. Specifically, the ability to deterministically finish an optical surface using a sub-aperture tool will allow optical glass fabricators to improve the figure of an optic in a less iterative and more repeatable, economical, and deterministic manner. We will also enhance the understanding of advanced sub-aperture polishing that can be used to remove damage and compensate for short-range internal flaws in optics artifacts that limit the suitability of small- and large-aperture optical materials in high-fluence, high-repetition-rate laser applications.

- Pre-MRF TWF correction interferogram
- PV: 175.4 nm PV_q (99%) 105.9 nm
- rms: 22.2 nm

- Post-MRF TWF correction interferogram
- PV: 34.6 nm PV_q (99%) 16.0 nm
- rms: 3.3 nm



- Scale up from 1/81 to 1/9 scale with respect to NIF-sized optics
- Improvement of 6x in TWF PV and 7x in rms

For our research into the manufacturing of precise, high-fluence optics for inertial-confinement fusion and other high-power laser systems, we successfully scaled up magnetorheological finishing polishing for potassium dihydrogen phosphate crystals from 51 mm to 152 mm.

Mission Relevance

This project supports the Laboratory's missions in national and energy security by expanding the knowledge base for fabricating optical components critical to the high-energy, high-power, fusion-class laser systems used to validate complex coupled-physics computer simulations, including codes used in lieu of nuclear stockpile testing.

FY11 Accomplishments and Results

We (1) successfully transitioned our MRF techniques for damage removal and the final polishing of fused-silica optics to our large-aperture MRF tool, verifying that the damage-removal protocols reduced processing time by a factor of three over previous techniques and that final finished optics had surface figures below 100 nm, peak to valley; (2) developed advanced software to provide for high figure control during MRF processing; (3) finished a demonstration large-aperture lens using MRF for damage performance and optical figure control, using our protocols to validate the ability of the processes to fabricate ultraprecise, damage-resistant optics; (4) conducted lifetime testing of the lens on the National Ignition Facility laser system; and (5) successfully scaled up MRF polishing for potassium dihydrogen phosphate crystals from 51 mm to 152 mm with ultraprecise figure control, as exhibited by surface figures of less than 35 nm, peak to valley—the next step would be to scale up our technique to 430-mm optics. Overall, this project enabled us to advance our understanding of MRF finishing technology and to develop and field new techniques for expanding its use in high-energy laser and novel optic fabrication. The developed technology continues to perform without issue, establishing that the advanced finishing protocols can produce damage-resistant optics for high-fluence laser applications and therefore is ready for production facilities, where it can be applied to state-of-the-art optical fabrication techniques for NIF and other high-power laser systems.

Publications

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Characterization of Tritium Uptake and Release by Inertial-Confinement Fusion Reactor Materials

James Fair (09-ERD-050)

Abstract

We will quantify the uptake and release of tritium by laser inertial-confinement fusion reactor materials for a range of surface preparation, exposure, and decontamination conditions. Data from this project will form the basis for designing and implementing tritium decontamination processes by systematically exploring its uptake and release in laboratory-scale packed-bed reactors and full-scale fusion-class laser optic enclosures. Key outputs will be mass transfer coefficients, other rate constants, and adsorption isotherms. The study will also characterize local electronic sensing as a method for directly measuring surface tritium activity and seek measures to maximize the availability of fusion facilities, significantly decreasing operating costs.

We expect to create the ability to (1) predict tritium uptake rates under known exposure conditions, (2) predict tritium release rates under controlled decontamination conditions, and (3) accurately and precisely measure surface tritium concentration on inertial-confinement fusion reactor components using direct electronic sensing devices. These results will form the technical basis upon which reliable and safe tritium decontamination subsystems and processes for laser inertial-confinement fusion reactors can be efficiently developed, implemented, and controlled.

Mission Relevance

This project supports LLNL's national security mission by providing experimental data from fusion-class laser systems to validate complex coupled-physics computer simulations for stockpile stewardship. In addition, our research will substantially increase our understanding of the behavior of tritium and tritiated compounds as they interact with optical components associated with large inertial-confinement fusion laser systems.

FY11 Accomplishments and Results

In FY11 we determined that for all materials studied except plastic (i.e., steel, aluminum, and glass), water-induced tritium release proceeded by rate-limiting mechanisms involving two dominant, concurrent surface reactions. The fastest surface reaction was found to follow first-order kinetics, while the second slower overall reaction was found to involve a chain mechanism. We determined further details of the mechanisms by measuring their thermal activation characteristics. At high surface-water coverage, the fastest reaction showed little thermal activation, while at low water coverage the reaction became strongly activated. The slower reaction showed only slight thermal reaction activation, but strong effects from thermally modulated water-surface coverage. Tritium release from plastic was found to be dominated by direct reaction with absorbed water and desorption of tritium oxide. We updated our numerical codes to include the new reaction models and extended the codes to allow simulation of systems composed of multiple materials. In addition, we implemented a new plug–flow version of the numerical-system-level model and developed an entirely new code for simulation of unsteady transport in arbitrary piping networks. With this project we developed a simple, unified, conceptual and calculation framework for the description of tritium transport to and from surfaces in enclosed systems containing water vapor. Research in this area will continue under program support from the National Ignition Facility and associated National Ignition Campaign.

Publications

Fair, J. E., and W. T. Shmayda, 2011. "A model for removal of surface-bound tritium using humid air." *Fusion Sci. Tech.* **60**(3), 1045. LLNL-CONF-457361.

Mitigation of Damage to Multilayer Mirrors

Christopher Stolz (09-ERD-051)

Abstract

The objective of our proposed study is to develop methods to arrest damage growth in optical interference coatings. This damage is caused by laser-induced fracture and absorption from sub-stoichiometric coating materials. Advancements in increasing mirror fluence survivability have focused on material selection, absorption and defect reduction, and laser conditioning. Currently, state-of-the-art mirror coatings are limited by a few growth sites, suggesting the possibility of a localized mitigation solution. We will explore laser- and mechanical-based mitigation methods coupled with an effort to understand thin-film properties, advanced electric-field modeling, and downstream-modulation modeling. We will validate site stability with laser damage testing and microscopy.

We expect to determine a thin-film mitigation process that will increase the resistance of laser transport mirrors to beam damage. Electric-field modeling will determine the proper film boundary to minimize electric-field effects. Beam-modulation modeling will determine the proper mitigation geometry for a reflective component. Once this process is established, it will be validated on mirrors suitable for use on fusion-class lasers. A second phase of this research will be devoted to reducing

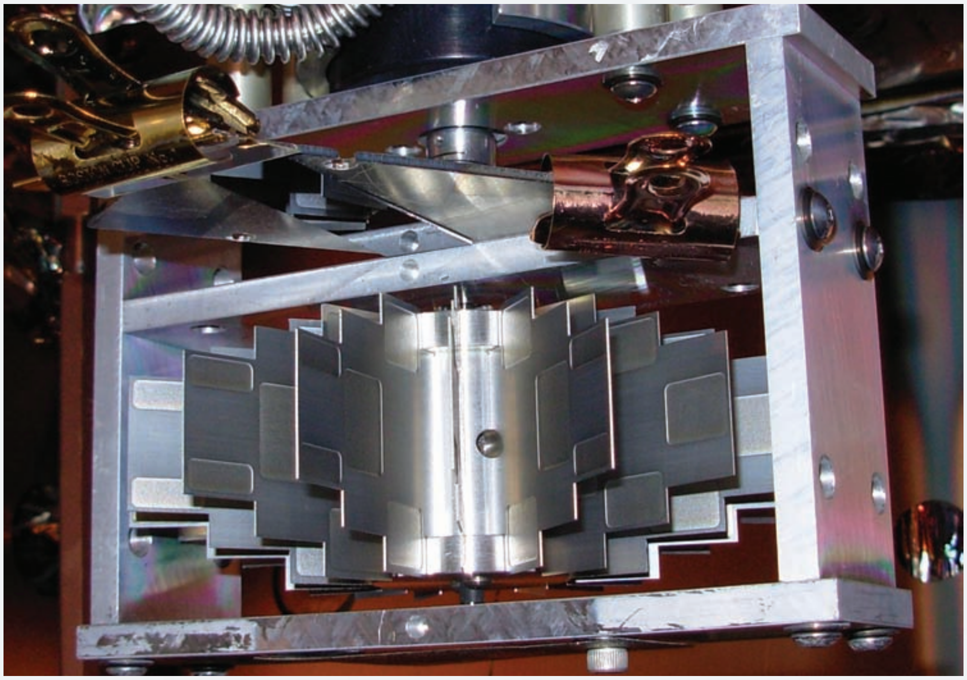
defects during coating deposition and an increased understanding of the role of substrate quality (scratches and digs) and subsurface damage on the resistance of mirrors to laser damage.

Mission Relevance

Transport mirrors are the fluence-limiting component in the fundamental wavelength section of advanced fusion-class lasers. Interest in reconfiguring these lasers to the second harmonic to realize even higher fusion gains elevates transport mirrors to the fluence-limiting optical component of the entire laser system. Fusion-class lasers are essential for the Stockpile Stewardship Program and other national security applications, as well as inertial-confinement fusion as an advanced concept for energy security and independence.

FY11 Accomplishments and Results

We achieved particle reduction during the electron beam deposition of silica films by incorporating a rotary vane velocity filter, which separates the slower velocity particulates from the atomic deposition plume. We also demonstrated directional change in particle momentum with laser plume heating—an achievement that could be combined with a directional filter to reduce particulate counts at the substrate plane. In summary, this project enabled the determination of the laser parameters—pulse length, wavelength, repetition rate, and pulse energy—needed in a femtosecond laser, scanning system, and associated diagnostic hardware to optimize mitigation protocols for multilayer mirror coatings. We successfully demonstrated particle reduction during coating deposition and the mitigation of defects in



The rotary vane velocity filter separates particulates from the atomic plume during deposition of multilayer thin films on laser transport mirrors.

multilayer mirrors that limit laser fluence. Our femtosecond machining parameters are being used to develop a mitigation protocol for higher-fluence transport mirrors for the National Ignition Facility.

Publications

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Direct Observation of Phase Transformations and Twinning under Extreme Conditions: In Situ Measurements at the Crystal Scale

Joel Bernier (10-ERD-053)

Abstract

We will develop an experimental technique for the direct three-dimensional observation of mechanical twinning and crystal-lattice-distortion phase transformations—both of which affect equation of state, strength, and stiffness—in polycrystalline materials under high pressure and temperature in situ. Such in situ observations at the crystal scale are essential for motivating, validating, and verifying

advanced constitutive models. We will study body-centered-cubic, hexagonal closely packed (HCP) phase transformations in iron by leveraging recent advances in synchrotron x-ray diffraction, and characterize the high-pressure HCP phase through direct observations. Experiments will take place at Argonne National Laboratory's Advanced Photon Source. Direct observations of these phenomena in situ at the crystal scale represent first-of-kind, discovery-class science.

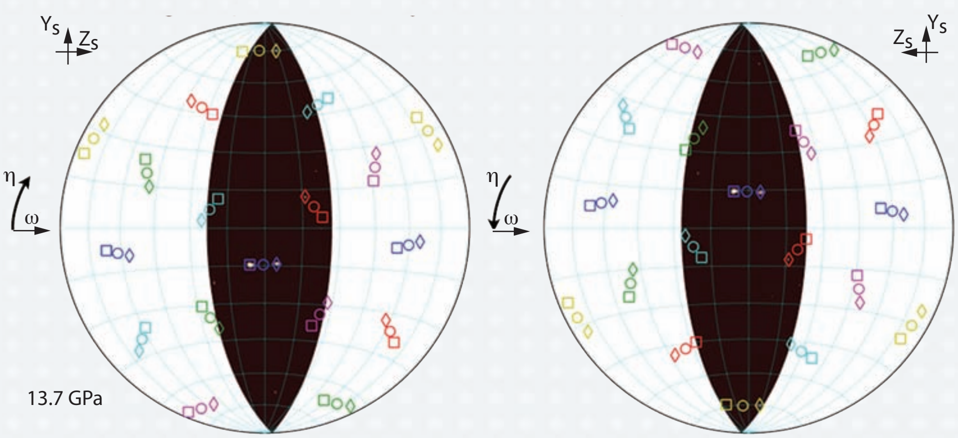
We expect to develop a three-dimensional in situ characterization technique that determines the (1) orientations and centers of mass of parent and transformed regions in individual embedded grains, (2) full strain and stress tensors averaged over these regions with a resolution of at least 10^{-4}, and (3) quasi-static thermal and mechanical loading in situ using a diamond anvil cell. This will provide direct variant selection, stress state, and growth measurements for the body-centered-cubic HCP phase transformation in individual grains embedded within an iron polycrystal, observations of mechanical twinning in HCP iron (if any), and material properties for the high-pressure HCP iron phase. These first-of-kind measurements will help determine the validity of constitutive models at high pressures.

Mission Relevance

High-fidelity materials models comprise a critical piece of the multi-physics simulation codes employed at LLNL in support of stockpile stewardship, particularly with respect to surety evaluation. This project will enable rigorous verification and validation of cutting-edge models at the crystal scale.

FY11 Accomplishments and Results

We (1) conducted experiments probing the effects of initial defect content, observing a retardation of the transformation with increasing dislocation density and subsequently determining that the observed transformation path strongly supports the Burgers-type mechanism, with the strength of different variants depending upon the stress deviator; (2) conducted additional iron studies and phase-transformation studies in zirconium and cerium at the Advanced Photon Source; (3) created code to model intensity distributions in heavily deformed materials; (4) made hardware



Pole figure projection for the measured intensities at the incipient phase transformation associated with the {010} planes in epsilon iron. Black is the background level, and the overlaid glyphs show locations of the predicted reciprocal lattice vectors for both the Bain and Burgers phase-transformation mechanisms with reference to orientation of the parent crystal. "Splitting" of the measured intensities is clearly visible and is the indelible signature of the Burgers mechanism. Because of the influence of deviatoric compression along the X axis, the blue and gold variants are most favored as the transformation begins.

improvements needed for our high-temperature experiments in FY12; and (5) made improvements to our software—particularly the interface—and demonstrated them in use at the Advanced Photon Source.

Proposed Work for FY12

Our primary objective in FY12 is to combine the high-angular-resolution, strain-sensitive technique we have developed with a high-spatial-resolution technique for grain and domain boundary mapping developed at Carnegie Mellon University, maintaining or improving upon our strain resolution while achieving a spatial resolution of at least 5 µm for in situ measurements. This should enable us to map grain and domain boundaries, particularly in deformed materials. In addition, we will also perform phase-change experiments with iron, zirconium, and cerium.

Publications

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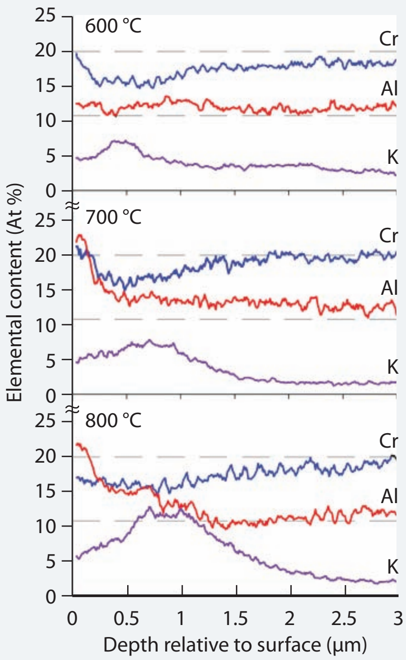
Material–Coolant Interactions in Fusion Reactors

Bassem El-Dasher (10-ERD-056)



The improved corrosion behavior of aluminum-containing oxide-dispersion-strengthened steels in FLiNaK molten salts has been attributed to the formation of a protective aluminum fluoride layer, as shown by the increase of aluminum (Al) near the surface.

Abstract

Inertial-confinement and magnetic fusion technologies require high-performance coolant systems for removal of heat from the chamber wall. In some cases, the coolants are multifunctional, enabling heat transport away from the fusion reactor wall and providing a medium for breeding tritium. Liquid-phase coolants suitable for use at the extreme temperatures and irradiations expected in these reactors pose a serious threat to reactor materials, which include advanced steels, refractory metal alloys, and ceramic composites. Determining coolant–material compatibility is critical for enabling fusion energy systems. We propose to evaluate materials damage and determine degradation mechanisms to provide an understanding of compatibility issues and how to resolve them.

We expect to (1) assess the threat of corrosion, stress corrosion cracking, and hydrogen embrittlement in candidate reactor materials; (2) elucidate the mechanisms involved in degradation of these materials; (3) develop predictive models to explain observed phenomena, thereby demonstrating scientific understanding of compatibility issues; and (4) identify viable strategies mitigating such attacks. Structural materials of interest include materials most likely to be used for the construction of fusion reactor systems, such as oxide-dispersion-strengthened (ODS) ferritic steels, refractory metal alloys, silicon and zirconium carbide, and composites of these materials. We will examine coolants of interest such as molten fluoride salts and liquid metals, including solutions of lead, bismuth, and lithium.

Mission Relevance

This project supports LLNL's mission of enhancing energy security for the nation and builds directly upon the Laboratory's world-class capabilities in fusion energy technologies. The proposed research is a key component of the Laboratory's strategic roadmap in energy security. Resolving key issues for practical, cost-effective fusion energy plants also supports the Laboratory mission in environmental security by enabling a source of abundant, clean power without issues of nuclear waste disposal, safety, carbon sequestration, or proliferation.

FY11 Accomplishments and Results

In FY11 we (1) studied the corrosion mitigation performance of ODS steels in FliNaK alkaline metal fluoride salt using beryllium sacrificial electrodes with our newly designed four-electrode high-temperature corrosion cell; (2) identified the mechanism of protection in aluminum-containing ODS steels in FLiNaK—namely, the formation of an aluminum fluoride layer; (3) performed high-temperature corrosion experiments on both ferritic–martensitic and ODS steels, measuring their performance with and without the presence of hydrogen (introduced electrolytically); (4) designed, built, and successfully deployed an electrochemical cell at Argonne National Laboratory's Advanced Photon Source, which was used to measure the kinetics of hydrogen bubble formation within the ferritic–martensitic and ODS steels; (5) began measurement of the mechanical behavior of ferritic–martensitic steels heat-treated to different conditions both with and without hydrogen; and (6) discovered excellent hydrogen-resistant behavior of silicon-containing ODS steels.

Proposed Work for FY12

For FY12 we will continue with research in our two main thrust areas: corrosion-control experiments focused on ODS steels and tantalum–tungsten alloys and tritium simulations focused on the effect of hydrogen on the metals. Specifically, we will (1) study the effect of the reduction–oxidation reaction approach of adding selected metals to molten salt, work that was originally planned for FY11; (2) examine the interaction of ODS steel and tantalum with lithium-based coolants; (3) continue hydrogen-charging experiments to study the effect of temperature on hydrogen retention, corrosion performance, and mechanical properties; (4) field an in situ hydrogen-charging setup at a synchrotron source; and (5) determine the parameters for joining ODS steels using friction methods.

Publications

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Thermodynamic and Kinetic Modeling of Advanced Nuclear Fuels

Patrice Erne Turchi (10-ERD-059)

Abstract

A key issue on the path to nuclear energy becoming an essential component of the U.S. clean energy strategy is complete burn of the nuclear fuel. We propose research that will enable high-burnup fuels by establishing the basic science for development and qualification of advanced nuclear fuel that will couple modern computational materials modeling, fabrication, and characterization capabilities with targeted performance-testing experiments using ion-beam facilities. This work will establish the scientific foundation and guide selection of the optimum fuel type for advanced reactor concepts.

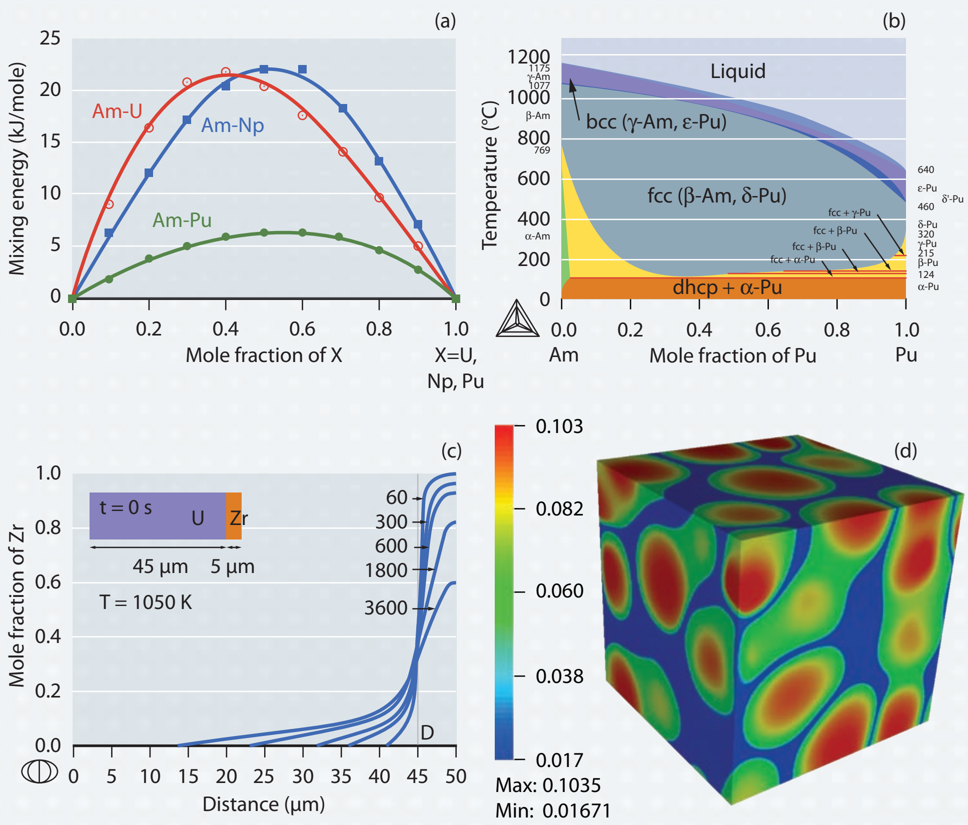
We expect to experimentally quantify phase stability and kinetics of phase transformations, interdiffusion, microstructural evolution, micromechanical properties, and the influence of severe radiation environments on fuel performance and, by the end of the project, develop a validated model for advanced nuclear energy materials under extreme conditions of radiation, temperature, and evolving chemistry. We will provide a science-based path forward to an optimized inert matrix fuel, while contributing to the development of a validated nuclear fuel database. We expect this project will establish LLNL's credibility within the nuclear energy community.

Mission Relevance

Our approach to development of the science of advanced nuclear energy fuels aligns well with the Laboratory's energy and national security missions. Development of both advanced fuel cycles and hybrid fusion–fission concepts is inexorably intertwined with the same scientific challenges. This research will extend LLNL capabilities and further enable actinide and high-energy-density science, high-performance computing and simulation, energy manipulation, and capabilities to develop materials on demand.

FY11 Accomplishments and Results

We (1) enhanced our modeling capabilities for assisting experiments by extending an LLNL phase-field code to account for thermal transport and make it compatible with the thermodynamic and kinetic data used in the CALPHAD (computer coupling of phase diagrams and thermochemistry) approach, to predict microstructural changes in the presence of a thermal gradient; (2) developed a search algorithm linked to the Thermo-Calc thermodynamic software for predicting the optimum composition of a multicomponent alloy with specific properties; (3) conducted ab initio studies on actinide alloys including uranium–molybdenum, uranium–titanium, plutonium–americium, and neptunium–zirconium and carried out phase diagram assessments of three of these alloys in collaboration with Texas A&M University; and (4) collaborated with Texas A&M University to prepare samples of nuclear fuels in bulk forms for diffusion couple studies and preliminary characterization. In summary, this project enhanced our theoretical capabilities for establishing the scientific basis of a high-throughput protocol for developing advanced nuclear fuel—a basis coupling modern computational materials modeling and simulation tools with fabrication and characterization capabilities and high-throughput performance-testing experiments. By upgrading state-of-the-art modeling codes and applying them to ab initio energetics and thermodynamic assessments of the phase diagrams of various mixtures of actinide alloys, we devised a tool for optimizing the composition of complex alloys for specific properties. In addition, our tool allows for predicting diffusion behavior in diffusion couples made of actinide and transition metals, including a new equation in the LLNL phase-field AMPE code, and predicting microstructure evolution during alloy coring. Next steps would include pursuing applications of this new capability in developing innovative fuels. For instance, the DOE Office of Nuclear Energy has expressed interest in our achievements.



(a) Phase stability trends in body-centered-cubic (bcc) americium alloys from ab initio calculations. The positive heat of formation in all three cases indicates a tendency towards phase separation. (b) Thermodynamic assessment of the americium–plutonium phase diagram based on the CALPHAD (computer coupling of phase diagrams and thermochemistry) methodology with input energetic results from ab initio calculations. (c) Theoretical prediction of diffusion behavior (mole fraction of zirconium versus distance) for a uranium–zirconium diffusion couple (the initial sample geometry is represented on the top left corner) maintained at 1050 K for various times. (d) Coring in alloys based on three-dimensional phase field simulations of the growth of plutonium–gallium's face-centered (fcc) cubic phase (green) from the bcc phase (blue). A higher composition of gallium appears in the center of the fcc grains.

Publications

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Turchi, P. E. A., P. Söderlind, and L. Kaufman, 2011. *Thermodynamic assessment of advanced nuclear fuel forms.* 10th Intl. Workshop on Fundamentals of Plutonium, Moscow, Russia, July 12–16, 2010. LLNL-PRES-438598.

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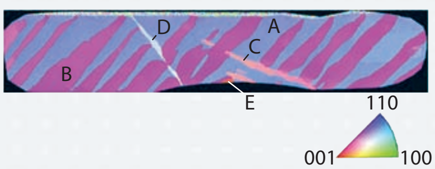

Twin boundaries forming in solidified single-crystal pure tin, shown by electron backscattering diffraction.

Engineered Lead-Free Solders for High-Performance Microelectronics

John Elmer (10-LW-002)

Abstract

The international effort to replace lead-containing solders has resulted in the development of a wide variety of solders based on low-melting-point alloys of tin. Intermetallic phase formation in these alloys and difficult nucleation of beta-tin during solidification result in large and undesirable anisotropic grains with poor mechanical properties. Long-term reliability of these joints is a serious concern for military and high-performance applications. We propose to develop synchrotron methods for in situ investigations of microstructure evolution in tin and lead-free solders. This work will be carried out at the Advanced Photon Source at Argonne National Laboratory, leveraging existing equipment from a previous program funded by the Office of Basic Energy Science.

We expect to develop a new characterization tool using in situ x-ray diffraction to directly observe solidification, phase transformation, and intermetallic compound growth between lead-free solders and substrates of copper and copper metallized with nickel and gold. We will investigate different commercial lead-free solders of tin alloys with silver and copper and compare them to pure tin to determine effects

of alloying content on the resulting microstructure. We anticipate that these tin-based solders will display large under-cooling (the difference between the melting temperature and temperature at solidification) that will be related to large grain sizes and nonequilibrium formation of undesired micro-constituents. Results will guide future work directed at developing high-performance lead-free solders.

Mission Relevance

Our project supports the Laboratory mission of ensuring the safety, security, and reliability of the U.S. nuclear deterrent. Mission-critical solder joints used in weapons systems must be extremely reliable over a much larger period of time than is required for conventional consumer electronics. Solder compositions, techniques, pad metallization, and intermetallic compound formation are all of concern. In addition, conventional solder packaging of diode arrays is insufficient for reliable operation of future fusion energy sources, and advanced laser targets and holders have important thermal and mechanical solder joints.

FY11 Accomplishments and Results

We continued work at the Advanced Photon Source that demonstrated the use of synchrotron methods to investigate microstructure evolution in tin and other lead-free solders, using our new characterization tool to measure solder solidification under-cooling to assist in the development of high-performance solders. In addition, the sintering kinetics of silver nanometer-scale ink, a lead-free solder alternative, was investigated with in situ synchrotron-based diffraction for the first time. The results provided in situ measurements of under-cooling in tin and indium, in situ sintering kinetics of silver nanoscale inks, and led to a record of invention for compound inoculants additions to reduce under-cooling and refine the microstructure of lead-free solders. This project successfully demonstrated our application of synchrotron methods to measure solder solidification under-cooling to enable development of high-performance solders. Major defense contractors and commercial vendors of microelectronic equipment are all actively pursuing development of new lead-free solders targeted for military and mission-critical applications, and continued research will be pursued through collaborations related to the Department of Defense.

Publications

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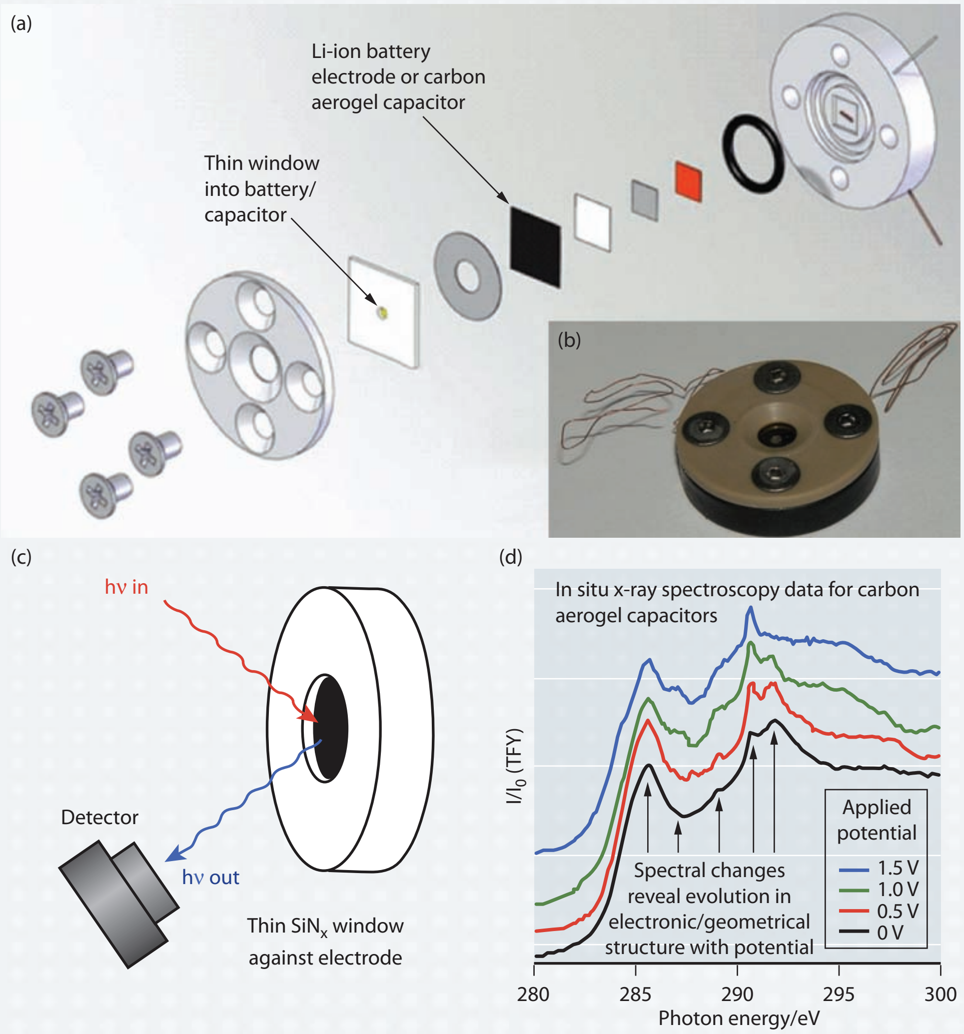
In Situ Spectroscopy and Microscopy for the Study of Advanced Materials for Energy Storage

Jonathan R. Lee (10-LW-045)

Abstract

We propose to apply a suite of in situ x-ray spectroscopy and electron microscopy techniques for investigating the performance of advanced materials for energy storage, specifically electrodes based on nanometer-scale group IV element crystals and capacitors based on nanoporous carbon. Developing new materials for energy storage is essential for securing this nation's energy future, and characterization of the chemical and physical phenomena that occur during charge transfer processes is a key component of this endeavor. Our studies will yield insight into the evolution in electronic and geometrical structures of the nanostructured materials during cycling, which is key to determining the mechanisms of energy storage and electrode degradation, as well as improving the design of next-generation materials.

If successful, we expect that our project will yield information regarding the mechanisms of energy storage in nanostructured electrodes and their degradation



The experimental cell displayed in (a) and (b) is used to investigate the physical behavior of electrical energy storage materials under operating conditions with soft x-ray spectroscopy, as shown in (c) and (d).

with repeated charge and discharge cycling. Complementary specific-charge measurements will allow determination of the effects of surface chemistry, structure, and interfacial properties on electrode performance. All of this information will have significant impact in the field of electrical energy storage and on the design of next-generation electrode materials. We will also gain insight into the modes and mechanisms of energy storage by nanoporous materials, which show great promise in capacitive energy storage applications. Finally, we will demonstrate the enormous potential of a suite of unique in situ analysis tools.

Mission Relevance

This project supports a core Laboratory mission to enhance the energy and environmental security of the nation by advancing the state of the art in electrical energy storage—specifically, new in situ techniques to investigate chemical, electrochemical, and physical processes at solid–liquid and solid–gas interfaces.

FY11 Accomplishments and Results

In FY11 we (1) developed an in situ soft x-ray spectroscopy technique to study materials for electrical energy storage with greater reliability and enable element-specific measurements of lower-atomic-number species in novel electrodes for electrical energy storage; (2) performed in situ soft x-ray spectroscopy studies to evaluate the mechanisms of charge transfer and storage and surface reactions of nanoporous carbon electrodes for capacitive energy storage; (3) successfully performed in situ x-ray spectroscopy measurements of novel electrodes for lithium ion batteries; (4) developed a method for achieving improved spatial resolution of in situ transmission electron microscopy imaging of electrical energy storage materials and used it to study nanocrystalline tin-based lithium ion battery electrodes; and (5) experimentally demonstrated the viability of using resonant soft x-ray emission spectroscopy for element-specific studies of lithium in electrode materials.

Proposed Work for FY12

In FY12 we propose to conduct a set of x-ray spectroscopy studies of metal oxide cathodes for lithium ion batteries with an emphasis on x-ray spectroscopy measurements to investigate the electronic and geometrical structure of lithium in cathodes as a function of charge–discharge cycling. In addition, we will make x-ray spectroscopy measurements of the transition metal elements and oxygen atoms and ions to determine their evolution in electronic and geometrical structure under the same cycling conditions. Comparison of our spectroscopy data with complementary studies of the capacity for electrodes to store charge should provide new insight into the modes and mechanisms of charge storage and electrode degradation.

Publications

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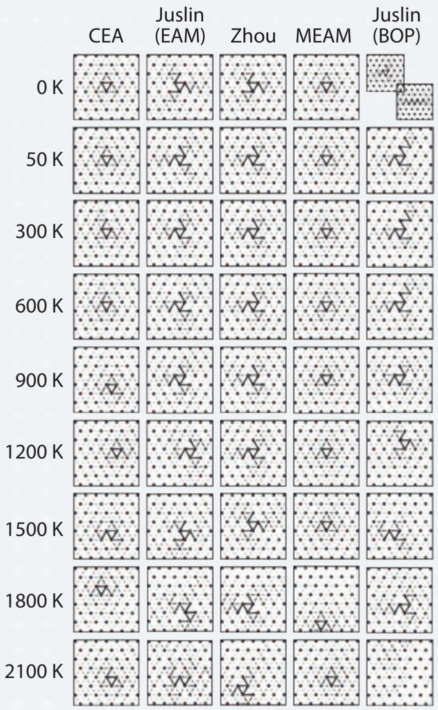


Temperature dependence of the screw-dislocation core structure for several tungsten potentials, such as the modified embedded atom method (MEAM).

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

Jaime Marian (11-ERD-023)

Abstract

Tungsten is an exceptionally suitable candidate for the first wall surrounding the central cell of a 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. In 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 candidate tungsten alloys 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, in which a suitable design for a first wall capable of sustaining the 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 Lawrence Livermore missions to enhance the energy and environmental security of the nation and to strengthen the nation's economic competitiveness, as well as supporting the Laboratory's strategic mission thrust in fusion energy to provide a sustainable and once-through, closed-fuel-cell nuclear energy option.

FY11 Accomplishments and Results

In FY11 we (1) tested five different interatomic potentials for tungsten and, on the basis of several static and dynamic properties, selected one—a modified embedded-atom method—as our end-member potential of choice to be used in a mixed-alloy potential construction method; (2) constructed the first version of a tungsten–titanium potential using this method and performed all the requisite density functional theory calculations to test its accuracy; and (3) computed screw dislocation mobilities for the modified embedded-atom method up to 2 GPa of stress and between 300 and 2100 K in temperature.

Proposed Work for FY12

In FY12 we will (1) parameterize kinetic Monte Carlo code using the potentials developed in FY11 and use the code to conduct kinetic Monte Carlo simulations of dislocation motion in tungsten–titanium alloys; (2) explore temperature, concentration, and stress parameters and provide the optimum ranges related to dislocation mobility using kinetic Monte Carlo simulations, which are much more efficient than direct atomistic simulations; (3) develop a tungsten–tantalum potential in the same fashion as the tungsten–titanium potential; and (4) use stochastic rate theory to analyze the effect of pulsed irradiation on these tungsten-based systems.

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 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 evaporation and condensation of material; and (3) explore the use of laser-based 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 laser-parameter-dependent energy absorption. We will establish a method for introducing controlled silica-rich vapor at lower temperatures to infill 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.

FY11 Accomplishments and Results

Using coaxial gas flow and infrared laser heating, we showed that reducing conditions (i.e., hydrogen-rich) increased silica removal rates up to fivefold over ambient conditions, even at relatively low hydrogen gas mixtures. Results were interpreted with a near-equilibrium model of the laser-based evaporation of silica. Experiments using variable flow rates revealed a mass-transport-limited evaporation process, further validating the near-equilibrium model. Based on these insights, we developed a semi-empirical predictive model for evaporation of silica dependent on temperature, gas flow rate, and gas composition. Reactive-force-field molecular dynamics computational models were also used to explain the effect of hydrogen on final surface shape through changes in surface energy caused by hydrogen reaction chemistry. Additionally, we demonstrated that silica could be polymerized directly on the surface of pristine silica under atmospheric conditions, with deposition rates one order of magnitude greater than those reported in the literature for the tetraethyl-orthosilicate silica precursor used. Initial damage tests of annealed, deposited films showed thresholds commensurate with etched and pristine levels.

Proposed Work for FY12

In FY12 we will (1) employ high-speed (megahertz) thermal, optical, and spectroscopic diagnostics to characterize evaporated material; (2) benchmark kinetic models and develop semi-empirical surface-shaping models in the fast evaporative regime; (3) develop and test evaporative mitigation methods using a tunable carbon dioxide laser; and (4) begin exploring localized silicon dioxide deposition using laser chemical-vapor deposition to re-plane damaged material.

Publications

Elhadj, S., et al., 2011. *Dynamics of silica particle heating and evaporation under CO_2 laser irradiation.* CLEO:2011 Laser Science to Photonics Applications, Baltimore, MD, May 1–6, 2011. LLNL-ABS-463454.

Elhadj, S., et al., 2011. *Evaporation kinetics of CO_2 laser heated fused silica.* 2011 MRS Spring Mtg. and Exhibit, San Francisco, CA, Apr. 25–29, 2011. LLNL-ABS-461515.

Feit, M. D., et al., 2011. *Densification and residual stress induced by CO_2 laser-based mitigation of SiO_2 surfaces.* SPIE Laser Damage 2011, Boulder, CO, Sept. 18–22, 2011. LLNL-PROC-461512.

Matthews, M. J., et al., 2011. *Microstructural relaxation phenomena on laser-modified fused silica surfaces.* APS March Mtg., Dallas, TX, Mar. 21–25, 2011. LLNL-ABS-462331.

Matthews, M. J., et al., 2011. "Synchrotron radiation infrared microscopic study of non-bridging oxygen modes associated with laser-induced breakdown of fused silica." *Appl. Phys. Lett.* **99**, 151109. LLNL-JRNL-493512.

Raman, R. N., et al., 2011. *Monitoring relaxation of electronic defects in SiO_2 following CO_2 laser heating.* 2011 MRS Spring Mtg. and Exhibit, San Francisco, CA, Apr. 25–29, 2011. LLNL-POST-485255.

