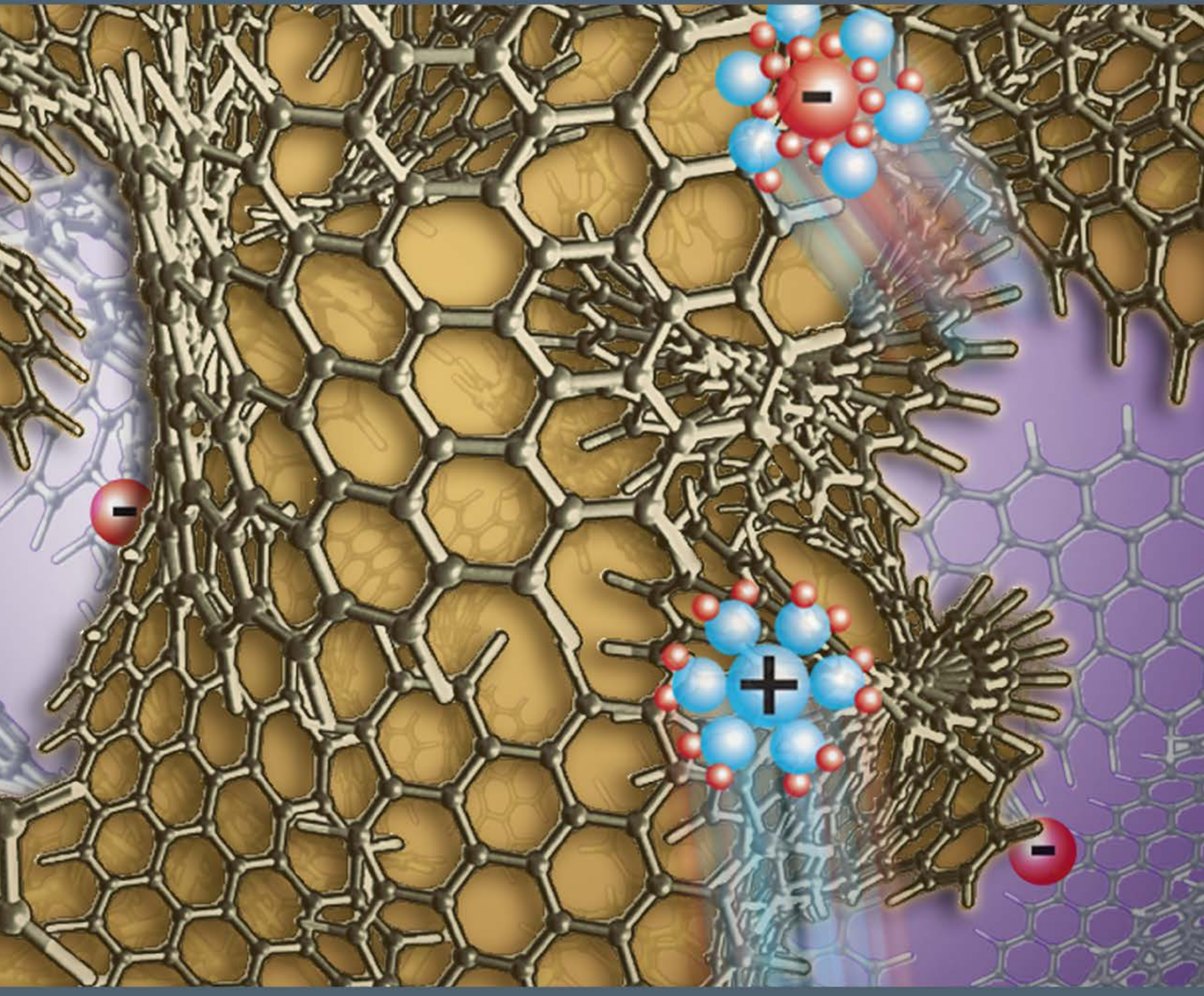
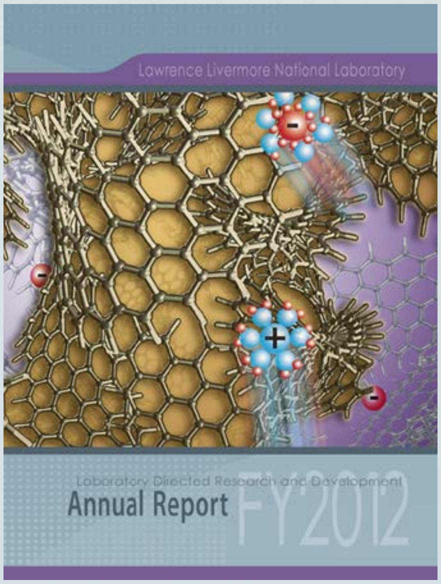
Lawrence Livermore National Laboratory



Laboratory Directed Research and Development

Annual Report FY2012



About the Cover

The future of sustainable energy strongly depends on scientific advances in materials for energy storage and conversion. The 2012 Laboratory Directed Research and Development project depicted on the cover, "Dynamically Tunable Nanometer-Scale Materials: From Atomic-Scale Processes to Macroscopic Properties" (12-ERD-035) is developing a fundamental understanding of electrical charge-transfer processes at material interfaces related to energy storage that is needed to develop the next generation of devices such as advanced batteries and fuel cells. Material interfaces are of particular importance, because all processes relevant to energy storage, whether physical or chemical, occur here. Principal investigator Juergen Biener is using a combination of experimental and theoretical tools in support of Lawrence Livermore Laboratory's energy and environmental security mission to develop technologies that will enable the nation to reduce its dependence on fossil fuels. The figure shows the three-dimensional structure of a new polymer-derived honeycomb material based on graphitic carbon (graphene) that consists of a network of single-layer nanometer-scale platelets. The material is mechanically robust and combines a graphene-like surface area with a porosity that allows macroscopic properties to be dynamically controlled through ion-induced electric fields between interfaces. This research was recently featured as the cover story in the scientific journal, *Advanced Materials*.

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Acknowledgments

The Laboratory Directed Research and Development Program extends its sincere appreciation to the principal investigators of fiscal year 2012 projects for providing the content of the annual report and to the publications team. The program also thanks the following members of the Laboratory Directed Research and Development Office for their many contributions to this publication: Barbara Jackson, administrator; Steve McNamara, computer specialist; Iris Lam, resource manager; and Kristen Croteau, business manager.

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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 $91.8 million for fiscal year 2012 supported 160 projects. These projects were selected through an extensive peer-review process to ensure the highest scientific quality and mission relevance.

The LDRD projects are consistent with the Laboratory's strategic 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 foundations 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 we have chosen to build or expand our influence
- Looking beyond the immediate challenges to future opportunities

The LDRD Program was conceived as a bold initiative, and scientific and technical risk 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

FY2012

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 *FY2012 Laboratory Directed Research and Development Annual Report*

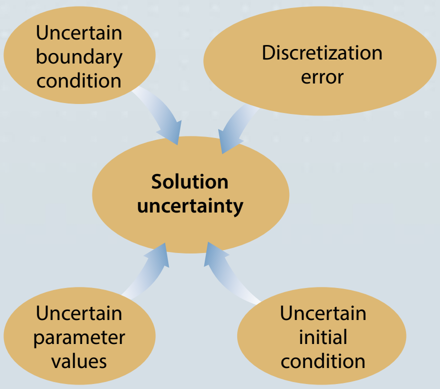
The LDRD annual report for fiscal year 2012 (FY12) provides a summary of LDRD-funded projects for the fiscal year and consists of two parts:

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 FY12, and a list of publications that resulted from the research.

Highlights of Accomplishments for the Fiscal Year

In FY12, 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.



Stockpile Stewardship

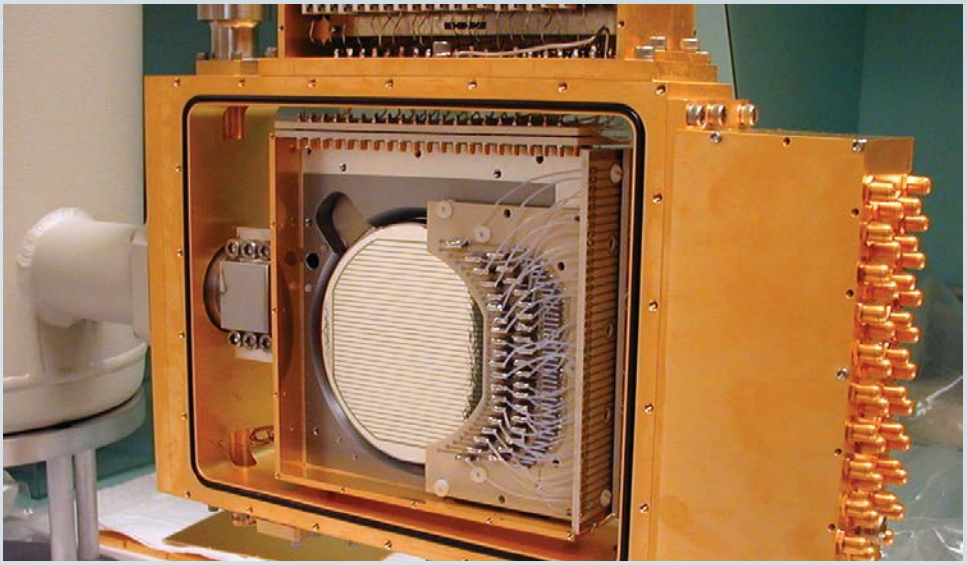
Uncertainty quantification is the science of determining uncertainties in the computational simulations of large and complex problems. Typically, there is no information that informs researchers of the uncertainty of predictions derived from their simulations and which of the many input uncertainties are important (e.g., a specific design or a physical location). Producing quantified and bounded uncertainties of simulations enables assessment of the reliability and accuracy of predictions. Furthermore, it enables scientists and decision makers to make informed choices on how to improve predictive capability (e.g., what types of data should be collected and to what accuracy or what experiments will be most informative). A just-concluded LDRD project created a focused, multidisciplinary scientific effort to investigate, develop, and apply uncertainty quantification science to high-impact scientific areas (10-SI-013). Researchers sought to develop

- Error and uncertainty propagation methods in multiple-physics and multiple-scale computer codes
- Novel methods for dimensional reduction of complex problems
- An advanced computational pipeline to enable complete uncertainty quantification workflow and analysis for ensemble computer runs at the extreme scale with self-guiding adaptation

The team developed a powerful set of uncertainty qualification methodologies to initially characterize the uncertainty for a combined ocean and atmospheric model to yield adaptively constructed simulations for past and future climate. The capability will ultimately be applied for placing increasingly precise uncertainty bounds on the performance of nuclear weapons. The NNSA Office of Advanced Simulation and Computing has provided support to continue this research to develop advanced uncertainty qualification approaches associated with numerical error in computer codes, topological adaptive sampling, sparse and scalable emulators, aggregation in adaptive sampling using regression models with multivariate response, and application of these uncertainty qualification methods to predictions accompanied by uncertainty in nuclear stockpile devices.