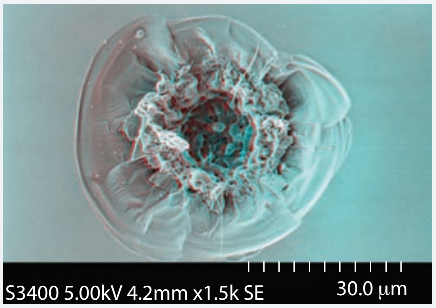
Shen, N., et al., 2011. *Evolution of nano- to micrometer-scaled surface structures in fused silica under high-temperature CO_2 laser smoothing.* 2011 MRS Fall Mtg. and Exhibit, Boston, MA, Nov. 28–Dec. 2, 2011. LLNL-POST-462915.

Shestakov, A. I., et al., 2011. *Simulation of infrared laser heating of silica using heat conduction and multifrequency radiation diffusion equations adapted for homogeneous refractive lossy media.* 2011 MRS Spring Mtg. and Exhibit, San Francisco, CA, Apr. 25–29, 2011. LLNL-CONF-461316.

Soules, T. F., et al., 2011. *An assessment of molecular dynamic force fields for silica for use in simulating laser damage mitigation.* SPIE Laser Damage 2011, Boulder, CO, Sept. 18–22, 2011. LLNL-PROC-461692.

Understanding the Stochastic Nature of Laser-Induced Damage

Christopher Carr (11-ERD-030)



A three-dimensional image of a damage site compiled from a stereo pair of scanning electron microscope images. The effect is created using red and blue three-dimensional glasses, with red on the left.

Abstract

Laser optic surface damage is of particular concern for laser facilities, because even small damage sites only a few micrometers in size initially 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, we will 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. 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 such facilities to operate at energy densities high enough to enable their full mission space—including inertial-confinement fusion and laser-ignition fusion energy—in support of the Laboratory's missions in stockpile stewardship and advanced lasers and applications.

FY11 Accomplishments and Results

In FY11 we developed the infrastructure and experimental methodology to determine the effect of factors on probability and rate of damage site growth. Specifically, we (1) we used polarimetry to determine the effect of residual stress, scanning electron microscopy to determine the effect of internal micro-structure, and optical coherence tomography to determine the effect of sub-surface fracture; (2) used magnetoreological fluid polishing to perform optical sectional tomography on growing damage sites to study the extent of the subsurface fracture network and how it is related to growth, and conducted preliminary thermal annealing experiments to isolate the effects of residual stress on growth rate; (3) completed scoping experiments and begun systematic experimentation measuring each of these factors; (4) performed controlled experiments to gauge how the depth of energy deposition affects damage site morphology, including direct imaging of luminosity caused by the absorption front; and (5) developed data-mining and machine-learning algorithms to identify, on the basis of site history, those sites likely to exhibit atypical growth behavior under controlled conditions.

Proposed Work for FY12

Building on our FY11 results, we will (1) use our previous findings to quantify the effects of parameters that affect growth rate and probability of growth, (2) begin a study to detail how shot history affects evolution of the physical aspects of damage sites that govern damage growth, and (3) apply the predictive data-mining and machine-learning algorithms developed in FY11 to identify individual sites that are likely to exhibit atypical growth and examine the physical features that our research has identified as important in damage growth.

Publications

Carr, C. W., J. D. Bude, and P. DeMangem, 2010. "Laser-supported solid-state absorption fronts in silica." *Phys. Rev. B* **82**, 184304. LLNL-JRNL-423847.

Carr, C. W., et al., 2011. "The effect of laser pulse shape and duration on the size at which damage sites initiate and the implications to subsequent repair." *Optic. Express* **19**(s4), A859. LLNL-JRNL-482133.

Cross, D. A., and C. W. Carr, 2011. "Analysis of 1ω bulk laser damage in KDP." *Appl. Optic.* **50**(22), D7. LLNL-JRNL-480645.

Liao, Z. M., et al., 2011. "Modeling max-of-N fluence distribution using measured shot-to-shot beam contrast." *Appl. Optic.* **50**(20), 3547. LLNL-JRNL-468532.

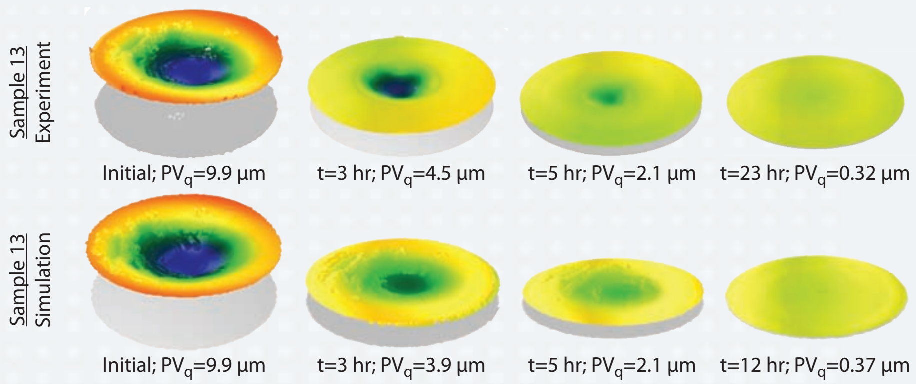
Negres, R. A., et al., 2011. "Exploration of the multiparameter space of nanosecond-laser damage growth in fused silica optics." *Appl. Optic.* **50**(22), D12. LLNL-JRNL-480354.

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

Tayyab Suratwala (11-ERD-036)

Abstract

This project will scientifically investigate critical phenomena affecting the full-aperture finishing of high-aspect-ratio optical components, conduct full-aperture optical polishing in rogue-particle-free environments by understanding and preventing key sources of these particles, and develop methods to enhance the lifetime and reduce the cost of inertial-confinement fusion and laser inertial fusion



Surface of a round workpiece as a function of polishing time illustrating convergence, after identifying and eliminating sources of nonuniform material removal (except for workpiece-lap mismatch). The top row represents the measured data and the bottom row shows simulation results using the SURF (Speeded Up Robust Feature) code.

optics. To this end, we will perform specifically designed polishing experiments and use models to understand the effects of pad wear, mechanical bending, residual stress, and thermal effects on nonuniform removal and to determine methods to control rogue particles, such as using 100% humidity to control particle size distribution. The fusion laser optics that we will target include main debris shields, continuous phase plate substrates, disposable debris shields, blue blockers, 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 decreased surface roughness.

Mission Relevance

The project supports the Laboratory's missions in advanced lasers and their applications as well as inertial-confinement fusion by helping to achieve 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 and thermal shock resistance and decreasing surface roughness. This project also furthers LLNL's mission in strengthening America's economic competitiveness by improving the precision of other optics and semiconductors.

FY11 Accomplishments and Results

In F11 we (1) conducted polishing experiments and modeling to quantify phenomena resulting from nonuniform pad wear and deformation; (2) studied the effect of these phenomena on nonuniform workpiece removal; (3) studied mechanical workpiece bending, residual stress, and post-polishing mechanical springing of the workpiece and their effects on nonuniform workpiece removal; (4) developed methods to mitigate effects on nonuniform removal; (5) used this new understanding and controls we developed to demonstrate, for the first time, convergent polishing of low-aspect-ratio workpieces; and (6) began initial filtration tests and slurry chemistry experiments to demonstrate control of particle size distribution during polishing.

Proposed Work for FY12

In F12 we will (1) continue to develop methods to mitigate the effects on workpiece removal of nonuniform pad wear and workpiece bending, with the goal of demonstrating figure convergence on high-aspect-ratio workpieces; (2) investigate a new phenomenon we discovered in FY11 called the "edge effect," which results in nonuniform wear; and (3) conduct polishing experiments and modeling to understand temperature distribution, how it can change the Preston coefficient (which relates material removal rate to the pressure and velocity applied during polishing) and cause workpiece deformation, and how thermal and mechanical properties and shape influence nonuniform workpiece removal.

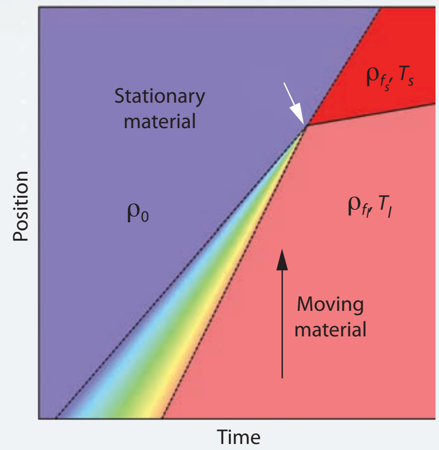
Hydrogen Melting and Metallization at High Density

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 (up to 100 GPa pressure) and low temperature. Through a combination of simulations and measurements, we aim to confirm or improve calculated shock Hugoniots—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 the shock-compressed behavior of hydrogen and deuterium 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.



An idealized schematic of a ramp wave steepening into a shock wave. We have observed the quasi-isentropic ramp compression of deuterium on an ultrafast timescale, which may enable the compression of deuterium to high density using a tabletop laser.

Mission Relevance

This research is aligned with LLNL 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.

FY11 Accomplishments and Results

In FY11 we (1) calculated shock Hugoniot points for hydrogen along the metallization transition at pre-compressions of up to 67 GPa; (2) calculated the shock Hugoniots for melting at pre-compressions of up to 36 GPa and verified that out calculations compare well to shock experiments; (3) observed under-100 ps quasi-isentropic compression, followed by a transition from ramp to shock wave, based on our data and simulations; and (4) calculated temperatures for deuterium shocked to near the melt and metallization transitions from pre-compressed states. Our results for FY11 demonstrate that isentropic ramp compression on an ultrafast timescale may provide a path to very high-density hydrogen with substantially lower experimental resources than have been previously used because the volume of compressed material (and thus compression energy) can be orders of magnitude smaller than previous work. This is a direct consequence of the compression rate used in this experiment and the observed ultrafast equilibration rate in deuterium.

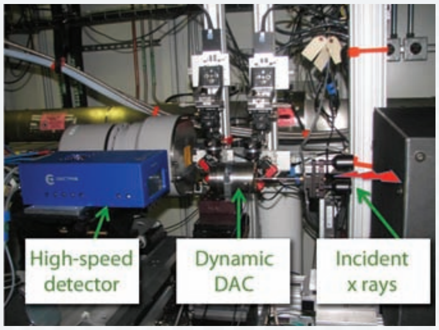
Proposed Work for FY12

Next year we will (1) extend measurements to pre-compressions greater than 50 GPa and experimentally determine the timescale of melting as a function of pre-compression and the final shocked state for hydrogen–deuterium mixtures, (2) determine the experimental conditions required to obtain metallization and perform metallization experiments, and (3) use experimental data on the superheated, solid, and liquid Hugoniots to experimentally estimate the temperature of phase transitions for hydrogen–deuterium mixtures based on calculations and experiments in FY11.

Publications

Armstrong, M. R., et al., 2011. "Shock compression of precompressed deuterium." *Proc. 17th APS Topical Conf. Shock Compression of Condensed Matter*. LLNL-PROC-491811.

Crowhurst, J. C., et al., 2011. "Invariance of the dissipative action at ultrahigh strain rates above the strong shock threshold." *Phys. Rev. Lett.* **107**(14), 144302. LLNL-JRNL-478314.



Our time-resolved x-ray diffraction experimental setup at Argonne National Laboratory's Advanced Photon Source enables time-resolved studies of the dynamics of pressure-induced phase transitions.

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

William Evans (11-ERD-046)

Abstract

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 microstructure, such as polycrystal or amorphous. 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 molecular system and liquid–solid transitions in the metals mercury, bismuth, and gallium.

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. Finally, success will establish LLNL as a pioneer in this new high-pressure science research capability.

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 dramatically enhance our technological ability to achieve "materials by design" for defense and other mission-relevant applications.

FY11 Accomplishments and Results

In FY11 we successfully addressed the goal of time-resolved measurements of the dynamics of pressure-induced phase transitions, concentrating our studies on bismuth and gallium. Specifically, we (1) enhanced the design of our dynamic diamond anvil cell for easier use; (2) performed time-resolved measurements (optical and x-ray diffraction) of pressure-induced transitions in bismuth and gallium; (3) collected, using the Advanced Photon Source at Argonne National Laboratory, time-resolved x-ray diffraction data of the phase transitions in bismuth and gallium in the range of 0 to 10 GPa; (4) initiated data analysis and evaluation of nucleation and growth parameters—although the time-resolution of our diffraction experiments was limited to approximately 10 ms because of limitations with the x-ray source and detection systems, this time resolution was adequate to identify transition dynamics; (5) formulated approaches to improve the performance of our diffraction experiments, which should reduce the time resolution to sub-millisecond levels; and (6) identified and hired two postdoctoral researchers to assist with our research.

Proposed Work for FY12

Using the dynamic diamond anvil cell, we will generate and dynamically vary the pressure conditions to induce a melt–freeze transition in mercury, bismuth, and gallium. In these experiments, we will selectively resolve stages in the phase transition by taking "snapshots" with x-ray pulses and will produce quantitative measurements of crystal growth and crystal structure evolution that will help understand dynamic processes by determining important kinetic parameters associated with phase transition, nucleation and melting, mechanical deformation, and crystal growth. We will also make time-resolved optical imaging and x-ray scattering measurements of pressure-induced melting and freezing reactions, with a time resolution greater than 100 ps, and use the resulting data for modeling.

Publications

Evans, W. J., 2011. *The dynamic diamond anvil cell (dDAC) innovations in high pressure science at LLNL*. LLNL-POST-485973.

Evans, W. J., H. Cynn, and M. J. Lipp, 2011. *Time-resolved studies of pressure-induced nucleation and growth*. Workshop Dynamic X-ray Diffraction/Spectroscopy Experiments at Extreme Conditions, Hamburg, Germany, May 5–6, 2011. LLNL-PRES-490396.

Evans, W. J., Z. Jenei, and J. H. P. Klepeis, 2011. *Static high pressure studies under non-hydrostatic/non-equilibrium conditions.* 17th Biennial Intl. Conf. APS Topical Group on Shock Compression of Condensed Matter, Chicago, IL, June 26–July 1, 2011. LLNL-ABS-475279.

Evans, W. J., et al., 2011. *Pressure-induced nucleation and growth*. LLNL-POST-450411.

Demonstration of Electron Spin Imbalance in Two-Dimensional States

Sung Woo Yu (11-LW-008)

Abstract

The field of spin transport electronics ("spintronics") faces a fundamental problem—the lack of a compact source of spin-polarized electrons compatible with integrated circuit technology for solid-state spintronics applications. Current sources use magnetic materials with external magnetic fields and are therefore incompatible with integrated circuits. It has already been shown that lower-dimensional structures of nonmagnetic materials can exhibit spin polarization through spin–orbit interaction by removal of spatial inversion symmetry. We propose a unique approach to creating a spin-polarized electron source that would leverage this finding by using an electric field, not a magnetic field, to create and change spin polarization at the edges of a two-dimensional (2D) nonmagnetic structure. We propose two new methods to

demonstrate spin polarization at the edges: measuring the magnetic moment as well as the voltage induced by spin current.

In this project, we will attempt to develop a new class of nonmagnetic dimensional materials suitable for future spintronics applications as spin injectors. This accomplishment will advance our understanding of the fundamental interactions between materials science and the quantum mechanical property called spin, and may also lead the way to a compact source of spin-polarized electrons, which is a prerequisite for realizing practical spintronics devices such as for quantum computing or communications. The novel idea of using an electric field on a thin structure of a nonmagnetic 2D material to spatially separate spin-polarized electrons would have a great impact in the field. Such a thin structure could become a critical component of integrated spintronic circuits.

Mission Relevance

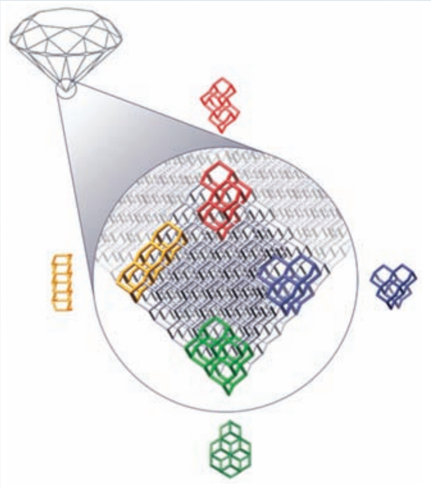
This work will provide fundamental information for the interaction between spin and materials that will advance technical understanding in the important field of spintronics, in support of the Laboratory's missions in basic science, advanced computing, and national security.

FY11 Accomplishments and Results

In FY11 we focused on the fabrication of low-dimensional nonmagnetic strips as materials with strong spin–orbit interactions for spintronics. Specifically, we (1) created ultrathin 2D nonmagnetic bismuth strips on a silicon nitride membrane by means of a magnetron sputter deposition system, (2) added platinum electrodes on the sides of the strip with a mask so that an electric voltage can be applied, (3) characterized the resulting strips by atomic force microscopy and by a full-field soft x-ray transmission microscope using circularly polarized synchrotron radiation at the Advanced Light Source at the University of California at Berkeley, and (4) developed high-quality single crystals of bismuth compounds Bi_2Se_3 and $TlBiSe_3$, and fabricated ultrathin 2D nonmagnetic strips by sectioning the crystals with a microtome and cutting with a femtosecond laser beam.

Proposed Work for FY12

We will conduct proof-of-principle experiments to demonstrate, for the first time, spin polarization at the edges of a 2D bismuth strip. Specifically, we will use magnetic microscopy with circularly polarized light (at the Advanced Light Source and Stanford Synchrotron Radiation Lightsource) to measure the magnetic moments induced by spin polarization at the edges. We will also investigate the possibility of measuring the voltage induced by the spin current by connecting two edges of a sample with a transverse strip made of a material with high spin–orbit interaction, which should allow a spin current to circulate and generate a transverse voltage.



Diamondoids are small structures where carbon atoms lie at diamond lattice positions and surface bonds are terminated in hydrogen. Diamondoids have unique properties: they bridge small hydrocarbons and bulk diamond properties.

Fundamental Properties of Diamondoids

Trevor Willey (11-LW-028)

Abstract

Diamondoid self-assembled monolayers are nanometer-scale engineered surfaces with the potential to revolutionize two drastically different technological regimes important to inertial-confinement fusion targets—their peculiar properties may help control the growth of hydrogen crystals, while diamond spherical shells are candidate capsule materials. Current deposition technologies for diamond thin films rely on defect-ridden, detonation-synthesis nanometer-scale diamond crystallites to nucleate the diamond layer, leading to highly polycrystalline films. This project seeks to explore the fundamental properties of diamondoid self-assembled monolayers and their potential to produce clean, thin, or template-created chemical-vapor-deposition diamond that is superior to what is possible with current technology. Using near-edge x-ray absorption fine structure spectroscopy, we will correlate the fundamental properties of diamondoid self-assembled monolayer substrates to hydrogen wetting and crystal nucleation, as well as to chemical-vapor-deposition diamond film quality. Our goal of developing a technology to produce more uniform crystalline and potentially template-created diamond films would greatly enhance technologies used to generate diamond inertial-confinement fusion capsules.

If successful, this project will provide the scientific foundation to overcome several technical hurdles to producing optimal inertial-confinement fusion target capsules. We expect to achieve a range of capabilities for nucleating dihydrogen on and wetting the surfaces of self-assembled monolayers, depending upon molecular structure, chemical functionality, and molecular arrangement and packing. We have already demonstrated the ability to present the diamondoid analogues of macroscopic diamond surfaces, but these previously formed monolayers were not robust enough for diamond chemical vapor deposition. By using more robust surface attachment, we expect to create diamondoid self-assembled monolayers conducive to chemical vapor deposition of diamond. If successful, this would revolutionize diamond thin-film growth for both inertial-confinement fusion and myriad other technologies.

Mission Relevance

This project supports LLNL's national security and energy security missions by developing a fundamental understanding of the nucleation and growth of hydrogen on self-assembled monolayers, which will provide a simple means for modifying the inner surfaces of inertial-confinement fusion targets, and by developing the capability to produce more uniform crystalline and potentially template-created diamond films for diamond inertial-confinement fusion target capsules.

FY11 Accomplishments and Results

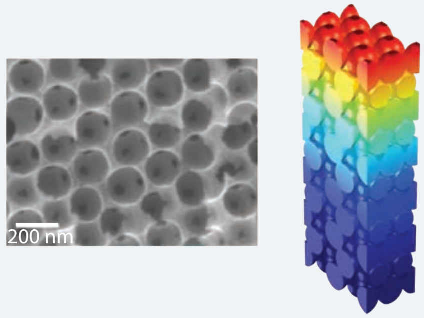
We focused on providing optimal monolayers for correlating hydrogen wetting to monolayer properties, which could transition to LLNL programmatic research on fusion target fabrication. In FY11 we (1) developed new and improved methods for forming self-assembled monolayers on a substrate of alkenes on silicon, for which we submitted a record of invention; (2) continued to characterize and develop this method for diamondoid monolayers; (3) investigated how structure (i.e., size as well as bond strain in diamond-like materials) affects electronic properties; and (4) redirected, with the fabrication and discovery of diamond aerogels, some of our efforts to investigate diamond nucleation from diamondoids and other materials using high-pressure, high-temperature methods, in addition to chemical vapor deposition.

Proposed Work for FY12

We will (1) continue fabricating and characterizing self-assembled monolayers with various structures, substrates, and chemical functionality; (2) correlate these parameters to dihydrogen wetting and crystal nucleation using contact angle and optical methods, with the goal of controlling hydrogen nucleation and growth; (3) verify that our diamondoid monolayers are sufficiently robust for chemical vapor deposition; (4) deposit diamond on these substrates using chemical vapor deposition; and (5) characterize the results using near-edge x-ray absorption fine structure, scanning electron microscopy, and other technologies.

Mass Transport through Porous Materials

Michael Stadermann (11-LW-037)



Left: A scanning electron microscope image of a carbon electrode template construct with polystyrene beads. The beads are removed before pyrolysis, leaving a periodic lattice of voids. Right: A scale model of the electrode used to model transport behavior. In the simulation, a fixed bias is applied to the electrode, and the steady-state concentrations of electrolyte are shown with a color scale.

Abstract

Porous systems are found in a wide variety of surface-reaction energy applications, from storage to catalysis. The porous structures in such systems are used to increase available surface area and thereby enhance the reaction rate. However, mass transport limitations caused by the distribution and shape of pore size can limit mass flow and cause local potential changes that reduce the efficiency of the electrical reactor and may permanently reduce its performance. We propose to address this problem by studying mass transport of well-defined porous systems through model and experiment to create design rules for an optimized electrode that will deliver a substantially increased power density without reducing energy density.

The primary result of this work will be microscopic understanding of mass transport through a porous scaffold. This understanding can be used to optimize battery

electrodes and other energy storage materials as well as catalyst beds. In addition, we will produce a proof-of-principle battery that is expected to have more than twice the energy density of a commercial battery. The technology is transferable to more energetic battery chemistries and has the potential to create a paradigm shift in battery fabrication, providing the U.S. with a competitive advantage and securing national energy needs.

Mission Relevance

This project supports the Laboratory's core strategic mission thrust area of delivering energy solutions to enhance national energy security. New materials for energy storage will be developed, along with a transferable electrode design relevant to all solid battery types. The design can leverage other battery technology advances and will have a major impact on the fabrication of batteries.

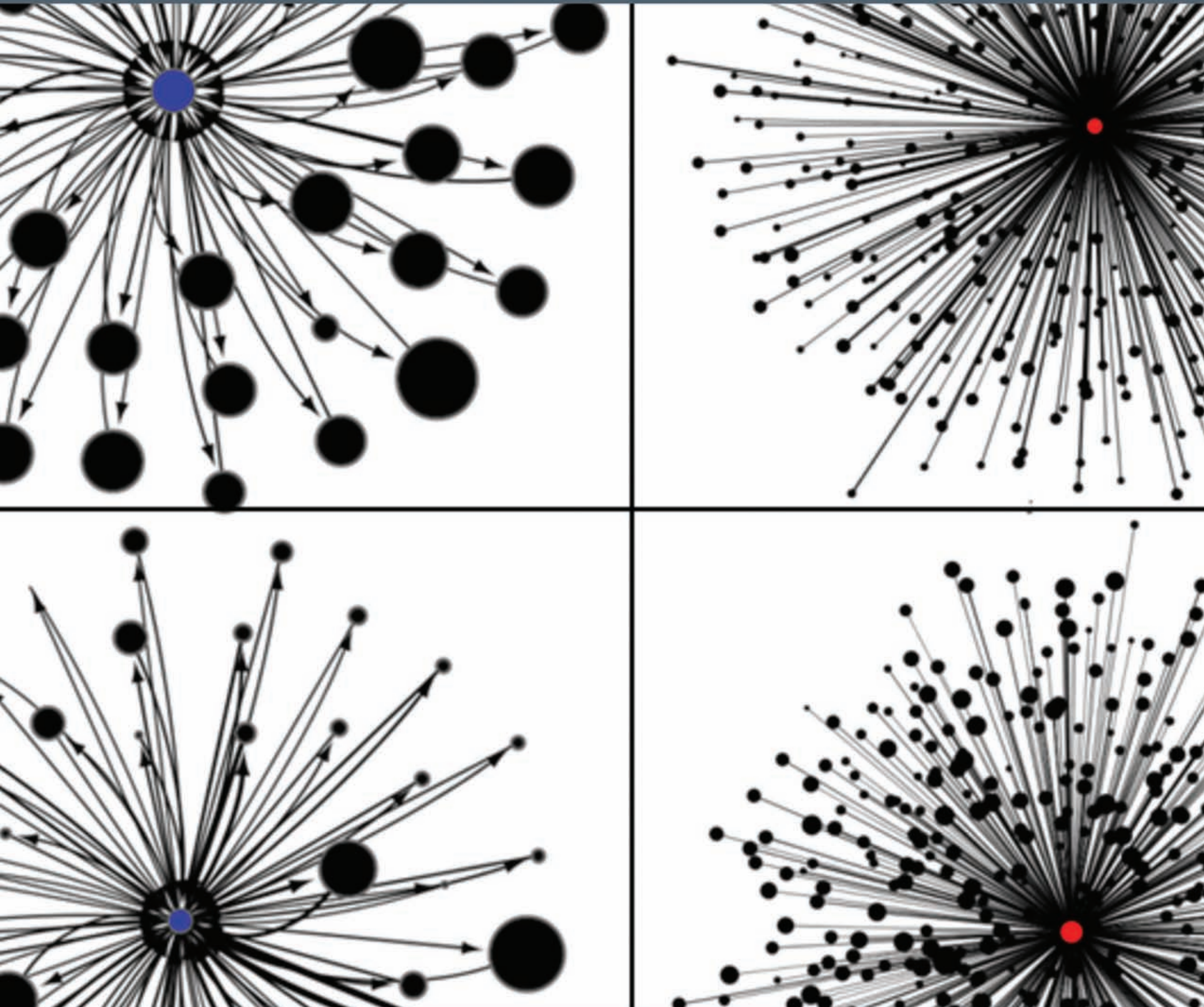
FY11 Accomplishments and Results

In FY11 we (1) decided to switch our experimental focus from zinc oxide to nickel oxide as an electrode material for this study because of its higher electrode stability, (2) studied the deposition and cycling behavior of nickel oxide material on flat carbon substrates, (3) created a model for material transport and tested it with a variety of parameters to determine scalability, and (4) developed a template-construct electrode and began to conduct tests to determine its performance.

Proposed Work for FY12

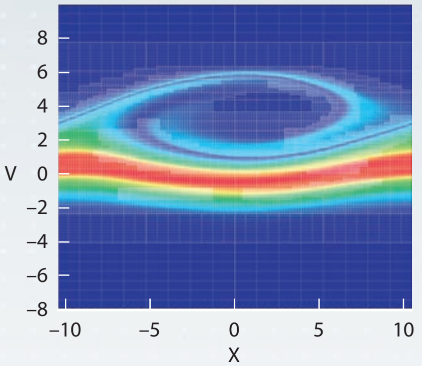
In FY12 we will (1) deposit nickel oxide onto the carbon template construct and measure the diffusion performance and compare it to the model, adjusting the model if necessary; (2) alter the structure to determine impact of the length scales on the transport behavior, and synthesize alternate structures with improved performance; and (3) attempt to build a nickel–zinc battery with optimized electrodes.

Mathematics and Computing Sciences



Laboratory Directed Research and Development

FY2011



The electron distribution function for a standard 1D + 1V Vlasov–Poisson bump-on-tail instability calculation at t = 22.5 s. The adaptively refined mesh is shown in white. Four levels of refinement reduced the mesh needed by more than 50%.

Efficient Numerical Algorithms for Vlasov Simulation of Laser–Plasma Interactions

Jeffrey Hittinger (08-ERD-031)

Abstract

The objective of this project is to develop efficient, high-fidelity continuum algorithms for the Vlasov–Maxwell system that will facilitate routine laser–plasma interaction simulations. For more efficient designs of laser-driven high-energy-density experiments, noiseless simulations based on continuum models are required to predict the nonlinear onset and behavior of stimulated plasma instabilities. Continuum Vlasov simulations are very computationally expensive because they require discretization in high-dimensional phases. We will investigate the use of adaptive mesh refinement in phase space and the use of nonlinear, high-order algorithms to reduce the cost and improve the robustness of Vlasov simulations.

We expect to reduce the computational cost of four-dimensional (4D) Vlasov simulations of laser–plasma interactions by two orders of magnitude and the cost of 6D Vlasov simulations by at least three orders of magnitude. This will enable feasible, routine 4D Vlasov simulations (120- to 48-hour runs on 128 processors) and groundbreaking 6D Vlasov simulations on Livermore's massively parallel computers. With the ability to perform these routine simulations, in conjunction with existing particle- and fluid-plasma models, plasma physicists will be able to predict the behavior of nonlinear stimulated-plasma instabilities, leading to more optimal designs of laser-driven high-energy-density experiments at future fusion-class laser facilities.

Mission Relevance

Developing the algorithms required to routinely simulate laser–plasma interactions typical in high-energy-density regimes will advance experiments at future high-power lasers. These high-energy-density experiments have direct application to Laboratory missions in stockpile stewardship, fusion energy research for long-term energy needs, and fundamental science such as astrophysics.

FY11 Accomplishments and Results

We (1) applied adaptive mesh refinement and single-grid codes to physically motivated verification problems, achieving the first-ever demonstration of Vlasov simulation using block-structured adaptive mesh refinement; (2) performed performance studies of the adaptive mesh refinement implementation and developed improved algorithms to reduce mesh refinement overhead, demonstrating promising performance and fidelity improvements; (3) conducted studies on a physically relevant 4D problem using the single-grid code; (4) produced new physics results on transverse particle motion effects on nonlinear electron plasma waves; and (5) wrote publications on algorithmic performance and new physics results. In summary, this project successfully developed and demonstrated new, efficient techniques for direct-discretization Vlasov equations—including the first-ever demonstration of block-structured adaptive mesh refinement for Vlasov simulation—that have already enabled novel physics investigations. Our 4D single-grid Vlasov code has been adopted to investigate higher-dimensional effects in nonlinear plasma wave evolution. A next step would be to investigate instabilities in collision-less shock formation, which would support further development and application of the Vlasov adaptive mesh refinement code.

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Supercomputing-Enabled Transformational Analytics Capability

Celeste Matarazzo (09-SI-013)

Abstract

We propose to develop fundamentally new analytical approaches for cyber security situational awareness for protecting and defending our nation's computing networks. Our supercomputing-enabled transformational analytics capability (SETAC) will focus on transforming the analysis process to provide real-time situational understanding by enhancing our current signature-based analysis with distributed machine-learning behavioral analytics that can be applied as data travels through the network. SETAC requires research in hardware that is powerful enough to apply analytic algorithms to multiple gigabit-per-second data streams, and also requires new analytic algorithms that can provide high-fidelity, real-time behavioral-view of the network through distributed "intelligent agents."

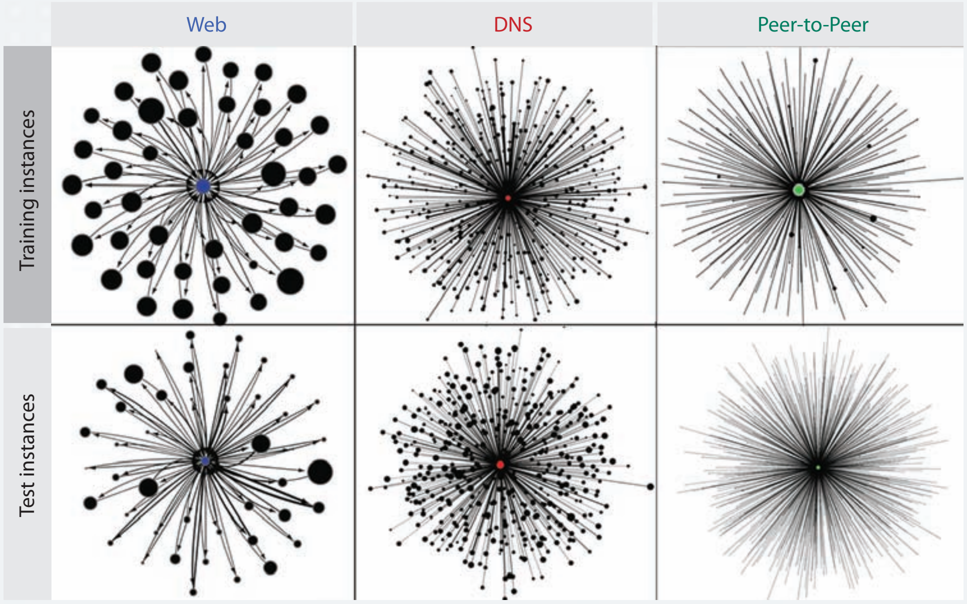
If successful, SETAC will provide real-time behavioral analytics for cyber security data. Our proposed system capabilities will include (1) identification and classification of high-volume streaming network traffic, (2) high-performance sensor nodes to increase processing capabilities and provide distributed detection across the network, (3) a distributed computing test bed to develop advanced global collection and analysis approaches, (4) new analysis algorithms to analyze data across the distributed system, and (5) system- and component-level metrics for performance assessment.

Mission Relevance

This proposal supports LLNL missions in nonproliferation and homeland security as well as supporting efforts in advanced defense capabilities. Our research will enhance Laboratory threat prevention and response technologies by addressing thrust areas in knowledge discovery, advanced analytics and architectures for national security and actionable situational awareness, and information dominance.

FY11 Accomplishments and Results

In the final year of this project we (1) increased data collection to over 1,000 agents deployed throughout LLNL, providing data over an extended period of time; (2) used the Deterlab test bed to create an instrumented malware analysis environment for a detailed understanding of real malware; (3) ported a compact, random forest classifier to field-programmable gate arrays and graphics processing units and applied it to a URL (universal resource locator) reputation dataset from the University of California at Irvine, achieving a rate of 100 million samples per second with the field-programmable arrays and a sampling rate of 22.4 million samples per second with the classifier algorithm running on graphical processing units; (4) developed a series of novel algorithms for network traffic and host classification based on statistical relational learning, community discovery, network structural characteristics, and role discovery; (5) conducted a comparative study of traffic and host classification algorithms on real network traces of relevance; (6) developed a novel algorithm for modeling the behavior of network hosts over time—demonstrating utility for predicting future behavior and anomaly detection—and discovered shared patterns in network-active process usage across all machines, identifying machines with anomalous combinations of activity using our latent Dirichlet allocation algorithm; and (7) coupled dynamic population models for describing the vectors by which a



Connectivity patterns among network hosts can, if properly analyzed, serve as network behavioral indicators. A Web client connects to a number of high-volume servers (left). A DNS server connects to other DNS servers with similar traffic volume (middle). A peer-to-peer host connects to many other low-volume peer-to-peer hosts (right). In this project we developed algorithms for automatically extracting such traffic patterns from network data and using them to infer the behavior of previously unseen hosts. The training instances (top row) were collected from three hosts on a single day and the test instances (bottom row) from three different hosts on another day in a live enterprise network. Node and edge size indicates communication volume relative to the central node in each frame.

threat moves through a compromised network with statistical learning methods for discovering and quantifying those transmission routes, and used this new tool to obtain results for a new class of dynamic population models to characterize the level of compromise suffered by a network under an advanced persistent threat scenario. In summary, our project developed SETAC and demonstrated its suitability for distributed analytics for anomaly detection and cyber situational awareness, providing a robust melding of network traffic classification, host-based learning tasks, and distributed sensor architecture.

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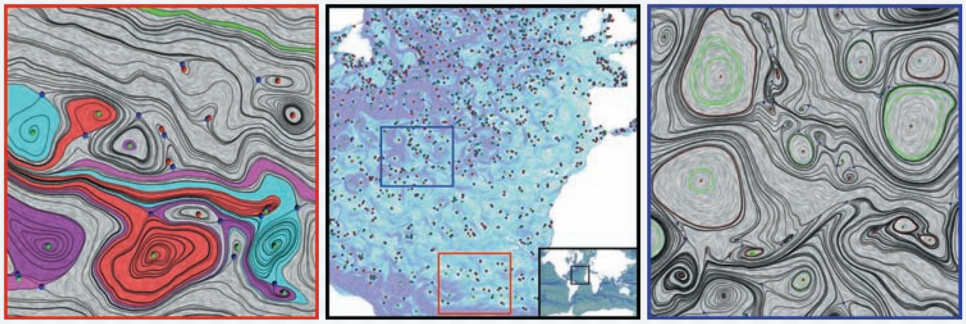
Quantitative Analysis of Vector Field Topology

Peer-Timo Bremer (09-ERD-014)

Abstract

Today's scientific computer simulations produce data of increasing complexity, resulting in an increased need for qualitative and quantitative analysis of complex features of physical phenomena to understand those phenomena in detail. Topology provides powerful tools to define and extract such features. We will focus on techniques to quantitatively analyze vector fields, such as the Earth's magnetic field and water flow in oceans, based on their topological structure. We will develop a mathematical framework for robustly extracting topology from sampled vector fields while maintaining rigorous error bounds. The extracted topology will allow quantitative analysis of topological structures. The hierarchical encoding we develop will allow us to analyze, simplify, and compare vector fields on multiple scales. These techniques will provide opportunities to find explicit definitions for secondary features such as vortices.



Simulation of North Atlantic Ocean currents. The center image is a 600 by 600 tile from the larger simulation of oceanic currents. Images on the left and right are magnified views of the topology for the tile. On the left, black lines represent separatrices grown from all saddles (blue balls) and the colored regions are unstable manifolds grown from all the sources (green balls). On the right, different classes of cycles are shown in different colors.

We will develop the computational theory necessary to generate the complete topology of general vector fields using entirely combinatorial algorithms unaffected by numerical errors. Using metrics tailored to specific application domains, we will construct a hierarchical representation of the topology to permit multiscale analysis. Finally, we will use topological techniques to enhance the ability to track features in dynamic vector fields. The resulting tools will represent fundamentally new analysis capabilities directly applicable to the most ubiquitous form of simulated data.

Mission Relevance

Vector fields best describe many phenomena relevant to Livermore research such as combustion, fluid flow, or magnetic fields. However, because of their inherent complexity, vector fields are often analyzed using derived scalar fields, which only partially describe these processes. Our direct quantitative analysis, simplification, and comparison of the structural properties of vector fields will provide LLNL with unique capabilities to more effectively analyze large simulations relevant to the national security mission areas of stockpile stewardship and global security. Our advances will also lead to new scientific insights.

FY11 Accomplishments and Results

In FY11 we (1) developed a new quantized vector field representation that allows complete extraction of a wide range of topological features, (2) received a best paper award at the IEEE Pacific Visualization conference for work supported by this project, and (3) developed and published a new theory and algorithm for the simplification of two-dimensional Jacobi sets used in differential topology. A Lawrence Scholar fellowship has been awarded to a research collaborator to continue this research as part of several DOE Office of Science projects for large-scale data analysis.

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Rapid Exploitation and Analysis of Documents

David Buttler (09-ERD-017)

Abstract

Exploitation of unstructured document text is vital to the national intelligence mission. Analysts face millions of existing relevant documents and a rapidly increasing volume of new ones that must be quickly assessed. We will develop advanced analytic tools to rapidly triage documents, which will allow analysts to focus on the most important ones. This proposal addresses specific research gaps in identifying concepts and automatically creating a concept hierarchy for summarizing a corpus. We propose to develop cross-language topic hierarchies because documents of interest are often in foreign languages, the translation of which is a major bottleneck. Because analysts are overloaded with streams of documents, we will monitor analyst interests and provide a personal ranking system.

Our research focuses on understanding approaches and techniques that promote efficient and effective discovery and analysis of large document sets given the limitations of current natural language-processing technologies. We will work with intelligence analysts to identify conditions where different methods assist or fail in supporting their work. Furthermore, we will identify a number of targeted research activities that cover technology gaps in document exploitation. In particular, we will explore approaches that assist in the rapid exploitation and analysis of document corpora. We expect to dramatically improve the ability of analysts to discover the most relevant documents for a particular line of inquiry.