Nuclear Threat Reduction

Researchers for an LDRD project developing an imaging approach for verification use (11-ERD-051) seek to create 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



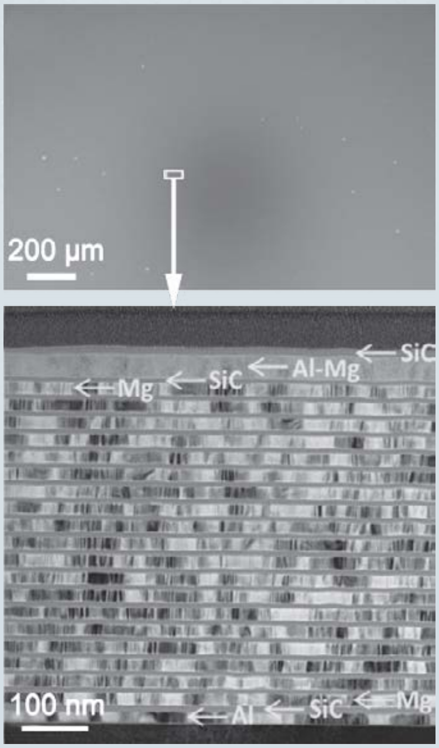
as a nuclear warhead, into a set of unclassified metrics. These metrics would be sufficiently accurate to identify a warhead from a gamma-ray image without divulging secret information, and include density and isotopic profile of materials that are used in warhead construction, which is a new concept that has not been realized for objects such as a nuclear warhead. Researchers have successfully validated computer simulations of radiation transport in three-dimensional objects and in a gamma-ray imager with comparisons of actual images from a Livermore gamma-ray spectrometer. In the coming year, they will design a prototype code-aperture gamma-ray system that is optimized for arms-control applications.

Controlling Fusion and High-Energy-Density Matter

Most astronomical shocks, such as those traveling through the explosion of a massive star in a collapsing supernova, do not involve collisions between bodies. The mechanism involved is thought to be plasma instabilities. Researchers supported by LDRD are developing a new class of experiments to observe the creation of collisionless shocks and the generation of magnetic fields as a by-product in a high-energy-density realm that can only be created on Earth by the National Ignition Facility (11-ERD-054). The experiments will study astrophysical shock-generation mechanisms using laser-created high-velocity plasma flows. The work will produce new diagnostic capabilities for measuring

- Magnetic fields
- Electron spectra
- Density and structure of plasma capabilities

while creating expertise and understanding for follow-on applications of fusion-class laser facilities for the Laboratory's mission in stockpile stewardship. This year the team has discovered, using a proton probe beam, unexpected stable field structures from small-scale plasma processes, and studied possibilities of how these structures are generated as well as plasma conditions when two plasma flows intersect.



Optical Materials and Targets

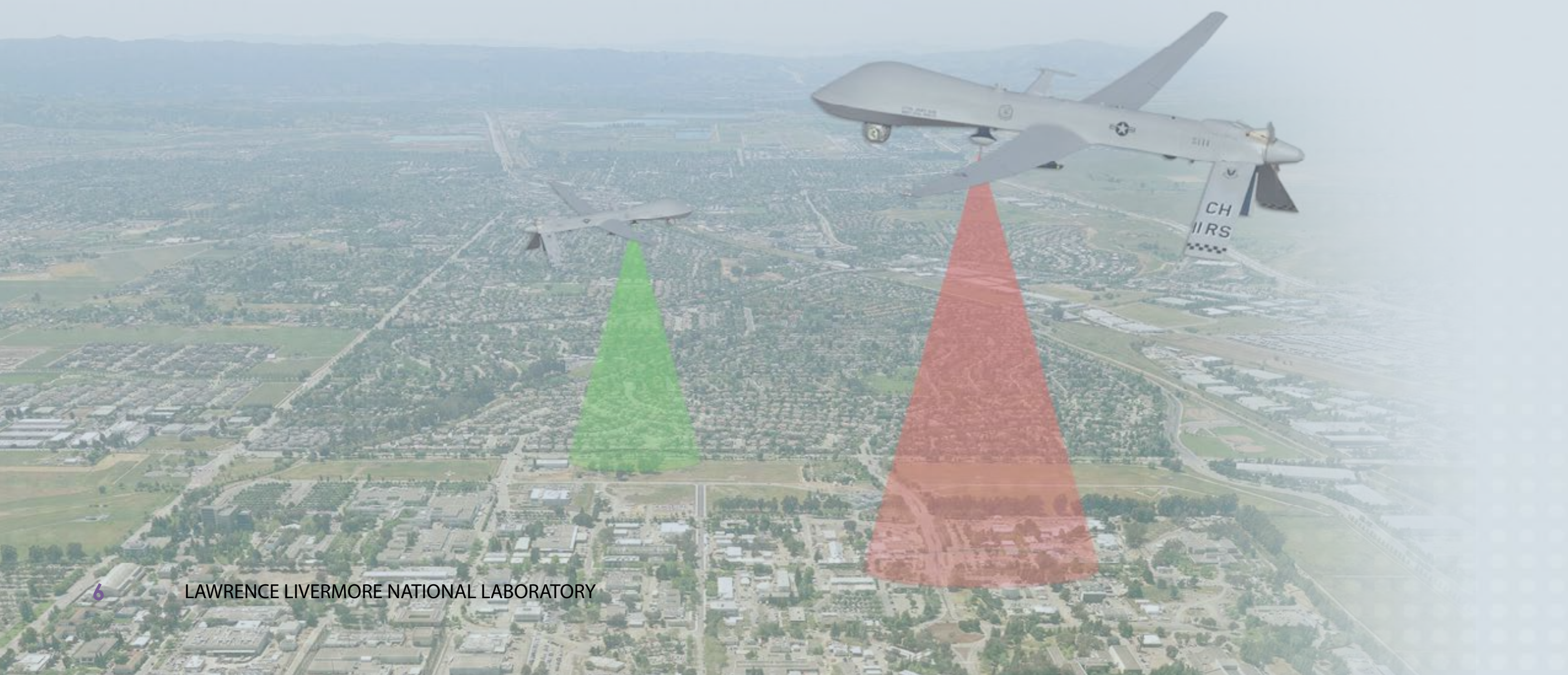
An FY12 LDRD project is devoted to multilayer thin-film science for core missions (12-ERD-055) including

- Roughness and microstructure manipulation
- Corrosion mitigation
- Defect reduction
- Smoothing

Researchers have determined the origins and mechanisms of corrosion propagation in magnesium and silicon carbide multilayers, whose potential to be the best-performing reflective multilayer coating relevant to solar observatories and space weather satellites is hampered by corrosion of the magnesium layer. Based on their analysis, they developed and demonstrated an efficient and simple-to-implement corrosion barrier for the multilayers that consists of nanometer-scale magnesium and aluminum layers that intermix spontaneously to form a partially amorphous aluminum and magnesium layer.

Cyber, Space, and Intelligence

A central concern in cyber security and intelligence is continuous surveillance, dependent on processing of high-volume data streams. An LDRD project's primary deliverable will be novel information theoretic approaches for the adaptive sub-sampling of very-high-throughput data streams to effect orders-of-magnitude increases in ingestion rates for filter-based learning software (11-ERD-035). 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. The end result will enable conclusions from data that is subject to random variation to be conducted in real time on data streams previously addressed only by retrospective techniques. Researchers have thus far developed dynamic particle filters capable of analyzing time-evolving, variable data. In the coming year they will implement their particle filters in Storm, a massively parallel, data processing software system for processing enormous amounts of data on the fly.

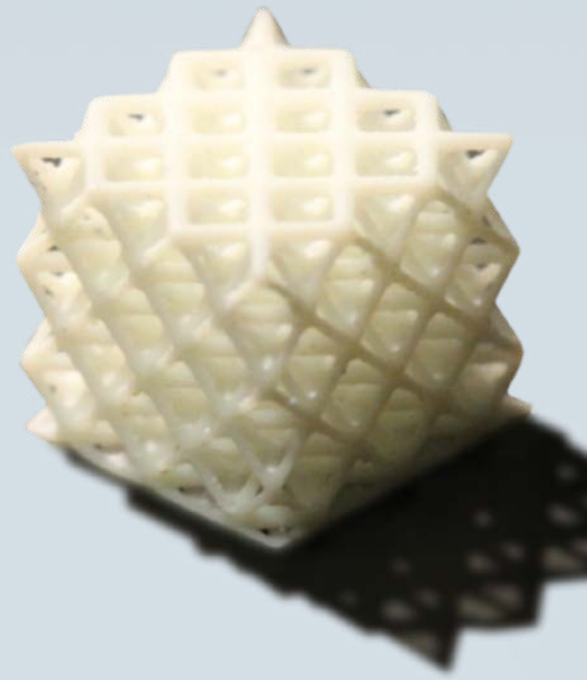


Materials on Demand

An LDRD project that is devoted to fundamentally understand and develop, from the ground up, new microscopic manufacturing techniques applicable to a variety of materials such as polymers, metals, and ceramics would enable production of three-dimensional large-scale geometries with micrometer-scale precision and scalable to achieve high manufacturing volumes at low cost (11-SI-005). This capability 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. Researchers have advanced the state of the art for three additive manufacturing fabrication technologies including

- Projection micro-stereolithography
- Electrophoretic deposition
- Direct ink writing

and have already produced microscopic-lattice materials with previously unachievable material property combinations. In the coming year the team hopes to advance their micro-manufacturing techniques by scaling up to produce larger parts, fully incorporating multiple materials into their manufacturing systems.

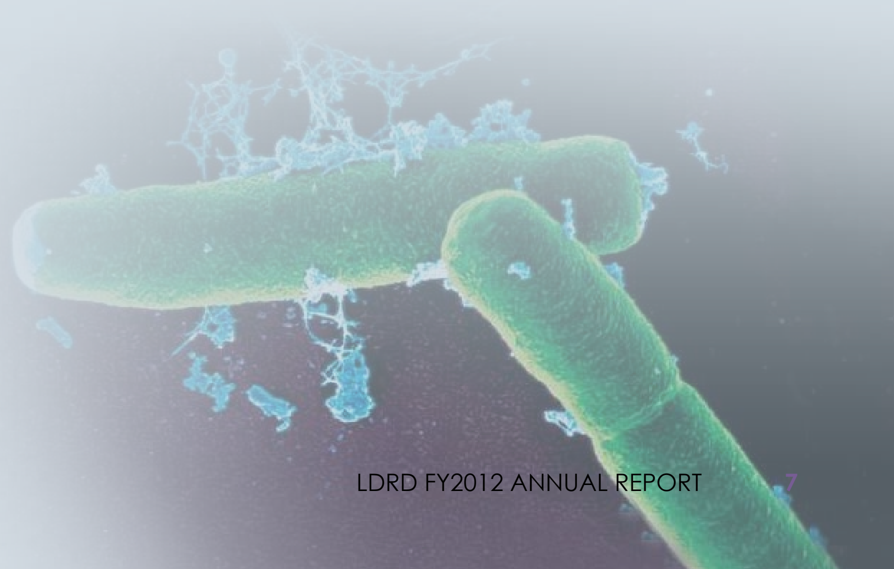


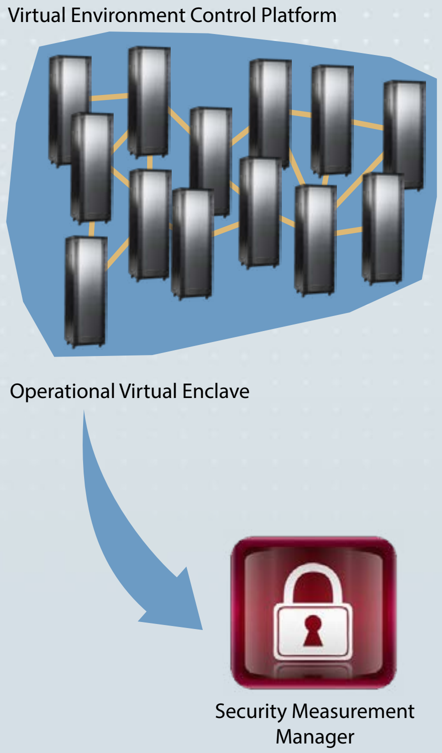
Biosecurity

An LDRD team is producing selected antigens that stimulate the production of antibodies such as small molecules, peptides, and membrane proteins for development of a new antibody selection approach—the display of antigens on nanometer-scale particles for synthetically generating antibodies (11-ERD-037). Using this approach, they will be able to rapidly select recombinant antibodies generated using cell-free methodologies directed against selected antigens related to host cell interactions with pathogens. The technology has the potential to

- Extend and revitalize LLNL's detection capabilities relevant to biosecurity
- Provide rapid mitigation of evolving and unknown biothreats
- Expand basic research in bioscience to improve human health

Researchers have thus far perfected a process for selecting, purifying, and analyzing single-chain antibodies that identify and neutralize secretion proteins used by the bacteria pathogen *Yersinia pestis* to introduce virulence factors into a host cell. In the coming year the team will test the ability of their synthetically generated antibodies to disrupt interactions between pathogen and host cells that lead to disease.



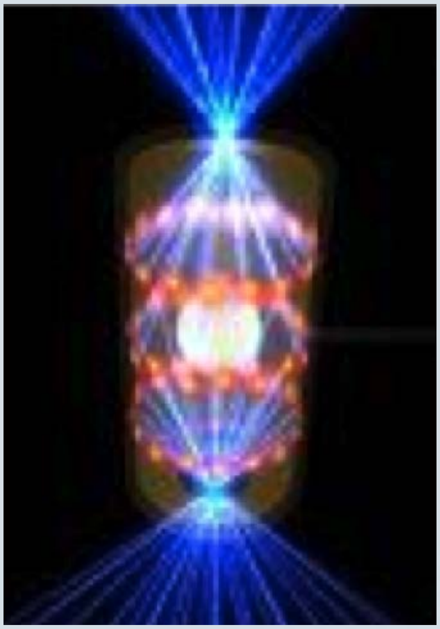


Information and Network Systems

Creating temporary computing environments that provide a means of conducting secure computational operations free from external probing, interference, and attacks is the goal of a current LDRD project on secure virtual network enclaves (12-ERD-016). The pervasive threat posed by

- Overseas adversaries
- Industrial espionage
- Insider threats
- Denial-of-service attacks
- Aggressively spreading malware

has substantially increased the risk of conducting business operations on traditional network infrastructures. Military services operating under hostile engagement scenarios face even greater threats to the security of their network-based information systems. The LDRD team is developing the capability to dynamically create secure network enclaves consisting of fully virtualized sub-networks that operate collectively and are capable of operating securely in a potentially compromised network. In FY12 the team developed the capability to automatically build a virtual enclave environment from a security-focused intermediate representation, and explored and identified characteristics that are unique to purely virtualized environments and that form the basis for novel security measurement approaches.

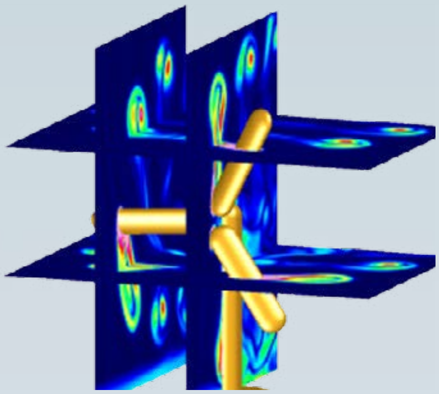


Laser Inertial Fusion Energy (LIFE)

Laser inertial fusion energy, which uses laser energy to ignite fusion fuel through inertial confinement of a high-energy plasma, can potentially help meet the world's growing demand for energy. Unlike conventional fission nuclear energy, it

- Generates only negligible nuclear waste
- Avoids the concerns of proliferation
- Poses little environmental risk

An LDRD project is addressing the question of how to successfully launch a tiny fusion target into a reaction chamber and design it to arrive at the chamber's center in the precise structural configuration necessary for a fusion implosion reaction to occur (10-SI-004). The team is achieving these objectives through experimental and modeling efforts to study specific high-risk aspects of the target cycle, including target layer properties, the fragile support structure of the target capsule, and the thermal reflectivity of ultrathin metal films used as energy reflectors. This past year, researchers have constructed various diagnostic devices to determine representative target material characteristics. This information is being used in computer simulations to model optimum target materials and configuration.

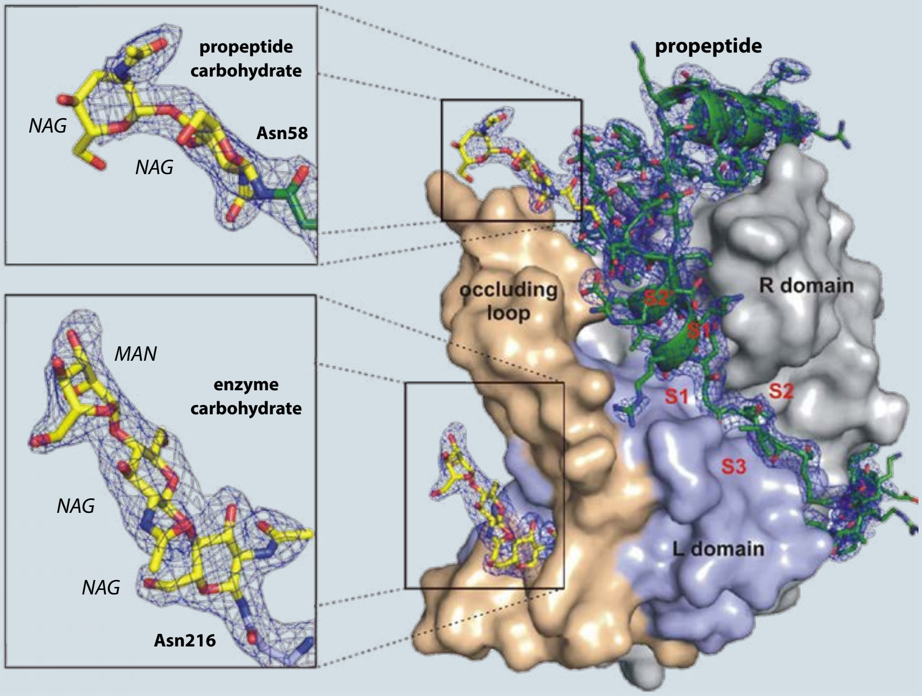


High-Performance Computing and Simulation

CgWind, developed by LDRD researchers, is a high-fidelity simulation of wind turbines and their rotating blades, as well as the simulation of turbine arrays in a wind farm (10-ERD-027). It will account for the effects of topography as well as regional wind flow patterns. The tool is orders-of-magnitude faster than existing approaches and will enable simulations that will provide new insight into the physics of complex turbulent flow problems experienced in energy-producing wind farms to enhance development of a clean and renewable energy source. As part of this work, the team addressed incompressible turbulent flows around complex and moving geometries and the combination of very-large-scale flows (meteorological) and very-small-scale flows (turbine-blade boundary layers). The successful conclusion of the project has led to a cooperative research and development agreement with an industrial partner for wind-park modeling and evaluation of new large-eddy turbulence models and near-field turbine models in collaboration with Caltech, Colorado University, and the University of Wyoming.

Bioscience and Biotechnology

An international team of LDRD researchers has for the first time used an ultra-intense x-ray laser to determine the previously unknown atomic-scale structure of a protein (12-ERD-031). The work was reported in the online edition of *Science*. The team determined the structure of an enzyme key to the survival of the single-celled parasite, *Trypanosoma brucei*, responsible for African sleeping sickness, a disease that kills 30,000 people each year. This new structural information should help guide the search for drugs that act like the propeptide, tying up the enzyme and killing the parasite. To determine the structure of the precursor form of the protein—which does not form crystals large enough for traditional x-ray diffraction—submicron crystals produced by the parasite were analyzed by the "diffraction before destruction" technique, in which individual nanometer-scale crystals are passed, one by one, through the x-ray beam at the Linac Coherent Light Source at Stanford University, then "stacking" the resultant diffraction data—in this case, from 178,875 individual nanocrystals. The achievement also demonstrates that the approach can provide otherwise unobtainable biomolecular information, potentially ushering in a new era of protein crystallography.