Mission Relevance

This proposal is a step towards creating a world-class information exploitation capability for LLNL's missions in national and homeland security by boosting the ability to process the increasingly massive data sets collected for nonproliferation, intelligence, and military missions. We will develop new algorithms to organize vast document sets and alert analysts of new information relevant to their analytic tasks. This capability will have direct applications for Livermore programs in counterproliferation analysis that are of interest to the Department of Defense.

FY11 Accomplishments and Results

In FY11 we (1) created a system to cluster search results by document similarity and user tags, allowing users to manually edit the clusters and enabling fast and large-scale tagging; (2) created an open-source system for word sense disambiguation and extended WordNet; (3) finished integrating our personalization code with our distributed infrastructure, allowing us to run scoring results over large data sets; (4) devised a method to use unsupervised topic models to assist users in formulating queries that are significantly more likely to find relevant results in a target corpus, and created a packaged system that allows the method to be easily deployed; (5) created a system to browse entity co-occurrence relationships by topic; and (6) explored the quality of three popular latent topic models using new semantic coherence measures to inform the choice of fundamental tools and future research directions. This project has provided a wealth of capability in large-scale text analysis that is currently being exploited for different Laboratory missions: the built-out infrastructure has made its way into production systems, and the different research components are providing a roadmap for capability deployment. We have also demonstrated new capabilities to enable analysts to understand large corpora quickly. Our results will guide the next round of production system development and deployment.

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Adding Validation and Novel Multiphysics Capabilities to the First-Principles Molecular Dynamics Qbox Code

Erik Draeger (09-ERD-019)

Abstract

We propose to increase the accuracy and the temperature and pressure range of the Qbox materials simulation code. Qbox currently relies on approximations that can break down at extreme temperatures and pressures. We will integrate Qbox with high-accuracy quantum Monte Carlo codes and develop automated validation tools to quantitatively assess these approximations on the fly. We will also develop a flexible, parallel multiphysics framework to couple different quantum simulation codes to allow researchers to easily implement and test new multiphysics algorithms. Lastly, we will use this framework to develop new multiphysics methods to deal with extreme conditions such as quantum effects of light nuclei and electron exchange and correlation effects.

We expect this project to result in (1) rebuilt, sustainable quantum Monte Carlo capabilities; (2) robust, on-the-fly validation tools for Qbox; (3) a flexible multiphysics framework to simplify development of new multiphysics quantum algorithms; and (4) new multiphysics algorithms to extend predictive materials simulations capabilities. These results will allow us to perform validated, accurate calculations of materials at extreme conditions with unprecedented accuracy. They will also pave the way for new breakthroughs in predictive materials modeling.

Mission Relevance

Predictive materials simulation has long been a cornerstone of NNSA and LLNL missions. Qbox is currently being used for calculations in the Stockpile Stewardship Program and the National Boost Initiative to the limit of its current capabilities. Increasing the accuracy, validation capabilities, and range of accessible conditions would be of direct benefit to national security efforts. As a general materials simulation tool, new capabilities would also benefit many basic research efforts performed at Lawrence Livermore.

FY11 Accomplishments and Results

In FY11 we applied our newly developed path-integral Monte Carlo (PIMC) approach to hydrogen plasma systems and demonstrated that the method can predict the lower-temperature region of the shock Hugoniot, a region previously inaccessible to many-body finite-temperature methods. Because obtaining more sophisticated nodal surfaces from direct linkage to Qbox was found to be computationally prohibitive, we instead developed a method whereby nodal surfaces could be constructed indirectly by analyzing Qbox wave functions and then constructing trial nodal surfaces with augmented basis functions, which are then optimized to minimize the total free energy within a many-body PIMC run. This systematic approach represents a major breakthrough in fixed-node PIMC and could open the door to extending fermion PIMC to materials beyond hydrogen and helium. In summary, this project successfully developed several new algorithms that increase the accuracy and range of application of first-principles materials simulations. These algorithms have already been integrated into an open-source code used by researchers at LLNL and other institutions.

Publications

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Role Discovery in Dynamic Semantic Graphs

Brian Gallagher (09-ERD-021)

Abstract

Role discovery arises in many intelligence and defense applications. Our objective is to develop algorithms that capture the formation and evolution of roles and functions in noisy and incomplete dynamic semantic graphs, which are used to uncover relationships hidden in streams of data from multiple sources. To model this complex process, the need exists for new algorithms that take into account both the heterogeneity and dynamicity in data. These new algorithms will generate statistical models that combine probabilistic predictive models (that handle data heterogeneity) with stochastic process models (that handle data dynamicity). For the first time, this work will unite two modeling categories on graphs. New statistical models will be judged based on their accuracy, run time, and space complexity and tested on publicly available as well as sponsor-provided data.

If successful, our work will provide viable solutions to some of the current analysis needs in cybersecurity as well as homeland and national security. In particular, the work will produce new algorithms that (1) model both the heterogeneity and

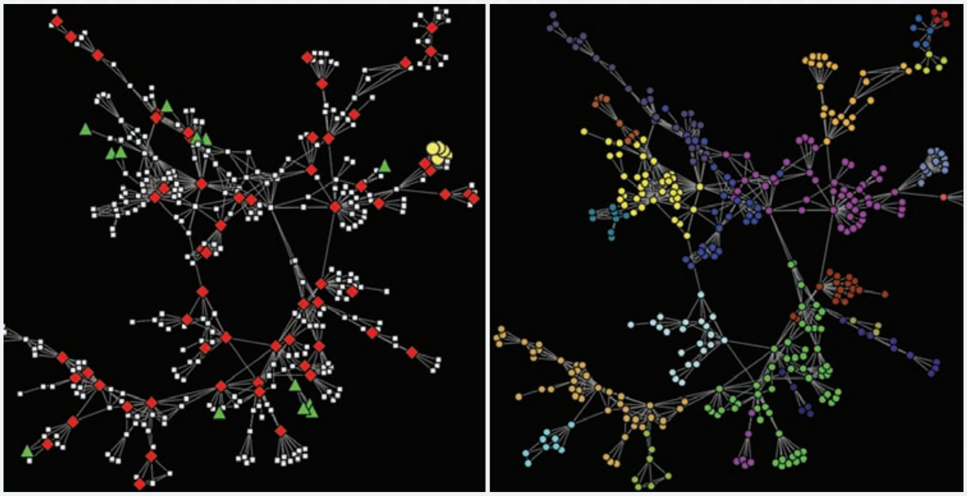
dynamicity in data, (2) allow analysts to directly examine the roles of entities in data sets, and (3) provide insights on the generative mechanisms of relationships. The discovered knowledge can subsequently be used for behavior modeling, anomaly detection, and other graph analytics problems. For instance, in the cybersecurity domain, the work will model the function of hosts over time to detect suspicious activities. These expected results are significant because they provide a different view of the asymmetric threats facing the nation.

Mission Relevance

This work supports LLNL missions in nonproliferation and homeland security and in advanced defense capabilities. By tracking the formation and evolution of roles (i.e., functions) in dynamic semantic graphs, the roles of entities of interest can be directly examined. This abstraction contributes to the understanding of hidden hostile elements and is well suited for identification of the asymmetric nature of threats faced in homeland and national security domains.

FY11 Accomplishments and Results

In FY11 we (1) developed a completely automated and unsupervised algorithm for role discovery in networks, using an approach that captures behavior encoded both in network interactions and individual expression through local attributes—that is, multifaceted information diffusion; (2) extended our role-discovery algorithm to model dynamic temporal behaviors; (3) developed role-based approaches for visualizing and understanding behavior in large networks; (4) developed a novel approach to network transfer learning based on generalizing roles across networks; (5) conducted a comparative study of role-discovery algorithms on network data sets from a variety of problem domains; (6) developed a maximal ancestral graph model-network generator that allows for fast generation of realistic networks and a scalable parameter estimation approach, which we validated against real-world networks; (7) developed efficient algorithms for computing network properties such as



Role discovery and community discovery are complementary approaches to network analysis pursued in this project. Roles capture the behavior or function of nodes and generalize across networks. Communities are densely connected groups of nodes and are specific to a single network. Our RolX algorithm automatically discovers four underlying roles in the Network Science Co-authorship Network (left)—bridge nodes (red diamonds), mainstream nodes (white squares), "path" nodes (green triangles), and tightly knit nodes (yellow circles). Our fast modularity community-discovery algorithm finds 22 communities in the same co-authorship network (right).

eccentricity, radius, and diameter and demonstrated a speedup of up to three orders of magnitude on real-world networks; and (8) completed a working prototype system for role discovery in dynamic semantic graphs. This project has resulted in novel algorithms for automatically learning predictive and descriptive models of behavior in networks and in a deeper understanding of the science behind these algorithms. Our role-discovery algorithm capability has been adopted by multiple mission-relevant projects, and this work is being supported by LLNL's Cyber Defenders Program, by the Knowledge Discovery and Dissemination Program at Intelligence Advanced Research Projects Activity, and through continuing collaborations with Carnegie Mellon, Rutgers, and Purdue Universities.

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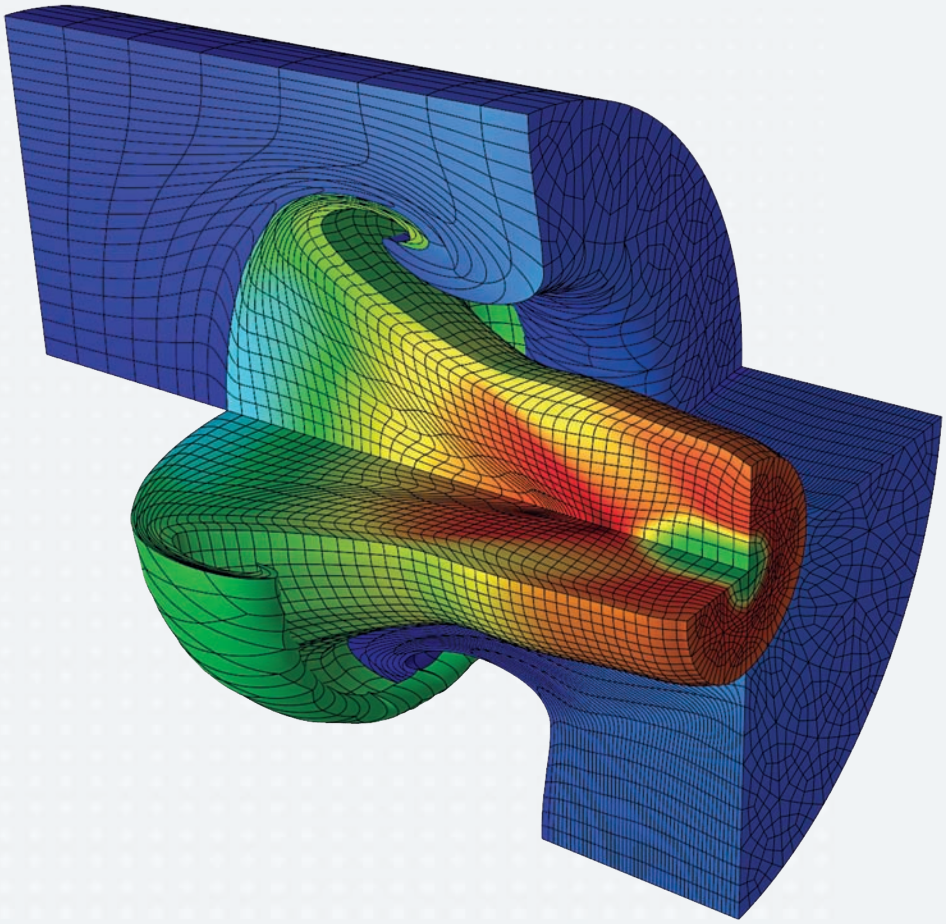
Modern Finite Elements for Lagrangian Hydrodynamics

Tzanio Kolev (09-ERD-034)

Abstract

The objective of this project is to develop new finite-element discretization algorithms for compressible Lagrangian shock hydrodynamics. Our methods will be more accurate and robust than the currently used computational schemes and will address several longstanding issues with the hydrodynamics simulation codes at LLNL, including symmetry preservation on unstructured grids, exact total-energy conservation, artificial viscosity discretization in multiple dimensions, and handling of hourglass-mode instabilities. This will be achieved by the application of modern finite-element technology for discretizing Euler equations on a moving grid, coupled with new mathematical theory and numerical algorithms.

If successful, this project will produce new finite-element discretization schemes that will enable Lagrangian simulations with LLNL's hydrodynamics codes with a higher degree of fidelity than is currently possible. This research could also allow the finite-



Parallel high-order curvilinear finite-element Lagrangian calculation of a shock triple-point interaction on unstructured three-dimensional mesh. Note the robustness of the method with respect to highly deformed three-dimensional zones.

element methodology, which has been very successful for elliptic problems, to be extended to general systems of hyperbolic conservation laws in a Lagrangian frame.

Mission Relevance

Hydrodynamics simulations are of critical importance in numerous LLNL applications, including stockpile stewardship, inertial-confinement fusion, and other mission-relevant efforts. The proposed research will develop finite-element simulations technology to improve the predictive capability of these simulations while requiring fewer user-adjustable parameters. This project therefore supports LLNL's missions in national and energy security.

FY11 Accomplishments and Results

In FY11 we (1) completed our general curvilinear, high-order finite-element framework for solving Euler's equations in a moving Lagrangian frame by demonstrating its application to problems in two-dimensional, axisymmetric, and three-dimensional geometries, as well as to problems involving materials with strength; (2) further demonstrated the benefits of our approach by performing studies with increased polynomial degree in several problems with high vorticity, including velocity-driven Rayleigh–Taylor instability; (3) implemented and tested the new methods in our BLAST high-order finite-element Lagrangian hydrocode and its core open-source visualization libraries MFEM (Multiscale Finite Element Method) and GLVis; (4) demonstrated that the local computationally intensive high-order finite-element approach is ideally suited for future computer architectures by developing a two-layered parallelization of the BLAST routines based on domain-level concurrency employing message passing interface tasks plus a layer of zone-level parallelism on a graphics processing unit; and (5) performed a large number of numerical experiments to investigate performance and scalability of our algorithms. The successful conclusion of this project provided new high-order curvilinear finite-element discretization techniques for more accurate, robust, and reliable Lagrangian hydrodynamic simulations. These techniques will continue to be developed to address components of the full arbitrary Lagrangian–Eulerian framework, including curvilinear mesh relaxation, high-order field remap, and handling of high-order mixed zones.

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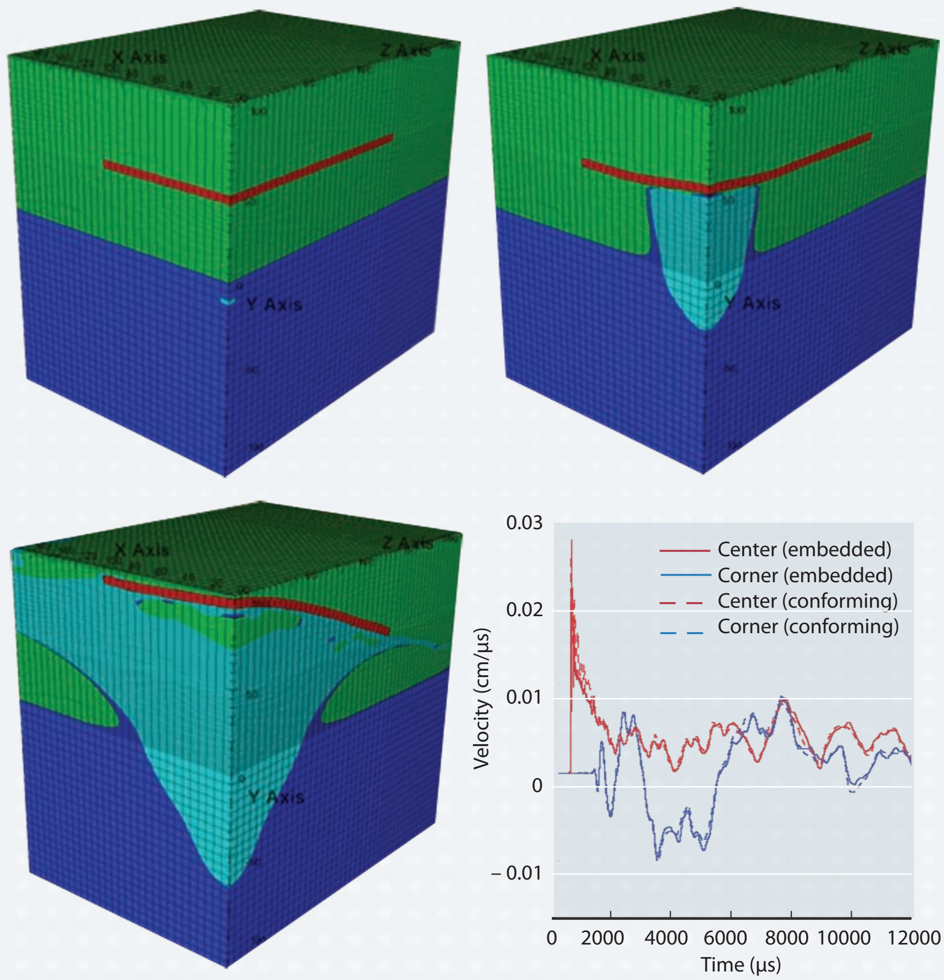
Lagrange Multiplier Embedded-Mesh Method

Michael Puso (09-ERD-044)

Abstract

Many engineering and physics analysis problems could be simplified by applying an embedded mesh. For example, a blast problem with a shell structure overlapping a rectangular air grid typically requires a conforming mesh between the shell and air mesh. However, the analysis may fail because of mesh tangling, even with arbitrary Lagrange–Eulerian (ALE) codes. We propose to develop an embedded-mesh method with a shell (or solid) mesh independent of the rectangular air grid and with appropriate constraints applied at shell–air boundaries. Embedded-mesh methods were originally developed for finite-difference schemes. We will develop a new embedded-mesh scheme that will efficiently allow monolithic coupling for explicit finite-elements.



Simulation of an aluminum plate subjected to a buried TNT explosives mine. Results obtained with a new embedded-grid method are shown along with a comparison of velocity to standard conforming finite-element results, shown in the graph. The plate mesh (red) is superposed on a grid composed of air (green), TNT (light blue), and soil (dark blue). Notice that plate mesh and background grid do not match.

We will develop a computational tool in which the embedded mesh eliminates the need for a conforming mesh and thus requires no mesh motion in the types of problems considered. This general constraint tool will be built to interface with different LLNL codes such as ALE3D and the Diablo ALE hydrodynamics code and will be demonstrated on two model problems: the blast rupture of shell structures and moving solid conductors in air grids. The new method may drastically simplify model building and improve code robustness in many instances.

Mission Relevance

The tools developed in this project will support LLNL's national security and defense mission, such as work involving moving solid conductors for flux-compression generation and rail guns. In addition, the proposed embedded-mesh method has applications for understanding the effects of high explosives, a core Livermore competency.

FY11 Accomplishments and Results

In FY11 we (1) extended our theory for handling fine-foreground and coarse-background meshes by using a multiplier field and constraints defined on the background mesh in place of the foreground mesh, which provided response with robustness that met our requirements; (2) extended our implementation to handle structural shell elements; (3) began extending the implementation to specific, Nédélec-type, finite elements; and (4) verified our new code using conforming mesh models on a variety of blast-related models, reproducing the conforming ALE code results very accurately—results tantamount to passing a standard validation process. This project has led to a new software capability in the production codes PARADYN and ALE3D for simplifying weapons and global security analysis problems by eliminating costly and time-consuming model development and providing more robust solution procedures. This new capability led to a six-year work-for-others project with the Army Research Laboratory for simulating the blast effects of improvised explosive devices on armored vehicles.

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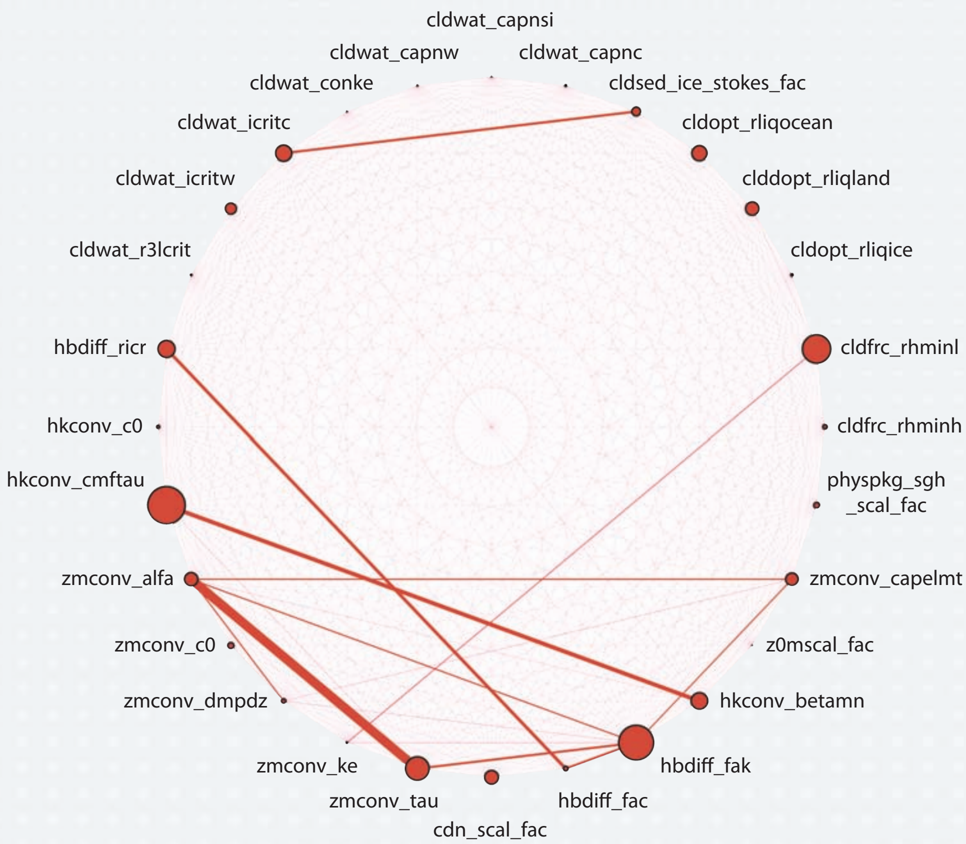
The Advance of Uncertainty Quantification Science

Richard Klein (10-SI-013)

Abstract

Uncertainty quantification deals with the propagation of uncertainty in multi-physics codes that depend in a highly nonlinear fashion on uncertainties in the underlying physics models, algorithms, databases, inputs, and output observables. We propose to build a focused multidisciplinary scientific effort that will investigate, develop, and apply uncertainty quantification science to high-impact scientific areas. We will make major advances in two leading areas of uncertainty quantification—high dimensionality and error and uncertainty propagation—by developing powerful intrusive and nonintrusive uncertainty quantification approaches. Finally, we will use Laboratory applications as a test bed for our new methodologies with an initial focus on climate prediction.

If successful, we will make significant advances in uncertainty quantification science by (1) developing error and uncertainty propagation methods in multi-physics and



The variations in global annual surface temperature caused by changing model parameter values are decomposed and illustrated using a network graph. Node sizes and edge widths are proportional to variance contributions from individual parameters and pairs of parameters, respectively. The graph shows that many important parameter co-variations drive variability in global temperature.

multi-scale codes; (2) tackling the "curse of high dimensionality" with development of novel methods for dimensional reduction; and (3) developing an advanced computational pipeline to enable complete uncertainty quantification workflow and analysis for ensemble runs at the extreme scale with self-guiding adaptation. We will apply these new methods to carry out uncertainty quantification for a coupled ocean–atmospheric model, yielding an adaptively constructed ensemble of simulations for past and future climate; characterize the uncertainty for important global climate variables; and carry out regional uncertainty quantification, with a focus on precipitation and evaporation changes.

Mission Relevance

Our work for this project will support the Laboratory mission in enhancing the nation's environmental security by focusing on reducing uncertainty in the parameters of climate models that cannot currently be constrained by available observations. Our research will also have a large impact in stockpile stewardship by providing methods that will enable increasingly precise uncertainty bounds to be placed on the performance of nuclear weapons. In addition, quantifying and reducing uncertainty in the inertial-confinement fusion design process will optimize target designs so that they are more likely to perform as intended and are less damaging to laser operations.

FY11 Accomplishments and Results

In FY11 we (1) developed error-transport methods for linear advection, the inviscid Burger's equation, Euler's equation, and implicit linear diffusion; (2) developed theory for adjoint error estimation, allowing flexibility in forward and adjoint solutions including linear discontinuities, and investigated error-estimation methods for advection-diffusion equations; (3) developed advanced topological methods for visualizing high-dimensional output; (4) developed new adaptive sample refinement methods, including initial topologically based methods; (5) developed research platforms to implement and compare adaptive sample refinement and dimensional reduction with other uncertainty qualification methods; (6) developed a set of election-voting-based aggregators that aggregate recommendations of developed adaptive sample refinement and dimensional reduction methods, with the goal of supporting automated decision analysis for the uncertainty qualification pipeline; (7) completed a set of studies showing that the performance of the aggregators is approximately the average of the individual adaptive sample refinement methods' performance; (8) performed input-parameters uncertainty qualification on atmosphere and sea-ice models to produce simulations of doubled atmospheric carbon dioxide, thereby characterizing uncertainty in global climate sensitivity, identifying important regional data sets, and quantifying specific regional uncertainty; and (9) began preparing for coupled ocean–atmospheric uncertainty quantification with the Community Earth System Model.

Proposed Work for FY12

In FY12 we will (1) develop adjoint error estimates and direct evolution methods for linear and nonlinear hyperbolic systems of equations and Euler and compressible Navier–Stokes equations; (2) continue researching methods to combine adaptive sample refinement approaches with dimensional reduction methods and demonstrate in a high-dimensional global (atmospheric and ocean) climate model study; (3) research techniques to develop an integrated codex for the combined adaptive sample refinement and dimensional reduction methods as a tool for decision analysis, and develop metrics that facilitate the decision feedback mechanism in the decision analysis engine; (4) apply the decision analysis engine to climate model study; and (5) carry out uncertainty quantification for the coupled ocean–atmospheric model and characterize the uncertainty for important global climate variables.

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ExaCT: Exascale Computing Technologies

Bronis de Supinski (10-SI-014)

Abstract

We propose to prepare for the large-scale supercomputer systems anticipated in the near future, with capabilities of multiple petaflops (one quadrillion floating point operations per second) and, in the longer term, of exaflops (one quintillion floating point operations per second) total performance. This will herald a new era of predictive simulation. Specifically, our ExaCT (exascale computing technologies) project will dramatically improve the scalability and performance of Laboratory applications,

particularly in the presence of much more frequent soft faults such as bit flips, through innovative algorithms and automated adaptation to systems with huge numbers of nodes, each with much less memory and less memory bandwidth per core. Overall, we will create fundamentally new approaches not only to algorithmic design, including integrated fault-tolerance strategies, but also to debugging and large-scale performance automation.

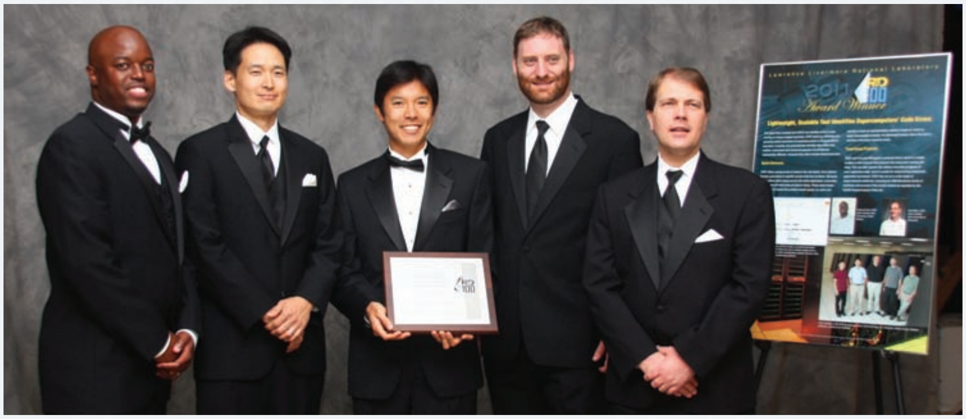
Our ExaCT project will produce tools and strategies to diagnose and overcome difficulties arising in large-scale supercomputer systems. We will (1) identify models of fault vulnerability and design strategies to overcome it, (2) design and develop sparse linear solvers to address memory constraints, (3) explore automated code transformations that reduce memory bandwidth requirements and asynchronously compute load redistributions to improve performance on millions of cores, and (4) investigate a debugging methodology to locate the source of programming errors automatically and to track algorithmic behavior. Our work with application scientists will demonstrate enhanced productivity, enabling effective use of large-scale systems and thus providing critical predictive simulation capabilities.

Mission Relevance

This research supports LLNL's broad national and energy security missions by providing the needed underpinnings for predictive simulation on large-scale supercomputers through extensions to our world leadership in algorithmic design and systems software and tools and by directly applying them to simulation applications for materials modeling, fusion energy science, and the physics relevant to stockpile stewardship.

FY11 Accomplishments and Results

In FY11 we (1) refined our detailed model of the performance of sparse linear-solver algorithms and developed resilience strategies for these algorithms based on the impact on their convergence of soft errors, (2) developed a novel approach based on Markov models of application control flow to identify the source code region in which errors occur in message passing programs, (3) developed a framework



The STAT (stack trace analysis tool), partially funded by LDRD exascale computing technologies research, won an R&D 100 Award. Pictured are team members Dorian Arnold, Dong Ahn, Gregory Lee, Matthew LeGendre, and Bronis de Supinski. Not shown: Barton Miller and Martin Schulz.

that asynchronously redistributes computational loads and continued our work on detailed statistical metrics for the relationship between measured application load and application-centric load models, and (4) implemented checkpoint-compression tools that exploit relationships between applications and data. In addition, we won an R&D 100 Award for our work on STAT (stack trace analysis tool), which was partially funded by this project.

Proposed Work for FY12

In FY12 we will (1) develop a benchmark for multicore-aware, sparse linear solvers; (2) demonstrate the automated root-cause analysis of coding errors in message-passing interface programs at scale; (3) demonstrate techniques that asynchronously redistribute computational load to our test bed applications; and (4) develop and implement an asynchronous-tree-based checkpoint offload engine as another load-redistribution strategy.

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Parallel Discrete-Event Simulation of Cyber Attack and Defense Scenarios and Automated Rollback Code Generation

David Jefferson (10-ERD-025)

Abstract

Widespread cyber attacks that overwhelm Web sites are a prime national security concern. A powerful 2009 attack that targeted dozens of government and private sites underscore how unevenly prepared the U.S. government is to block such multipronged assaults. We propose to develop a prototype discrete-event simulation using the NetWarp platform for executing cyber attack and defense code on a network of virtual machines. The code will be synchronized using a discrete-event simulator to accurately replicate, for the first time, the timing of events in the modeled system. In parallel, we will develop the BackStroke system to automatically generate a companion method that exactly reverses unwanted side effects of specialized computer algorithms. This is a capability critical for "optimistic" parallel discrete-event simulation and will be developed using LLNL's ROSE compiler technology.

We expect to develop NetWarp as the first simulation platform to enable realistic studies of cyber attack and defense scenarios that are scalable and accurate in both

behavior and performance. It will enable ensemble studies to optimize cyber defense systems against an array of attack codes and do so without prior understanding of how the attack codes work. By developing the BackStroke system, we will dramatically lower barriers to the use of optimistic parallel discrete-event simulation techniques. Programmers will not be forced to write inverse methods for every forward-event method, which doubles the amount of code required, nor will they face the extremely difficult debugging required when inverse methods are hand generated.

Mission Relevance

Our project supports the Laboratory's mission to reduce or counter threats to national security by allowing us to simulate cyber attack and defense scenarios much more accurately and speedily and in some cases at a larger scale than ever before. In addition, these simulations can be done early, before the attack code is even understood. The BackStroke system will support several Laboratory missions because it is a fundamental improvement to optimistic parallel discrete-event simulation for most applications, especially network simulation.

FY11 Accomplishments and Results

We (1) continued our proof-of-concept implementation of NetWarp on Linux clusters at small scale; (2) demonstrated the running of NetWarp on a mixed Windows and Linux network; (3) began investigating the possibilities of the Qemu virtual machine monitor, which has a notion of virtual time built into it, as an alternative to Xen and demonstrated that we can scale a simple Qemu virtual network to 1,000 nodes; (4) demonstrated the deterministic repeatability of NetWarp simulations; (5) achieved a deep understanding of the semantic issues in C++ as they relate to BackStroke and how the issues affect the possibility of generating reverse code for C++ functions such as classes, inheritance, polymorphism, and dynamic allocation; (6) developed sophisticated new algorithms for reverse-code generation with scalar data; and (7) added several program analysis tools to ROSE to enable reverse computation.

Proposed Work for FY12

In FY12 we will (1) complete our proof-of-concept implementation of NetWarp to run a large virtual network of greater than 1,000 nodes, although with a greater than tenfold slowdown; (2) conduct extensive performance testing and validation on small networks to verify that NetWarp duplicates the behavior of an actual network and measure how close the timing values on a simulated network come to actual values on a real network; and (3) integrate BackStroke with a parallel discrete-event simulator and demonstrate that it enables both easier optimistic simulation development and faster runtime performance than other optimistic methods and that it can create an inverse method for almost any realistic C++ forward method.

Publications

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CgWind: A Parallel, High-Order Accurate Simulation Tool for Wind Turbines and Wind Farms

William Henshaw (10-ERD-027)

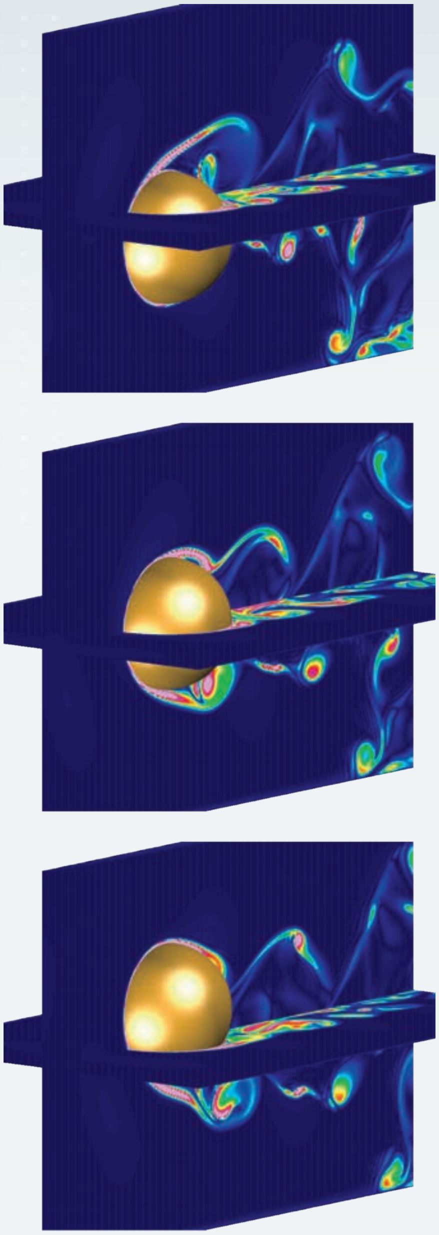
Abstract

Our objective is to develop a next-generation computational tool, called CgWind, for the high-fidelity simulation of wind turbines with rotating blades and for the simulation of arrays of turbines in a wind farm, including the effects of topography and regional winds, through coupling to mesoscale atmosphere models. CgWind will solve the incompressible Navier–Stokes equations and will be based on the development of high-order accurate, compact approximations; nonlinear large-eddy simulation models for turbulent flows; adaptive mesh refinement; matrix-free multi-grid algorithms; and parallel moving-grid-generation algorithms.

We expect that CgWind will be used by the wind turbine community to design, optimize, and predict the performance and power output of wind turbines and wind farms for energy generation. This tool will be orders of magnitude faster than existing approaches and will enable simulations that will provide new insight into the physics of these complex turbulent flow problems. CgWind, by bridging the gap between the larger scale atmospheric modeling being performed at LLNL and turbine-scale flows, will be an important component of an end-to-end wind-modeling capability. As part of this work, we will also develop new mathematics and algorithms, such as fast, high-order accurate parallel matrix-free multi-grid algorithms and fast, parallel moving-grid-generation algorithms.

Mission Relevance

This proposal supports the Laboratory's energy and environmental security mission by enabling and enhancing development of a clean and renewable wind energy source. Our research also addresses the grand challenge goal of simulating high Reynolds-number incompressible turbulent flows around complex and moving geometries and the multiscale coupling of very large-scale flows (meteorological) with very small-scale flows (turbine-blade boundary layers) in support of LLNL's cutting-edge science, technology, and engineering in the area of high-performance computing and simulation. The new parallel, high-order accurate matrix-free multi-grid algorithms and moving-grid-generation algorithms developed under this proposal will impact various other simulation fields.



Simulation of an incompressible flow past a sphere oscillating up and down, computed with new parallel moving algorithms developed as part our CgWind computational tool. Contours of the vorticity are shown at three different times.

FY11 Accomplishments and Results

We (1) continued developing high-order compact schemes that accurately treat no-slip wall boundary conditions, as well as support for parallel computations and moving grids, by producing a new class of compact operators that treat first- and second-order derivatives in a more uniform way; (2) continued developing a high-order multi-grid solver, including accurate treatment of Neumann- and Dirichlet-type boundary conditions in three dimensions, achieving the first-ever fourth-order-accurate and parallel multi-grid solver for overlapping grids; (3) improved parallel grid generation for moving-grid problems, with promising results for solution of the difficult task of generating overlapping grids for parallel computers; (4) developed and incorporated large-eddy simulation turbulence models for overlapping grids; and (5) performed demonstration calculations of a unique capability to model moving turbine blades in parallel.

Proposed Work for FY12

In FY12 we will continue each of our ongoing lines of work, including (1) developing high-order compact schemes that will focus on improving performance of the approximate factored compact scheme, and validation of this using a number of configurations relevant to wind energy; (2) work on a high-order multi-grid solver, which will involve efforts to improve efficiency and robustness for moving grids; (3) improvements for moving-grid problems that will include developing grid-generation capabilities specific to wind turbine blades and work on the speed and efficiency of grid-generation algorithms for configurations typical of wind turbine applications; and (4) developing large-eddy simulation turbulence models for overlapping grids that focus on incorporating selected models into the approximate-factored compact scheme and validation of the accuracy of these models.

Publications

Chand, K. K., and M. A. Singer, 2011. *Verification and validation of CgWind: A high-order-accurate simulation tool for wind engineering.* 13th Intl. Conf. Wind Engineering (ICWE13), Amsterdam, the Netherlands, July 10–15, 2011. LLNL-CONF-483351.

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Chand, K. K., et al., 2010. *CgWind: A high-order-accurate simulation tool for wind turbines and wind farms.* 5th Intl. Symp. Computational Wind Engineering (CWE2010), Chapel Hill, NC, May 23–27, 2010. LLNL-CONF-424863.

Chand, K. K., et al., 2011. *Implicit high-order compact schemes for incompressible flow.* SIAM Conf. Computational Science and Engineering, Reno, NV, Feb. 28–Mar. 4, 2011. LLNL-PRES-471491.

Binary Analysis

Daniel Quinlan (10-ERD-039)

Abstract

We propose to develop new binary analysis techniques required for analysis of software and, in particular, for evaluation of specific behavioral properties that are critical for cyber security classification software. Our work will also address the use of parallel machines for the analysis of binaries and will result in tools for the analysis of binaries, comparative analysis, evaluation of properties, and application of formal-method technologies. We intend to leverage existing work in LLNL's open ROSE compiler infrastructure to support the development of specialized mixed static and dynamic analysis tools. This work will be used to build new forms of binary analysis and support domain-specific binary analysis tools.

We expect to develop new tools, new forms of binary analysis, external collaborations with industry and government, and new opportunities related to programs in cyber security. Our work is significant for the Laboratory and others throughout the DOE and elsewhere in the government. Our new tools will allow for a deeper look into binary forms for software and provide transparency to commercial off-the-shelf software—the most dominant software use within government and industry.

Mission Relevance

Our proposal is directly relevant to LLNL's strategic mission thrust in cyber and space security and intelligence. Our proposed research is central to cyber security, building a research-level of expertise in binary analysis at LLNL and especially evaluation of software and its use on Laboratory networks. An ability to analyze the software that defines network traffic will be key to understanding and predicting network behavior and providing situational awareness of ongoing network attacks in the future. Furthermore, the general identification of properties within binary software is one of a range of ways of establishing bounds on its capabilities.