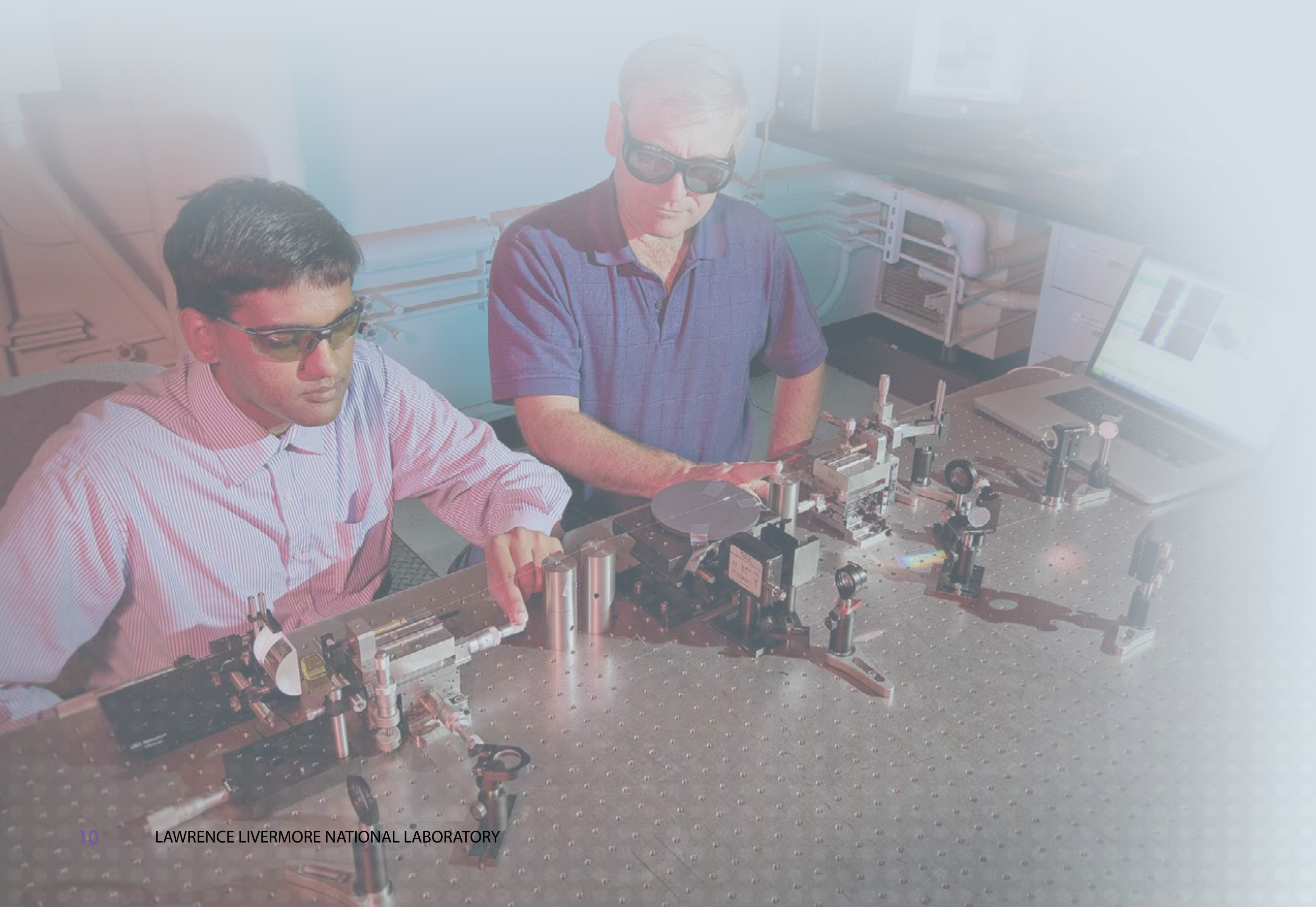


Measurement Science and Technology

Researchers are working to develop a new, transformative carbon-14 measurement science that is applicable to environmental and biomedical research (11-ERD-044). This technology will be less expensive and simpler to operate and maintain than accelerator mass spectrometry and will be similar in size to current optical spectrometers, allowing for routine radiocarbon-based studies in any laboratory and in unattended use in remote field locations. Although accelerator mass spectrometry has the proven sensitivity to measure carbon-14 dioxide at ultralow levels, the technology's costs, complexity, and throughput limit its usefulness in developing large-scale programs to monitor fossil fuel emissions. The team has completed a tabletop prototype system based on coupling two distinct laser spectroscopy approaches, and in the coming year will determine and optimize the

- Range of sample size
- Range of carbon-14 concentration
- Measurement precision
- Accuracy
- Throughput

with the goal of defining the operational parameters that would be expected for a commercial system.



Advanced Optical Systems and Applications

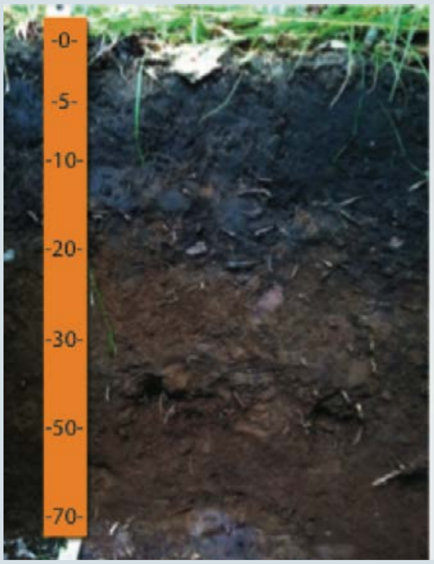
Fiber lasers are a critical technology for 21st-century energy and defense missions. Fiber lasers have scaled in output power and pulse energy in the last 25 years and are reaching their limits imposed by the physics of a technology originally developed for telecommunications systems. A team of LDRD researchers has successfully developed new optical fibers and fabrication techniques, for which three patents have been filed (10-SI-006). The team not only created a comprehensive model of a ribbon-shaped (flattened) optical fiber structure, or waveguide, but also developed new fabrication techniques and constructed a three-story optical fiber draw tower that reduces the time to produce a new fiber design from four months to about a week. They then constructed and tested a pulsed laser system that can amplify light beams to powers well beyond the fundamental limits of conventional round fibers used in telecommunications. Extending these lasers to higher pulse energies and average powers will support national missions as diverse as

- Directed-energy laser weapons
- Laser range finders and remote sensors
- Mono-energetic gamma-ray generation
- Laser-based particle acceleration
- Extreme ultraviolet sources
- Advanced machining

Earth and Environmental Sciences

Forest soils represent a significant pool for carbon sequestration and storage, but the factors controlling soil carbon cycling are not well constrained. Researchers are comparing the soil carbon storage and dynamics in five broadleaf forests in the eastern U.S. that vary in climate, soil type, and soil ecology in a current LDRD project (11-ERD-053). They found that

- Total soil-carbon inventory in the top 60 cm of soil were lowest at the site with the coarsest soil texture and at the warmest site.
- Differences in climate only partly explained differences in the soil organic matter content and mean turnover times, which also spanned a large range of 75 to 480 years.
- The turnover rates, inferred from carbon-14 measurements made at Livermore's Center for Accelerator Mass Spectrometry, were fastest at the warmest site but slowest at sites in the northeast, rather than the coldest sites in the upper Midwest.



The team speculates that soil texture, mineralogy, drainage, and biological activity may be at least as important as climate for controlling the soil carbon dynamics in temperate broadleaf forests. Studies of this type are essential to understanding how the carbon sequestered in soils will respond to changes in climate, and how those changes should be incorporated into coupled ecosystem and atmosphere models used to predict behavior of Earth's climate.

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.

American Association for the Advancement of Science

Christopher Keane

Christopher Keane was one of two Lawrence Livermore scientists awarded the distinction of fellow of the American Association for the Advancement of Science in 2012. Keane is being recognized for "distinguished technical and scientific leadership in developing inertial confinement fusion and high energy density science, and leading a robust global science community in this area." He is the author of more than 100 scientific publications and has given numerous invited talks at major international conferences and other meetings. Keane is also a member of the American Physical Society and a recipient of the NNSA Silver Medal, NNSA Defense Programs Award of Excellence, and Fusion Power Associates Special Award. He has participated in numerous LDRD research projects, including six current studies related to astrophysics, plasma, and shock physics of materials subjected to high temperatures and pressures with applications in stockpile stewardship, fusion energy production, and understanding origins of the universe (11-FS-004, 005, 006, 007, 008, and 009).

The Presidential Early Career Award embodies 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.

Presidential Early Career Award for Scientists and Engineers

Heather Whitley

For her work using path-integral Monte Carlo techniques for calculating results from random sampling to produce very accurate quantum statistical potentials; for applying these methods to a basic, foundational understanding of thermal conductivity in fusion ignition capsules for the National Ignition Facility; and for service to Lawrence Livermore's postdoctoral association, Heather Whitley received a 2012

Presidential Early Career Award. Whitley is currently a co-investigator for an LDRD project examining the transport properties of dense plasmas and a new hybrid simulation technique for matter at extreme conditions by developing advanced capabilities to model plasma physics uncertainties about thermal conductivity and stopping power from ion and electron collisions (12-SI-005). She is also involved in research for identifying and characterizing the electronic structure of defects and interface structures in the fundamental building block of quantum supercomputers, known as qubits (12-ERD-020).

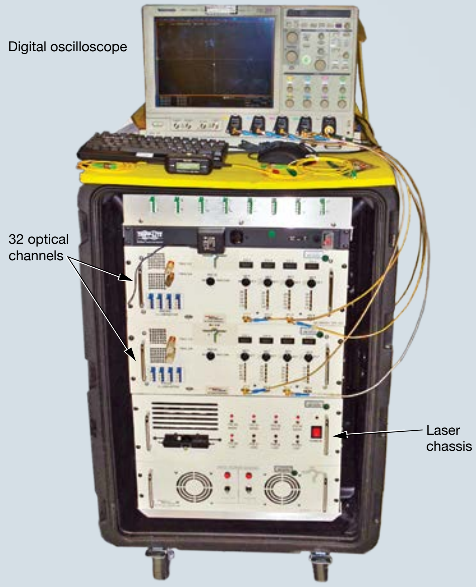
Jeffrey Banks

Jeffrey Banks was also honored for his work in computational physics, scientific computation, and numerical analysis, especially pioneering contributions in numerical approximations to hyperbolic partial-differential equations—which can be used to describe a wide variety of phenomena such as sound, heat, electrostatics, electrodynamics, fluid flow, or elasticity—focusing on the development and analysis of nonlinear and high-resolution finite-volume and finite-difference methods, as well as for his service in high schools and the scientific community. Banks has served on LDRD research teams striving to make significant advances in uncertainty quantification science for application to important global variables to strengthen our models of climate change (10-SI-013), and is currently on a team that is examining processes that affect laser light propagation and the effects on distribution of electrons from plasma waves excited by laser light relevant to fusion energy (12-ERD-061).

R&D 100 Awards

It is noteworthy that about two-thirds of our R&D 100 Award-winning technologies over the past decade have had their roots in LDRD projects, including three of this year's awards. Each year, *R&D Magazine* presents these "Oscars of Invention" to the top 100 technological advances that contribute to meeting an important national or societal need. This year's R&D 100 Award winners bring the Laboratory's total to 143 since Livermore began participating in 1978.

- ***Multiplexed Photonic Doppler Velocimeter.*** For years, scientists conducting shock-physics experiments were limited to measuring sudden velocity increases at only a few discrete points on a target's surface, used to address diagnostic needs of the NNSA nuclear design laboratories. Today, scientists are routinely recording 96 channels of optical velocity data at a fraction of the former cost to acquire data from just a few channels. The new enabling technology, for which its developers earned an R&D 100 Award, is called multiplexed photonic Doppler velocimetry. First demonstrated in LDRD work at Livermore, it has since been used at Los Alamos National Laboratory and the Nevada National Security Site to collect key data in hydrodynamic



A compact multiplexed photonic Doppler velocimetry system includes a digital oscilloscope for recording data, 32 optical channels for simultaneously measuring 32 discrete surface velocities, and a chassis for powering the lasers that are used to determine the beat frequency—a combination of the Doppler-shifted light from one laser and the reference light from another laser.

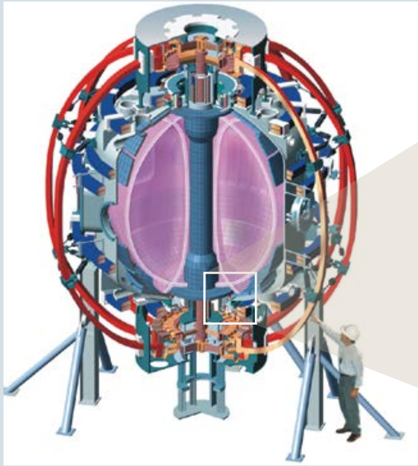
Livermore physicist Natalia Zaitseva leads a research team that has developed the first plastic material capable of efficiently distinguishing neutrons from gamma rays.

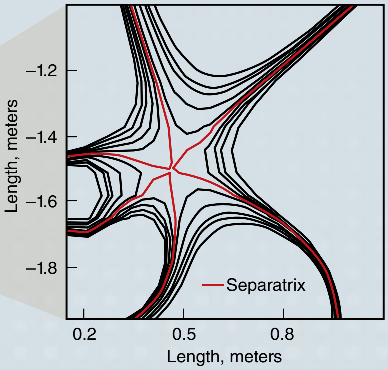
experiments in support of national security work. NNSA's National Security Technologies, LLC, is our partner in the award, which had roots in an LDRD project headed by Paul Sargis that worked to replace the Fabry–Perot velocimeter—which had a low channel count because of size, complexity, and cost—with a novel-fiber-optic implementation of laser Doppler velocimetry (98-ERD-016).

- ***Plastic Scintillator.*** In 2007, the Laboratory launched a three-year LDRD exploratory research project aimed at improving capabilities to effectively detect and identify fissile materials. Better detectors are needed to counter nuclear smuggling and proliferation. With LDRD support, systematic surveys were completed of 150 materials, leading to a better understanding of the physics of scintillation and the development of efficient, low-cost materials. This highly successful effort was followed up with programs at the Laboratory funded by the NNSA's Office of Nonproliferation Research and Development and the Department of Homeland Security's Domestic Nuclear Detection Office. The R&D 100 Award is a testimony to the importance of the breakthroughs that were made and the substantial progress that is being made toward enhancing our nation's security. The LDRD project, devoted to exploring the scintillator material candidates best suited to detection and monitoring of fissile materials and developing the scintillators for large-crystal growth (07-ERD-045), was led by Natalia Zaitseva.

- ***Snowflake Divertor.*** The "snowflake" power divertor, an ingenious method for dissipating exhaust power from hot plasmas in tokamak fusion test reactors, has been tested at institutions that are partner recipients of the award, including the Princeton Plasma Physics Laboratory, the École Polytechnique Fédérale de Lausanne Center for Research in Plasma Physics in Switzerland, and Oak Ridge National Laboratory. The concept for the divertor was developed under an LDRD project to explore innovative divertors for future fusion devices led by researcher Dmitri Ryutov (08-ERD-019). The snowflake divertor reduces heat flux to the divertor plates by an order of magnitude, does not affect the core plasma behavior at the edge (a critical result, because the plasma edge can strongly affect the core), and reduces harmful impurity concentration in the core.

A "snowflake" power divertor is installed at the bottom of the tokamak at the National Spherical Torus Experiment at Princeton Plasma Physics Laboratory. A snowflake magnetic configuration shows the characteristic hexagonal shape of the separatrix (inset), which is the singular magnetic field line that determines overall field geometry.





American Geophysical Union Fellows

Qingyun Duan was elected in 2012 as an American Geophysical Union fellow for "his fundamental contributions to hydrological and meteorological modeling including physical process representation and efficient, effective model parameterization." Duan—from the College of Global Change and Earth System Sciences, Beijing Normal University, Beijing, China—was a co-investigator for an LDRD project predicting effects of climate change and variability on water availability to determine how global warming and year-to-year natural climate variability will affect surface temperatures, precipitation amounts, rain and snow partitioning, soil moisture content, the moisture content of the snow pack, and the amount and timing of flow through rivers (03-ERD-042). In addition, he participated in a project devoted to predictive knowledge systems for large, complex data sources to enable computations capabilities that allow analysts to extract knowledge in a meaningful and timely way using pattern discovery, learning and prediction, and data-intensive computational architectures (06-SI-006).

American Geophysical Union Ascent Award

Stephen Klein

Stephen Klein became one of the inaugural recipients of the Ascent Award from the Atmospheric Science's Section of the American Geophysical Union for "elucidating the role of clouds in climate change and the fidelity with which climate models simulate clouds." The award aims to reward exceptional mid-career scientists in the field of atmospheric and climate sciences. The 93-year-old American Geophysical Union's goal is "to promote discovery in Earth and space science for the benefit of humanity." Klein is a co-investigator for an LDRD project on enhancing climate model diagnosis and model comparisons that seeks to develop advanced performance metrics for model simulations of climate processes, including resolving uncertainties in cloud feedbacks and other key mechanisms (10-ERD-060).

Department of Energy Early Career Research Program

The Early Career Research Program 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. This year, the Office of Science awarded 68 recipients out of a total of 850 proposals, and LDRD researcher Celine Bonfils was selected for her project to improve understanding of the nature and causes of past changes in droughts for identifying potential onset of future drought (11-ERD-006). Instead of solely focusing on changes in drought characteristics, Bonfils and her team are investigating the naturally driven and externally forced components of known large-scale drought precursors such as specific ocean temperature patterns or shifts in atmospheric circulation.

Celine Bonfils (second from left) is shown with the other Laboratory scientists awarded the Early Career Research Program award (Andreas Kemp, Gianpaolo Carosi, and Jaime Marian).

John Dawson Award for Excellence

A far-reaching discovery about the interactions between laser and matter with important implications for the National Ignition Facility has led to the selection of a team of researchers as recipients of the 2012 John Dawson Award for Excellence in Plasma Physics Research presented by American Physical Society, Division of Plasma Physics at their annual meeting in Providence, Rhode Island. The team's work led to the conclusion that the energy-transfer process during laser and matter interaction could be controlled using slight adjustments to the laser beams' wavelengths, and even be used as a novel tool to tune the implosion symmetry of National Ignition Facility targets. A member of their team, Robert Kirkwood, is an LDRD researcher leading a project on high-energy beams for simulating high-yield nuclear events (12-FS-012).

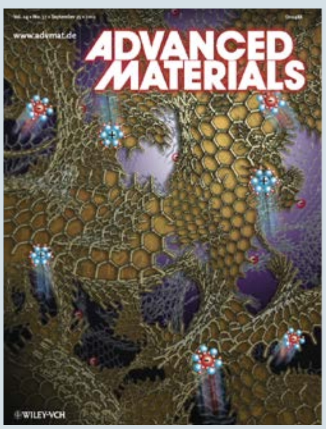
LDRD researcher Robert Kirkwood (back row, center) was part of a team of LLNL researchers that received the 2012 John Dawson Award for Excellence in Plasma Physics Research.

Associate Editor for Frontiers in Microbial Immunology

Amy Rasley

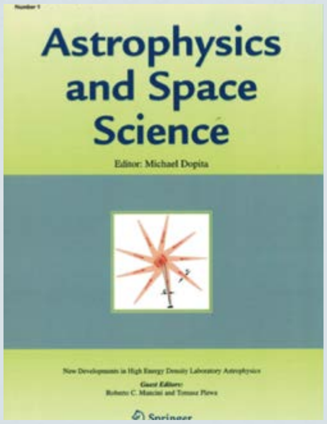
Amy Rasley has been named as the associate editor for the scientific journal Frontiers in *Microbial Immunology*, which publishes articles on the most outstanding discoveries across the research spectrum of microbial immunology. Rasley also received a Fitzpatrick Postdoctoral Award from the University of California, Davis for her work. She is an LDRD investigator working to develop an innate immunity for biodefense applications with targeted immune-modulation to counter emerging threats (11-ERD-016). She has also served as a co-investigator or led several other LDRD projects devoted to the understanding, prevention, and therapeutic countermeasures for pathogens and potential biological threat agents (09-ERD-054, 09-LW-036, and 08-FS-011).

Journal Covers



The cover art from the September 2012 issue of *Advanced Materials* represents the three-dimensional structure of a new polymer-derived material that consists of a three-dimensional network of single-layer nanometer-scale platelets of graphitic carbon. The material is mechanically robust and enables researchers led by Juergen Biener to dynamically control its properties through ion-induced interfacial electric fields (12-ERD-035).

A scanning electron micrograph of a protist recorded by LDRD researcher Kevin Carpenter is featured on the cover of the December 2011 issue of *Microbe*. Protists, simple micro-organisms without specialized tissues, are responsible for half of all photosynthesis on the planet and play a major role in the natural cycling of carbon and nutrients. Carpenter's LDRD-funded research focuses on the often-symbiotic relationship between protists and bacteria, with the ultimate goal of improving efficiency and reducing cost in converting plant biomass to biofuels (11-LW-039).



The November 2011 issue of *Astrophysics and Space Science* featured on its cover a paper by LDRD researcher Dmitri Ryutov. The paper, "Using intense lasers to simulate aspects of accretion disks and outflows in astrophysics," describes how some aspects of accretion disc physics can be experimentally simulated by using an array of properly directed plasma jets created by intense laser beams. Ryutov has served as a principal or co-investigator on numerous LDRD projects extending back to 1996, including astrophysical collisionless shock generation by laser-driven laboratory experiments (11-ERD-054), innovative divertors for future fusion devices (08-ERD-019), the creation of a neutron star atmosphere (04-ERD-028), and x-ray optics and applications for fourth-generation light sources (00-ERD-025). He is also a 2010 recipient of the Fusion Power Associates Distinguished Career Award.

A paper reporting the results of the initial implosion experiments on the National Ignition Facility in late 2010 and early last year was featured on the cover of the April 2012 issue of *Plasma Physics and Controlled Fusion*. The photograph is of a cryogenic ignition target before being fully enclosed by the shroud. Lead author Siegfried Glenzer was joined on the paper by a large group of collaborators from Livermore's National Ignition Campaign, the Massachusetts Institute of Technology, and the U.K. Atomic Weapons Establishment. Glenzer is a frequent LDRD researcher on projects related to plasma characterization, shock turbulence physics, laser interactions with dense plasmas, and characterizing compressed matter (02-ERD-013, 02-ERD-023, 03-ERI-070, and 08-ERD-002). He has most recently served as co-investigator for a project on advanced inertial fusion target designs and experiments for transformative energy applications (11-SI-002) and as the principal investigator for research on the dynamics of ultrafast heated matter (11-ERD-050).

Best Paper Awards

At the Ninth International Symposium on Special Topics in Chemical Propulsion in Quebec, Canada, LDRD co-investigator Kyle Sullivan was awarded Best Paper for his work on a current LDRD project on scalable high-volume micro-manufacturing techniques for three-dimensional mesoscale components (11-SI-005) headed by LDRD researcher Christopher Spadaccini. The team intends 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. The project also garnered a Best Poster Award for both the 2011 Fall and 2012 Spring Materials Research Society Meetings for team members Jennifer Lewis and Xiaoyu Zheng, respectively.

Research efforts on a sub-wavelength plasmon laser with potential applications in ultra-dense information storage, multiple-channel parallel data communication, multiplexed miniature spectroscopy, and enhanced metrology (12-ERD-065) by LDRD researchers Tiziana Bond, Allan Chang, and Mihail Bora was awarded a Best Paper Award by the International Academy, Research, and Industry Association. The association's aim is to promote scientific and industrial interchanges between members of existing associations and standardization bodies and to establish bridges between different scientific, academic, and industrial cultures. They are focused on advanced technologies, tomorrow's products, and inventions, and promote them through various events and publications.