FY11 Accomplishments and Results

We continued our work with the Department of Homeland Security's Computer Emergency Readiness Team (CERT) and our focus on tools. Specifically, we

(1) developed new techniques to support analysis of untrusted binaries, including formal-methods-based approaches supporting a new level of disassembly and verification of binary executables; (2) developed new methods to support the static analysis of binaries using concrete and symbolic techniques, creating techniques to capture the local semantics of the binary executable; (3) developed emulation techniques to support more advanced static analysis and mix of both the static and dynamic analysis of binary executables; and (4) developed techniques to rewrite the binary representation in ROSE to make it structurally inert and non-executable.

Proposed Work for FY12

In FY12 we will (1) continue our analysis of samples in the CERT database using some of the new tools built in FY11, and on function identification for the binary executable files, which we will then compare with alternative analysis tools; (2) analyze samples from the CERT database with LLNL supercomputers to use new forms of analysis that mix static and dynamic methods developed in FY11; (3) define new formal-methods analysis technologies, building on data-flow technologies being developed for the ROSE compiler software, which will be important for identifying security properties if successfully applied to binary executable files; and (4) continue to work with CERT and focus on tools to support their work, which can be provided to their collaborators elsewhere within the government.

Publications

Saebjoernsen, A., Matzke, R., and Quinlan, D., 2011. *A practical mixed analysis framework for arbitrary binary fragments*. 16th Intl. Conf. Architectural Support for Programming Languages and Operating Systems (ASPLOS-XVI), Newport Beach, CA, Mar. 5–11, 2011. LLNL-CONF-512886.

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Uncertainty Visualization

Peter Lindstrom (10-ERD-040)

Abstract

We will develop new techniques for management and visualization of high-dimensional and high-volume data sets generated in petascale ensemble runs for uncertainty quantification. Our techniques will enable compressed data storage for post-simulation analysis without expensive reruns. This will provide unique visualization capabilities for online monitoring of ensembles to aid uncertainty

quantification parameter sampling for exploration of high-dimensional merit functions and for displaying probability distributions resulting from invasive uncertainty quantification techniques. Our approach relies on wavelet statistics for concise feature description, data clustering, and highly compressed storage, and on statistical summaries and new spatiotemporal interpolation schemes for visualizing complex and uncertain data.

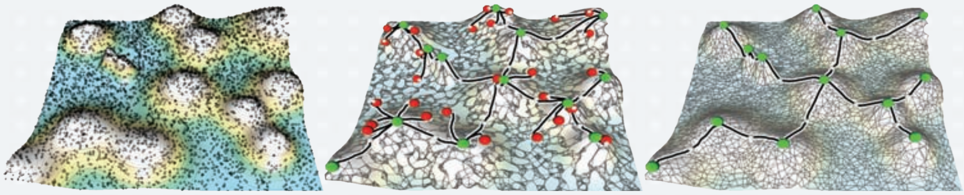
We expect to enable offline data analysis and evaluation of new merit functions with two-orders-of-magnitude lossy but error-bounded compressed storage of time-varying simulation fields. Our methods will provide the first-ever automated tools for selective visualization of the range of behavior in huge ensembles to facilitate decision making with regards to early termination of runs and uncertainty quantification parameter selection, thereby reducing wasted computation time. We will also provide visual aids for understanding the complex relationships between high-dimensional input and output spaces and how sensitive the outcome of a simulation is to individual or combined parameter settings. In combination, these techniques will provide entirely new capabilities critical to the success of the Laboratory's uncertainty quantification efforts.

Mission Relevance

Uncertainty quantification is a critical component of computer simulation across many mission-relevant applications, with an identified need in stockpile stewardship, climate modeling, and many other applications of high-performance computing. By helping to create an advanced capability to manage, analyze, visualize, and ultimately gain insight from the complex and large quantities of data generated in uncertainty quantification codes, this project supports the Laboratory's missions in national and energy security.

FY11 Accomplishments and Results

In FY11 we (1) developed a structure-preserving topological representation of high-dimensional scalar functions for visualization and analysis; (2) devised a new, robust approach to defining neighborhoods for sparsely and irregularly sampled high-dimensional data; (3) applied these neighborhood graphs in spectral clustering



Irregularly sampled two-dimensional scalar function (left). The detected true (green) and false (red) maxima and ridges connecting them given by six-nearest-neighbor (middle) and relaxed Gabriel (right) neighborhood graphs. Our relaxed neighborhood definition ensures robust detection of topological features by eliminating the false maxima (red) implied by other, denser neighborhood graphs.

together with nonlinear diffusion to improve the quality of solutions to both low- and high-dimensional classification problems; (4) designed a new diagram for extracting and visually presenting nonlinear correlations between numerical and categorical data based on information-theoretic quantities; (5) invented a fast algorithm for numerically robust contouring of piecewise quadratic scalar functions; (6) developed a method for locating critical points of functions on Riemannian manifolds embedded in high dimensions; and (7) devised density estimation techniques for piecewise linear and quadratic functions.

Proposed Work for FY12

In FY12 we will (1) complete work on visualizing and analyzing distribution fields for qualitative and quantitative assessment of spatial uncertainty, (2) develop compressive sensing techniques for the online compression of spatiotemporal data, (3) design optimal rectilinear-distance reconstruction bases for a compressed representation of climate and other application data, (4) investigate applications of relaxed and stochastic empty-region graphs such as high-dimensional clustering and gradient estimation, and (5) develop a theoretical foundation for "gradient graphs" that correctly encode the topology and segment the domain of high-dimensional scalar functions.

Publications

Correa, C., and P. Lindstrom, 2011. *Improving spectral clustering with empty region graphs*. LLNL-TR-513768-DRAFT.

Correa, C., and P. Lindstrom, 2011. *The mutual information diagram for uncertainty visualization*. IEEE VisWeek Working with Uncertainty Workshop, Providence, RI, Oct. 24, 2011. LLNL-CONF-513738.

Correa, C., and P. Lindstrom, 2011. "Towards robust topology of sparsely sampled data." *IEEE Trans. Visual. Comput. Graph.* **17**, 1852. LLNL-JRNL-479881.

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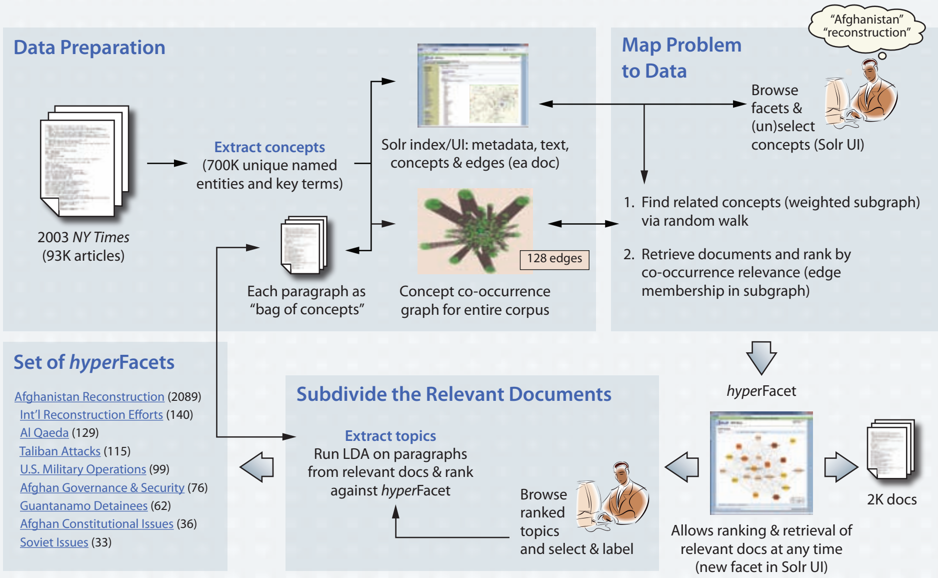
M3Net: Taxonomy Construction and Topic Mapping for Multimedia Message Manager Traffic

Douglas Poland (10-ERD-041)

Abstract

Intelligence analysts need to discover, access, and exploit intelligence information in a secure, tailored manner. Our objective is to connect analysts' domain models to multimedia message manager (M3) traffic in a way that allows them to more effectively exploit this vital data source. We are motivated by recent research in topic extraction, lexicon and taxonomy development, and faceted search. A pathway is needed for putting the power of these new text analytics at analysts' fingertips. To this end, we will construct a taxonomy known as M3Net from M3 data and develop a capability for dynamic updating. We will then develop a methodology for mapping analyst models, such as the existing FactWeave or Elecent software tools. Archived and incoming messages will be indexed via M3Net, and the mapped models will then facilitate creation of domain-specific meta-indices that are instantiated as personalized facet structures.

We expect to develop an intuitive, multifaceted view of message traffic derived from analyst models of those messages for the intelligence community. Instead of a keyword-filtered list of hundreds of messages per day, our M3Net will connect analysts to the data they need and enable them to focus on higher-level analytics. If successful, this methodology lends itself to fusion with other data sources and is a step towards plug-and-play model and data compatibility.



A flowchart describing the generation, in minutes, of a set of hyper-dimensional facets realized as weighted sub-graphs that combine the results of graph analytic and topic modeling operations. The combined results—in this example, *New York Times* reporting in 2003 on reconstruction in Afghanistan—are then organized and ranked.

Mission Relevance

This project supports the Laboratory mission in national and global security. The success of M3Net will be a significant step forward in the Laboratory's mission thrust in cyber and space security and intelligence, particularly enabling end users to efficiently exploit intelligence information. In addition to advancing a more effective and efficient analytic paradigm at LLNL, with direct benefits to ongoing programs and sponsors, this project would establish the Laboratory as a leader in the development of new analytic technology that will be needed for national security.

FY11 Accomplishments and Results

In FY11 we (1) conducted experiments on facet generation, determining that reliance on a generic language model leads to both rigidity and noise that cannot be mitigated; (2) addressed deficiencies in lexicon acquisition and taxonomy mapping, learning that by combining lexicon acquisition and co-occurrence graph construction, we can bring very powerful graph analytics to bear in the search for related documents; and (3) built on the preceding lessons to construct a novel way for analysts to interactively generate arbitrarily complex, quantifiable facets by first transforming each paragraph in the corpus into a bag of keywords and phrases, constructing links for each in-bag co-occurrence, adding the links to a standard faceted search index as properties of the source documents, and then using the faceted search application as a front end for graph analytic and topic model algorithms. These algorithms may be employed iteratively to generate weighted subgraphs ("hyperfacets") that can be employed to rank documents, both old and new. The ideas and expertise developed in this project represent a unique capability—the interactive generation of complex quantitative facets—and will continue to be proposed to end users needing rapid, deep analysis of large collections of text.

Eigensolvers for Large-Scale Graph Problems

Van Henson (10-ERD-054)

Abstract

The eigenvalues and eigenvectors of graph matrices used in computer science are crucial to several applications—ranking graph nodes, clustering, identifying communities and common interests, and computing message times between nodes. Using multilevel technology, we will produce scalable parallel algorithms for computing the most critical eigenvalues and eigenvectors of matrices of extremely large graphs (with billions of vertices) and for swiftly updating the eigensystems as the underlying graphs rapidly evolve. For the former, we will use graph characteristics such

as power-law distributions to generate optimal search strategies and starting points. For the latter, we will restrict the computation to a carefully chosen *d*-dimensional subspace to capture the most significant variations in two-dimensional operations.

We expect to produce new mathematical knowledge about how the eigensystems of graph matrices are found and how they evolve, and about the nature of graphs arising from networks and information-processing applications. We will produce research software implementing the scalable algorithms on large Livermore computer clusters such as Hera, develop an understanding of implementations for the BlueGene family of systems, and explore the possibility of implementing an eigensystem-updating algorithm under a streaming computing infrastructure. When hardened and produced, these tools will enable the application of popularity, clustering, community identification, commute time, and other important analytical tools for graphs whose sheer size have heretofore precluded such treatment.

Mission Relevance

Large-scale graphs are used in many cyber security and intelligence applications involving, for instance, the Internet, network intrusions, funding flows on financial networks, email message traffic, power grids, information propagation, information retrieval, and data mining, which this research can impact importantly. The fundamental technology we develop can be transferred and developed for a host of related applications in data mining and analysis, such as the singular-value decomposition, principal component analysis, low-rank factorization and approximation, and linear and logistic regression analysis. These applications are of great interest to external sponsors working in cyber security and intelligence.

FY11 Accomplishments and Results

In FY11 we (1) perfected and implemented the scalable MatVec (matrix–vector multiplication) developed last year, and used it to produce eigenpairs for large graphs—our largest run on 768 cores of the Ansel system at LLNL, took 5 hours to produce 21 converged eigenpairs on a graph with 125 million vertices and 1 billion edges, the largest eigenpair calculation we are aware of on scale-free power-law graphs; (2) developed disaggregation as a parallel accelerator for MatVec and employed it to implement a multilevel eigensolver; (3) developed a theory on locally supported eigenvectors and made considerable progress toward parallelization of the multilevel eigensolver with a code design and partial implementation; and (4) performed a thorough study of low-rank approximations to commute time and are documenting the resulting theories we developed.

Proposed Work for FY12

For FY12 we will (1) continue researching multilevel methods, finding effective aggregation schemes; (2) complete parallelization of our multilevel solver; (3) determine whether low-rank approximations of the Laplacian matrix of a graph can be used to accurately compute commute times and implement the approximations in codes that can be used in allied projects; (4) work with Sandia National Laboratories to implement and analyze low-rank updating methods for eigensolvers and tensors on the Cray XMT shared-memory supercomputer; (5) determine the efficacy of rapidly updating eigenspectra for evolving graphs; and (6) begin to use the technology developed in this project in actual mission-relevant applications at LLNL.

Publications

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Unifying Memory and Storage with Persistent Random-Access Hardware

Maya Gokhale (11-ERD-008)

Abstract

Today, the ten-thousandfold difference in latency between disk and main memory creates a dichotomy between memory-based objects and external files. Flash memory reduces that latency to a factor of 1,000, but most likely cannot serve as slower dynamic random-access memory. We will develop a programming model and system software to use low-latency, random-access, persistent memory in computation nodes. Our new approach is to unify transient and persistent memory using flash memory and phase-change memory, a new persistent-memory technology similar to dynamic random-access memory. Using hybrid flash and phase-change memory subsystems, we will design, prototype, and evaluate software systems that remove the distinction between program variables in transient memory and external data files, enabling high performance in concurrent large-memory algorithms on future many-core computation nodes.

If successful, our proposed persistent-memory approach will have a transformative impact on computer programming by enabling highly multithreaded applications to access persistent objects in dynamic random-access memory as easily as transient data structures, increasing system reliability and greatly reducing the burden of inserting fault tolerance into the computing system, and enabling information science algorithms to operate on very large data structures without having to use out-of-core or distributed memory techniques, reducing the number of nodes required for the task. For instance, our proposed architecture and system software would enable graph algorithms used in monitoring applications to run on individual many-core computation nodes having large, persistent-memory subsystems with better performance than on clusters.

Mission Relevance

This project supports LLNL's national security mission by developing a transformative memory technology for many applications relevant to national security such as graph processing for intelligence-gathering applications. It also supports LLNL's exascale initiative by designing and evaluating a fault-tolerant, scalable node architecture incorporating node-local storage in the memory hierarchy.

FY11 Accomplishments and Results

We (1) wrote a new simulation device driver to measure the quantitative performance of future persistent-memory storage arrays, (2) used the driver to study and optimize performance of two applications—a metadata store manager and an asynchronous graph-traversal framework—as they process very large data sets in simulated persistent memory, (3) collaborated with the University of California at San Diego to design the hardware architecture of next-generation storage controllers that include a processor to perform application-specific computing in place, and

(4) incorporated a new checkpoint method using memory-mapped checkpoint files into two applications—a parallel molecular dynamics simulator LAMMPS and a hydrodynamics simulation application. Our graph traversal algorithm using a flash storage array and one compute node won seventh place in the international Graph500 competition, in contrast to all other entries that used traditional supercomputers with thousands of nodes. Our work in storage processor design resulted in a record of invention, and we are incorporating the new checkpoint mechanism into a production code.

Proposed Work for FY12

In FY12 we will (1) improve and further validate the persistent-memory simulator, and continue to transition the code base into a more general memory-mapped fault handler; (2) develop a dynamic-memory allocator for persistent memory, incorporate the allocator into two data-intensive applications, and optimize performance of the allocator; (3) develop two application-specific modules (data stream filter and priority queue manager) that run on a persistent-memory controller; (4) improve our graph algorithm for large-scale graphs; and (5) incorporate the memory-mapped checkpoint file into the scalable checkpoint and restart library.

Publications

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Compressive Sensing for Wide-Area Surveillance

Paul Kidwell (11-ERD-022)

Abstract

With this project we will, for the first time, address and 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 the quality of the refined intelligence produced.

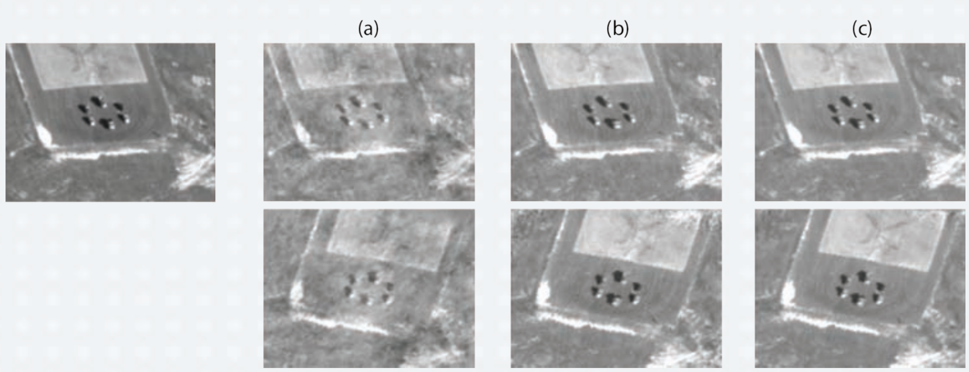
If successful, this project will help achieve the data-compression factors in multispectral data sets that are necessary to transmit surveillance data with current bandwidth-limited technologies and to reduce the onboard processing costs of wide-area surveillance. Using compressive sensing, we expect 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.

FY11 Accomplishments and Results

We (1) completed modeling and simulation for generating synthetic wide-area data sets, both with and without platform motion, and obtained measurements of compression potential in settings with a two-dimensional (2D) and 3D wavelet basis;



We reconstructed a sequence of 600 video frames with varying ratios, and determined that compressive sensing recovery of unmanned aerial vehicle video is possible at compression ratios of more than 160:1. The top row shows the initial frames at varying compression, with the "ground truth" image collected on location in the upper left corner. The bottom row shows the last frame at the same varying compression ratios. The ratios shown left to right are (a) 667:1, (b) 167:1, and (c) 20:1.

(2) demonstrated the reconstruction quality of 23-dB peak signal-to-noise ratio at a compression ratio of 167:1 using 2D wavelets under translation-based video motion, also finding that higher compression ratios would be possible with 3D wavelets if the video motion could be correctly predicted; (3) studied "noise folding" effects on reconstruction from compressive measurements; and (4) designed two strategies for motion prediction: progressive encoding for coarse reconstruction, from which to estimate motion parameters for full reconstruction, and an iterative method to jointly reconstruct data and refine motion parameters.

Proposed Work for FY12

Efforts in the second year will address technical challenges with processing real data sets. Specifically, we will (1) acquire short- and mid-wave infrared video data sets and perform upfront sensor calibration and image conditioning to compensate for fixed pattern defects, image vignetting, and non-uniformities; (2) apply existing camera stitching and video stabilization pipelines to the color video data to produce full field-of-view stabilized data sets; (3) perform compressive sensing of the foreground channel of a stabilized video data set and compare with the results from compression by a video encoder; (4) survey a variety of feature and mutual information and entropy-based methods to register various types of images into a common reference coordinate system; and (5) examine the feasibility of adapting existing methods and developing new techniques for stabilizing multi-spectral video data.

Data Abstractions for Portable High-Performance Computing

James McGraw (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 ported to newer supercomputers without requiring major rewriting and while maintaining performance.

This research will result in C++ code data abstractions relevant to Advanced Simulation and Computing codes that, when implemented with the ROSE compiler, automatically re-write 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.

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 our explored notations and transformations to the actual control and data structures of important Advanced Simulation and Computing codes and will focus on current and explicitly proposed exascale machines.

FY11 Accomplishments and Results

In FY11we (1) examined existing array notations for use in this project, concluding that the A++ array notation language (already in ROSE) would be adequate, as well as created nine small codes using A++ for testing basic layouts and developed several automated layout transformations using ROSE; (2) developed the necessary legality checks to ensure that new layouts would result in correct code; (3) applied transformations to the nine test codes and ran them on three computer platforms—results demonstrated that changing the data layout can have significant impact on performance, with factors of improvement ranging from 3.39 to 4.46, which show that automatic transformations of data layouts are feasible and will allow significant flexibility in tuning codes for performance; and (4) wrote two new versions of the unclassified code LULESH (Livermore Unstructured Lagrangian Explicit Shock Hydrodynamics)—the rewrite to array notation was done to create a more complex code for experimentation, and LULESH was reduced from 2888 to 1637 lines and was significantly more readable. The second new code version created a more direct scheme for computing "force" that will expose far more parallelism and introduce no extra synchronization. Both codes will enable significant parallel platform studies in FY12.

Proposed Work for FY12

We will (1) revise and extend the data abstractions to handle other key physics algorithms, (2) define and implement optimization techniques and directives to demonstrate the range of data layouts that can be supported for these data abstractions, and (3) measure the performance gain achieved by flexible data-layout options.

Publications

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

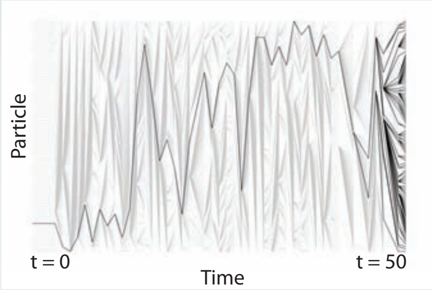
Adaptive Sampling Theory for Very-High-Throughput Data Streams

Daniel Merl (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 represented in the form of statistical learning algorithms—of novel information theoretic approaches for adaptive sub-sampling of very-high-throughput data streams to effect orders-of-magnitude increases in ingestion rates for filter-based learning algorithms. This will serve as a key component of analytic surveillance systems and will be accomplished while both minimizing the effects of uncertainty introduced by sub-sampling as well as maintaining mathematical guarantees of estimation consistency.



Our research borrows ideas from evolutionary biology to develop new computational statistical methods in which prediction accuracy is used as the selective pressure driving novel sequential Monte Carlo algorithms.

Mission Relevance

This research will provide a suite of capabilities that will support virtually all manner of large-scale streaming data analysis at LLNL, and in particular supports the Laboratory's strategic cyber, space, and intelligence 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.

FY11 Accomplishments and Results

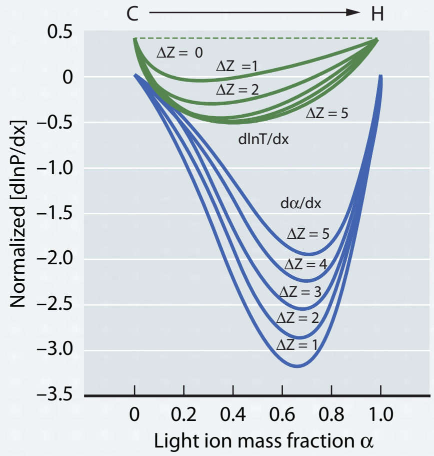
After a midyear start, we made significant progress towards our goals of exploring hybrid strategies for combining full-particle filters with smart-particle approaches (a type of machine-learning algorithm) and developing novel methods based on different underlying state-space models. Specifically, we (1) implemented a new modeling framework to facilitate inference and prediction for arbitrary constellations of data types; (2) developed new filter-based learning algorithms for probabilistic, deterministic finite automata, which enables the online learning of simplistic grammars for text data; (3) integrated the new filter into our composite mixture model to create a unique ability to simultaneously model arbitrary configurations of numerical, categorical, and text features; and (4) began using this composite mixture model as the core machine-learning infrastructure for researching novel parallel and hybrid filter strategies.

Proposed Work for FY12

In FY11 we will develop self-adapting particle filters that automatically adjust themselves to balance the tradeoff between estimation variance and data ingestion. Specifically, we will (1) explore hybrid strategies for combining full-particle filters with smart-particle approaches to obtain the accuracy of full-particle filters with the speed of smart-particle approaches, (2) develop mathematical bounds on the estimation error incurred by a smart particle as a function of the frequency with which the smart particle can be recalibrated against the full-particle filter, and (3) rigorously compare the empirical performance of these novel approaches, using existing filtering methods as benchmarks.

Publications

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



Temperature gradients (green) and concentration gradients (blue), normalized to the logarithmic derivative of pressure (proportional to acceleration) versus light-ion mass fraction in a binary ion mixture for several charge state differences between hydrogen and carbon. This highlights the strong change in the temperature gradient compared with mainline, average-ion simulations (top dashed green line) that neglect this important effect.

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 fusion energy. 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 incorporating ion diffusive phenomena into 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 caused by pressure gradients) and thermal diffusion, with eventual incorporation into the Livermore's suite of radiation-hydrodynamics production codes.

Much 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, 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 nonfluid (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.

FY11 Accomplishments and Results

In FY11 we (1) obtained a plasma analog of an adiabatic lapse rate (the degree of temperature variation with height) for atmospheric physics, (2) identified a new source of plasma temperature gradient and self-generated electric field in a binary ion species mixture that is proportional to the product of concentration gradient and difference in average ionization states, and (3) determined that application of the theory to inertial-confinement fusion implosions could have a strong impact on average hydrodynamic profiles as compared with predictions from mainline production codes that neglect diffusion.

Proposed Work for FY12

In FY12 we will (1) use the hybrid particle-in-cell simulation code LSP to assess shock morphology in a spherically converging geometry, (2) study multi-ion species distributions in LSP to assess the strength of mass diffusion in high-energy-density plasmas in the presence of electric fields, and (3) continue analysis and proposals for shots on the OMEGA laser at the University of Rochester in strong collaboration with the Massachusetts Institute of Technology.

Publications

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

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 solve 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 LLNL'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 grid model to accurately make long-term planning decisions.

Mission Relevance

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

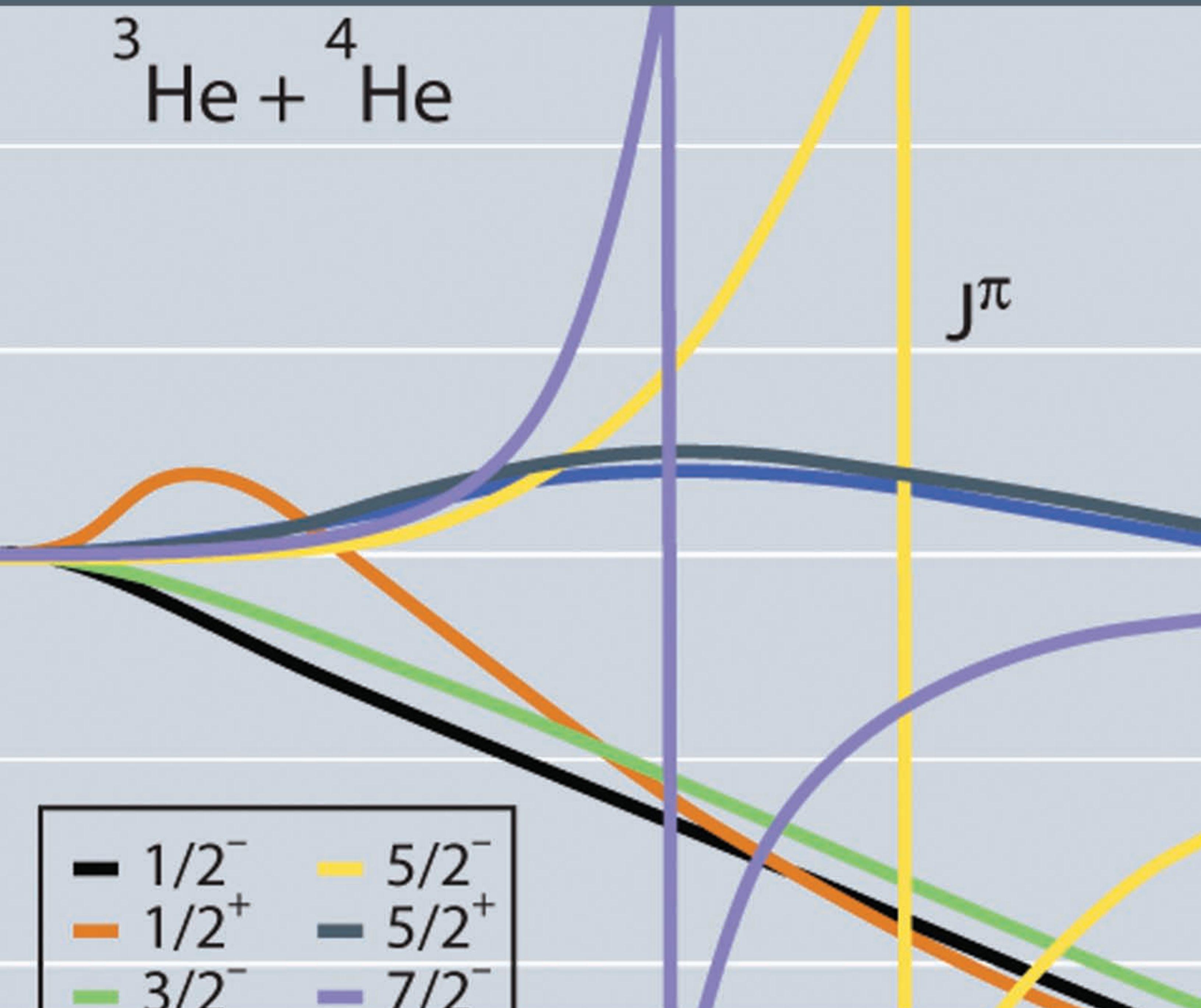
FY11 Accomplishments and Results

In FY11 we (1) acquired open-architecture distribution-system modeling software (Gridlab-D) from Pacific Northwest National Laboratory and initiated development of a high-resolution model of distribution systems; (2) developed an approach to modeling intermittent wind and solar generation using the open-architecture Weather Research and Forecasting model; (3) began developing decomposition-based optimization approaches for parallel solution of large-scale problems, such as decomposing a 1-month grid-planning problem into 30 separate 1-day sub-problems and including a time axis decomposition approach based upon Lagrangian relaxation; (4) developed an algorithm to solve the unit commitment problem (which generators to turn on and when) based on a novel approach inspired by a solution technique for processor scheduling; and (5) set up a high-performance computing environment for uncertainty quantification using the open-architecture code DAKOTA from Sandia, which will support analysis of the impact of intermittent resources and will help quantify impacts of some simplifying assumptions.

Proposed Work for FY12

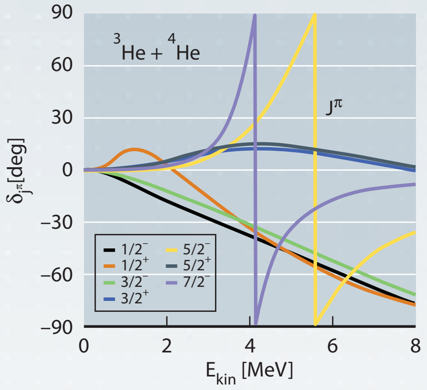
In FY12 we will (1) extend the grid model begun in FY11 to include mid- and large-scale models; (2) complete analysis of the impacts of simplifying assumptions in grid modeling, including the impacts of a simplified representation of alternating current power flow as direct current power; (3) work with collaborators at the University of California, Berkeley and Princeton University to adapt approximate dynamic programming and two-stage optimization algorithms to exploit high-performance computing platforms and profile their performance to gain insight to guide further algorithm improvement; and (4) begin developing processor-utilization methods to balance overall computing load by optimizing the process by which sub-problems from decomposition are assigned to separate processors, as well as methods to parallelize the process of solving the underlying large-scale optimization problem.

Nuclear Science and Engineering



Laboratory Directed Research and Development

FY2011



Nuclear phase shifts generated by the elastic scattering of a ^{3}He nucleus with an alpha particle (^{4}He nucleus), as obtained within an ab initio calculation that included the ground state and four 1/2+ pseudo-states (to simulate the virtual breakup) of ^{3}He. The ^{3}He + ^{4}He phase shifts are necessary to calculate the ^{4}He(^{3}He,gamma)^{7}Be radiative capture reaction important to astrophysics.

How Carbon and Oxygen Can Be Made in Stars: An Ab Initio Approach to Nuclear Reactions

Sofia Quaglioni (09-ERD-020)

Abstract

Predictions of stellar and nucleosynthesis modeling depend strongly on knowledge of the reactions that produce carbon-12 and oxygen-16. These thermonuclear reactions determine the carbon-to-oxygen ratio during stellar helium burning, with far-reaching consequences for the production of all heavier species. Current knowledge about the reactions is insufficient, and a breakthrough can be reached only with a fundamental theory. We propose to develop an ab initio many-body nuclear reaction theory to accurately predict the rates of alpha-capture reactions responsible for production of the two elements. In this project, alpha-capture rates will be calculated from first principles by means of an ab initio no-core shell model combined with the resonating-group method.

We will systematically develop formalism and codes to treat the scattering on light nuclei of increasingly heavier projectiles and perform the first-ever ab initio calculations of D(T,n)^{4}He and ^{4}He(^{3}He,gamma)^{7}Be cross sections as intermediate steps towards addressing the alpha-capture reactions leading to the formation of carbon-12 and oxygen-16. The successful completion of this project will result in (1) a long-awaited ab initio theory to explain alpha clustering in light nuclei and to calculate cross sections of reactions important for astrophysics from first principles; (2) an improved accuracy of the 2-alpha(alpha,gamma)^{12}C and ^{12}C(alpha,gamma)^{16}O reaction rates; and (3) an enhancement of the predictive capability in stellar modeling.

Mission Relevance

This project will establish the capability to perform calculations of light-ion reactions relevant to LLNL efforts in fusion energy generation and will result in a fundamental understanding of alpha clustering and improved alpha-burning cross sections for astrophysics, in support of the Laboratory's national security mission.

FY11 Accomplishments and Results

Because of budget reductions in FY10 and unanticipated major modifications of the code needed to address the increased computational challenge with three-nucleon projectiles, in FY11 we performed only initial steps toward development of the four-nucleon projectile formalism, necessary to describe alpha–nucleus scattering and alpha clustering in light nuclei. Specific work included (1) completing development of the formalism and codes for three-nucleon projectiles—implementation of the Hamiltonian kernel turned out to be particularly time consuming, requiring an unexpected major re-design of the code; (2) successfully performing ^{3}He–alpha and ^{3}H–alpha scattering calculations and preparing all necessary input and codes for the first ab initio calculation of the $^4He(^3He,gamma)^7Be$ and $^4He(^3H,gamma)^7Li$ capture reactions; (3) publishing our deuterium–alpha scattering and lithium-6 structure results and successfully completing our ab initio investigation of the $d(^3H,n)^4He$ and $d(^3He,p)^4He$ reactions. The successful completion of this project delivered novel theoretical approaches and computational tools for addressing fusion reactions that power stars and Earth-based fusion facilities such as the National Ignition Facility, and provided the research community with accurate evaluations and uncertainties for nuclear astrophysics and fusion diagnostics. The Office of Nuclear Physics in the DOE Office of Science through the Early Career Research Program will provide support for additional research efforts to continue development of a comprehensive framework that will lead to a fundamental description of reactions between light ions in a thermonuclear environment, as well as the structural properties of exotic nuclei.

Publications

Barrett, B. R., P. Navratil, and J. P. Vary, 2011. "Ab initio no-core shell model." *Nucl. Phys. News* **21**, 5. LLNL-JRNL-479988.

Navratil, P., and S. Quaglioni, 2011. *Ab Initio many-body calculations of the* $^3H(d,n)^4He$ *and* $^3He(d,p)^4He$ *fusion*. LLNL-JRNL-501012.

Navratil, P., and S. Quaglioni, 2009. "Applications of local chiral N2LO three-nucleon interaction to nuclear structure and reactions." *Bull. Am. Phys. Soc.* **54**, DH.00008. LLNL-ABS-414346.

Navratil, P., and S. Quaglioni, 2009. "Towards the first ab initio calculation of the deuterium–tritium fusion." *Bull. Am. Phys. Soc.* **54**, DM.00005. LLNL-ABS-414340.

Navratil, P., R. Roth, and S. Quaglioni, 2010. "Ab initio many-body calculations of nucleon scattering on ^4He, ^7Li, ^7Be, ^{12}C and ^{16}O." *Phys. Rev. C* **82**, 034609. LLNL-JRNL-439759.

Navratil, P., S. Quaglioni, and R. Roth, 2011. "Ab initio theory of light-ion reactions." *J. Phys.: Conf. Ser.* **312**, 082002. LLNL-PROC-455934.

Navratil, P., et al., 2009. "Recent developments in no-core shell model calculations." *J. Physics G: Nucl. Part. Phys.* **36**, 083101. LLNL-JRNL-411567.

Quaglioni, S., 2011. *Solving the long-standing problem of low-energy nuclear reactions at the highest microscopic level*. LLNL-ABS-480859.

Quaglioni, S., and P. Navratil, 2011. "Ab initio many-body calculations of deuteron-^4He scattering and ^6Li states." *Phys. Rev. C* **83**, 044609. LLNL-JRNL-469878.

Quaglioni, S., and P. Navratil, 2009. "Ab initio many-body calculations of nucleon–nucleus scattering." *Phys. Rev. C* **79**, 044606. LLNL-JRNL-409500.

Quaglioni, S., P. Navratil, and R. Roth, 2010. "Scattering of light nuclei." *Eur. Phys. J. Web Conf.* **3**, 01012. LLNL-PROC-421512.

New Target Concepts for Stockpile Science at the National Ignition Facility

L. John Perkins (09-ERD-036)

Abstract

We propose to address one of the most important scientific questions facing the Laboratory: How will achievement of ignition and thermonuclear yield at the National Ignition Facility (NIF) be utilized to provide unique weapons physics data for the Stockpile Stewardship Program? Accordingly, we propose to establish the technical basis of new target concepts that operate in the nuclear (high-energy-density) regime and provide stockpile stewardship data unattainable at any other facility outside of an underground nuclear test site. We will focus on target designs that have the potential to provide data for the most pressing perceived needs in primary design physics and emphasize a very-near-term target variant that could be fielded before, or in parallel with, the NIF ignition campaign.

We expect to produce a significant answer to the question of how NIF could be best utilized for stockpile stewardship once ignition is achieved. Livermore Advanced Simulation and Computing weapons simulation codes are the basis for high-consequence stockpile decisions, but they must be validated against real data in the nuclear regime. Accordingly, the results from this study will position the Laboratory to take full advantage of NIF ignition with technically credible proposals for advanced, stockpile-relevant targets that make full use of the high-energy densities stemming from ignition and thermonuclear burn.

Mission Relevance

This work supports the Laboratory's mission in national security by furthering development of advanced experimental platforms for NIF, advanced physical models for predictive capability, and advanced weapons physics simulation capabilities, as well as supporting high-energy-density burning-plasma science and diagnostics for stockpile science that are beyond state of the art.

FY11 Accomplishments and Results

In FY11 we (1) completed final design of the target and our assessment of key performance predictors including symmetry, stability, and laser–plasma interaction; (2) completed the comparison of those performance predictors, in conjunction with two of our principal weapon design codes; (3) finished an assessment of the shot schedule that would be required on NIF, including a definition of the requirements for late-time nuclear performance; and (4) analyzed proposed NIF experiments to understand the impact of data that would be produced on key program initiatives and codes. The successful completion of this research delivers both a unique target design and a complete understanding of how an experiment on this class of targets could be utilized to provide unique weapons physics data. The work, including actual experiments with designs developed as part of this research, will continue with Livermore program support.

Shock Temperatures from Neutron Resonance Spectroscopy

James McNaney (09-ERD-037)

Abstract

Temperature is a fundamental thermodynamic property, and accurate temperatures are necessary to understand and predict processes involving thermal activation such as chemical reactions, plastic flow, and phase transitions. However, temperature is extremely difficult to measure during dynamic loading. Optical techniques such as pyrometry are widely used but cannot probe the bulk volume of opaque materials such as metals. We intend to develop neutron resonance spectrometry for measuring temperature dynamically and in situ during shock loading by developing a neutron source based on a short-pulse laser.