Best Abstract

The "Genomics of Cell-to-Cell Communication: Identification of DNA Sensors in Humans" (10-ERD-020) was presented at the American Society for Bone and Mineral Research annual meeting and earned the Best Abstract Award for team member Bryan Hudson for an LDRD project headed by Gabriela Loots. Their research effort proposes to identify genomic signals that are activated in response to the presence of cancer cells. Also, for this work, Hudson was recognized with the Best Poster Award at Livermore's annual postdoctoral symposium.

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 FY12, LDRD costs at LLNL were $91.8M, which is 5.53% of total Laboratory costs. The number of patents resulting from LDRD-funded research since FY08 and the percentage of total patents that were derived from LDRD research and development are 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 FY12, LDRD projects generated 45% of Livermore's total patents, even though the LDRD program constitutes less than 6% of the Laboratory's budget.

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

Patents	FY08	FY09	FY10	FY11	FY12
All LLNL patents	57	46	54	60	78
LDRD patents	23	20	27	32	35
LDRD patents as percentage of total	40%	43%	50%	53%	45%

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 FY08 to FY12 account for 41% of the 741 total. In FY12, there were 162 records submitted at Livermore, with 79 (49%) of those attributable to LDRD-supported projects.

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

Record of Invention	FY08	FY09	FY10	FY11	FY12
All LLNL records	110	145	160	164	162
LDRD records	44	56	66	59	79
LDRD records as percentage of total	40%	39%	41%	36%	49%

Finally, LDRD plays a role in producing Laboratory copyrighted material. From FY08 to FY12, LDRD-supported projects accounted for over 26% of the 316 Livermore copyrights. In FY12, there were 74 LLNL copyrights, with 17 (23%) 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 FY12 there were 1,016 such articles, of which at least 230 (23%) resulted from LDRD projects. Over the last several years, the percentage of LDRD-supported articles has remained relatively consistent, with a five-year average of 20% of total Laboratory publications. The following table shows the number of journal articles per fiscal year resulting from LDRD-funded research since FY08, and the percentage of total articles that were derived from LDRD research and development.

Journal Articles	FY08	FY09	FY10	FY11	FY12
All LLNL articles	1,214	1,311	966	994	1,016
LDRD articles	233	225	227	207	230
LDRD articles as percentage of total	19%	17%	23%	21%	23%

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 FY12 portfolio included 91 formal LDRD-funded collaborations involving 59 LDRD projects (65% of the total projects funded). Collaborating institutions included the University of California (15% of total collaborators), other academic institutions (61%), DOE sites (3%), and other collaborators such as other government agencies and industry (20%). These statistics do not include the numerous informal collaborations that PIs 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 FY12, the LDRD Program supported 72% of the Laboratory postdoctoral researchers—there was an average of 207 postdoctoral researchers at LLNL in FY12, of which 149 were supported in some way by LDRD projects. The Laboratory continues significant recruitment efforts to maintain the total number of postdoctoral researchers.

Program Mission

To fulfill its missions, LLNL must continually invest in the science and technology that form the foundation of its signature capabilities. The LDRD Program, which was established by Congress at all DOE national laboratories in 1991, is LLNL's most important single resource for fostering 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, 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 2012, Laboratory Director Penrose Albright and the deputy director for science and technology (Tómas Díaz de la Rubia through August 2012) were responsible for the LDRD Program. Execution of the program was delegated to the director of the LDRD Program Office, William Craig. The LDRD Program at LLNL is in compliance with DOE Order 413.2B and other relevant DOE orders and guidelines.

[1] *U.S. Department of Energy Order 413.2B, Laboratory Directed Research and Development* (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).

[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).

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.

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

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

Strategic Context for the FY12 Portfolio

The FY12 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 is 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 FY12, the Laboratory's LDRD Program strongly supported DOE strategic themes:

[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).

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 foundations:

Mission Focus Areas

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

Science, Technology, and Engineering Foundations

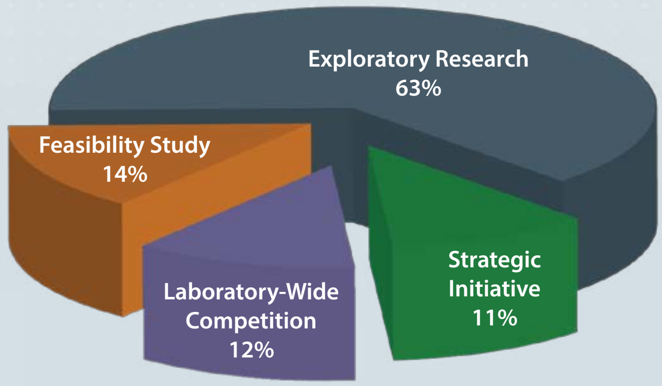
- Controlling Fusion and High-Energy-Density Matter
- High-Performance Computing and Simulation
- Materials on Demand
- Measurement Science and Technology
- Energy Manipulation
- Information and Network Systems
- Earth and Environmental Sciences
- Bioscience and Biotechnology
- Optical Materials and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

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

Structure of the FY12 Portfolio

The FY12 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.

In FY12 the LDRD Program funded 160 projects with a total budget of $91.8M. The distribution of funding among the LDRD project categories is shown in the following pie chart.



Distribution of funding among the LDRD project categories. Total funding for FY12 was $91.8M.

Strategic Initiative

In FY12, the LDRD Program funded 17 SI projects. Although the SI category represented just about 11% of the total number of LDRD projects for FY12, it accounted for over 33% of the budget. The SI projects ranged in funding from $1.2 to $3.0M.

Exploratory Research

The LDRD Program funded 101 ER projects for FY12. The largest project category, ERs accounted for over 63% of the number of LDRD projects and almost 60% of the budget for the fiscal year. Projects in this year's ER category ranged in budget from $140K to $1.7M.

Laboratory-Wide Competition

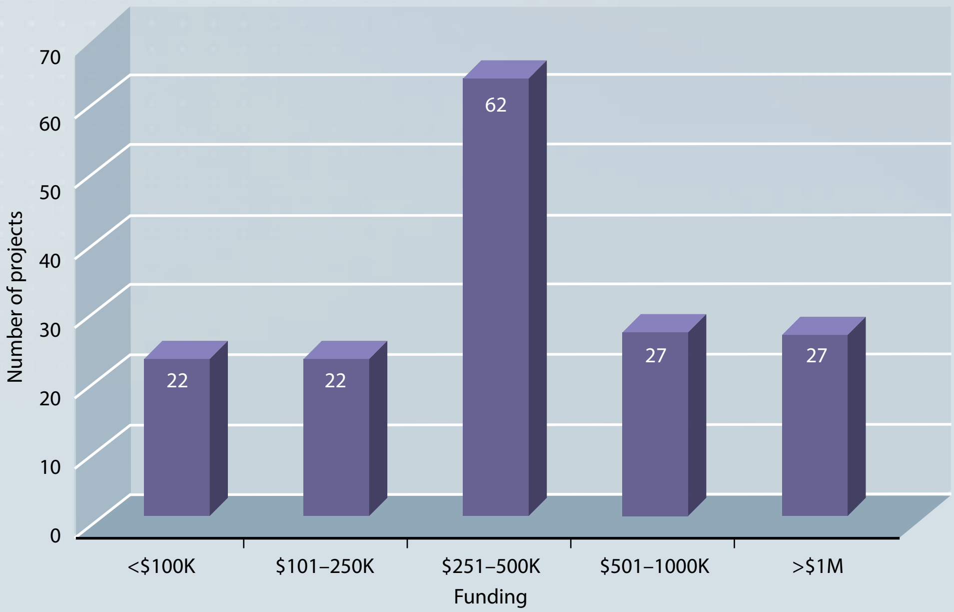
In FY12, 19 LW projects were funded, which represent about 12% of the LDRD projects for the year and approximately 5% of the budget. The LW projects for FY12 ranged in funding from $100 to $280K.

Feasibility Study

The LDRD Program funded 23 FS projects in FY12, which represent over 14% of the LDRD projects for the year but less than 2% of the budget. The FS projects for FY12 ranged in funding from $40 to $100K.

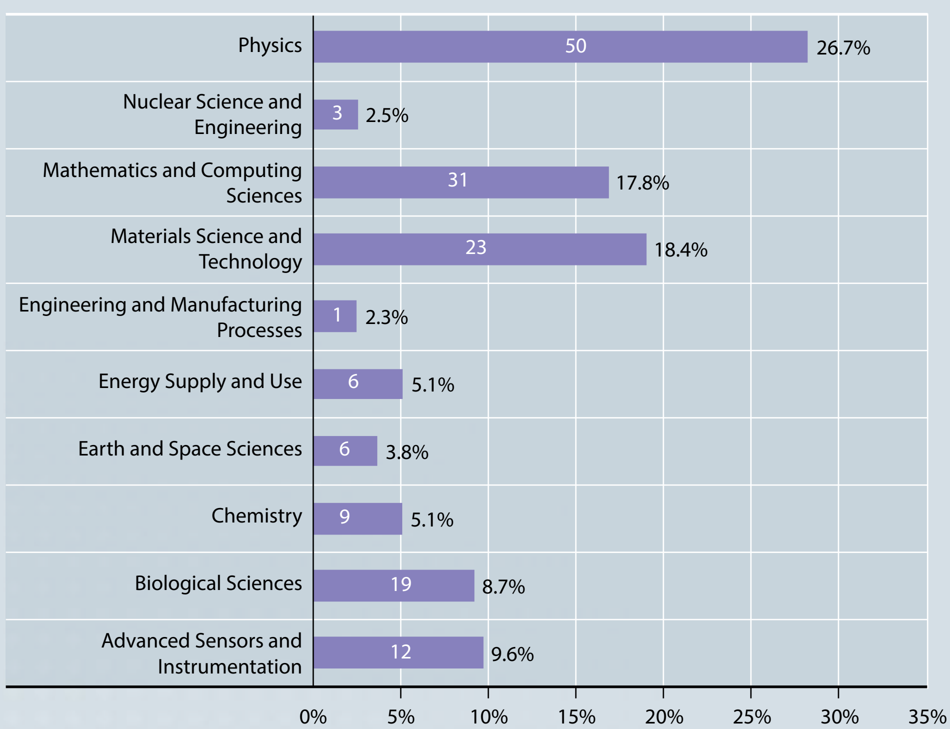
The following bar chart shows the funding distribution by dollar amount for the 160 FY12 projects—over 52% of the projects were in the $101 to $500K range, with about 14%

falling below $100K. Projects in the $501K to $1M funding range accounted for almost 17% of the total, and nearly 17% of the projects received more than $1M. The average funding level for the 160 projects was about $574K.



Number of projects and levels of funding. The average funding level for an LDRD project in FY12 was about $574K.

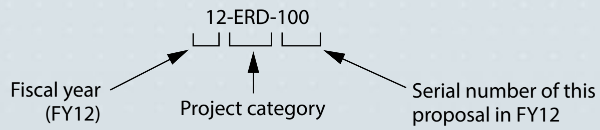
Percentage of LDRD funding and number of projects for each research category for FY12 are shown in the following chart. Exactly 65% of the total number of LDRD projects fall under the physics, materials science and technology, and mathematics and computing sciences categories, with costs for the three research categories at about 63% of the total LDRD budget.



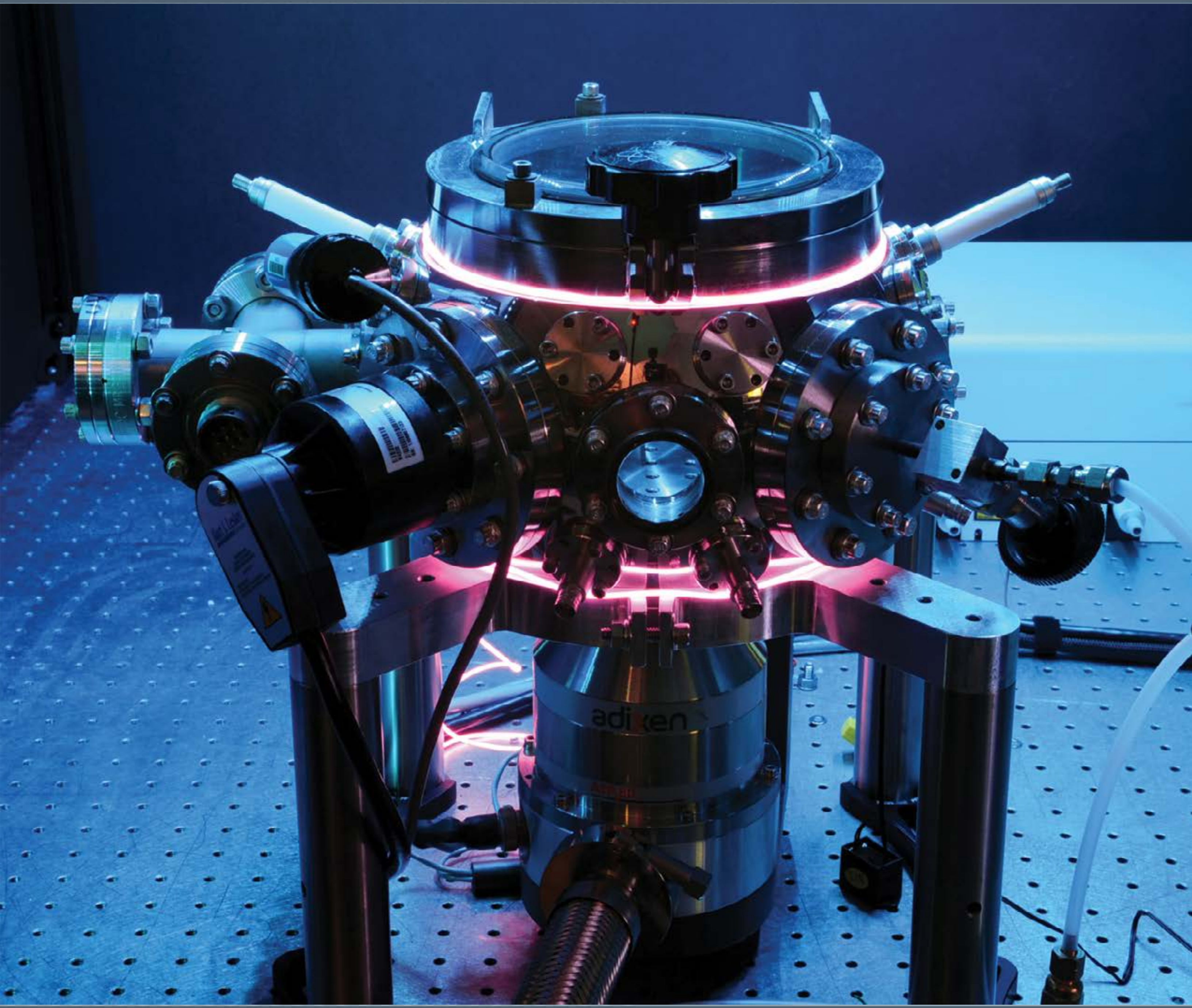
Percentage of LDRD funding and number of projects in each research category in FY12.

Organization of FY12 Project Summaries

Project summaries for the LDRD FY12 annual report are organized in sections by research category (in alphabetical order). Within each research category, projects appear for the various groups including SI, ER, LW, and FS. Each project is assigned a unique tracking code, an identifier that consists of three elements. The first is the fiscal year the project began, the second represents the project category, and the third identifies the serial number of the proposal for that fiscal year. For example:



Advanced Sensors and Instrumentation



Laboratory Directed Research and Development

FY2012

Real-Time Space Situational Awareness

Scot Olivier (10-SI-007)

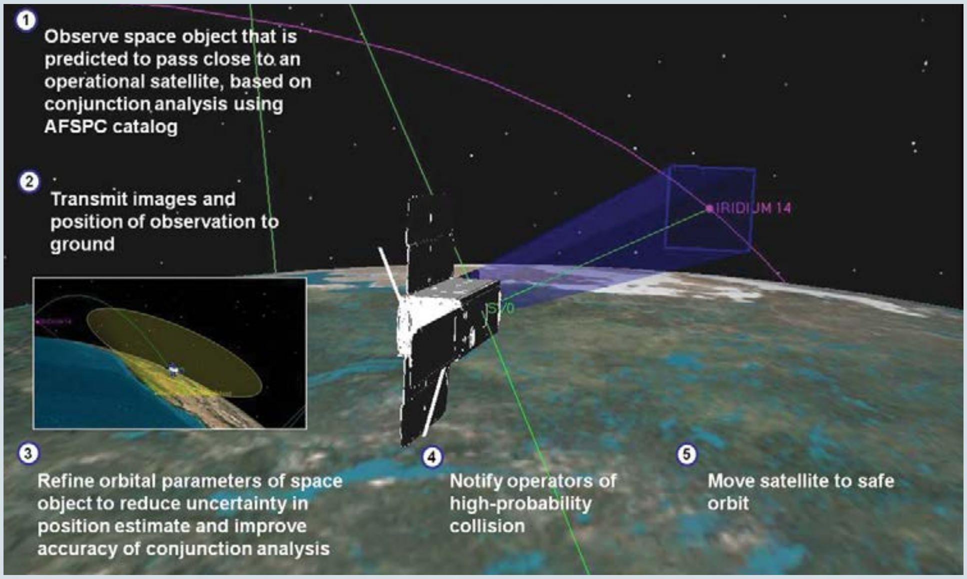
Abstract

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 are 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



The space-based telescopes for actionable refinement of ephemeris (STARE) concept uses very small satellites ("nanosatellites") to help prevent future collisions in space that could destroy valuable satellite systems. An initial STARE *Pathfinder* nanosatellite—built by LLNL, Boeing, the Naval Postgraduate School, and Texas A&M University—was launched successfully in September 2012. The above image shows the general process by which STARE data would be used to avoid collisions.

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.

FY12 Accomplishments and Results

In FY12 we (1) used data from multiple sensors to validate techniques for predicting the position and time that space objects will enter the Earth's atmosphere and for quantifying the uncertainty in these predictions; (2) calculated the quantitative benefit of using space-based sensors for providing the data needed to make collision warnings easier to use and act on by satellite operators; (3) calculated the quantitative effect of combining multiple space-based observations to refine orbital trajectories of space objects; (4) simulated methods for optimizing the cueing of space-based sensors to collect the data needed for enhanced collision warnings for satellite operators; and (5) completed the integration, testing, and launch of a prototype "nanosatellite" about the size of an large soda bottle with an optical payload designed to demonstrate techniques for collecting data for enhanced collision warnings. In summary, we established a new capability for the high-performance modeling and simulation of technologies for space situational awareness, developed a new methodology for using space-based sensors to collect data to enhance collision warnings for satellite operators, and demonstrated a novel approach for deploying space-based sensors using nanosatellite technologies that reduce costs by a factor of 100 to 1,000 compared with traditional space systems. Follow-on work from this project already includes U.S. Air Force support for developing operational space command-and-control tools based on the modeling and simulation capabilities developed for this project and a partnership with NASA Ames Research Center to deploy a constellation of nanosatellites to demonstrate the mission concept of using space-based sensors to collect the data that satellite operators need to better act on collision warnings. We also anticipate that the Defense Department and other government agencies will support work to extend the use of our nanosatellite technologies to other related national security applications.

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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 for 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.

FY12 Accomplishments and Results

In FY12 we (1) developed our laser design for improved performance and robustness, notably by improving the pump delivery; (2) enhanced the fidelity of our propagation model by adding cylindrical filters and the improved pump delivery; (3) completed a thermally robust, harmonic converter design that provides high conversion efficiency under thermal load; and (4) developed a prototype mounting fixture for a Pockels cell (an electro-optic modulator) that will enable deployment of a novel approach that avoids plasma electrodes. Because of funding reductions, we did not pursue development of the diffractive final optic. However, the successful conclusion of this project enabled a novel laser design for production of high-energy pulses at high-repetition rates. Our results are now being applied to development of the overall conceptualization of inertial fusion power plants and has attracted the attention of several potential external sponsors.

Publications

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Embedded Sensors for Monitoring Complex Systems

Jack Kotovsky (10-ERD-043)

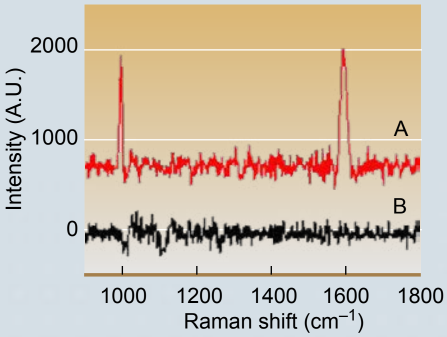
Abstract

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



Curve A is the surface-enhanced Raman scattering spectrum of a fiber probe developed for the remote detection of toluene vapor, with a laser power of 2 mW and an integration time of 10 s. Curve B, for comparison, is the spectrum obtained with a non-patterned optical fiber in toluene vapor with the same system configuration.