Successful completion of this work would result in a high-fidelity, time-resolved temperature measurement technique capable of probing the interior of a dynamically loaded material on the nanosecond timescale. Development of a high-fluence, single-shot, laser-based neutron source would represent a significant achievement, while resonance spectrometry at the nanosecond timescale would represent a groundbreaking achievement. The technique would have diagnostic application in mid- to large-scale fusion-class laser facilities, and measurements made using this approach would lead to a better understanding of temperature-dependent material behavior in general.

Mission Relevance

Development of a neutron resonance spectrometry capability would be of high value to Laboratory efforts in both weapons and photon science, in support of the Stockpile Stewardship Program and the national and energy security missions. In particular, the proposed work would support advanced physical model development, improved materials property understanding, advanced experiments and diagnostics, and high-neutron-flux science. This capability would have significant impact to understanding many aspects of material behavior under dynamic loading conditions, including those relevant to LLNL fundamental high-energy-density research.

FY11 Accomplishments and Results

In FY11 we successfully demonstrated neutron measurement in the presence of prompt radiation from a short-pulse laser. After preliminary measurements made it clear that sufficient neutron production would not be possible at small-scale facilities such as the Titan laser, we determined and verified appropriate scaling relationships to determine facility requirements, then conducted the experiments at the OMEGA Extended Performance laser. Our experiments there verified our scaling relationships and the production of neutrons with energies up to 18 MeV. In summary, this project demonstrated significant neutron production using a short-pulse laser system. This neutron source should have utility in materials-damage studies for future inertial-fusion reactors. We also developed the capability to characterize the number and directionality of the source, devised scaling relationships for assessing the likely production at a range of laser facilities, and generated very high-energy neutrons—up to 18 MeV—at a laser-based facility for the first time. The next steps will be to conduct materials-dynamics experiments using laser-based neutron spectrometry and to develop a high-energy pulsed neutron source.

Publications

Higginson, D. P., et al., 2011. "Production of neutrons up to 18 MeV in high-intensity, short-pulse laser–matter interactions." *Phys. Plasma.* **18**, 100703. LLNL-JRNL-485811.

Swift, D. C., et al., 2011. *Neutron resonance spectrometry for temperature measurement during dynamic loading*. 17th APS Topical Conf. Shock Compression of Condensed Matter, Chicago, IL, June 26–July 1, 2011. LLNL-PROC-492411.

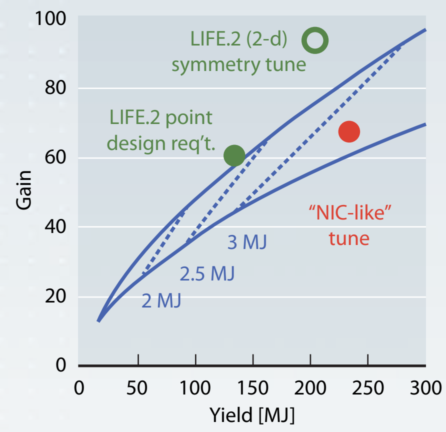
Advanced Inertial Fusion Target Designs and Experiments for Transformative Energy Applications

Peter Amendt (11-SI-002)

Abstract

Inertial fusion energy research is gaining momentum in the scientific world and the energy arena, and a key challenge for realizing fusion energy production is to develop a viable and experimentally validated "point design"—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 the laser inertial fusion energy (LIFE) concept, 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 LLNL scientists and engineers who developed the point design for NIF, will deliver a suite of LIFE target designs developed with radiation-hydrodynamic simulations and tailored to test several key physics issues on NIF.

The proposed project will deliver an ensemble of central-hot-spot ignition target designs for LIFE that leverage the current National Ignition Campaign point design. A collection of techniques to significantly increase the coupling efficiency of these targets for moderate (<100) energy gain 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 (>100) fast ignition using the high-efficiency hohlraums developed for the central-hot-spot ignition. Experiments at NIF to test the key physics uncertainties in the fast-ignition concept are also planned.



Thermonuclear gain-versus-yield analytic scaling laws for a high-efficiency laser target model (upper solid blue curve) with indicated laser energy curves (dashed lines). Solid green point is laser inertial fusion energy point-design requirement, red point denotes two-dimensional integrated hohlraum symmetry tune with conventional ("NIC-like") cylindrical hohlraum, and the open green circle depicts optimum simulation tune using advanced design features: a rugby-ball-shaped hohlraum, a pair of laser-entrance-hole shields, and oversized capsule relative to hohlraum radius. Simulations do not include expected performance degradation from three-dimensional effects, laser backscatter, and mix from hydrodynamic instability growth.

Mission Relevance

The proposed research could have a groundbreaking, transformative impact on LIFE, fusion energy in general, and high-energy-density matter research. The research will thus help enhance America's national security (developing proliferation-resistant advanced energy technologies), economic security (improving energy efficiency), and energy security (reducing energy imports and greenhouse gases).

FY11 Accomplishments and Results

In FY11 we made significant progress in both the central-hot-spot target design and the fast-ignition target design. Specifically, we (1) developed two-dimensional (2D) integrated models of several candidate central-hot-spot designs, including designs with hohlraum radiation shields, alternative hohlraum wall materials, and a rugby-ball-shaped hohlraum; (2) performed initial tests of the performance of these candidate targets at the OMEGA laser to validate the models; (3) developed new integrated simulation capabilities for fast-ignition fusion targets; and (4) used the new simulation capabilities to begin an initial high-gain, fast-ignition point design.

Proposed Work for FY12

In FY12 we expect to (1) complete 2D hohlraum modeling with radiation shields for testing at NIF at full scale with 3-omega laser light; (2) complete the first laser–plasma instability assessment of 2D advanced hohlraum designs (rugby-ball-shaped hohlraums with radiation shields) using National Ignition Campaign methodology for 3-omega and 2-omega full-scale point designs; (3) complete the design of a 2-omega NIF full-quadrant experiment to test laser–plasma instabilities with 2D integrated hohlraum modeling and laser–plasma instability post-processing, pending near-term NIF capability; and (4) complete a NIF full-scale experimental assessment of improved hohlraum efficiency at 225 eV and 3 omega, both with and without radiation shields.

Publications

Amendt, P., et al., 2011. "LIFE pure fusion target designs: Status and prospects." *Fusion Sci. Tech.* **60**, 49. LLNL-PROC-463711.

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 a stockpile 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 by demonstrating 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 hypothesize a set of metrics for this purpose and will test them with simulations of specific nuclear warheads from the U.S. stockpile. Assuming that this approach will be fruitful, we also propose to build, test, and optimize a prototype of the imaging system to perform the proof of principle for this verification approach.

We expect a successful project to establish the utility and optimization 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 will develop a prototype gamma-ray imager, for

which we will initially design and build a proof-of-concept prototype using only a small fraction of the number of active detector modules needed for a complete unit to allow for testing and demonstration of the concept at relatively low cost. Then, we will integrate a full hybrid nuclear-imaging module into a prototype for thoroughly validating system performance, and simulate the performance of a large, integrated system that would be composed of a large number of these nuclear-imaging modules.

Mission Relevance

This research supports the central Laboratory mission of ensuring national security, specifically, reducing the threat of nuclear attack by a rogue state or terrorists through technologies that support nuclear nonproliferation treaties.

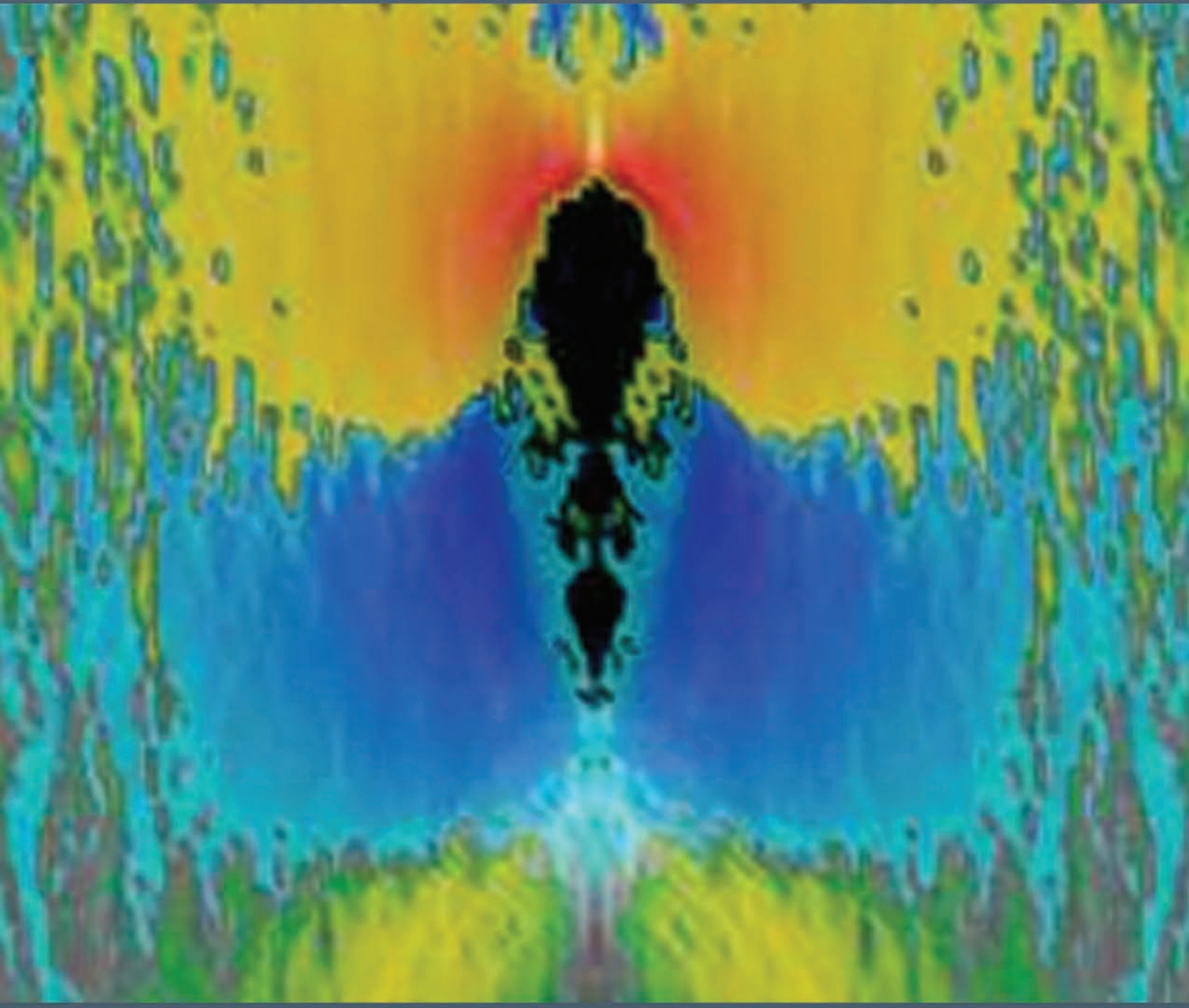
FY11 Accomplishments and Results

In FY11 we (1) developed a realistic three-dimensional radiation-source model based on analysis of a U.S. nuclear warhead, (2) modeled a Compton gamma-ray imaging system and began the necessary simulation benchmarking with an existing imaging system, and (3) calculated shape metrics derived from a three-dimensional image reconstruction of a radiation-source model.

Proposed Work for FY12

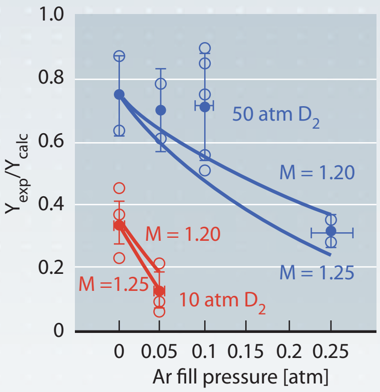
In FY12 we will (1) simulate the gamma-ray images that would be measured by a Compton imager or a coded-aperture imager from a realistic three-dimensional radiation-source model derived from analysis of a U.S. nuclear warhead, (2) calculate the sensitivity of features derived from three-dimensional image reconstruction, and (3) design, build, and benchmark a partial prototype of a gamma-ray imager.

Physics



Laboratory Directed Research and Development

FY2011



Ratio of measured to calculated neutron yield in the absence of fuel-pusher mix versus an argon fill pressure of 50 atm (blue) and 10 atm (red) for deuterium in plastic capsules. Solid points denote average over individual shots shown as open circles. Solid curves denote barodiffusion model for indicated Mach numbers *M* and average argon ionization state (+16).

Mesoscale Studies of Hydrodynamic Instability Growth in the Presence of Electric and Magnetic Fields

Peter Amendt (08-ERD-062)

Project Description

Recent proton backlighting data on laser-driven imploded capsules and rippled foils indicate the presence of strong self-generated electric (~1-GV/m) and magnetic (~1-MG) fields. Understanding their origin could be relevant to planned demonstrations of inertial-confinement fusion. Elucidating the nature of the fields and their effects on interfacial instability growth will require an approach that departs from standard single-fluid hydrodynamics and instead treats the system as an aggregate of coupled electron–ion fluids—that is, as a plasma. This project will explore plasma effects on important hydrodynamic instabilities such as Rayleigh–Taylor and Richtmyer–Meshkov, as well as hohlraum dynamics.

We expect to provide an evaluation of electric and magnetic field effects in imploding systems, an understanding of their origin and magnitude, and suggested remedial measures if the effects are deemed significant. Initially, we will evaluate methods to understand the underlying physics—both analytically and computationally—and to interpret the growing database for benchmarking our models and techniques. This research will potentially impact not only ignition on fusion-class lasers but also many high-energy-density studies with national security mission relevance.

Mission Relevance

This project supports LLNL's energy security mission by furthering the goal of robust ignition designs for inertial-confinement fusion, and also supports the national security mission by impacting investigations of high-energy-density imploding systems.

FY11 Accomplishments and Results

In FY11 we emphasized documenting and finalizing our understanding of barotropic diffusion in thermonuclear mixtures. Specifically, we demonstrated success of the barotropic diffusion hypothesis in helping to explain several outstanding anomalies in the inertial-confinement fusion database. Further investigations with the particle-in-cell Large Scale Plasma code of inertial-confinement fusion plasma shocks were also conducted. The successful conclusion of this project has led to growing national interest in exploring the role of electric fields in inertial-confinement fusion implosions, particularly the expanding National Ignition Facility database. This project has inspired a new research direction on the role of anomalous diffusive phenomena in high-energy-density states of matter that is now underway in a related LDRD project.

Publications

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Study of Kelvin–Helmholtz Instability in High-Energy-Density Hydrodynamic Processes

Hye-Sook Park (08-ERD-069)

Abstract

An unanswered question in high-energy-density hydrodynamics is how the Kelvin–Helmholtz instability can be studied in a controlled fashion. A controlled Kelvin–Helmholtz experiment would be valuable in demonstrating the ability to field a number of science studies on future fusion-class lasers, including eventual supernova experiments. In this project, we will conduct Kelvin–Helmholtz and related

supernova experiments on the OMEGA laser at the University of Rochester and the National Ignition Facility (NIF) laser at Livermore, leveraging LLNL expertise in target design, simulation, diagnostics, and other fields. This project will be conducted in collaboration with the University of Michigan.

If successful, this project will deliver an x-ray radiograph of the rolled-up vortex of a material during a controlled Kelvin–Helmholtz instability, demonstrating whether hydrodynamics experiments can resolve multiple interspersed layers of materials that absorb or transmit x rays or if these layers are too adversely affected by turbulent mixing and the recently observed phenomenon of mass stripping to serve as valid diagnostics. After experiments on OMEGA and modeling and simulation of the results, we will have a better understanding of how to implement a supernova experiment on NIF. Our experiments and modeling will provide new diagnostic requirements for the astrophysics experiments on NIF and show where the transition to turbulence occurs in Kelvin–Helmholtz vortex rollups.

Mission Relevance

Understanding Kelvin–Helmholtz instability is central to many hydrodynamic processes important to the Stockpile Stewardship Program. This project supports the Laboratory's national security mission by potentially producing an entirely new type of experimental data for the validation of stockpile stewardship codes.

FY11 Accomplishments and Results

In FY11 we performed a third series of Kelvin–Helmholtz experiments on the OMEGA laser to understand the sensitivity of Kelvin–Helmholtz evolution to interface roughness. Specifically, we (1) used our high-quality data to investigate turbulent mixing in a foam–methylidyne interface driven by Kelvin–Helmholtz instability, observing turbulence-like structures in shots with both flat interfaces and interfaces having pre-imposed two-dimensional modulations; (2) analyzed the data, finding that the mixed methylidyne material extends about 100 μm into foam and verifying that our simple mix model, based on simulation predictions of the interface velocity, was close to the observations; (3) used the three-dimensional massively parallel ARES code to analyze the bubble feature seen in data from our FY09 experiments, determining that the bubble resulted from shock imprinting on the shock-tube wall; and (4) continued to collaborate with the University of Michigan on both the Kelvin–Helmholtz and hydro-instability experiments. This project provided a detailed understanding of the hydrodynamics in linear, nonlinear, and turbulent-mixing regimes, and our high-quality experimental measurements are being used to test various hydrodynamics simulation tools at LLNL. The next step would be to continue experiments on OMEGA and perform high-power laser experiments on NIF.

Publications

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Precision Mono-Energetic Gamma-Ray Science for NNSA Missions

Christopher Barty (09-SI-004)

Abstract

We propose to study compact, mono-energetic gamma-ray (MEGa-ray) science for four high-impact NNSA missions: (1) isotope-specific nuclear resonance fluorescence imaging of stockpile and ignition components, (2) isotopic assays of fission products and for nuclear waste stream applications, (3) isotopic radiography and quantitative assays of DOE legacy waste, and (4) isotope-specific flash radiography of multicomponent turbulent hydrodynamic experiments and ignited plasmas. We will demonstrate the world's first MEGa-ray system capable of meeting these needs and provide a dramatic new ability for scientific investigation of photonuclear interactions.

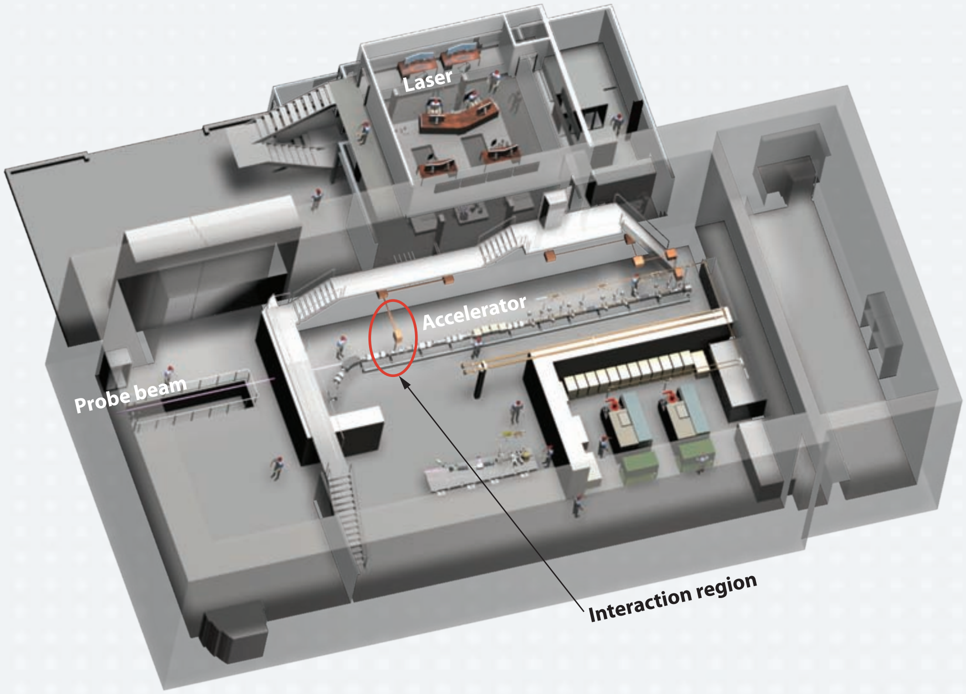
We expect to develop short-pulsed laser-based MEGa-ray sources for the static, isotope-specific imaging and assaying of highly enriched uranium, as well as for applications in stockpile stewardship, legacy waste, and dynamic nuclear resonance fluorescence imaging. The flash isotope radiography we envision would allow time-resolved observation of the turbulent mixing of stockpile-relevant materials using isotope tracer layers. With a modular compact design, the robust MEGa-ray source would also enable portable isotope radiography tools for other national and homeland security applications.

Mission Relevance

This project supports the Laboratory's national security mission by developing a new and potentially revolutionary dynamic, isotope-specific radiography capability for stockpile stewardship and fundamental weapons physics by increasing the proliferation resistance of the nuclear fuel cycle with precision isotopic monitoring and by demonstrating the potential for MEGa-ray sources for detection of highly enriched uranium at trade portals. The project also supports environmental management efforts by enabling quantitative assessment and reclassification of the DOE complex's nuclear waste.

FY11 Accomplishments and Results

Our primary accomplishment in FY11 was the invention of an entirely new configuration for Compton scattering, which we named asymmetric Compton scattering. This approach, for which a patent application was filed, operates at a higher effective repetition rate, increases the flux of the MEGa-ray source, decreases the bandwidth, and potentially simplifies much of the underlying accelerator and laser subsystems. Additionally, we (1) completed the fiber-based laser system for ultraviolet pulse production that enabled record pulses; (2) completed design of the 5.59-cell X-band photo-gun and fabricated the components for this system at the Stanford Linear Accelerator Center—further photo-gun experimental effort was redirected toward evaluation and design of an appropriate gun for asymmetric Compton scattering; (3) tested commercial 120-Hz, joule-class interaction laser heads and found them to be of insufficient beam quality—we developed a redesign that corrected the problem for production of 10-ps pulses via hyper-dispersion chirped-pulse amplification; (4) developed a model for Compton scattering that was used to determine not only optimal configurations for symmetric but also for asymmetric Compton scattering for picosecond-scale temperature measurements; (5) used models to evaluate efficacy of MEGa-rays for material assay of rare-earth elements, which has resulted in industrial interest in a MEGa-ray Cooperative Research and Development



Concept for a precision MEGa-ray capability at Lawrence Livermore National Laboratory for isotope-specific imaging and assaying of highly enriched uranium, as well as for applications in stockpile stewardship and legacy nuclear waste.

Agreement; (6) conducted integrated X-band laser tests of the photo-gun drive laser; (7) designed X-band accelerator sections and fabricated parts for symmetric Compton scattering; and (8) evaluated designs for asymmetric mode operation. The successful completion of this project resulted in discussions with industry regarding a Cooperative Research and Development Agreement for a MEGa-ray enabled material assay and in an entirely new configuration for Compton scattering that both increases MEGa-ray flux and decreases bandwidth and simplifies associated laser and accelerator hardware.

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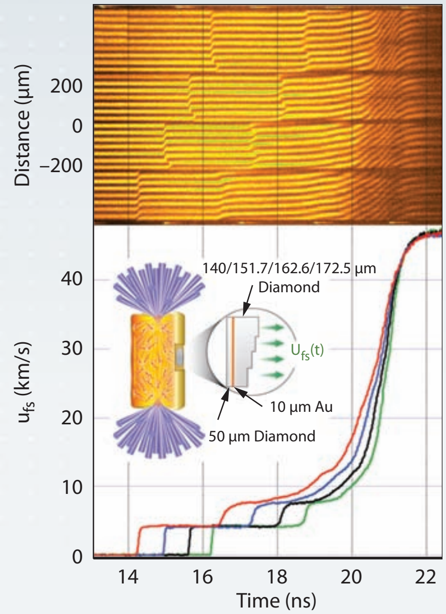
Physics and Chemistry of the Interiors of Large Planets: A New Generation of Condensed Matter

Gilbert Collins (09-SI-005)



We used the National Ignition Facility to study the velocity history of ramp-compressed diamond—one of three experimental results used to measure the quasi-isentropic equation of state for carbon up to 50 Mbar. Raw data from the velocity interferometer system for any reflector interferometer measurement is shown in the top image. The bottom graph is the analyzed velocity history. The velocities for several different thicknesses are used in a Lagrangian analysis to determine stress and density.

Abstract

We propose to employ Livermore's advanced laser facilities to systematically characterize condensed matter at extreme conditions of gigabar pressures and over tenfold compression. This research focuses on establishing a new generation of experiments accessing the unexplored regime of ultrahigh compression, with applications that range from understanding the origin and evolution of planets to testing and significantly extending fundamental theories of condensed matter. Our effort will form a community of the world's leading scientists in condensed matter and planetary science.

We expect to develop a new scientific frontier of extreme states of matter, from ultradense condensed matter to burning plasmas. We will determine the key constraints on planetary and stellar evolution models using hydrogen, helium, and mixtures at more than tenfold compression, from a low-temperature isentrope to a burning plasma. We will determine whether hydrogen becomes a Wigner crystal, superconductor, or superfluid at extreme densities, as well as characterize iron at low-temperature and ultrahigh-pressure conditions expected to exist at the core of extrasolar super-Earth planets. In addition, we will produce the first data on chemistry in the gigabar regime and determine the fundamental nature of and key physics enabling burning plasmas.

Mission Relevance

To successfully maintain an aging stockpile, the nation needs new predictive capabilities. Of particular importance to the Stockpile Stewardship Program is development of advanced capabilities to predict the physical properties of matter under an extremely broad range of dynamic conditions—specifically, equation-of-state and constitutive models. Our project will fully develop the required experimental capabilities as well as provide important data on metals and hydrogenic liquids in support of the Laboratory's mission in national security.

FY11 Accomplishments and Results

In FY11 we (1) mapped the melt curve of diamond to 11 Mbar; (2) discovered a new polymeric phase of liquid carbon; (3) developed a new platform for quasi-isentropically compressing matter to many terapascals on the National Ignition Facility and used the platform to measure the stress density of ramp-compressed carbon to 50 Mbar—the highest pressure solid ever studied on Earth and equivalent to the highest pressure conditions expected to occur at the center of Saturn; (4) developed a new technique for performing diffraction on ramp-compressed matter and used it to discover new phases in solid magnesium oxide, tin, tantalum, and iron at extreme pressures;

(5) developed picosecond extended x-ray absorption fine structure spectroscopy and used it to measure the temperature of ramp-compressed iron at conditions equivalent to those of the Earth's core; and (6) discovered that helium and hydrogen phase-separate at conditions deep inside Saturn. In summary, we have developed a number of new techniques to explore high-energy-density matter and made several important related discoveries that either have been published or are in the process of being published, including fundamental discoveries on the behavior of matter existing deep within giant planets, to provide the first benchmark for materials theory and planetary evolution models. This project has initiated a new frontier of material science at terapascal conditions at the National Ignition Facility, where we intend to pursue the next step in this work using the facility's cutting-edge capabilities. Finally, by holding and participating in workshops, conferences, and conference sessions, we helped facilitate the growth of this new field and helped build a new community of scientists intensely interested in high-energy-density materials at more than 20 institutions around the world. This community has written many successful proposals to explore high-energy-density materials on the Jupiter and OMEGA lasers and the Z Machine—work that should ultimately produce benefit to America's national and energy security and economic competitiveness.

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From Super-Earths to Nucleosynthesis: Probing Extreme High-Energy-Density States of Matter with X Rays

Bruce Remington (09-SI-010)

Abstract

We propose to develop advanced x-ray diagnostic systems to investigate materials at ultrahigh pressures (>10 Mbar), such as those found at the cores of newly discovered "super-Earth" exoplanets, and plasmas at ultrahigh densities (>100 g/cm^3) in nuclear burn in stars and supernovae. We will develop three types of diagnostic capabilities including a lattice diagnostic, an ultrafast x-ray diagnostic for investigating nuclear burn, and a multi-frame, single line-of-sight imager. We will also validate these capabilities through experiments at laser and calibration facilities. This project will enable precision science to be conducted on matter under the most extreme conditions of density and temperature accessible, which is important to both basic and applied science.

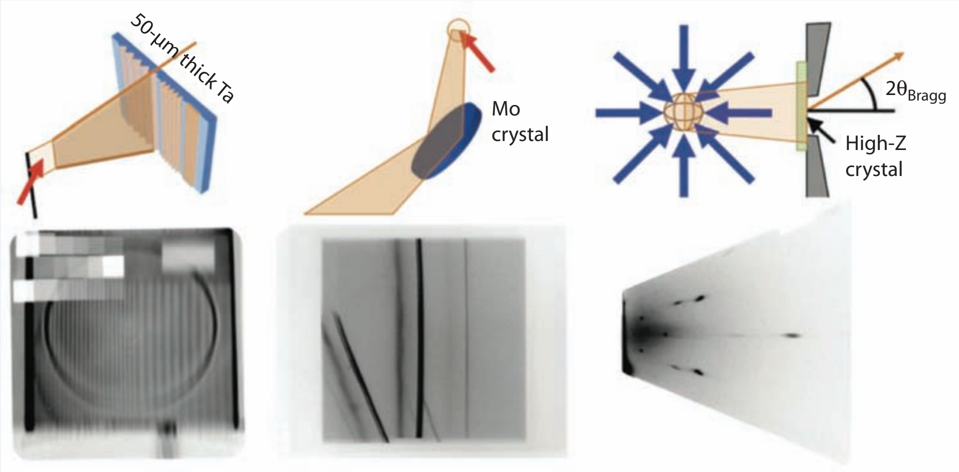
We expect to develop advanced x-ray diagnostic systems to probe extreme high-energy-density states of matter relevant to the science of super-Earths and nucleosynthesis, with the goal of enabling materials science at ultrahigh pressures and studies of burning plasmas at ultrahigh densities. These new capabilities will make it possible to probe super-Earth core conditions with a dynamic lattice-level diffraction diagnostic at 100 times higher pressures and 4 times higher x-ray energies than is currently possible. These capabilities will also enable researchers to probe nucleosynthesis plasma conditions by dynamic characterization of hot, dense burning plasmas at 100 times higher densities and with 100,000 times higher yield than is currently possible.

Mission Relevance

By developing capabilities to measure material phase and lattice response at ultrahigh pressures and conditions in burning plasmas at ultrahigh densities, which are relevant to stockpile stewardship and high-energy-density science, this project supports the Laboratory's national security mission.

FY11 Accomplishments and Results

During FY11 we (1) successfully developed dynamic Laue diffraction at 10 to 30 keV on single-crystal samples of tantalum shocked to approximately 2-Mbar peak pressures—the Laue patterns are used to determine the sample phase and strength, the Laue spot shapes contain information about the defect density, and the relative brightness of the various Laue spots contains information about the sample temperature; (2) developed dynamic Bragg diffraction at 15 to 22 keV in shocked single-crystal molybdenum and tantalum at pressures of 0.1 to 1 Mbar—this allowed a determination of the one- to three-dimensional lattice compression threshold for molybdenum and an estimate of its strength, which led, in part, to an Edward Teller award; and (3) worked, in collaboration with the Massachusetts Institute of Technology,



Experiments utilizing greater than 10-keV line emission or broadband x-ray sources. High-energy radiography to measure the growth of Rayleigh–Taylor unstable features as a sensitive test of strength models of tantalum requires a line-emission backlighter at 22 keV to penetrate the approximately 50-µm-thick tantalum targets (left). Time-resolved Bragg diffraction of molybdenum to probe the lattice dynamics of shocked body-centered cubic metals requires a 17-keV backlighter to probe a sufficient number of lattice planes (middle). White-light or broadband transmission Laue diffraction of tantalum or molybdenum to study the ultrahigh-pressure phase and twinning in body-centered cubic metals requires a bright broadband source between 10 and 25 keV (right).

to develop an ultrahigh-speed multi-frame single line-of-sight x-ray camera. Specific work included design and simulation verification of the imaging, image storage, and timing control portions of the design. What remains to be completed are the read-out and integration portions of the design as well as fabrication of the prototype chips and design and production of ancillary electronics to test the design. The successful conclusion of our project resulted in a 1-ps gated all-optical framing camera with less than 10-μm detector resolution, which won a 2010 R&D 100 award, and a multi-time-frame solid-state (all optical) imaging detector, a continuous-time x-ray power detector, and a one-dimensional continuous-time record camera. This project has enabled novel diagnostic techniques for probing extreme high-energy-density conditions suitable for National Ignition Facility experiments. Variants of the diffraction and burning plasma techniques are under active consideration for implementation at the facility for experiments in FY12 and 13.

Publications

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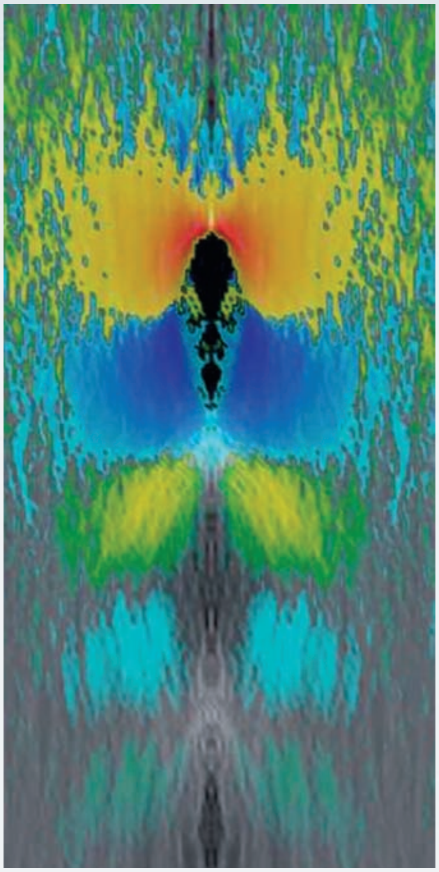

One-component plasma wake field caused by a projectile ("anti-neon") in electron gas. The projectile is centered in the upper third of the field of view, moving upwards. The density response is shown in gray-scale, with the wake trailing behind. The log of the energy transfer field is shown in color, truncated at a small value.

The Microphysics of Burning, Hot Dense Radiative Plasmas

Frank Graziani (09-SI-011)

Abstract

We intend to develop a detailed microphysical understanding of the physics of burning, hot dense radiative plasmas by building a validated state-of-the-art N-body simulation capability. The extreme conditions of these plasmas mean that experimental data for validation are difficult to obtain. What is needed for this data-starved environment is a quantitative way of telling whether or not the physics used to describe burning, hot dense radiative plasmas is correct. We will address this critical need by building a world-class massively parallel *N*-body simulation capability in conjunction with experimental validation experiments. The microphysics of burning, hot dense radiative plasmas will be simulated without the assumptions that underlie current models used to calculate inertial-confinement fusion or astrophysical problems.

We expect to (1) develop an *N*-body simulation capability using molecular dynamics to investigate the properties of hot dense matter undergoing thermonuclear burn; (2) employ the new simulation capability to test microphysical foundations of widely accepted theoretical models; and (3) validate the new simulation capability using the ultrashort-pulse lasers Titan at Livermore, the OMEGA Laser at the University of Rochester, and the Linac Coherent Light Source at Stanford.

Mission Relevance

The results obtained with this project directly address the issue of predictive capability, which is at the core of the Stockpile Stewardship Program. Furthermore, the validated simulation capability for burning, hot dense radiative plasmas that we develop will extend LLNL's world leadership in high-energy-density physics to include not only experimental expertise but computational modeling as well. It will help establish LLNL as the world center for high-energy-density-physics, in support of the Laboratory's mission in fundamental science breakthroughs.

FY11 Accomplishments and Results

We (1) completed development of a new time-dependent simulation capability for dense plasmas centered around a massively parallel molecular dynamics (MD) code capable of simulating material conditions of high temperatures, high densities, significant electromagnetic fields, mixtures of high- and low-atomic-number elements, and non-Maxwellian particle distributions in inertial-confinement fusion experiments and many stellar interiors; (2) incorporated into our new code key atomic, radiative, and nuclear processes, so that their interacting effects under non-ideal plasma conditions can be investigated; (3) developed new concepts to deal more efficiently with the very disparate dynamical timescales that arise in fusion plasmas; (4) carefully compared quantum effects on electron trajectories as predicted with our method to those predicted by more elaborate dynamical methods; and (5) began mining the potential of this new MD tool for investigating a wide range of physical processes in plasmas, obtaining new results for electron–ion coupling and charged particle stopping. In summary, this project has, in addition to producing an MD simulation capability for gaining insights into the behavior of hot dense plasmas, yielded advances in computational methodology, elucidated the strengths and weaknesses of quantum statistical potentials as effective interactions for MD, explained the model used for quantum events possibly occurring in a collision, and advanced new experimental approaches that play a central role in validation. Next steps include evaluating thermal conductivities, diffusivity, and equations of state; using our new MD capability to investigate plasma mixtures in which a high-atomic-number ion component is strongly coupled while the proton–proton component is weakly coupled, thereby providing a valuable test of the assumptions underlying current theoretical treatments of plasma mixtures; and extending our MD tool to the warm dense matter regime.

Publications

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More, R., and F. Graziani, 2010. *Emission of visible light by hot dense metals.* 19th Intl. Toki Conference (ITC19) on Advanced Physics in Plasma and Fusion Research, Toki City, Japan, Dec. 8–11, 2009. LNL-CONF-423472.

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Improved Spectral Line-Shape Models for Opacity Calculations

Carlos Iglesias (09-ERD-004)

Abstract

Opacity, or the measure of impenetrability to electromagnetic or other radiation in a medium, is important for energy-transport simulations used in the design and analysis of Laboratory applications such as inertial-confinement fusion. However, theoretical opacities vary by as much as a factor of two because of uncertainties in models of spectral line shape. Our main goal is to improve line-shape models in opacity codes by applying line-broadening theory to complex electronic configurations that have been heretofore neglected by the scientific community, but which are essential for opacity calculations. The results will be extended to the super-configuration array concept, which is vital for calculations involving myriad configurations. Finally, we will synthesize this gained knowledge into a suite of routines for incorporation into opacity codes.

The project will produce improved opacity codes by addressing three uncertain aspects of line-shape models: line-width calculation using approximate formulas, far-wing behavior, and line broadening in complex multi-electron ions. We will produce fast computer subroutines that parameterize these complex processes and incorporate those subroutines into opacity codes. These achievements will result in improved theoretical opacities, which will in turn resolve uncertainties in many LLNL applications that depend on accurate simulation of energy-transport mechanisms.

Mission Relevance

This project supports the Laboratory's missions in national and energy security by addressing an important knowledge gap in advanced physical models for predictive capabilities for weapons science, nuclear physics, astrophysics, and high-energy-density science.

FY11 Accomplishments and Results

The performance of several algorithms, not previously applied to Stark line-broadening calculations, were compared in FY11. We determined that model-reduction schemes were by far the most computationally efficient. These schemes rely on either the Lanczos or Arnoldi algorithms to reduce large matrices to smaller effective ones that conserve relevant properties. In addition, we developed a Stark line-broadening code using fast algorithms that can be readily inserted into other codes. Our goal was to replace older Stark broadening codes that are not only inefficient but also relied on uncontrolled approximations to speed the calculations. Our new Stark broadening code will be used in radiation transport codes to analyze experimental spectra obtained at the National Ignition and other facilities.

Publications

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First-Principles Planetary Science

Kyle Caspersen (09-ERD-012)

Abstract

The objective of this project is to provide a first-principles understanding of the interiors of the four gas giants in our solar system—Jupiter, Saturn, Uranus, and Neptune. The lack of experimental data on the interior of these four planets leads to uncertainty and unanswered questions about their formation, current structure, and how they will evolve. We will address these questions using the predictive capability of first-principles calculations. In particular, we will calculate planetary isentropes to predict the pressures and temperatures in these gas giants and transport properties along the isentropes to provide insight into the planetary dynamics.

We intend to construct thermodynamically consistent isentropes that are free from most current assumptions. We will also calculate transport properties along these isentropes. This will be the first time that planetary isentropes and their corresponding transport properties have been calculated completely with first-principles calculations. The unequaled precision and predictive power of these calculations have, and will, allow us to address fundamental questions in the field of planetary science and in the more general field of high-energy-density science. This project will also directly benefit many ongoing efforts at LLNL, including equation-of-state table development, the generation of mixing and plasma models, and dynamic compression experiments.

Mission Relevance

This project supports LLNL's national security mission by developing predictive models for examining the properties of materials under extreme conditions. Such techniques, if successful, will have direct application to equation-of-state models for materials of interest for stockpile stewardship and other Laboratory missions.

FY11 Accomplishments and Results

In FY11 we (1) completed calculation of the free-energy grid for constructing an arbitrary isentrope for hydrogen–helium planets, (2) continued the calculation of transport properties and published our results, and (3) continued our study of the kinetics of helium condensation. The successful conclusion of this project provided data for, and insight into, the equation of state and transport properties of planetary mixtures from electronic-structure theory. The project impacted existing planetary models, as well as resulted in numerous publications. We are seeking support from the National Aeronautics and Space Administration to continue our study of planetary materials under extreme conditions using electronic-structure techniques.

Publications

Chau, R., S. Hamel, and W. Nellis, 2011. "Chemistry in the deep interior of Uranus." *Nat. Comm.* **2**, 203. LLNL-JRNL-426271-DRAFT.

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An Imaging X-Ray Line-Shape Diagnostic for Burning Plasmas

Peter Beiersdorfer (09-ERD-016)

Abstract

We intend to investigate the suitability of neon-like tungsten as a tracer ion for measuring ion temperature and velocity in a hot, ignited plasma. Such measurements are crucial for achieving fusion ignition and burn. We will build a new type of imaging spectrometer, which will operate in a vacuum and use a spherically bent crystal in its Johann geometry. We will also test key aspects of a tungsten-based spectrometer,

including throughput issues, window design, performance over a large temperature range, and power dissipation of the detector in a vacuum. We will utilize LLNL's Electron Beam Ion Trap facility, which is uniquely suited to produce the relevant radiation.

If successful, we will produce novel atomic data and perform world-class science experiments such as measurements of excitation-rate coefficients, ultrafast radiative lifetimes, and dielectronic rate coefficients, which have never been measured for such high-atomic-number systems but are needed to determine ion temperature and motion. We will also provide the scientific and technological basis for choosing a tungsten-based instrument design and create a prototype instrument for hot plasmas with which we will assess the technological hurdles of measuring radiation from an ignited plasma.

Mission Relevance

This project supports the Laboratory's missions in energy security by developing groundbreaking technologies for achieving fusion-driven energy sources. This project will also help attract top talent to the Laboratory in this topical, cutting-edge field.

FY11 Accomplishments and Results

In FY11 we successfully performed high-resolution spectral measurements of tungsten atomic L-shell lines, which represent the highest-resolution measurements of these lines so far, and we were able to determine the wavelength of salient features of the tungsten spectrum with high accuracy. We also were able to assess blends with neighboring charge states that may affect line shapes that can serve as burning plasma diagnostics. Our measurements validated the scientific and technical concepts of the burning plasma ion-temperature spectrometer. The successful conclusion of this project demonstrated the suitability of neon-like tungsten as a tracer ion for spectroscopic measurement of temperature in burning plasma. The newly developed concepts and instrumentation are now being applied to programmatic measurements and we will further develop the concept for implementation on the ITER tokomak fusion reactor in France.

Publications

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Ultrafast Nanoscale Dynamic Imaging Using X-Ray Free-Electron Lasers

Stefan Hau-Riege (09-ERD-023)

Abstract

By delivering ultrashort pulses of unprecedented brightness, x-ray free-electron lasers will revolutionize x-ray science. Our goal is to take advantage of the timely coincidence of the advent of these lasers, such as the Linac Coherent Light Source (LCLS) at Stanford, and our team's recently gained experience at LCLS in molecular-resolution imaging with x-ray free-electron lasers to answer several questions concerning the properties and dynamics of high-energy-density matter of importance to NNSA programs. Experiments at LCLS will directly complement results obtained at the National Ignition Facility by providing a very high-fidelity probe for materials in only moderate high-energy-density states. Energy densities accessible at the National Ignition Facility will be substantially higher, but the probe is less well suited for imaging.

Besides developing new capabilities for LLNL such as a novel plasma probe with high penetration power and a ultrahigh-precision high-energy-density lattice and microstructure diagnostic, we will perform fundamental research in the field of x-ray and material interaction. This will benefit a wide variety of other areas, including ultrafast atomic molecular physics, cluster physics, and coherent x-ray imaging of nanoscale objects.

Mission Relevance

By exploiting an entirely new generation of x-ray sources, this project supports the Stockpile Stewardship Program, which is highly dependent on high-quality data describing the properties and processes of high-energy-density matter. National security and weapons science are served through the proposed study of dynamic systems with broad application to the investigation of material dynamics under extreme conditions. Novel imaging techniques are also at the cutting edge of photon science applications, which will be developed at LCLS.

FY11 Accomplishments and Results

In FY11 we (1) finalized the evaluation of the LCLS diffraction data obtained in 2010, which included performing additional semiclassical molecular dynamics simulations; (2) used our preliminary results to devise new schemes to simulate the interaction of LCLS radiation with matter, including using tight-binding molecular dynamics; (3) performed time-delay x-ray pump–probe experiments at the LCLS on nanocrystalline solids, using Bragg powder diffraction instead of x-ray Thomson scattering as the diagnostics to increase the signal; and (4) performed an experiment to study the atomistic dynamics in laser-driven shocked and melted materials through x-ray diffraction at the LCLS. This project conducted the first-ever set of high-energy-density state experiments at LCLS and has laid the groundwork for an experimental program for the end station of the LCLS Matter in Extreme Conditions Instrument in 2012. The work performed in this project has attracted the interest of the Office of Science's Basic Energy Sciences Program and the Office of Science–NNSA Joint Program in High Energy Density Laboratory Plasmas.

Publications

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Scrape-Off-Layer Flow Studies in Tokamaks

Thomas Rognlien (09-ERD-025)

Abstract

Boundary flows are presently unexplained but play a critical role in tritium accumulation via co-deposition in devices using carbon, such as the ITER international tokamak fusion reactor project, which will operate with a strict tritium-inventory limit. We propose to develop new techniques to measure boundary-plasma flows in tokamak reactors and assess and implement new physics and computational methods in the UEDGE code to understand and predict the flows. We will design an optical imaging system that permits measurements in high-power plasmas to be tested in the DIII-D tokamak in San Diego. To make reliable predictions of boundary flows with UEDGE, we will implement and validate new algorithms and physics such as kinetic effects distilled from our kinetic TEMPEST code.

We expect to deliver a prototype optical system for the DIII-D tokamak that can reliably measure plasma flows in at least one poloidal segment of the scrape-off layer, the outer layer of a tokamak plasma that is affected—"scraped off"—by a divertor or limiter. In addition, other suitable diagnostic techniques may be identified. The upgraded version of UEDGE will be implemented, and comprehensive validation will be performed to determine the physics of measured flows that can be applied to optimize device operation and future design. Synergy of experimental and theoretical and simulation work will lead to major advancement of boundary-plasma physics, positioning LLNL as a leader in efforts for an anticipated new device design. Results will be published in journals relevant to magnetic-confinement fusion.

Mission Relevance

Development of a new diagnostic system and its use to validate advanced simulations of the boundary plasma in magnetically confined fusion devices supports the Laboratory's mission in energy security through development of fusion energy in tokamaks. The project will help to establish fusion as an abundant, reliable, and clean energy source. In addition, our research supports the Laboratory's emerging science and technology plan of research in high-energy-density and burning plasmas, as well as efforts in high-fidelity simulations.

FY11 Accomplishments and Results

In FY11 we (1) developed a procedure to eliminate, by careful alignment of the spectrometer and analysis of the complex viewing geometry, most spurious artifacts from the large-area Fourier transport spectrometer signal on DIII-D, allowing a systematic reduction of flow velocities of higher-charge-state carbon from Fourier transport spectrometer data; (2) obtained good comparison between large features of the experimental carbon flow and the UEDGE edge-plasma transport code, which simulates density, velocities, and energies of all species, while including the cross-magnetic-field charge-particle drifts and identifying signal noise as a limiting factor; and (3) obtained and used a radiation-hardened camera with a signal intensifier to further improve signal-to-noise ratio. This project demonstrated, for the first time ever, that impurity in the ion flow at the edge of a magnetic fusion energy tokamak can be measured by Fourier transport spectrometer, a nonperturbative, advanced optical technique, with the basic features of the flow agreeing with results obtained with the UEDGE edge-plasma simulation code. The Fourier transport spectrometer system has become a valued diagnostic on the DIII-D National Fusion Facility, with other international facilities such as the Joint European Torus expressing interest in utilizing this type of diagnostic. The Australian National University has also expressed interest in partnering with LLNL in an ITER-related proposal that would make use of the new capabilities.

Publications

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Critical Enabling Issues for Burning-Plasma Diagnostics

Steven Allen (09-ERD-030)

Abstract

This project will study and define two diagnostic techniques for tokamak burning plasmas: a motional Stark effect technique to measure current profile and an infrared radiation fast-scanning technique to measure plasma wall temperature. For the motional Stark effect diagnostic, we will address several important issues of measurement including a polarization-preserving vacuum window, techniques to minimize double refraction caused by vacuum forces and temperature gradients, and whole-spectrum measurement of the Stark spectrum. For the infrared diagnostic, we will investigate the feasibility of developing a fast, two-color scanning technique for conducting measurements in burning plasma experiments such as the ITER international tokamak fusion reactor project.

In collaboration with the University of Arizona, we will pursue motional Stark effect polarization engineering and develop a polarization-preserving vacuum window. Measurements of the whole Stark spectrum will be used to compare an alternate measurement technique with motional Stark effect and to evaluate the effects of different plasma conditions on spectrum details. We will implement and test a novel data-acquisition scheme that will allow real-time monitoring of both plasma instabilities and optical properties of the polarimeter. Lastly, we plan to develop a prototype fast two-color infrared radiation line scanner (two colors make the measurements less susceptible to changes in surface emissivity) for measurement of tokamak wall temperatures.

Mission Relevance

Our research supports LLNL's mission in energy security by making important contributions to the study of the physics of magnetic-fusion burning plasmas, such as those in the ITER project, and by applying cutting-edge tools to the development of fusion energy as an abundant, reliable, and clean energy source.

FY11 Accomplishments and Results

In FY11 we (1) analyzed data from the new digital data-acquisition system; (2) determined the effect of magnetohydrodynamics fluctuations on the motional Stark effect measurements—specifically, there was development of a magnetic island because of "flux-pumping" of an edge-localized mode in the General Atomics DIII-D tokamak in San Diego; (3) demonstrated the interaction of plasma oscillations at the same frequency as the motional Stark effect polarimeter; (4) studied the DIII-D motional Stark effect polarimeters, including detailed Mueller matrix measurements of mirrors exposed to the plasma, and showed that they preserved the polarization state of the light—in situ measurements of the whole DIII-D polarimeters were obtained and the derived Mueller matrix elements were sometimes inconsistent; and (5) initiated design of a two-part vacuum window with both plus and minus polarizations—the validity of the concept was demonstrated, but the thicknesses of the two materials were very different. Our project addressed several issues of the extension of the motional Stark effect (to measure plasma current profiles) from its use on current tokamaks to its application on burning plasma experiments: digital data acquisition of the complete polarimeter output, measurement of magnetohydrodynamics fluctuations, polarization response of the optics (Mueller matrix), and transmission of the light through a vacuum window. The motional Stark effect diagnostic has been assigned to the U.S. by the international ITER reactor project diagnostic team, and our research will be continued as part of the development of the diagnostic for the next burning plasma experiment.

Publications

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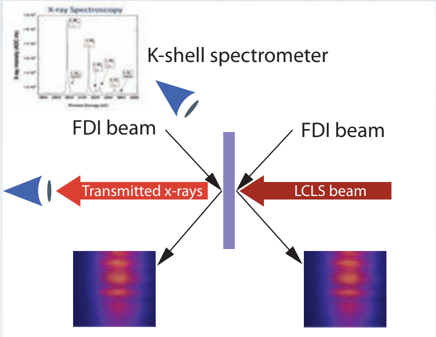

Experimental layout to measure the effects of high pressure on bound states of plasmas performed at the Linear Coherent Light Source at Stanford.

Experimental Determination of Dense Plasma Effects on Bound States in Extreme States of Matter

Ronnie Shepherd (09-ERD-032)

Abstract

We propose a series of experiments coupled with calculations and simulations to study well-characterized plasmas to determine the conditions in which the outer electrons are no longer bound to the ionic core in a dense plasma. We will measure K-shell absorption spectra while varying the electron density with the use of aerogels and shock-compressing solids. In addition to absorption, we will measure target temperature to account for ionization. These measurements will test the effects of continuum lowering and pressure ionization in dense plasma—conditions that occur in stellar interiors and nuclear explosions.

We expect to provide detailed data on the average occupation number for outer bound states in plasmas ranging from weak to strong coupling regimes. We expect these data to benchmark pressure ionization and continuum-lowering models. This groundbreaking work is expected to appear in high-profile journals and set a new standard in plasma physics experiments.

Mission Relevance

This project supports LLNL's national and energy security missions by improving advanced physical models and enabling critical calculations for weapons physics and inertial-confinement fusion.

FY11 Accomplishments and Results

During our third year we (1) analyzed the data from FY10 experiments, (2) fabricated additional aerogel targets for sub-critical experiments, (3) completed development of the time-resolved transmission grating spectrometer, and (4) performed a high-pressure experiment at the Linear Coherent Light Source at Stanford, which we used because of problems at Livermore's Jupiter Laser Facility. As a result of this project, we created high pressure by isochoric heating of silver, and simulations suggest the silver reached a pressure of 20 Mbar. The L-shell spectrum was measured to determine the effects of high pressure on the outer bound states. We successfully demonstrated our technique of measuring the effects on bound states by x-ray backlighting of short-pulse laser-heated matter. Using short-pulse laser-heated solids, we monitored the change in ionization balance as the density dropped from hydrodynamic decompression. We were, however, unsuccessful in acquiring data in the compressed matter regime, and the field still needs this data to complete the understanding of density effects on bound states. We suggest further experimentation on compressed matter, and will attempt these measurements as part of a larger programmatic campaign at Livermore.

Publications

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Arc Initiation of High Explosives

James McCarrick (09-ERD-042)

Abstract

Recent experiments have shown unique aspects of arc initiation in high explosives that cannot be predicted or reproduced with existing computational models, such as those with low thresholds that scale inversely with input power. These issues pose significant implications for the safety and surety of existing and future weapon systems. Our objective is to develop a fundamental understanding of the physical mechanism of arc initiation in high explosives. To this end, we will perform experiments required to support the physical model for specific explosives. We will also develop a rigorous, nonempirical physical model appropriate to the unique combinations of plasma energy transport and high-temperature, high-explosives kinetics. Time-resolved infrared spectroscopy and microcalorimetry will supply data to support this model.

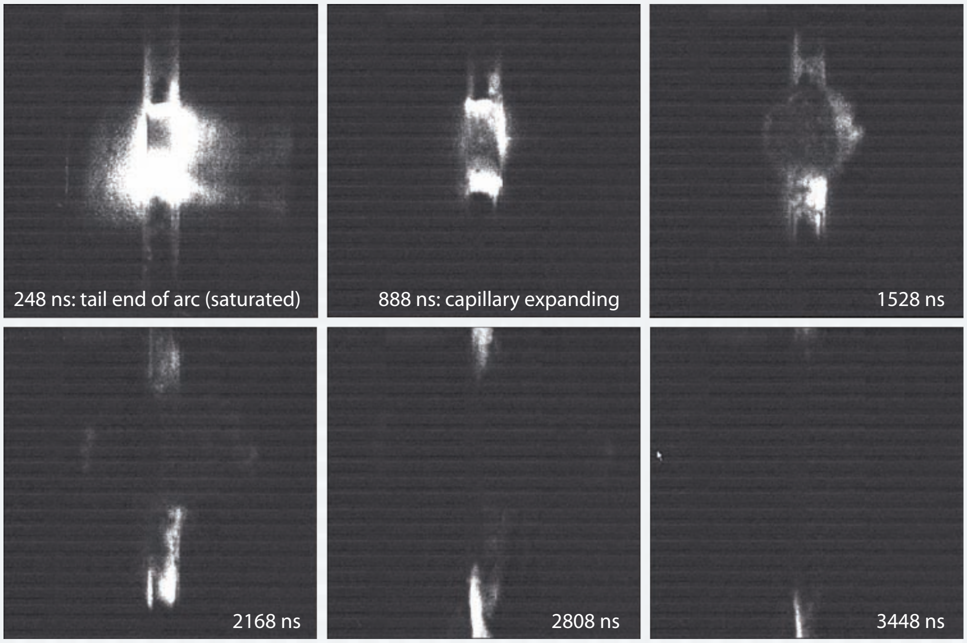
This project will result in a fundamental understanding of the (1) processes governing energy transport in confined high-power arcs, (2) kinetics of high explosives in the unique limit of temperatures of the same order as activation energies, and (3) transition process from a static volume of overdriven reactive material to a propagating detonation. This combination of knowledge will yield a physical model that will provide previously nonexistent predictive capability for safety analysis and future engineered applications. We expect publications in peer-reviewed journals and patent opportunities relating to the use of high explosives for mining operations.

Mission Relevance

This work supports the Laboratory's mission in national security by achieving better understanding of the safety of the existing nuclear stockpile and by improving basic high-explosives science, which in turn enables improved safety and surety of the future designs that will ultimately be necessary for a more efficient weapon complex. In general, this work provides a strong head start in the Laboratory becoming a high-explosives center of excellence as part of NNSA's nuclear weapons complex transformation.

FY11 Accomplishments and Results

In our modeling work, we analytically demonstrated the existence of a self-sustaining "pumped convective burn" wave, a newly identified type of wave that is capable of fast energy release—much faster than a thermal explosion, albeit still slower than a full detonation—in porous high explosives without requiring shock compression of the solid phase or compaction of the porous material. In our time-resolved infrared spectroscopy experiments, we performed capillary tube experiments in which a high-power arc completely fills the test volume. In low-density pentaerythritol tetranitrate, we observed rapid energy release, with 1.5-mg samples able to drive capillary expansion speeds of about 1.7 mm/µs in a geometry that precludes any sort of run-up as would occur in deflagration waves driven by more traditional thermal insults. We also tested triamino-trinitro-benzene and confirmed that it is insensitive to this class of insult, at least at the fixed test level of 100 mJ/mg. The pumped convective burn modeling and small-scale testing capabilities developed in this project have been adopted for nuclear weapons engineering at LLNL. The next step would be to investigate the applicability of these capabilities and our new knowledge about the phenomenon of pumped convective burn to a more general class of prompt thermal insults, including lightning.



Frames from a high-speed video of 1.5 mg of low-density pentaerythritol tetranitrate subjected to an electrical arc in a glass capillary tube. The energy deposition is roughly uniform throughout the volume, indicating that the observed violence of the response is from the fast thermal insult and not from a propagating run-up. The input energy is much less than the stored chemical energy. Expansion speed is estimated at 1.7 mm/µs.

Publications

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An Atomic Inner-Shell X-Ray Laser Pumped by the Linac Coherent Light Source

Alexander Graf (09-LW-044)

Abstract

We propose to develop a discretely tunable x-ray laser that will produce femtosecond pulses of nanometer wavelengths by using Stanford's Linac Coherent Light Source (LCLS) to pump an atomic inner-shell x-ray laser. In contrast to other methods, our inexpensive technique can be directly implemented on one of the LCLS user instruments. Diagnostic techniques such as x-ray photoelectron spectroscopy and diffraction would benefit from femtosecond-time resolution. The new source would enable the study of nonlinear quantum optical effects in the x-ray regime for the first time, opening avenues to new basic science. We will assess the optimal pumping regime by numerically simulating the time-dependent gain and fielding a first experiment at LCLS, demonstrating lasing at 1.4 nm in singly ionized neon.

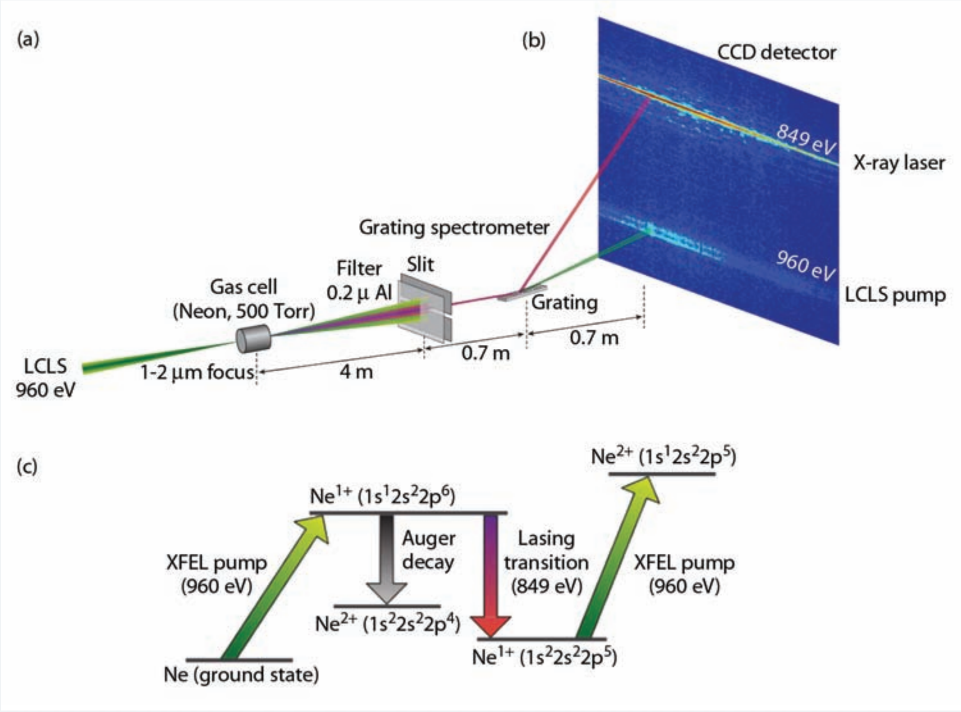
We intend to demonstrate our proposed lasing scheme of inner-shell photoionization with LCLS on atomic neon to achieve x-ray pulses at nanometer wavelengths, pulse durations of a few femtoseconds, and intensities reaching 5×10^{17} W/cm. With our method, LCLS pulses will be transformed into x-ray pulses of shorter duration and better longitudinal coherence. The increased time resolution would enable pump–probe studies of chemical reactions of an ensemble of molecules or phase transitions in solids. The longer coherence time and higher shot-to-shot spectral stability will make possible, for the first time ever, the direct study of nonlinear quantum optical effects in the x-ray regime.

Mission Relevance

The proposed work supports LLNL's mission in frontier science and advanced technology by developing a beyond the state-of-the-art instrumental capability studying ultrafast, nonlinear physics and the physics of interactions of high-intensity x rays and atoms.

FY11 Accomplishments and Results

In FY11 the data from the first experiment in FY10 was evaluated and interpreted. Our x-ray laser was shown to have a narrow width (determined by the excited-state lifetime broadening) and no spectral jitter when compared to the LCLS x-ray free-electron laser (XFEL) beam. Doubling the XFEL energy showed an average increase in the x-ray laser line energy of four orders of magnitude, which exceeds the brilliance of extreme ultraviolet plasma lasers by two to three orders of magnitude. A one-dimensional, time-dependent, self-consistent gain model was developed to estimate the gain parameters of our x-ray laser. In addition, we performed a second experiment at LCLS that measured resonant photon pumping in neon. This project resulted in the first realization of an x-ray laser in the kiloelectronvolt regime based on atomic population inversion, driven by rapid K-shell photoionization using pulses from an XFEL. Our scheme provides greatly increased wavelength stability, monochromaticity, and improved temporal coherence compared to present-day XFELs. These x-ray lasers may prove useful for high-resolution spectroscopy and nonlinear x-ray studies, and we expect our research will continue at the DESY German research center for particle physics.



In our experimental scheme (top) the Linac Coherent Light Source (LCLS) free-electron x-ray beam is focused into a gas cell filled with neon to a focus spot of 1 to 2 µm. A flat-field x-ray grating spectrometer was fielded at about 4 m beyond the cell and a charge-coupled device (CCD) captured the transmitted laser images. The bottom image shows population inversion of the $1s^12s^22p^6$ to $1s^22s^22p^5$ transition that is created by K-shell photoionization of neutral neon. The Auger decay time of the inverted state (2.4 fs) dominates the kinetics of the system in the small-signal gain regime. The lower lasing state is depleted by K-shell photoionization.

Publications

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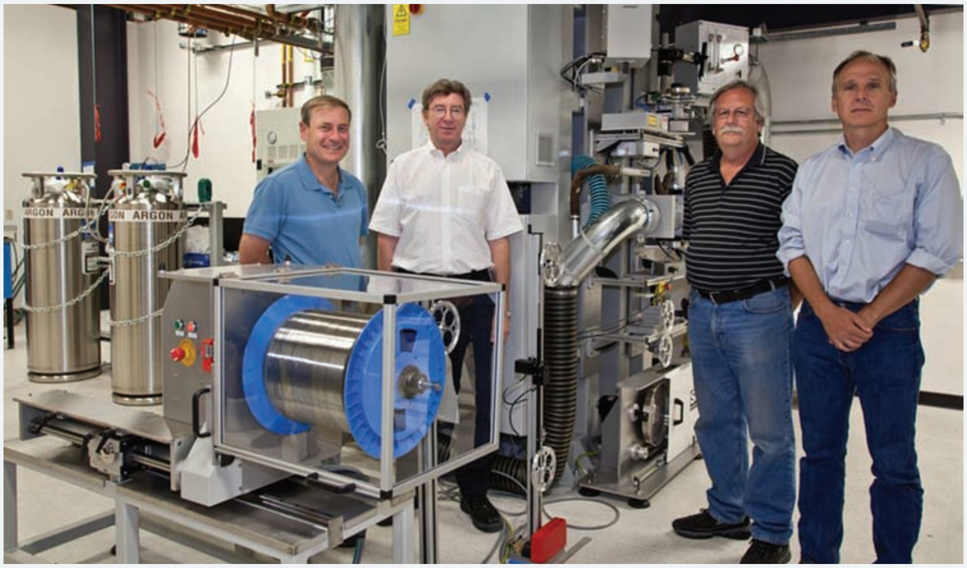
Science and Technology of Unconventional Fiber Waveguides for Emerging Laser Missions

Jay Dawson (10-SI-006)

Abstract

Fiber lasers are a critical technology for 21st-century energy and defense missions. Fiber lasers have scaled in output power and pulse energy in the last 25 years and are reaching limits in output power (~20 kW) and pulse energy (~4 mJ) imposed by the physics of a technology originally developed for megawatt and picojoule telecommunications systems. Livermore theoretical models show the physical reasons for the limits, which suggest that radical, new optical-fiber designs based on rectangular ribbon cores are needed to enable fiber lasers with up to two orders of magnitude more pulse energy and output power. We propose to provide the research and development to enable these new waveguide designs with flexible, efficient transport of ultrahigh-power light from any laser source to the end application.

We expect to develop and demonstrate a breakthrough ribbon-fiber waveguide with the potential to achieve 100-kW to 1-MW continuous-wave laser power from a single fiber-laser aperture. The ribbon fiber will enable higher-energy laser pulses from fiber sources. A range of wavelengths will be explored, from 900 to 2000 nm, and we will construct a comprehensive model of the waveguide and laser physics. We intend to (1) develop new optical fibers and fabrication techniques; (2) model, fabricate, and test mode-conversion techniques; and (3) construct and test a pulsed system demonstrating all but the thermal physics for scaling fiber lasers beyond 10 kW. Numerous publications and patents are anticipated and the new technology will enable new missions.



Optical-fiber draw-tower installation team after initial fiber draw. From left to right: Jay Dawson, Will Hardman (SG Controls, tower vendor), John Tassano, and Mike Messerly.

Mission Relevance

Development of megawatt-power fiber lasers falls squarely within the LLNL mission thrust area of advanced laser optical systems and applications. Fiber lasers are an important technology needed to produce efficient, high-average-power beams with good beam quality in a compact, robust form at a variety of wavelengths. Extending these lasers to higher pulse energies and average powers will support national missions as diverse as directed-energy laser weapons, laser rangefinders and remote sensors, mono-energetic gamma ray generation, laser-based particle acceleration, K-alpha x-ray sources, extreme ultraviolet sources, and advanced machining.

FY11 Accomplishments and Results

In FY11 we (1) successfully incorporated thermal and laser gain effects in a beam-propagation method model—the code is currently being employed to explore ribbon-fiber laser parameters; (2) completed the draw tower installation; (3) purchased spatial light modulators and used them to demonstrated the initial concept—we successfully converted a low-order Gaussian mode into a higher-order ribbon mode using our design; (4) investigated long-period gratings—it was determined that in the case of the ribbon fiber, a useful long-period grating would need to be over 1-m long, which is not practical; (5) successfully showed, as part of our long-period grating investigation, that we could use a point perturbation to create a similar effect, although with lower efficiency than desired; and (6) developed higher-order large flattened mode waveguide designs and filed patent applications.

Proposed Work for FY12

We will (1) use our draw tower to fabricate photonic crystal ribbon fibers, including gain-selective ribbon fibers and possibly flattened, higher-order-mode fibers; (2) complete development of our modeling tools for designing and characterizing photonic crystal fibers, as well as studying laser effects of ribbon fibers; and (3) demonstrate a high-power ribbon-fiber laser amplifier operating in the 1-kW regime with the goal of achieving these power levels in a single-frequency laser, which would exceed the current state of the art in fiber technology.

Publications

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Advanced Rare-Event Detectors for Nuclear Science and Security

Adam Bernstein (10-SI-015)

Abstract

Remote nuclear reactor monitoring using antineutrino detectors could revolutionize global nonproliferation efforts by providing continuous and near-real-time monitoring of plutonium production at its source. We propose to lay the foundation for a program to develop ultrasensitive rare-event neutral particle detectors for global nuclear security. We will collaborate on five international, next-generation physics experiments to enable reactor monitoring at hundreds of kilometers and fissile material detection at a standoff of hundreds of meters. We hope to provide neutral-particle detector capabilities with dramatically improved standoff distance, energy spectroscopy, directionality, particle identification, cost effectiveness, and robustness.

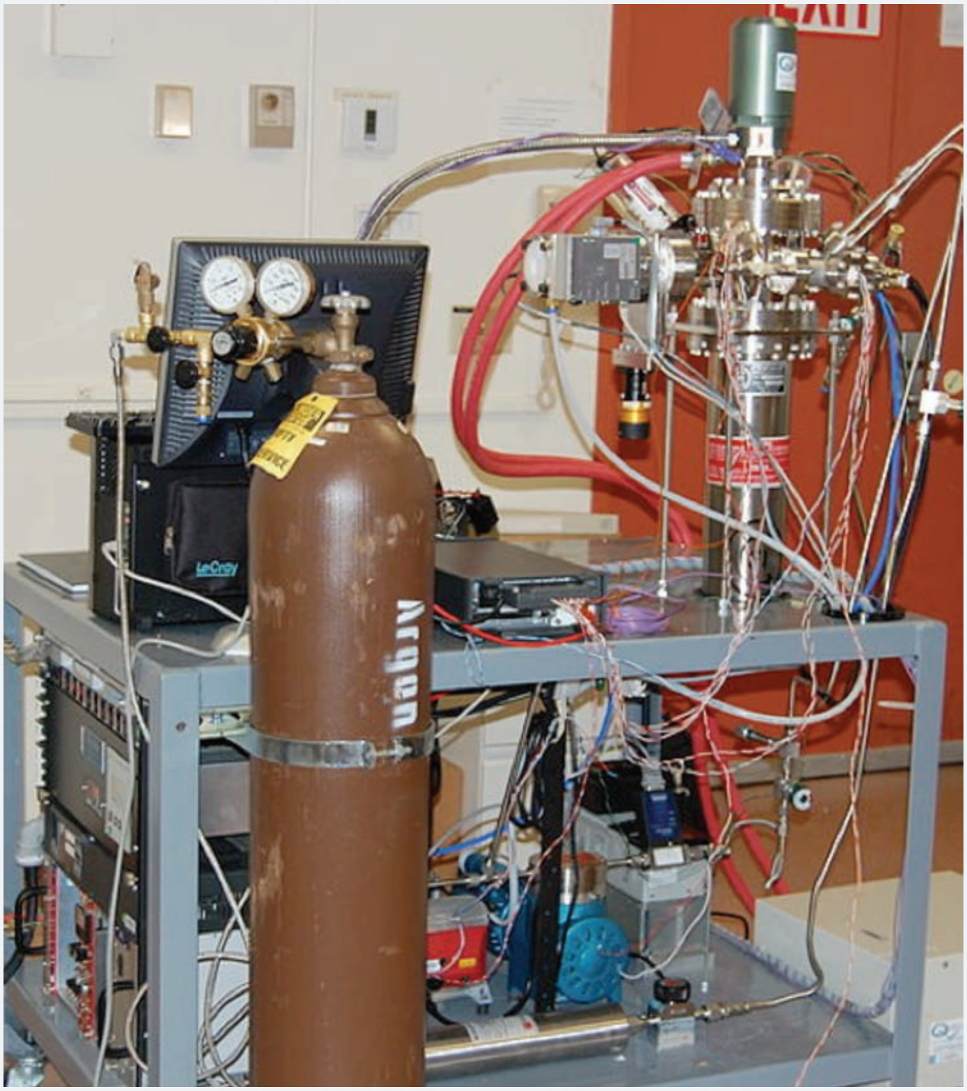
We expect to transform two core global nuclear security missions that require breakthrough research and development in rare-event detection: standoff monitoring of nuclear reactors and detection of noncritical special nuclear materials. We will apply transformative capabilities from fundamental science and establish Livermore as a world center for rare-event detection. We intend to demonstrate technology for the standoff detection of nuclear reactors with antineutrino detectors, which will enable discovery of operating reactors across borders, as well as provide reactor safeguards. In addition, we will develop technologies for fissile material search and monitoring by addressing essential needs for practically deployed systems, including neutron and gamma-particle identification, directionality, nanosecond-scale timing resolution, spectroscopy, and low-cost, large-solid-angle coverage.

Mission Relevance

The detection of rare gamma rays, neutrons, and antineutrinos emitted by nuclear materials and reactors directly supports Laboratory efforts in nonproliferation and homeland and national security. This project integrates the high-profile fundamental science of dark matter and neutrino physics with the very practical research required to achieve breakthroughs in reducing the global nuclear threat. In addition, it will provide the Laboratory access to cutting-edge technology, expertise, and potential hires.

FY11 Accomplishments and Results

In FY11 we began collecting data with a neutrino detector at the Double Chooz experiment in France, collaborating with the Massachusetts Institute of Technology to provide a predicted antineutrino rate based on a reactor simulation; (2) completed the next phase of data collection at the Cern Axion Solar Telescope in Switzerland, including design of a large-scale follow-on detector for possible deployment in the U.S.; (3) completed aboveground deployment of the world's largest xenon detector, the Large Underground Xenon Detector at the Sanford Underground Laboratory site in South Dakota; (4) achieved the first-ever operation of a 1-kg argon dual-phase



Livermore's argon dual-phase detector, built to detect coherent scatter using a reactor neutrino source—an essential precursor for further lowering the energy threshold for dark matter detection.

detector built to measure ionization quenching in liquid argon and ultimately detect coherent scatter using a reactor neutrino source—this measurement is an essential precursor for further lowering the energy threshold for dark matter and coherent scatter detection in argon and xenon; and (5) published several important articles on the performance of xenon detectors—including a detector with a stringent limit on low-mass weakly interacting massive particles—and presented our results at conferences, including talks on groundbreaking work on the properties of xenon detectors at low ionization thresholds by a postdoctoral researcher.

Proposed Work for FY12

In FY12 we will (1) publish the world's most precise limit of the neutrino mixing angle theta-13; (2) perform a dark matter search using the Large Underground Xenon Detector, which we expect to result in discovery of the world's most stringent limit on weakly interacting massive particles of dark matter; (3) continue to investigate scaling properties for large scintillator and doped-water Cerenkov detectors in the context of a wide-area search for exclusion of small (10-MWt) reactors; and (4) continue to study scintillators capable of advanced pulse shape discrimination to improve sensitivity and reduce costs for near-field reactor monitoring devices.

Publications

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Sorensen, P., 2010. "A coherent understanding of low-energy nuclear recoils in liquid xenon." *J. Cosmol. Astropart. P.* **9**, 033. LLNL-JRNL-454331.

Sorensen, P., 2011. "Anisotropic diffusion of electrons in liquid xenon with application to improving the sensitivity of direct dark matter searches." *Nucl. Instrum. Methods* **635**, 41. LLNL-JRNL-471078.

Sorensen, P., and C. E. Dahl, 2011. "Nuclear recoil energy scale in liquid xenon with application to the direct detection of dark matter." *Phys. Rev. D* **83**, 063501. LLNL-JRNL-471079.

Sorensen, P., et al., 2010. "Lowering the low-energy threshold of xenon detectors." *Proc. 8th Intl. Workshop on the Identification of Dark Matter (IDM2010)*. LLNL-PROC-463991.

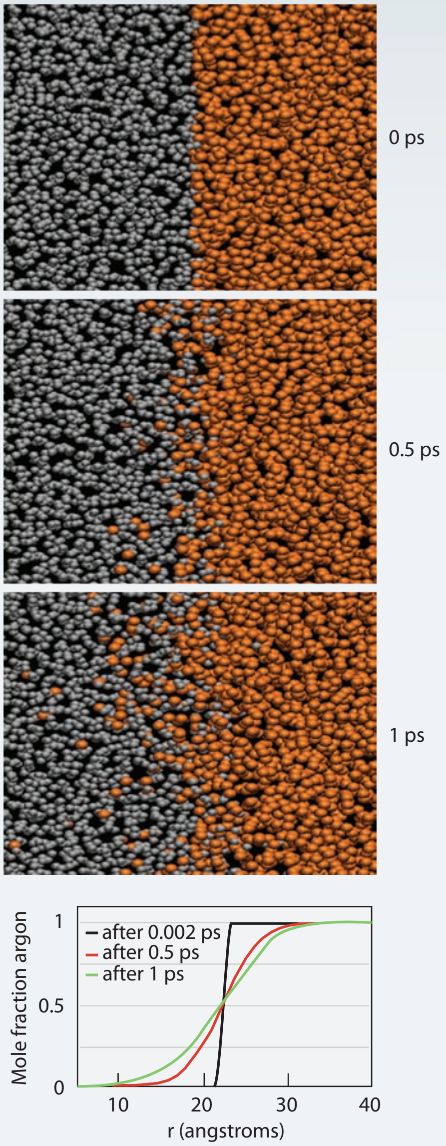
Mix at the Atomic Scale

Paul Miller (10-ERD-004)

Abstract

The need to compute the effects of mixing of materials in a dynamical system is central to a wide variety of mission-relevant work at LLNL. Mixing involves diffusion and is dependent on processes occurring at the atomic scale. Approximations are employed in continuum simulations of mixing that simplify or ignore complications and subtleties of diffusion and transport coefficients. We aim to formulate better mixing models by employing both continuum hydrodynamics and molecular dynamics codes—both Livermore strengths. We will develop metrics and a framework for the interchange of information between the two approaches, use molecular dynamics methodology to uncover atomic-level transport properties, and use a continuum hydrodynamics code to define limitations of the continuum approach and explore strategies for improving models.

We expect to develop a transport theory with the aid of molecular dynamics and continuum simulations. Large-scale simulation problems will be used for discovery and exploration, to gauge the adequacy of the methods, and to assess model improvements. Through an analysis of these simulations, we will begin developing a more complete, atomically informed model of mixing for use in continuum calculations. By the end of this project, we expect to have made significant improvements in our ability to model complex mixing processes at both atomistic and continuum levels of resolution, resulting in substantial advancements in our understanding of diffusion and mixing.



Snapshots of an isothermal and isobaric interface broadening of a hydrogen and argon mixture modeled with the Yukawa potential. Transport coefficients can be extracted by analyzing the composition profile in these results.

Mission Relevance

By improving our ability to simulate nuclear weapon-related mixing phenomena more accurately, this work supports the Laboratory's mission in stockpile stewardship.

FY11 Accomplishments and Results

In FY 11 we (1) continued to develop the particle physics Yukawa potential (screened Coulomb potential), conducting a first round of simulations, and improved the continuum modeling algorithms; (2) conducted simulations of diffusivity and viscosity in molten aluminum and copper alloys as a function of concentration, pressure, and temperature; (3) compared metrics for atomistic and continuum models; and (4) derived, from molecular dynamics simulations, an equation of state for the continuum code and carried out large-scale molecular dynamics simulations of interface broadening in mixed plasmas using the Yukawa interaction.

Proposed Work for FY12

We will (1) use our Yukawa capability to simulate diffusivity and viscosity in a hot, dense mixed-plasma test case; (2) complete work on the equation of state derived from the molecular dynamics code and submit the results for publication; (3) complete a comparison of our molecular dynamics and continuum results; and (4) explore, if our preliminary results warrant, use of a large-scale Yukawa plasma simulation to further examine transport processes in the presence of fluid mixing.

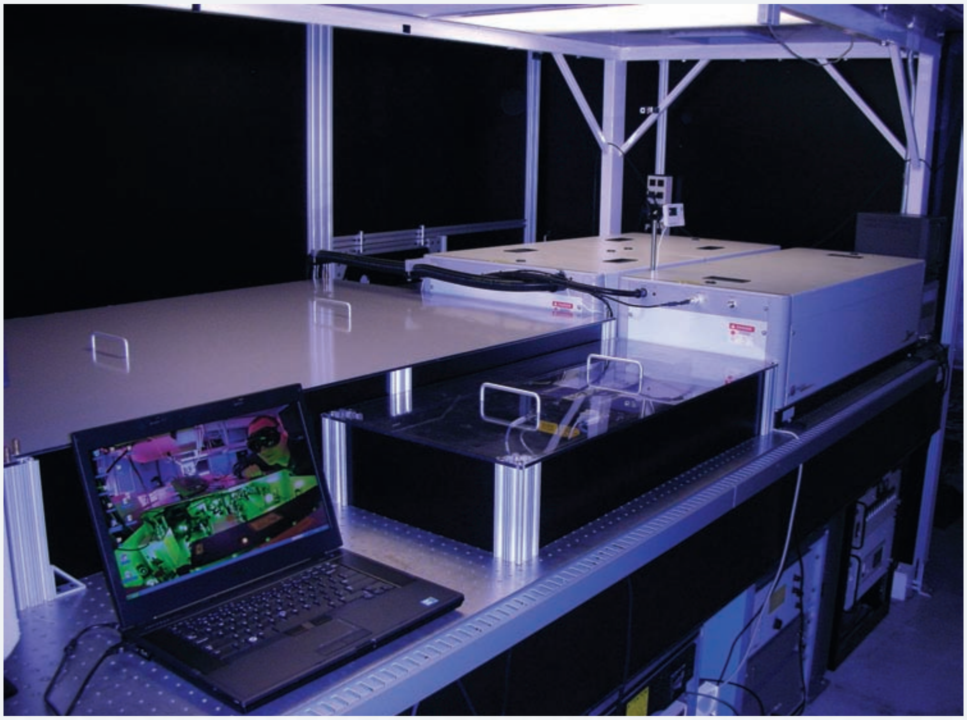
High-Gradient Inverse Free-Electron Laser Accelerator

Scott Anderson (10-ERD-026)

Abstract

We will employ the inverse free-electron laser (IFEL) advanced acceleration technique to prove the feasibility of its use to create compact light sources. The IFEL is a leading contender to demonstrate high-gradient, high-quality acceleration at medium-energy ranges, and scales naturally to megahertz repetition rates with advancing laser technology. This technology has the potential to serve as the acceleration mechanism in deployable, compact light sources for special nuclear materials detection, or to provide tabletop, gigaelectronvolt-class accelerators for academia and industry. Our IFEL will capture a 50-MeV beam and accelerate it to 125 MeV through a 50-cm undulator installed at Livermore's high-brightness electron facility.

A successful IFEL demonstration—accelerating a 50-MeV beam to 125 MeV—will change the approach for future accelerator design for applications needing from



The titanium-doped sapphire inverse free-electron laser drive produces 500-mJ, 100-fs, 785-nm pulses at a 10-Hz repetition rate with a tabletop footprint.

50 MeV to a few gigaelectronvolts. The tabletop footprint of our IFEL opens the door for academic and industrial applications needing compact, high-quality, low-emittance gigaelectronvolt beams not currently available. Compared to conventional accelerators, this IFEL will enable a reduction in size by a factor of 20 and cost by a factor of 10. Meter-scale IFEL accelerators have the potential to produce greater than 350-MeV high-brightness electron beams with increasingly high repetition rates made possible by ever-advancing laser technology. As such, IFELs are attractive for future gamma-ray sources. Our IFEL will lead the advanced accelerator community, demonstrating compact, high-quality, high-gradient acceleration.

Mission Relevance

The accelerator technology developed in this project supports the Laboratory's mission to reduce or counter threats to national and global security. Our project specifically impacts Livermore's strategic mission thrust of nuclear threat elimination by providing capability for standoff detection and identification of nuclear materials. In addition, it supports efforts in cutting-edge science—specifically, energy manipulation to reduce accelerator footprints by three orders of magnitude at fixed energy. Also, the LLNL strategic thrust of advanced laser optical systems and applications will be supported through the potential to upgrade the Laboratory's mono-energetic gamma-ray source for nuclear resonance fluorescence science and applications.

FY11 Accomplishments and Results

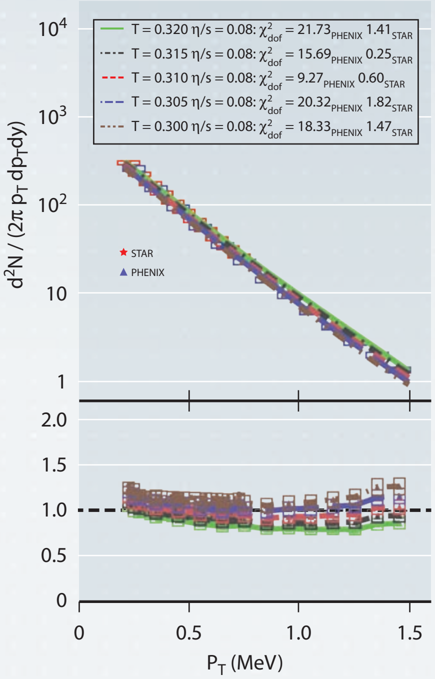
We (1) installed the IFEL front-end system and verified that it met design specifications: 25 mJ of infrared radiation, compression to a pulse duration of 100 fs, and 200 µJ of ultraviolet light for photo-cathode drive; (2) acquired an IFEL high-power amplifier; (3) designed and began construction of the IFEL electron beam line; (4) demonstrated 720-mJ laser operation; (5) finalized high-power laser transport design; and (6) presented our work at the 2011 Particle Accelerator Conference.

Proposed Work for FY12

In FY12 all tasks necessary to complete the experiment will be performed. Specifically, we will (1) finish acquiring laser system components and construct the high-power vacuum transport system; (2) perform IFEL acceleration experiments, which include spatially and temporally overlapping the laser and electron beams, optimizing beam size and focal position, and characterizing IFEL energy gain, energy spread, emittance, and beam stability; and (3) communicate our research at conferences and in publications in collaboration with the University of California, Los Angeles.

Publications

Anderson, S. G., et al., 2011. *The LLNL/UCLA high gradient inverse free electron laser accelerator.* 2011 Particle Accelerator Conf. (PAC'11), New York, NY, Mar. 28–Apr. 1, 2011. LLNL-PROC-478111.



STAR and PHENIX experimental data at the Large Hadron collider compared with results obtained with the CHIMERA (comprehensive heavy-ion model evaluation and reporting algorithm) simulation package developed under this project.

Modeling and Measuring Quark–Gluon Plasma Shock Waves

Ron Soltz (10-ERD-029)

Abstract

The possibility of developing shock waves in heavy-ion collisions was first suggested as a consequence of the compression of nuclear matter during the initial stages of collision. The discovery of jet suppression has renewed interest in the possibility that high-energy partons—quark or gluons—generated by hard scatterings early in the collision process might induce shock waves in the surrounding medium. We propose to collaborate with theorists to model the jet energy loss and medium response with sufficient accuracy to determine whether measurable signatures exist. This project leverages an LLNL-developed capability that has already been successfully used to identify non-Gaussian, long-range components in the space–time emission of pions and kaons in heavy-ion collisions.

We expect to use hydrodynamic models to predict the space–time signatures of shock waves in heavy-ion collisions, and to measure these signatures at the ALICE (A Large Ion Collider Experiment) detector facility during lead-ion collision runs at the Large Hadron Collider near Geneva. This project leverages the Laboratory's high-performance computing capabilities and will bring new talent to LLNL by hiring a postdoctoral researcher to run hydrodynamic models.

Mission Relevance

Heavy-ion collisions, which elucidate some of the most fundamental physics of matter, are studied with some of the most complex detectors and models ever developed. By advancing capabilities in modeling heavy-ion collisions, this project extends the capability for modeling the transport of particles and photons in matter, a required capability for radiation detection systems in national security and many other applications. This project also extends capabilities in high-energy-density science and very-large-data manipulation tools, in support of the Laboratory's national security mission.

FY11 Accomplishments and Results

In FY11 we (1) continued to develop our two-dimensional hydrodynamic model with microscopic transport, increasing the list of final-state resonances to match the known resonances of particle data from the Large Hadron Collider; (2) completed the CHIMERA (comprehensive heavy-ion model evaluation and reporting algorithm) software tools, enabling a quantitative comparison between model results and multiple data measurements; (3) added a Monte Carlo Glauber model to accommodate nucleon position fluctuations in the initial state, which is important because initial-state fluctuations have been suggested as a possible alternative to shock-wave measurements; and (4) initiated a collaboration with a group that authored a new three-dimensional hydrodynamic model that incorporates a more faithful representation of fluctuations in the initial state and will therefore be better suited to incorporate energy-loss mechanisms.

Proposed Work for FY12

In FY12 we will finish the two tasks remaining to complete the project's proposed research: incorporating energy loss into our hydrodynamic model to predict the magnitude of the momentum and space–time shock-wave signatures, then comparing them to initial-state fluctuation effects, and analyzing data from the Large Hadron Collider to begin the search for this signature.

Publications

Garishvili, I., 2010. *Modeling hydrodynamic properties of the QGP at RHIC*. 2010 Fall Mtg. APS Division of Nuclear Physics, Santa Fe, NM, Nov. 2–6, 2010. LLNL-CONF-461619.

Soltz, R., 2010. *Quantifying properties of the QGP*. Institute for Nuclear Theory Workshop: Quantifying the Properties of Hot QCD Matter, Chicago, IL, June 14, 2010. LLNL-PRES-418133.

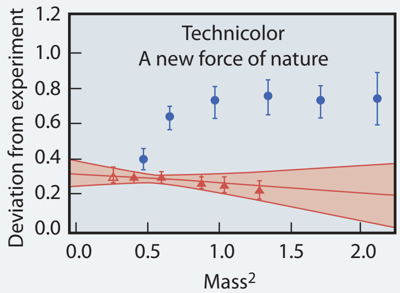
Unlocking the Universe with High-Performance Computing

Pavlos Vranas (10-ERD-033)

Abstract

We propose to use large-scale numerical simulations on LLNL's BlueGene supercomputers to unravel the inner workings of the universe—from the physics beyond the standard model of elementary particles created a trillionth of a second after the Big Bang, to the quark–gluon plasma created a millionth of a second after the Big Bang, to the stable nuclear matter that comprises most of the visible universe today. We will compare and contrast our simulation results with current and upcoming experiments at the Large Hadron Collider near Geneva, Switzerland. We also intend to study the underlying theories of quantum chromodynamics (QCD) and the Yang–Mills theories, which can only be studied numerically using simulations on a discretized space–time (lattice) mapped onto the nodes of massively parallel supercomputers.

We expect to develop a theoretical understanding of the inner workings of nature spanning three layers of physical phenomena: (1) the physics of mass generation of elementary particles, (2) the quark–gluon plasma and its transition to nuclei, and (3) the interactions of nuclei and the emergence of nuclear physics. The overall deliverable is the prediction, using the most fundamental scientific starting point, of particles and phenomena that will be observed in extremely high-energy ion collisions. Our work will help understand and guide the largest and highest-energy experiments of our time—those performed at the Large Hadron Collider. No other groups in the world are pursuing this type of research at this scale.



Technicolor is a possible theory for particle physics at energies that are currently being accessed for the first time ever at the Large Hadron Collider. Technicolor introduces a new force of nature, and numerical simulations on Livermore's BlueGene supercomputers indicate that it is consistent with established experimental results, and therefore is a viable theory for the new physics being probed at the collider. As the particle mass is lowered, a Technicolor theory with six types of particles (blue points) deviates less from experiment. A theory with two types of particles (red points) deviates even less (a value of ~0.3) and does not change much with varying mass.

Mission Relevance

The use and development of high-performance computing underlies all aspects of this proposal and impacts multiple Lawrence Livermore missions. For example, methods and algorithms we develop will be relevant to the new multicore architectures on which the most advanced stockpile stewardship simulations are now run. In addition, first-principles calculations of the nuclear force at the QCD level are directly relevant to light-ion reaction cross sections in fusion energy. By establishing Livermore as a world leader in this cutting-edge area of physical theory, this project will also attract leading postdoctoral researchers.

FY11 Accomplishments and Results

In FY11 we tested theories and made important predictions about particle physics at the energies currently being accessed at the Large Hadron Collider, where our predictions could be verified. Specifically, we (1) performed simulations to measure the spectrum and scattering parameters of the 2- and 6-flavor SU(3) Yang–Mills theory using lattice configurations generated in FY10; (2) simulated the 10-flavor SU(3) Yang–Mills theory at low pion mass; (3) began simulations of the SU(2) Yang–Mills theory; (4) investigated the thermal transition of QCD using nearly physical pions (~200 MeV) on 163 × 8 space–time lattices; (5) began measurements of the QCD equation of state using the highly improved staggered quark discretization method for fermions, enabling LLNL to provide important input to experiments at the Relativistic Heavy Ion Collider at Brookhaven National Laboratory; and (6) completed finite-volume many-body calculation techniques within a harmonic oscillator basis that will help integrate our lattice QCD results into ab initio many-body methods and enable us to study many important nuclear physics problems.

Proposed Work for FY12

In FY12 we will (1) complete 2- and 4-flavor simulations of the SU(2) Yang–Mills theory and measure condensate enhancement as well as the spectrum and scattering parameter, which will demonstrate feasibility and properties of technicolor theories for mass generation for teraelectronvolt physics at the Large Hadron Collider; (2) complete our investigations of the thermal transition region of QCD using physical pions on large (323 × 8) space–time lattices and complete our study of the QCD equation of state using highly improved staggered quark fermions, which will enable us to determine, within a few percent, values of the relevant quantities in the QCD thermal transition region; and (3) complete studies of our new 230-MeV pion clover lattices, which will provide results on nuclear interactions with a few-percent accuracy.

Publications

Appelquist, T., et al., 2011. "Parity doubling and the S parameter below the conformal window." *Phys. Rev. Lett.* **106**(23), 231601. LNL-JRNL-508011.

Buchoff, M., 2010. *Extracting scattering parameters using the isospin chemical potential.* 28th Intl. Symp. Lattice Field Theory, Sardinia, Italy, June 14–19, 2010. LLNL-PROC-461612.

Wasem, J. V., in press. "First lattice calculation of nuclear parity violation." *Phys. Rev. Lett.* LLNL-JRNL-491671.

Discovery and Synthesis of Materials for High-Energy-Density Science

Stanimir Bonev (10-ERD-038)

Abstract

We propose to develop a first-principles computational framework that will enable prediction of the phase stability of materials at finite temperature, thereby accelerating discovery and synthesis of high-energy-density materials. Specifically, we will study nitrogen- and carbon-rich molecular compounds that, under the application of pressure, transform from weakly bound molecular crystals to covalent solids. Large amounts of energy can be released when a transformation is induced from a covalent crystal—a metastable high-energy state at low pressure—to the stable low-energy molecular phase. The potential use of such materials for practical applications depends on the ability to produce significant amounts of the high-energy-density state and to preserve the attained modifications when the materials are cooled to near-ambient conditions.

Successful project completion will result in development of a computational toolkit for examining similarities between low-atomic-number, high-pressure liquid and solid phases and for carrying out automated phase searches that are specifically designed to predict finite-temperature phase stability. We will apply these methods, in close collaboration with experiments, to examine the effect of impurities on phase transformations in nitrogen- and carbon-rich materials, which are good candidates for achieving metastable high-energy-density states of matter. The primary benefit of this work will be development of a first-principles-based approach that can be used to accelerate discovery and synthesis of novel materials for high-energy-density science.

Mission Relevance

The proposed project will result in new computational capabilities at LLNL that are directly applicable to the cross-cutting challenges of the Laboratory's science, technology, and engineering foundation. In particular, by combining sophisticated methods for automatic phase exploration with first-principles simulation methods, we will develop an integrated approach that will enable a detailed understanding of high-energy-density matter and will accelerate the discovery of new materials relevant to LLNL's mission of ensuring the safety, security, and reliability of the U.S. nuclear deterrent. In addition, continued development and application of predictive simulation capabilities will make full use of the Laboratory's investments in high-performance computing and simulation.

FY11 Accomplishments and Results

We (1) computed and characterized the phase diagram of carbon dioxide up to 200 GPa and 10,000 K, including regions of stability for polymeric liquid phases; (2) completed our simulations and analysis of nitrogen-rich mixtures and started those of mixtures rich in carbon dioxide; (3) began searches for novel nitrogen-rich polymeric solids as high-energy-density material candidates, using input from liquid simulations as our starting point; (4) worked on developing phase search methods applicable directly at finite temperature; and (5) developed and tested a novel approach for computing the free energy of liquids, with the same accuracy but ten times the efficiency of the ab initio thermodynamic integration technique.

Proposed Work for FY12

In FY12 we will (1) complete the liquid preconditioned crystalline searches for nitrogen-based high-energy-density materials; (2) perform simulations of carbon dioxide liquid mixtures; (3) use these simulation results to apply crystalline searches to selected systems of interest as potential high-energy-density candidates; (4) develop and apply efficient search methods for finite-temperature phase stability using molecular dynamics; and (5) examine, if time permits, whether the presence of a surface or an interface can enhance the stability of covalently bonded nitrogen- and carbon-rich solids, selected based on the results of our crystalline searches.

Publications

Boates, B., and S. A. Bonev, 2010. *Metallic nitrogen at high pressure and temperature.* APS March Mtg., Portland, OR, Mar. 15–19, 2010. LLNL-ABS-420448.

Boates, B., and S. A. Bonev, 2010. *Metallic nitrogen*. Gordon Conf. Research at High Pressure, Holderness, NH, June 27–July 2, 2010. LLNL-POST-438642.

Boates, B., and S. A. Bonev, 2011. *Phase transitions in carbon dioxide at high pressures and temperatures.* 22nd Congress and General Assembly Intl. Union of Crystallography, Madrid, Spain, Aug. 22–30, 2011. LLNL-PRES-494816.

Boates, B., and S. A. Bonev, 2011. "Structural and electronic properties of dense liquid and amorphous nitrogen." *Phys. Rev. B* **3**, 174114. LLNL-JRNL-470580.

Boates, B., et al., 2011. *Structural and optical properties of liquid* CO_2 *up to 1 terapascal.* APS March Mtg., Dallas, TX, Mar. 21–25, 2011. LLNL-ABS-465272.

Boates, B., et al., 2011. "Structural and optical properties of liquid CO_2 for pressures up to 1 TPa." *J. Chem. Phys.* **134**(2), 064504. LLNL-JRNL-458115.

Teweldeberhan, A. M., and S. A. Bonev, 2010. *Alkali and alkaline earth metal alloys at high-pressure.* APS March Mtg., Portland, OR, Mar. 15–19, 2010. LLNL-ABS-420450.

Teweldeberhan, A. M., and S. A. Bonev, 2010. "Comment on 'Structural prediction and phase transformation mechanisms in calcium at high pressure'." *Phys. Rev. Lett.* **104**, 209610. LLNL-JRNL-421347.

Teweldeberhan, A. M., and S. A. Bonev, 2011. "Structural and thermodynamic properties of liquid Na–Li and Ca–Li alloys at high pressure." *Phys. Rev. B* **83**, 134120. LLNL-JRNL-465275.

Teweldeberhan, A. M., and S. A. Bonev, 2010. *Thermodynamic stability of alkali and alkaline earth metals at high-pressure.* Gordon Conf. Research at High Pressure, Holderness, NH, June 27–July 2, 2010. LLNL-POST-438596.

Teweldebrhan, A. M., J. L. Dubois, and S. A. Bonev, 2011. *High-pressure phases of calcium*. APS March Mtg., Dallas, TX, Mar. 21–25, 2011. LLNL-ABS-465215.

Teweldebrhan, A. M., J. L. Dubois, and S. A. Bonev, 2011. *Thermodynamic properties of solids and liquids at extreme conditions.* 17th Biennial Intl. Conf. APS Topical Group on Shock Compression of Condensed Matter, Chicago, IL, June 26–July 1, 2011. LLNL-ABS-473853.

An Intense Laser-Based Positron Source

Scott Wilks (10-ERD-044)

Abstract

Our goal is to characterize and investigate a new high-brightness positron source based on ultra-intense laser–matter interactions. This work will allow us to understand the underlying science, assess potential applications, and result in the addition of a brand new class of positron sources. We will further development of the world's most advanced direct hot-electron diagnostic for high-energy-density physics research using experiment, theory, and simulation. We will use state-of-the-art simulation codes to design experiments, execute several experimental campaigns on various high-intensity lasers around the world, and compare data with predictions.

We expect to lay the science foundation for a new class of intense positron sources. In particular, we will (1) develop an understanding of the detailed physics behind laser-to-positron coupling, (2) investigate physics of the sheath electric field behind the target and its effect on average positron beam energy, (3) measure beam emittance, (4) increase the accuracy with which laser-generated fast-electron distributions are measured, and (5) assess the possibility of generating gamma-ray radiation at 511 keV. Significantly, we will have determined the applicability of this source for researchers specializing in electron–positron jets, particle accelerators, gamma-ray generation, and noninvasive measurements of materials using positrons.

Mission Relevance

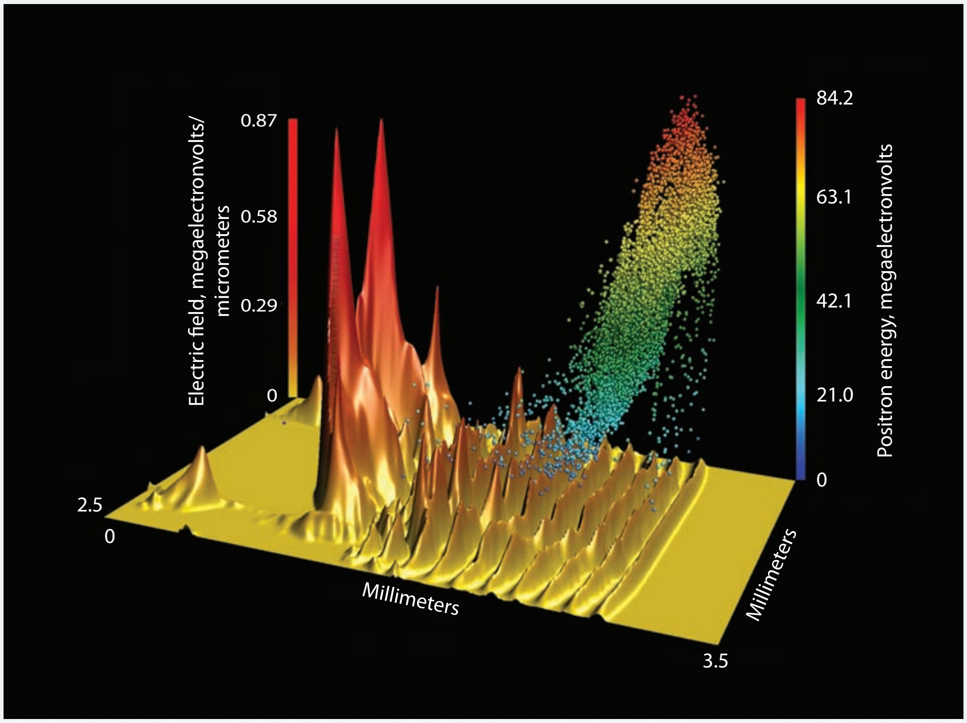
This project will lay the science foundation for a new and potentially powerful source of positrons, which may lead to new approaches for diagnosing experiments in plasma and atomic physics, fusion science, high-energy-density physics, accelerator and particle-beam science, nondestructive interrogation of materials, and astrophysics. These areas of science are foundational to the Stockpile Stewardship Program.

FY11 Accomplishments and Results

In FY11 we (1) invented, and filed a provisional patent on, a novel positron source for conventional and plasma accelerators; (2) carried out experiments to measure the emittance of laser-generated positrons; (3) built additional positron spectrometers; (4) assessed that a gamma-ray spectrometer for 511-keV radiation is not feasible; and (5) exercised previously developed advanced simulation capabilities that greatly refined our picture of positron generation using ultra-intense lasers, with emphasis on effect of the sheath field.

Proposed Work for FY12

In FY12 we will (1) finish designing our experiments to measure positron beam emittance and annihilation radiation and to focus the positrons, (2) complete work on additional positron spectrometers, (3) conduct several experimental campaigns on ultra-intense lasers around the world, and (4) use our simulation capabilities to analyze the data from these campaigns, with the goal of further refining our understanding of positron generation with ultra-intense lasers.



An integrated simulation that includes, for the first time, all the physical effects believed to occur in laser-generated electron–positron pair production. The illustration shows that the escaping positrons (particles) can gain tens of megaelectronvolts from the electric field (surface plot) generated by escaping electrons.

Publications

Chen, H., 2010. *Electron and positron generation in intense short pulse laser solid interactions.* 11th Intl. Workshop on Fast Ignition of Fusion Targets, Shanghai, China, Oct. 17–21, 2010. LLNL-ABS-444531.

Chen, H., et al., 2011. "Emittance measurement of laser produced positrons." *Bull. Am. Phys. Soc.* **56**(16), GO7.00010. LLNL-ABS-491101.

Chen, H., et al., 2010. *Laboratory produced relativistic electron-positron jets.* HEAD 2010 High Energy Astrophysics Division, Hawaii, HI, Mar. 1– 4, 2010. LLNL-ABS-423467.

Chen, H., et al., 2009. "Making relativistic positrons using ultraintense short pulse lasers." *Phys. Plasma.* **16**(12), 122702. LLNL-JRNL-416496.

Chen, H., et al., 2010. "Relativistic quasimonoenergetic positron jets from intense laser–solid interactions." *Phys. Rev. Lett.* **105**(1), 015003. LLNL-JRNL-418061.

Park, J., et al., 2010. *The effect of preplasma on intense laser produced positrons.* 52nd Ann. Mtg. APS Division of Plasma Physics, Chicago, IL, Nov. 8–12, 2010. LLNL-ABS-442939.

Szabo, C. I., et al., 2010. *Spectroscopy of positron annihilation gamma rays from laser-exited media*. 52nd Ann. Mtg. APS Division of Plasma Physics, Chicago, IL, Nov. 8–12, 2010. LLNL-ABS-450390.

Wilks, S. C., 2011. "Electron-positron jets created by ultra-intense lasers." *Bull. Am. Phys. Soc.* **56**(16), UI2.00001. LLNL-ABS-491095.

Multi-Scale Polymer Flows and Drag Reduction

Todd Weisgraber (10-ERD-057)

Abstract

The reduction of 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 multi-scale simulations of drag reduction that will increase our understanding of the underlying physics.

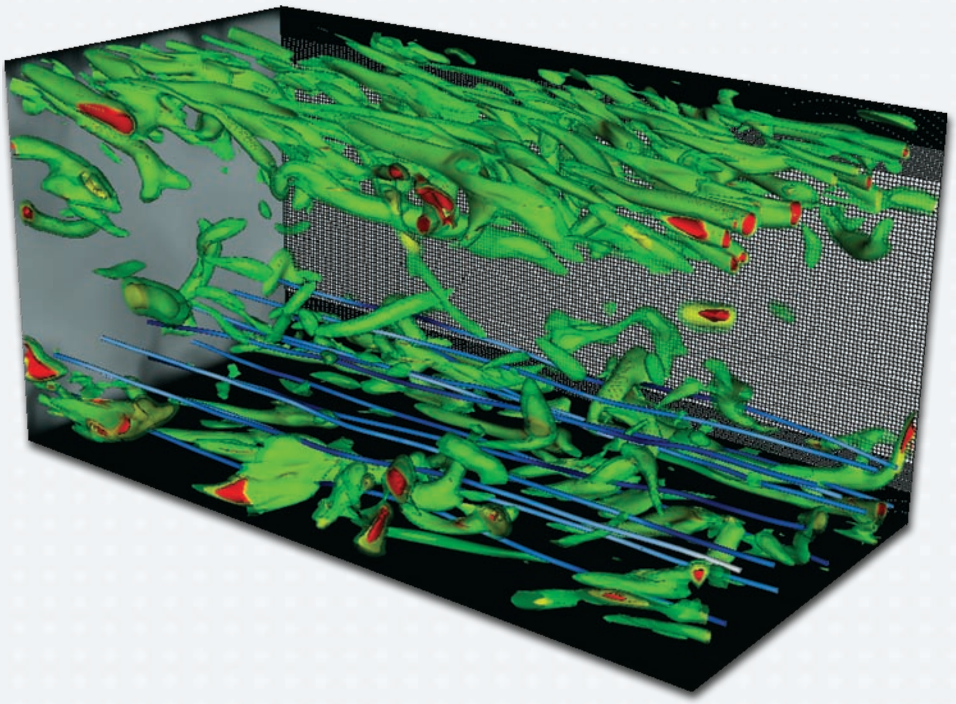
We expect to perform a series of large-scale simulations to provide valuable insight into a fundamental hydrodynamics problem, including a more detailed understanding of the complex dynamics of polymers and turbulence. Additional knowledge of drag reduction could make it possible to optimize this effect in more practical applications. For instance, 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.

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, part of the LLNL mission focus area of biosecurity.

FY11 Accomplishments and Results

In FY11 we emphasized completing the adaptive mesh refinement code for use with the lattice-Boltzmann method, accessing the Chombo library developed at



In this simulation of a turbulent channel flow, the contours (green) of a vortex structure are affected by stream tubes (blue) near the bottom wall. The fixed grid resolution can be seen on the background plane, parallel to flow direction.

Lawrence Berkeley National Laboratory. Specifically, we (1) developed a mesh-refinement algorithm for the lattice-Boltzmann method that is more accurate than previously proposed algorithms and does not require modification for steady-state problems, (2) validated our approach with several benchmark problems, and (3) began simulations of transition and turbulent flow in smooth and roughened channels. Our mesh-coupling innovation relies on fully conservative spatial and temporal interpolation algorithms at coarse–fine grid interfaces and can reduce the numerical error by a factor of ten compared to existing techniques.

Proposed Work for FY12

In FY12 we will conduct a series of large-scale turbulent drag-reduction simulations. Specifically, we will investigate flow instability, turbulent transition, and the onset of drag reduction and study how the size of roughness elements compared to the coiled polymer affects flow stability, transition to turbulence, and the magnitude of drag reduction.

Publications

Weisgraber, T., and B. Alder, 2010. *Polymer flows in channels with roughened walls.* 19th Intl. Conf. Discrete Simulation of Fluid Dynamics (DSFD 2010), July 5–9, 2010, Rome, Italy. 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 Lawrence 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 controllably 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 basic research program aimed at estimating and reducing systematic errors within the framework of ground-state and finite-temperature quantum Monte Carlo codes.

We intend to develop a robust toolset 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.

Mission Relevance

Accurate first-principles simulations of materials are essential for current and future goals of LLNL 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.

FY11 Accomplishments and Results

In FY11 we (1) successfully tested our novel Fermion path-integral Monte Carlo method by simulating low-temperature helium-3 and found good agreement between our results and known experimental values; (2) implemented Coulomb action in the Monte Carlo code and began using it to study the temperature-dependent properties of electron gas; (3) introduced an enthalpy term in our Monte Carlo code to study the constant-pressure, ground-state properties of solid hydrogen; (4) laid the groundwork to begin searching for candidate structures for the solid hydrogen III phase at high pressure; and (5) developed, coded, and began testing an unbiased released-node quantum Monte Carlo method using nonlocal pseudo-potentials.

Proposed Work for FY12

In FY12 we will (1) complete unbiased simulation of finite-temperature electron gas and begin studying the low-temperature phase diagram of hydrogen using our Fourier path-integral Monte Carlo method; (2) use this Monte Carlo method to determine the thermal conductivity of fermion-type systems, with helium-3 as a test case; (3) begin searching for candidate structures for the solid hydrogen III phase using our new constant-pressure, ground-state, path-integral methodology; and (4) perform exact released-node quantum Monte Carlo calculations of the ground-state properties of candidate structures for compressed hydrogen using the results of our candidate search, as well as structures previously proposed in the literature.

Publications

DuBois, J. L., 2011. *Algorithms for exact quantum simulations at the exascale.* LLNL-PROP-517051.

DuBois, J. L., 2011. *Developing a comprehensive simulation capability for warm dense matter.* LLNL-PROP-510091.

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. 15–19, 2010. LLNL-PRES-426109.

Tubman, N., J. L. Dubois, and B. Alder, 2011. "Recent results in the exact treatment of fermions at zero and finite temperature." *Advances in Quantum Monte Carlo.* Oxford University Press, New York, NY. LLNL-BOOK-493754.

Tubman, N., et al., 2011. "Prospects for release-node quantum Monte Carlo." *J. Chem. Phys.* **135**(18), 184109. LLNL-JRNL-473852.

Tubman, N., et al., 2010. *Transient methods in quantum Monte Carlo calculations.* Quantum Monte Carlo in the Apuan Alps VI, Vallico Sotto, Tuscany, Italy, July 24–31, 2010. LLNL-PRES-444177.

Tubman, N., et al., 2010. *Transient quantum Monte Carlo investigations of few-electron systems.* APS March Mtg. 2010, Portland, OR, Mar. 15–19, 2010. LLNL-PRES-427939.

Whitley, H. D., J. L. DuBois, and K. B. Whaley, 2011. "Theoretical analysis of the anomalous spectral splitting of tetracene in ^{4}He droplets." *J. Phys. Chem. A* **115**(25), 7220. LLNL-JRNL-464446.

Feasibility of a Hybrid Rubidium Resonance and Exciplex Pump Laser

Raymond Beach (10-FS-002)

Abstract

We propose to examine a new concept for relaxing the tight wavelength tolerance on diode arrays used for excitation of alkali resonance lasers. This involves the use of a recently demonstrated excitation pathway in which the photo-association of an alkali metal atom and a polarizable buffer gas atom results in broad absorption-wing features on certain alkali spectral lines. These absorption features can easily accommodate the line widths of conventional laser diode arrays. The fundamental issue that must be addressed is pump irradiance required for efficient operation. We therefore propose to generate data and a companion laser model that permits us to reliably project laser system performance over a broad range of parameters.

We intend to evaluate the feasibility of a hybrid concept that uses the LLNL-developed hydrocarbon-free rubidium resonance laser scheme with a rubidium and rare-gas exciplex (excited-state complex) laser that can accommodate conventional diode arrays for pump excitation. This is a high-risk, but if proved feasible, potentially

very high-payoff investigation that could alter the landscape of high-average-power diode-pumped lasers envisioned for a broad range of defense activities, and already used in commercial material processing applications. This activity will contribute to maintaining LLNL's technical vitality and world dominance in the field of advanced diode-pumped laser development.

Mission Relevance

Our research supports the Laboratory's missions in advanced lasers and applications as well as national security. It is directly relevant to laser-based defense needs in addition to addressing future mission needs within the defense complex.

FY11 Accomplishments and Results

In FY11 we developed a new model of exciplex-pumped lasers and completed supporting experiments that illustrate its use. Specifically, we (1) performed a side-by-side comparison of a five-level rubidium–helium–xenon exciplex-pumped alkali laser, a four-level rubidium–xenon exciplex-pumped alkali laser, and a conventional rubidium–helium diode-pumped alkali laser system; (2) developed an alternative methodology for modeling far-detuned excitation in collision-broadened gaseous media; and (3) applied the methodology to specific laser systems using experimentally measured absorption line shapes. Using the same methodology for both types of lasers enables a clear and straightforward comparison between the two systems and facilitates easy and quick comparative studies. Based on our experimental line measurements and model development, we have determined that the exciplex-based approach will be of little practical value in the near term because of the irradiance limitations of laser optics today. However, if the limits of optics could be extended to megawatt per square centimeter levels, the approach might then be a viable path to efficient, high-power laser sources.

Coherence-Preserving X-Ray Adaptive Optics

Michael Pivovaroff (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 (LCLS) and the National Synchrotron Light Source II at Brookhaven National Laboratory. This new class of new 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 LCLS 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 LCLS, 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, the 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.

FY11 Accomplishments and Results

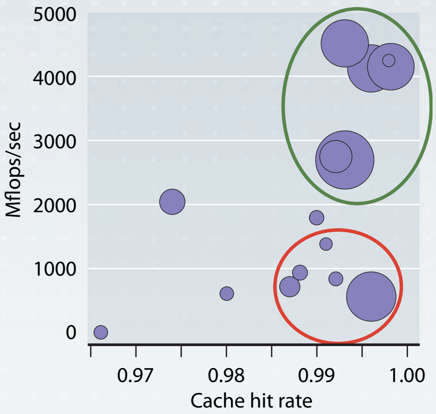
In FY11 we developed a number of simulation and modeling tools for predicting how x-ray optics can influence the wave front and intensity of an x-ray beam as it propagates through a beam line, from source to target. Specifically, we (1) wrote new codes, built interfaces to existing software packages, and created a benchmark of our capability by modeling well-defined problems and comparing our results to those obtained using independent tools; (2) carefully considered a number of options for wave-front sensing and selected a Hartmann-based approach for detailed study; and (3) initiated collaborations with LCLS and Brookhaven National Laboratory and partnerships with commercial companies to explore manufacturing options for mirror actuation.

Proposed Work for FY12

For FY12 we will (1) choose one or two approaches for mirror actuation and partner with a company or university group to construct a working prototype x-ray mirror, (2) complete our investigation of possible concepts for wave-front sensing and select at least one approach for laboratory testing, (3) finish developing our modeling and simulation capability, and (4) finish fabricating prototype subsystems and begin integrating a complete system that will be ready to test with an appropriate light source in FY13.

Publications

McCarville, T. J., M. J. Pivovaroff, and R. Soufli, 2011. *LCLS x-ray mirror measurements using a large-aperture visible light interferometer.* 4th Workshop on Adaptive and Active X-ray and XUV Optics, Oxfordshire, UK, Apr. 4–5, 2011. LLNL-ABS-510933.



Metric analysis for exascale architectures is inherently multidimensional. In the figure, each bubble represents a routine within a Lagrangian hydrodynamics simulation, sized according to runtime. The traditional thinking is that a high cache hit rate indicates high performance, but this is not always true. In the above example, dependency analysis shows that inefficiency (red circle)—in contrast to the high-performance cluster (green circle)—results from a poor choice of data structure.

Advanced Algorithm Technology for Exascale Multi-Physics Simulation

Charles Still (11-ERD-017)

Abstract

We propose to develop computational algorithms that will enable multi-physics simulation codes to efficiently utilize exascale computers. As early as 2018, NNSA could site an exascale computer at LLNL. With a thousandfold performance increase and only a tenfold electricity requirement, the system architecture is a dramatic departure from the hardware of today, and codes will have to change drastically to utilize it. We will characterize which multi-physics algorithms extend to exascale hardware, which do not, and how to determine this in general. A successful project will result in metrics and characterization techniques for others to use and in exascale-tuned hydrodynamics and diffusion reference codes as proof of principle on exascale surrogate hardware.

Current multi-physics 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 node, leading to restricted memory and message-passing bandwidth per node. To take advantage of these systems, algorithms must exhibit significant intranode concurrency while limiting data motion. Using a coupled hydrodynamics–diffusion reference code as an example, we will develop computational algorithms that will enable multi-physics simulation codes to effectively and efficiently utilize exascale machines.

Mission Relevance

A pressing need exists 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 for transitioning multi-physics codes to exascale systems, along with a coupled arbitrary Lagrangian–Eulerian (ALE) hydrodynamics–diffusion reference code. This work furthers the Laboratory's stockpile stewardship science mission and will bolster the foundation in high-performance computing and simulation.

FY11 Accomplishments and Results

In FY11 we (1) verified the suitability of ALE3D as the donor multi-physics code using a coupled ALE hydrodynamics–diffusion code; (2) began defining an initial suite of metrics for data motion and parallelism and characterization techniques for exascale performance, using existing surrogate hardware and performance tools; (3) extracted the relevant components from ALE3D into a reference code, named xALE; (4) began to identify problem areas for exascale performance; (5) began exploring data-streaming and compression techniques applicable to Lagrangian hydrodynamics;

and (6) completed an initial implementation of the Lagrangian hydrodynamics kernel for graphical processing units. Throughout this process of exploring transformation strategies in the context of a mini-application, we used metrics to determine the degree of success, and then incorporated lessons learned into the full code xALE.

Proposed Work for FY12

By the end of FY12 we will complete a key decision as to whether the optimized kernels can be combined into an efficient code on the Sequoia supercomputer at Livermore. Specifically, we propose to (1) continue to identify problem areas for exascale performance, (2) extract those problem areas into kernels, (3) identify data structures and transformations amenable to exascale architectures, (4) identify massive multithreading and computational transforms, and (5) optimize the kernels using identified data structures and computational transforms.

Publications

Brunner, T. A., 2012. *A new multigroup thermal radiation diffusion miniapplication*. SIAM Conf. Parallel Processing for Scientific Computing (PP12), Savannah, GA, Feb. 15–17, 2012. LLNL-ABS-494432.

Still, C. H., 2011. *Challenges beyond the software pipeline.* Salishan Conf. High-Speed Computing, Gleneden Beach, OR, Apr. 25–28, 2011. LLNL-PRES-481274.

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 to predict 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 (bcc) phases that all actinide elements adopt prior to melt.

Our results will predict and explain the occurrence of the bcc 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

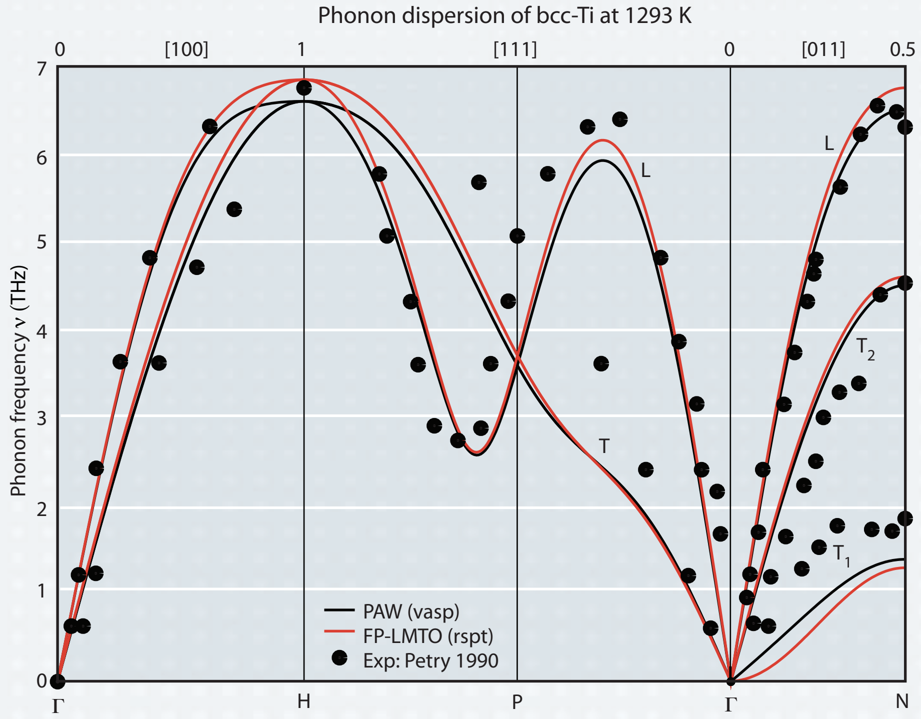
the phase diagrams for actinides—and other metals—can be computed from first principles with the reliability that already exists for the low-temperature region. This is important for fundamental scientific reasons and is also an extremely important technical component in describing equation-of-state properties that are now very difficult to assess, either with 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.

FY11 Accomplishments and Results

In FY11 we (1) successfully implemented and tested self-consistent phonon codes for the simple metal titanium; (2) defined and tested a total-energy approach to approximate forces in a scheme to calculate temperature-dependent lattice dynamics using titanium as a model case with a 27-atom super cell—demonstrating that our new model system can readily calculate forces through any robust plane-wave code—and compared the results to force components obtained from the derivative with respect to displacement of the total; (3) compared the obtained components



Phonon dispersion for body-centered-cubic titanium (bcc Ti) calculated with traditional techniques (forces calculated with the Projector Augmented Wave code PAW) and with a new, all-electron approach (total energies determined with our new Full-Potential Linear Muffin-Tin Orbital code FP-LMTO).

with that of an all-electron code to be used for actinide systems; and (4) applied our methodology with an all-electron theory to the prototype actinide metal uranium. We learned through our FY11 computations that the relativistic spin–orbit interaction is important for the phonon stability of bcc uranium, although our approach allows us to accurately calculate total energies while accounting for spin–orbit coupling.

Proposed Work for FY12

In FY12 we will (1) continue to implement and optimize our self-consistent phonon codes; (2) develop an "exact muffin-tin orbitals" method capable of treating actinide alloys in random phases, which will enable us to work on predicting the high-temperature and high-pressure phase diagrams of actinide fuels; and (3) calculate the temperature-dependent phonons for select f-electron metals (i.e., rare earths and actinides) in their bcc phase to address stabilization of high-temperature phases in f-electron materials.

Dynamics of Ultrafast Heated Matter

Siegfried Glenzer (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 material 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 temperature and density of free electrons, which in turn determine the width and absolute intensity of the scattering spectrum. We will also

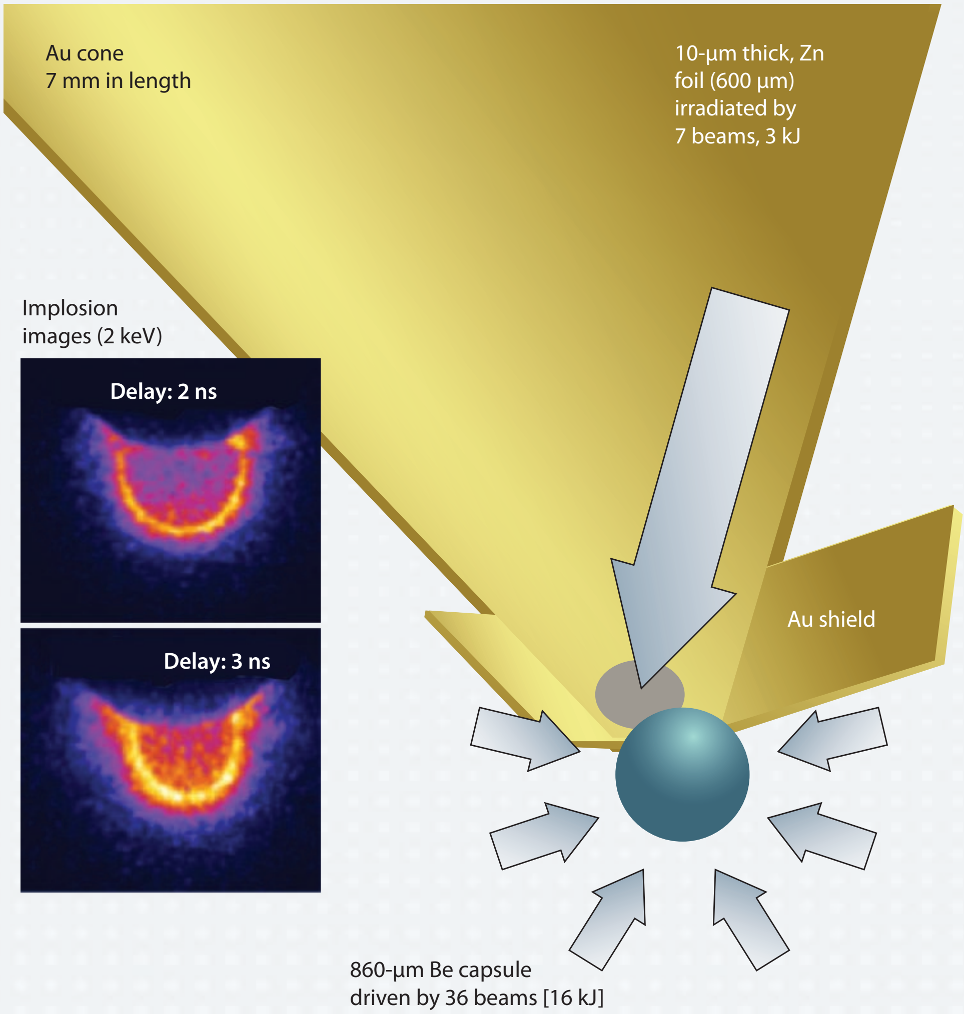
determine the collective (forward) scattering regime of the plasmon shift, which determines collective electron Langmuir oscillations and provides insight into the quantum correlations of dense matter.

Mission Relevance

Ultrafast heating of matter is an important phenomenon for understanding physical processes relevant to LLNL astrophysics and high-energy-density science programs. This newly proposed application of x-ray scattering will provide definite data important to several Laboratory missions. For instance, equation of state and collision processes are important for successful modeling of experiments on the NIF, in support of Livermore's missions in stockpile stewardship, climate and energy challenges, and laser ignition fusion energy.

FY11 Accomplishments and Results

In FY11 we demonstrated new capabilities to achieve accurate measurements of dense-matter properties. Specifically, we (1) demonstrated the capability to perform



Our LDRD project on the dynamics of ultrafast heated matter enables us to measure the properties of matter assembled in spheres compressed with x-ray Thomson scattering.

precision adiabatic measurements in capsule implosions on the OMEGA laser at the University of Rochester and developed a plan for our first demonstration on NIF; (2) demonstrated the capability to measure temperatures with x-ray scattering on cryogenic hydrogen, boron, and boron nitrate with laser-produced Lyman-alpha x-ray sources on the Titan and Janus lasers at LLNL; (3) demonstrated x-ray scattering on carbon using a free-electron laser x-ray source at the Linac Coherent Light Source at Stanford; and (4) developed techniques for x-ray scattering on cryogenic hydrogen targets with a free-electron laser.

Proposed Work for FY12

In FY12 we will (1) perform precision measurements of the ultrafast melting process involved in x-ray scattering on free-electron lasers, (2) assess the material properties of shocked deuterium and beryllium and determine their conductivity and equation of state, (3) incorporate the influence of detailed balance on proton-heated matter into a temperature diagnostic that is independent of the velocity distribution function, and (4) develop a new experiment on NIF to measure properties of highly compressed matter.

Publications

Davis, P. F., 2011. *X-ray Thomson scattering measurements from shock-compressed deuterium*. 17th Intl. Conf. Atomic Processes in Plasmas, Belfast, UK, July 19–22, 2011. LLNL-CONF-491044.

Davis, P. F., et al., 2012. "An apparatus for the characterization of warm, dense deuterium with inelastic x-ray scattering." *J. Instrum.* **7**, P02004. LLNL-JRNL-496611.

Fortmann, C., 2011. *Adiabatic index in shock-compressed Be.* LLNL-JRNL-507611.

Fortmann, C., et al., 2011. "Measurement of the heat capacity ratio in shock compressed matter." *Bull. Am. Phys. Soc.* **56**(16), BAPS.2011.DPP.NO8.10. LLNL-JRNL-489961.

Hau-Riege, S., 2011. *Ultrafast dynamics in isochorically-heated nanocrystalline solids.* LLNL-JRNL-501252-DRAFT.

Kritcher, A., 2011. *Measurements of the properties of highly compressed degenerate matter using x ray Thomson scattering.* 17th Intl. Conf. Atomic Processes in Plasmas, Belfast, UK, July 19–22, 2011. LLNL-ABS-491018.

Kritcher, A., et al., 2011. "Development of x-ray Thomson scattering for implosion target characterization." *High Energ. Density Phys.* **7**(4), 271. LLNL-JRNL-471879.

Kritcher, A., et al., 2011. "In-flight measurements of capsule shell adiabats in laser-driven implosions." *Phys. Rev. Lett.* **107**(1), 01500. LLNL-JRNL-473096.

Le Pape, S., 2011. *Structure of compressed liquid boron*. LLNL-JRNL-491498.

Ross, J. S., et al., 2011. "Ultraviolet Thomson scattering measurements of the electron feature with an energetic 263 nm probe." *J. Instrum.* **6**, P08004. LLNL-JRNL-489959.

Astrophysical Collision-Less 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. Collision-less 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 collision-less 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 (NIF)—to observe the creation of collision-less shocks and generation of magnetic fields.

If successful, this project will generate a wealth of knowledge about the physics of collision-less shock dynamics. Our observations and experimental results will be important for understanding plasma processes occurring in astrophysical collision-less shocks, such as the generation of magnetic fields and the 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 later NIF experiments. This project will also attract the world's foremost experts in this field to participate in those NIF 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 for the Laboratory's core mission in stockpile stewardship. In addition, the project will leverage investments in NIF to further missions in energy security.

FY11 Accomplishments and Results

In FY11 we performed high-power laser experiments at the OMEGA Laser Facility to demonstrate our concept. Specifically, we (1) measured the characteristics of streaming plasma using Thompson-scattering diagnostics and validated that conditions for collision-less shock formation were achieved; (2) demonstrated weak Weibel-mediated current filament and magnetic field generation most likely began to occur in our experiment, which was the first-ever accomplishment of this type of quantitative measurement of a plasma flow, although further verification of our conclusion is still required; and (3) further characterized the laser-created flow and developed magnetic-field diagnostics using proton deflectometry and proton radiography to directly measure magnetic fields present before and during the collision-less interaction.

Proposed Work for FY12

In FY12 we will (1) continue to work on identifying collision-less shock signatures by probing the plasmas from different flow directions while attempting to create higher flow velocities and higher ion densities; (2) refine and enhance our diagnostic probes to measure magnetic fields and distinguish their generation mechanisms, such as fountain fields in non-interacting flow and self-generated fields in interpenetrating interacting flows; (3) use the OMEGA Extended Performance laser's optical diagnostics, such as the optical interferometer, to capture images of collision-less shock formation; and (4) compare our results with radiation hydrodynamics and particle-in-cell simulations.

Publications

Park, H. S., et al., 2012. "Studying astrophysical collisionless shocks with counterstreaming plasmas from high power lasers." *High Energ. Density Phys.* **8**(1), 38. LLNL-JRNL-510851.

Ryutov, D. D., et al., 2011. "Collisional current drive in two interpenetrating plasma jets." *Phys. Plasma.* **18**(10), 104504. LLNL-JRNL-491452.

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 induced by mitigation techniques. We will extend the capability of our BOUT++ plasma turbulence code to include the full ELM cycle. We propose to conduct experiments on the DIII-D reactor at General Atomics to understand ELM control by three-dimensional non-axisymmetric perturbation fields and on the National Spherical Torus Experiment (NSTX) at Princeton University to understand ELM control by novel two-dimensional divertor geometries, then use the 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 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.

FY11 Accomplishments and Results

In FY11 we (1) extended the BOUT++ code to magnetic X-point geometry and plasma shear flow; (2) calculated the linear instability of X-point DIII-D and NSTX plasmas, including $E \times B$ shear and ion diamagnetic flows, and found unstable edge modes for finite flows; (3) studied a drift-ordered two-fluid model of reconnection driven by resonant magnetic perturbations and found that diffusion smooths $E \times B$ and ion drift resonances and that reconnection occurs primarily at the electron resonance; (4) analyzed ELM size scaling with hyper-resistive-layer width and found that it increases with hyper-resistivity; (5) showed ELM size dependence on the frequency of collisions at both low and high values; and (6) acquired narrow-bandpass filters and temperature-controlled ovens to upgrade the DIII-D edge motional Stark effect diagnostic for pedestal current measurements, as well as a high-gain, gated image intensifier to be installed on the DIII-D periscope for ELM imaging.

Proposed Work for FY12

We will (1) incorporate new motional Stark effect edge data into the ELM model in BOUT++; (2) extend comparisons of the ELM model and experimental data to a lower frequency of particle collisions; (3) upgrade our resonant magnetic perturbation penetration models in BOUT++; (4) verify linear ideal and nonlinear island solutions and begin to assess the impact on turbulence; (5) compare interpretive and predictive simulations with data for transport by resonant magnetic perturbation at a high frequency of collisions; (6) measure simultaneous changes in heat and particle fluxes caused by rotating magnetic perturbations using injected impurities, if necessary, to enhance imaging resolution; and (7) use new NSTX Thomson-scattering channels to obtain high-spatial-resolution, high-electron-density, and high-temperature data for the snowflake divertor configuration.

Publications

Joseph, I., 2011. *Edge-localized mode control and transport generated by externally applied magnetic perturbations*. LLNL-JRNL-512873.

Joseph, I., 2011. *ELM control using external magnetic perturbations. 13th Intl.* Workshop on Plasma Edge Theory, South Lake Tahoe, CA, Sept. 19–21, 2011. LLNL-PRES-499531.

Joseph, I., 2011. *Theory of the generation of non-axisymmetric scrape-off-layer perturbations for controlling tokamak edge plasma profiles and stability*. 53rd Mtg. APS Division of Plasma Physics, Salt Lake City, UT, Nov. 14–18, 2011. LLNL-PRES-512836.

Joseph, I., and X. Xu, 2011. *The effect of anomalous electron viscosity on edge transport*. TTF2011 Joint EU–US Transport Task Force Workshop, San Diego, CA, Apr. 6–9, 2011. LLNL-ABS-471500.

Joseph, I., et al., 2011. *Experimental overview of ELM control using external magnetic perturbations*. 13th Intl. Workshop on Plasma Edge Theory, South Lake Tahoe, CA, Sept. 19–21, 2011. LLNL-PRES-499357.

Waelbroeck, F. L., et al., 2011. *Plasma response to resonant magnetic perturbations*. LLNL-JRNL-507991.

Xu, X. Q., et al., 2011. "Nonlinear ELM simulations based on a non-ideal peeling-ballooning model using the BOUT++ code." *Nucl. Fusion* **51**(10), 103040. LLNL-JRNL-464898.

Xu, X. Q., et al., 2010. *Nonlinear ELM simulations based on peeling-ballooning modes using the BOUT/BOUT++ code*. 23rd IAEA Fusion Energy Conf., Daejeon, South Korea, Oct. 11–16, 2010. LLNL-CONF-457792.

Investigation of Fast Z-Pinches for Scalable, Large-Current, High-Gradient Particle Accelerators

Vincent Tang (11-ERD-063)

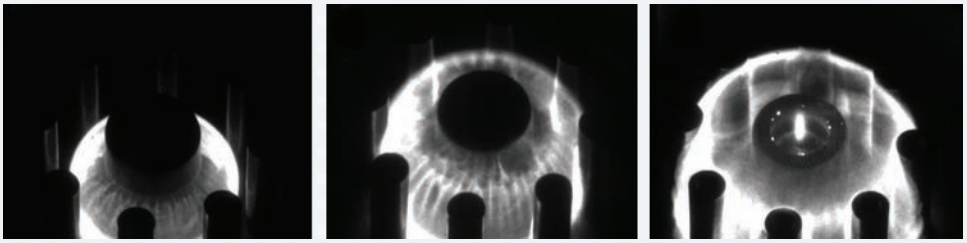
Abstract

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 created by using 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 scalable 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 the acceleration gradients in DPF Z-pinch plasmas and to 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 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 missions in national and homeland security—specifically nonproliferation and accelerator science—by providing the technological and



A typical Z-pinch accomplished with a dense plasma focus device. As shown on the left, a coaxial plasma sheath is formed by flashover breakdown when a capacitor bank is switched to the central anode surrounded by eight cathode rods. The plasma sheath is accelerated to approximately 100 km/s in the coaxial gun geometry (middle). A dense Z-pinch plasma forms on top of the anode once the sheath reaches the end of the gun and collides onto itself (right).

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 the physics of plasmas important to many LLNL programs.

FY11 Accomplishments and Results

In FY11 we (1) designed, fabricated, and assembled a 4-kJ DPF test stand, along with base diagnostics; (2) initiated DPF operations with successful formation and diagnosis of Z-pinch plasmas; (3) transferred and refurbished the 4-MeV radio-frequency quadrupole accelerator for the probe beam—a significantly improved beam that will replace the originally planned proton probe beam; (4) completed simulations for our transport beam line and associated diagnostics; (5) completed the first fully kinetic particle-in-cell simulations of DPF using the LSP (Large Scale Plasma) code; (6) compared our LSP simulations with fluid and hybrid models and parameter scans and began comparing the simulation results with experimental data; and (7) hired a postdoctoral researcher for our modeling and analysis efforts.

Proposed Work for FY12

In FY12 we will achieve the first-ever use of an ion probe beam to explore acceleration mechanisms in a high-energy-density Z-pinch plasma. Specifically, we will (1) finish characterizing the DPF Z-pinch plasma and beam output for both deuterium and argon gas fills, (2) conduct the first set of experiments using our 4-MeV radio-frequency quadrupole ion probe beam, (3) perform simulations of the DPF plasma and beam output through our fully kinetic particle-in-cell and hybrid models, and (4) compare our new experimental data with the leading models through both direct and synthetic diagnostic approaches.

Publications

Schmidt, A., et al., 2011. *PIC simulations of dense plasma focus Z-pinch*. 53rd Ann. Mtg. APS Division of Plasma Physics, Salt Lake City, UT, Nov. 14–18, 2011. LLNL-ABS-490260.

Tang, V., et al., 2011. *The LLNL high-gradient Z-pinch ion probe experiment*. 53rd Ann. Mtg. APS Division of Plasma Physics, Salt Lake City, UT, Nov. 14–18, 2011. LLNL-ABS-490433.

Tang, V., et al., 2010. *The LLNL Z-pinch ion probe experiment*. 37th Intl. IEEE. Conf. Plasma Science, Norfolk, VA, June 20–24, 2010. LLNL-ABS-471576.

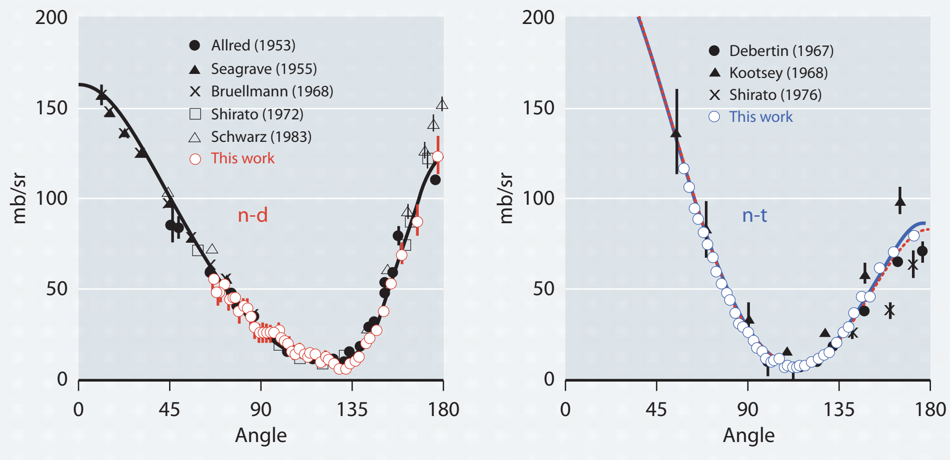
Nuclear Plasma Physics

Dennis McNabb (11-ERD-069)

Abstract

To understand the evolution of the universe and the fundamental underpinnings of high-energy-density plasmas, as well as fully realize 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 to help improve the 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–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 on short-lived isotopes are important to a detailed understand of the s-process (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.



Data and calculations for neutrons elastically scattering from deuterium (left) and tritium (right), with data from the OMEGA Laser Facility shown in red (deuterium) and blue (tritium). The deuterium data are compared with calculations from Epelbaum, et al., *Phys. Rev. C* **66**, 064001 (2002). The tritium data are compared with calculations from Hale, et al., *Phys. Rev. C* **42**, 438 (1990) and with new ab initio calculations performed as part of our project. These figures were first published in *Physical Review Letters* in 2011.

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 of 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.

FY11 Accomplishments and Results

In FY11, to investigate thermonuclear fusion in inertial-confinement fusion plasmas, we (1) established a collaboration with the Laboratory for Laser Energetics at the University of Rochester, the Massachusetts Institute of Technology, and Indiana University; (2) made precise measurements of a fundamental nuclear process—the elastic scattering of neutrons off heavy forms of hydrogen, deuterium, and tritium—a first for a high-energy-density laser facility; (3) performed initial measurements of the particle spectra of $T(t,2n)^4He$ (the fusion of two tritium nuclei into an alpha particle and two neutrons) and $^3He(^3He,2p)^4He$ (an important fusion reaction relevant to hydrogen burning in stars), with our initial work demonstrating that thermonuclear fusion in inertial-confinement fusion plasmas is different from measurements performed with conventional accelerators; (4) performed initial nuclear theory calculations for the thermonuclear reactions that we are studying experimentally; and (5) performed calculations to design laser blowoff plasma experiments to achieve an integrated understanding of nuclear excited states in burning plasmas and hopefully achieve the first-ever observation of plasma-induced nuclear excitations.

Proposed Work for FY12

In FY12 we will (1) perform detailed measurements and calculations of spectral shapes to better understand continuum states in the ^4He,2n and ^4He,2p systems; (2) improve our theoretical models of the nuclear–plasma interactions that lead to nuclear excited-state populations, specifically nonlocal thermodynamic equilibrium effects; (3) use our improved models to plan and develop an experimental approach to validate the theory of plasma–nuclear coupling; and (4) perform initial measurements on the capture gamma-ray spectra needed to measure low-energy neutron spectra with the gamma-reaction-history diagnostic device at NIF.

Publications

Casey, D. T., et al., 2012. "Evidence for stratification of deuterium–tritium fuel in inertial confinement fusion implosions." *Phys. Rev. Lett.* **108**(7), 075002. LLNL-JRNL-515951.

Frenje, J. A., et al., 2011. "Measurements of the differential cross sections for the elastic n-^{3}H and n-^{2}H scattering at 14.1 MeV using an inertial confinement fusion facility." *Phys. Rev. Lett.* **107**, 122502. LLNL-JRNL-489019.

Kritcher, A. L., et al., 2011. *High-energy-density and nuclear physics experiments at NIF.* LLNL-PRES-490287.

McNabb, D. P., 2011. *Measurement of thermonuclear reactions with charged particles.* LLNL-PRES-480632.

McNabb, D. P., et al., 2011. *Measuring $^3He(^3He,2p)^4He$ and $^3H(^3H,2n)^4He$ reactions near 10 keV at inertial confinement facilities.* LLNL-PRES-466074.

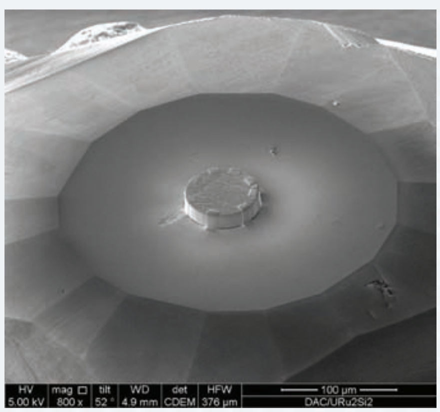
McNabb, D. P., et al., 2011. *Nuclear physics possibilities at NIF.* LLNL-PRES-466071.

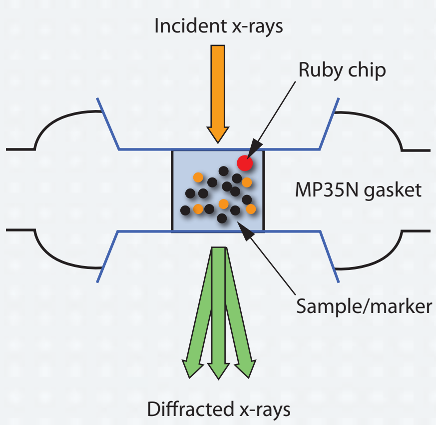
Structure–Property Relationships in Ferropnictide Superconductors at Extreme Pressures

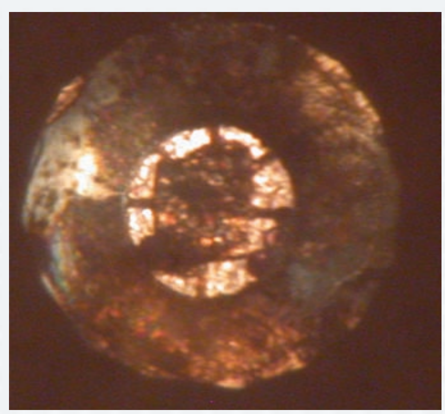
Jason Jeffries (11-LW-003)

Abstract

We propose to illuminate, for the first time ever, the integral relationships between structure and superconductivity in the newly discovered ferropnictide







We are using designer diamonds and synchrotron x-ray techniques to gain unprecedented access to structure–property relationships over a wide range of phase space. A scanning electron microscopy image of a sample attached to a designer diamond anvil using a focused ion beam (top left). A single crystal seated against embedded microprobes within the high-pressure sample space of a designer diamond anvil cell is shown on the right. A schematic illustration of the configuration of high-pressure synchrotron x-ray diffraction is seen at bottom left.

superconductors—iron-based superconductors comprised primarily of a material containing one of the pnictide elements such as arsenic. Elucidating their structure–property relationships will profoundly alter the understanding of superconductivity, providing the first platform from which theoretical models can be expanded from their descriptive formulations to more-predictive capabilities. These new predictive theories will be critical to the discovery of next-generation superconducting materials for the electrical power grid. We propose to couple high-pressure capabilities unique to LLNL with state-of-the-art methods for growing the highest quality samples possible to experimentally determine structural properties conducive to superconductivity.

We expect to grow the highest quality single crystals of ferropnictide compounds to date and to map the pressure conditions that maximize the onset of superconductivity in these materials. Using x-ray diffraction, we will reveal a never-before-seen correlation between structural properties and superconductivity. Furthermore, we will exploit the newly discovered structure–property relationships as feedback for maximizing superconductivity in ferropnictides at ambient pressure. Close collaborations with theorists will provide an avenue for interpreting fundamental physical mechanisms linking the structural and electrical properties in this class of materials. When successfully implemented, superconductors in the electrical grid could save nearly a trillion kilowatt-hours of energy per year and provide energy storage solutions for variable-availability renewable energy sources.

Mission Relevance

With this research, we aim to determine the structure–property relationships of superconductors with the future goal of engineering materials suitable for use in the electrical power grid, in support of the Laboratory's core mission to enhance national energy security. In accomplishing this proposed work, we plan to support a graduate student, thus training a future scientist in mission-critical techniques that encompass many Laboratory thrust areas.

FY11 Accomplishments and Results

In FY11 we successfully synthesized new compounds and investigated their pressure-dependent structural and electrical properties. Specifically, we (1) successfully synthesized and investigated a binary iron–arsenic compound under pressure using high-pressure electrical transport and synchrotron-based x-ray diffraction, revealing that although the crystal structure did not change, the lattice compressed under pressure, destroying magnetism presumably through a change in electron–phonon coupling combined with a possible stiffening of the phonon modes; (2) completed a high-pressure structural characterization of a compound in the $(Sr,Ca)Fe_2As_2$ series and found a structural volume-collapse transition that is driven by arsenic–arsenic bonding and that seems to destroy superconductivity near 4 GPa; and (3) performed additional experiments on bismuth telluride and bismuth selenide, discovering new superconducting phases in both compounds after structural phase transformations that lead to metallization.

Proposed Work for FY12

In FY12 we will focus on further measurements of electrical transport and x-ray diffraction to investigate another member of the (Sr,Ca)Fe_2As_2 system, with a goal of illuminating the relationship, if any, between the structural volume collapse and the destruction of superconductivity. We will also investigate a related binary compound, platinum arsenide, which is a major in-growth of samples of platinum-doped $CaFe_2As_2$ that leads to confusion about the intrinsic properties of the doped system.

Publications

Hamlin, J. J., et al., 2011. "Low-temperature electrical resistivity of praseodymium at pressures up to 120 GPa." *Phys. Rev. B* **84**, 033101. LLNL-JRNL-508476.

Jeffries, J. R., et al., 2011. *Distinct superconducting states in the pressure-induced metallic structures of the nominal semimetal* Bi_4Te_3. Intl. Conf. Strongly Correlated Electron Systems (SCES 2011—Commemorating 100 Years of Superconductivity), Cambridge, UK, Aug. 29–Sept. 3, 2011. LLNL-POST-492951.

Jeffries, J. R., et al., 2011. "Distinct superconducting states in the pressure-induced metallic structures of the nominal semimetal Bi_4Te_3." *Phys. Rev. B* **84**(9), 092505. LLNL-JRNL-508452.

Jeffries, J. R., et al., 2011. "Interplay between magnetism, structure, and strong electron–phonon coupling in binary FeAs under pressure." *Phys. Rev. B* **83**(13), 134520. LLNL-JRNL-463133.

Jeffries, J. R., et al., 2011. *The behavior of semi-metal* Bi_4Te_3 *under pressure.* American Physical Society March Mtg. 2011, Dallas, TX, Mar. 21–25, 2011. LLNL-PRES-474259.

Demonstrating Precision Delayed-Neutron Spectroscopy Using Trapped Radioactive Ions

Nicholas Scielzo (11-FS-014)

Abstract

Neutrons emitted following the beta decay of fission fragments can provide information for many fields of basic and applied science such as stockpile stewardship, nuclear energy, and nuclear structure and astrophysics. The existing data, however, is quite poor. We propose to perform, for the first time, delayed-neutron spectroscopy using trapped radioactive ions—the daughter nucleus emerges unperturbed from the trap, and the time of flight to an ion detector can be measured. The energy of the emitted neutron can be precisely reconstructed from the large nuclear recoil imparted

by the neutron using conservation of energy and momentum. This approach will enable a revolutionary way to study delayed-neutron decay with high efficiency, few backgrounds, and excellent energy resolution.

We expect to demonstrate, by performing measurements using trapped radioactive ions, that delayed-neutron spectroscopy can be revolutionized. The technique will be successfully demonstrated by measuring decay branching ratios and energy spectra for the beta decay of iodine-137 (a very well-characterized fission product) with high precision, efficiency, and signal-to-noise ratio. Ultimately, a unique program of study can be initiated using the techniques developed in this proposal at the new Californium Rare Isotope Breeder Upgrade facility at Argonne National Laboratory, which for the first time will produce high-intensity, high-quality fission-fragment beams of all elements from zinc with an atomic number of 30 to dysprosium with an atomic number of 66.

Mission Relevance

This work represents innovative fundamental research at the frontier of nuclear science that will revolutionize delayed-neutron spectroscopy and is well aligned with the Laboratory's five-year strategic plan. Delayed-neutron data provide a unique opportunity to significantly reduce the uncertainties of neutron-capture reactions on fission-fragments of interest for stockpile stewardship, and will play a major role in understanding high-energy-density environments and determining the origin of elements in the cosmos. In addition, delayed-neutron data is needed for next-generation nuclear reactors that have the opportunity to deliver energy solutions while reducing greenhouse-gas emissions, in support of LLNL's commitment to energy and environmental security.

FY11 Accomplishments and Results

In FY11 we (1) completed the design for a new detector array for the existing ion trap at Argonne National Laboratory—simulations developed for this project show that these new detectors will increase the detection efficiency of the technique by an order of magnitude and will improve resolution of the neutron energy measurement; (2) analyzed, using simulations, data collected during an initial proof-of-principle; (3) designed and fabricated micro-channel plate detectors; and (4) began fabricating beta detectors.

Proposed Work for FY12

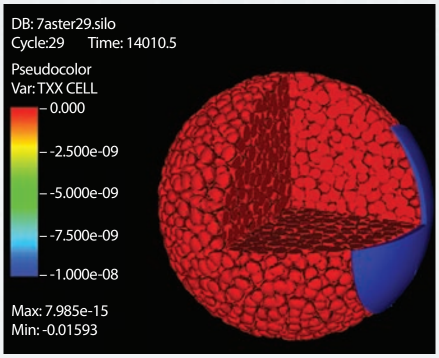
In FY12 we will (1) complete instrumentation of an existing ion trap with an array of micro-channel plate detectors and plastic scintillator detectors, (2) load the trap with iodine-137 from an offline californium-252 fission-fragment source that is available for this effort, and (3) perform data collection for statistics and thoroughly investigate potential systematic effects, with analysis of the data being conducted at LLNL. We expect the results of this research will be published in high-visibility scientific journals.

Publications

Yee, R., et al., 2011. *Beta-delayed neutron spectroscopy using trapped radioactive ions.* LLNL-JRNL-511460-DRAFT.

Asteroid Deflection

Paul Miller (11-FS-015)



Initial Lagrangian setup of an asteroid comprised of 6,250 rock masses (each with 256 elements). The blue cap on the right-hand side represents all of the energy imparted to the asteroid from a nuclear explosive, and would push the agglomerated mass out of its Earth-bound trajectory.

Abstract

Several major national reports have identified nuclear energy coupling as an important strategy for deflecting near-Earth objects. In fact, the U.S. Nuclear Regulatory Commission has reported to Congress that nuclear explosives are the only current technology available to defend Earth against large asteroids, or when the time before a collision is short. We will develop a matrix of parameter variations to investigate for such scenarios, including a range of threat compositions, sizes, dynamics, and times to impact. We will then optimize parameters such as height of burst and yield. We will collaborate with expert material scientists to determine material models for asteroids.

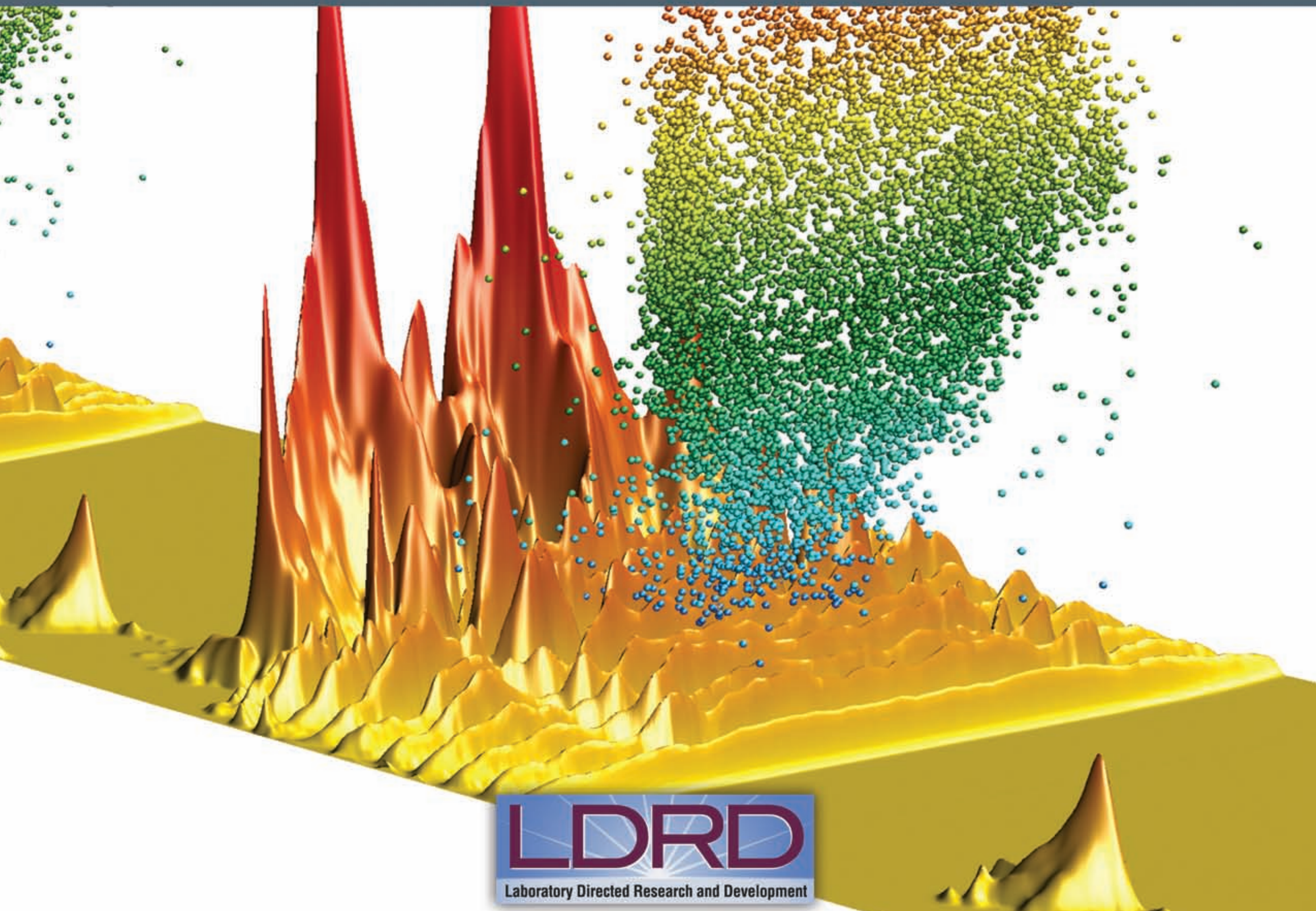
By the end of this feasibility study, we will have developed a matrix of important parameters for developing energy coupling for deflecting near-Earth objects. We will also forge collaborations with material experts in this area. Our work will determine the feasibility of further investigations and will help prepare LLNL and the NNSA complex for a role in future threat response.

Mission Relevance

This project sets the stage for further investigations into an important and challenging application area relevant to national security—leveraging LLNL nuclear design capabilities to investigate the potential use of the nation's stockpile to defend against near-Earth objects.

FY11 Accomplishments and Results

After a mid-year start, we (1) developed a matrix of parameter variations, (2) determined material-science considerations, (3) constructed a preliminary set of impact threat scenarios, and (4) conducted several modest simulations as a means of defining future large-scale simulations. An LLNL postdoctoral researcher also participated and was mentored in this approach. This successful feasibility study has laid the groundwork for future investigations, which are now expected to proceed.



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