FY12 Accomplishments and Results

We (1) improved performance to detect toluene at concentrations as low as 250 ppm at reduced temperatures; (2) used kinetics modeling to reveal the process to be a recoverable physisorption one; (3) demonstrated the Raman detection of three organic vapors together through hollow photonic crystal fibers, transferring the nanometer-scale pillar substrates onto a multimode fiber tip after nanoparticles failed to yield improvements and using SERS to successfully detect a target substance with parts-per-billion sensitivity; (4) developed, in collaboration with an industrial partner, a custom, single-fiber system with flexible, miniature lenses less than 1 mm in diameter and which couple directly into the resonant chamber of the gas test cell; (5) verified exceptional performance with the vibrometer after abandoning the Fabry Perot system; (6) used new models to design final prototypes that performed more than an order of magnitude better than previously, measuring with a sensitivity of 100 ppm with a very strong signal, leading us to estimate that in an acoustically shielded environment, we could detect a target molecule at 1 ppm with the current design of 2 by 70 mm; and (7) developed and tested dual- and single-laser noise-cancellation systems. In summary, a miniature, all-optical photoacoustic spectrometer was developed and demonstrated to have a performance suitable for mission-relevant work. The next step is to continue work for weapons applications. We also successfully demonstrated the capability of SERS and nanopillar substrates for multiplexed vapor sensing with a sensitivity of hundreds of parts per million and furthered its potential by expanding it to a fiber-based format. Interest has been expressed by companies interested in SERS technology for drug testing (by breath analysis) and by a company interested in fiber-based systems for environmental monitoring.

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Quantification of Carbon-14 by Optical Spectrometry

Ted Ognibene (11-ERD-044)

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

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

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

FY12 Accomplishments and Results
In FY12 we (1) finished assembling the prototype cavity ring-down spectroscopy system, (2) constructed and tested a gas sample-handling apparatus, and (3) discovered reliability issues with the laser, which precluded use for continued

testing. While the laser was undergoing repair, we assembled a cavity ring-down spectroscopy system using a different laser, which was unable to obtain the optimal line for best carbon-14 sensitivity, but capable of measuring elevated samples.

Proposed Work for FY13

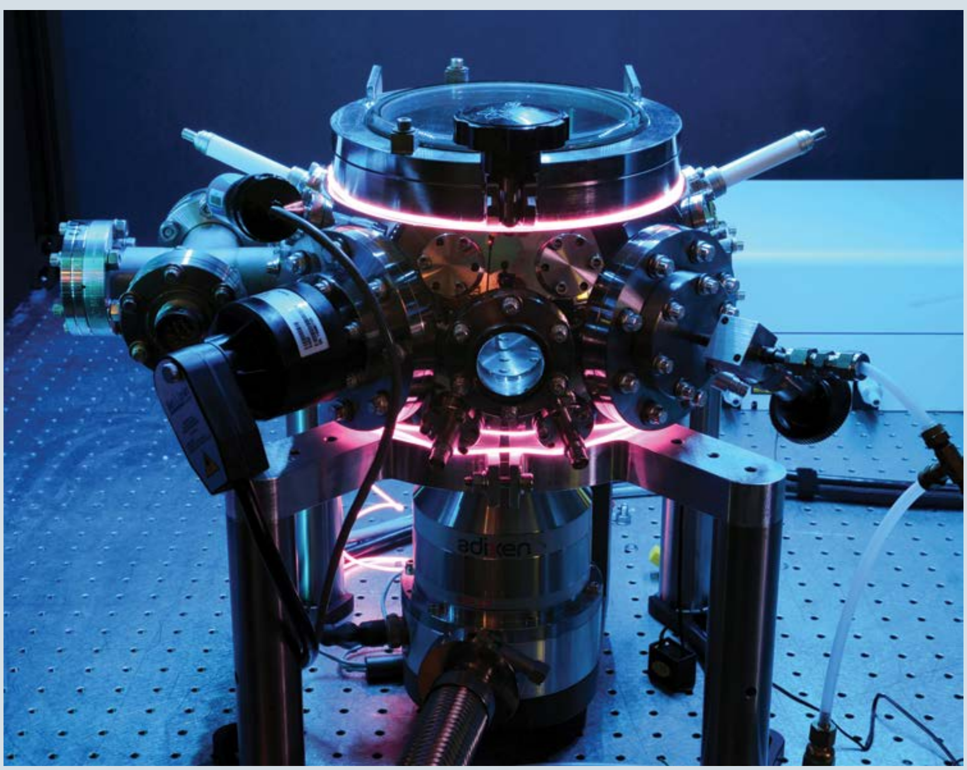
In FY13 we plan to continue evaluating the prototype cavity ring-down spectroscopy system for measuring $^{14}CO_2$. Specifically, we will determine and optimize the range of sample size, the range of carbon-14 concentration, measurement precision, accuracy, and throughput with the goal of defining the operational parameters that would be expected for a commercial system. We will also continue benchmarking the system through comparison to accelerator mass spectrometry measurements of split samples.

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,



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

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 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.

FY12 Accomplishments and Results

In FY12 we (1) extended models for electron cascades in gallium arsenide, creating the ability to accurately treat electron scattering to states with energies greater than 100 eV; (2) developed a model of the generation and transport of Cherenkov radiation in our gamma sensor; (3) successfully developed in situ deposition controls to fabricate optimized radoptic sensors; (4) designed, fabricated, and began validating the performance of the prototype electron optics system 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 (5) began follow-on experiments to demonstrate an integrated detector consisting of a radoptic sensor coupled to an optimized electron demagnification system for ultrafast detection of concentrated electron flux.

Proposed Work for FY13

In FY13 we will (1) refine the neutron and gamma-ray converter designs using the Geant and MCNPX particle and photon transport codes and MatLab models developed in FY11 and FY12, (2) extend our models of electron cascades in gallium arsenide and indium gallium arsenide phosphide to treat electron relaxation processes up to 10 eV to improve modeling of the sensor temporal response, and (3) conduct experiments to improve radoptic electron sensor sensitivity, refine the electron optics system to minimize dispersion of electron transit times, and integrate the electron detection system with an ultrafast optical recorder to produce an integrated system with picosecond temporal resolution.

Publications

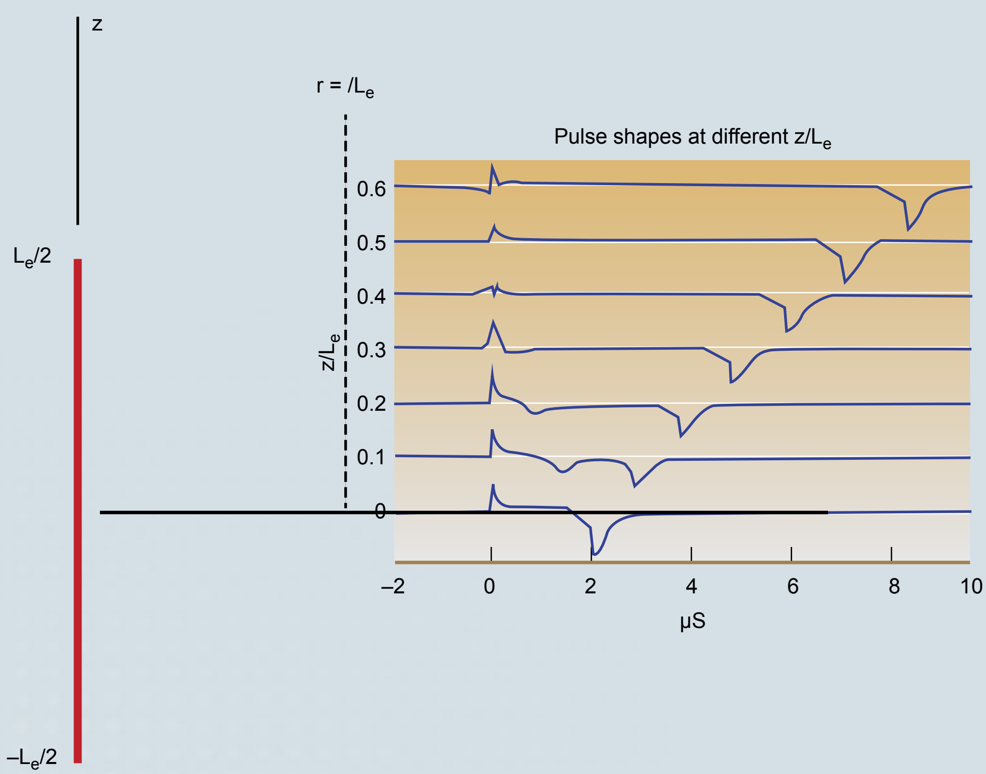
Vernon, S. P., et al., 2012. "X-ray bang-time and fusion reaction history at picosecond resolution using Radoptic detection." *Rev. Sci. Instrum.* **83**(10), 10D307. LLNL-CONF-555292.

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

Jerry Carter (11-ERD-061)

Abstract

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 reduced size, ruggedness for operation in a wide range of environments, high sensitivity with low sample volume, and immunity to environmental noise.



Calculations with our photo-acoustic Raman spectroscopy model of laser pulses at different locations relative to the laser focal region (red line).

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 uses, we expect our tuning fork and acoustic resonator PARS will have significant benefits in terms of sensitivity, selectivity, noise suppression, and form factor. We also anticipate that this technology combined with optical fibers will enable remote sensing for national security applications such as weapon material lifetime diagnostics for enhanced surveillance or optical lifetime predictions for advanced fusion-class laser systems.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we solved several key technical challenges, improved our experimental approach, and completed development of the first-ever dynamic PARS model for fluids. Specifically, we (1) updated the PARS model for fluids by adding viscous dissipation and thermal conduction; (2) extended our PARS model to solids using an approach based on an impulsively heated line source inside an infinite solid; (3) modeled and verified the resonance frequencies of our acoustic resonator cell; (4) significantly lowered (to 20 db) the noise of our acoustic detection setup by using filtering, shielding, and pre-amplification modifications; and (5) used a custom Raman converter (developed in FY11) pumped with a pulsed neodymium-doped yttrium–aluminum–garnet laser to demonstrate first Stokes Raman conversion at a high repetition rate (3 kHz) in hydrogen up to 600 psi while suppressing other Stokes output orders, although we found the resulting low-intensity beams insufficient to drive the stimulated Raman for generating a PARS signal of hydrogen, and so switched to a different laser having higher peak-power pulses. We also filed a record of invention.

Proposed Work for FY13

In FY13 we propose to (1) complete the resonantly detected photo-acoustic Raman analysis of model solid-phase materials and to validate the PARS theoretical performance model developed as part of our FY12 effort, (2) determine the feasibility of optical-fiber-based resonantly detected PARS as a micro-volume probe, and (3) incorporate a laser vibration measurement device into some of our PARS measurements on single crystals to investigate the feasibility of performing PARS as a completely non-contact method.

Publications

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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 micropower radar technology and a traditional microphone, 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 intend to 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 will 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.

FY12 Accomplishments and Results
In FY12 we (1) achieved our human subjects data acquisition, data analysis, and documentation requirements; (2) demonstrated and documented the feasibility of a background noise-removal algorithm; (3) examined the capabilities of speech-silencing methodologies; and (4) demonstrated the potential and requirements to enable real-time voice signature-alteration capability with a series of experiments using our radar neck sensor to obtain accurate, noise-free information on speech signals. Our project led to an improved understanding in hardware, data recording, and an understanding of voiced speech signals. It has enhanced our knowledge of micro-power ultrawideband impulse radar applications, and is leading to improved antennas, new algorithms, and human communications applications. Several new ideas were examined ranging from real-time speech cancellation to trachea-instrument improvements. We were successful in understanding the processing of voiced speech periods for several applications ranging from speaker verification to speech sound reduction (e.g., "cancellation"), and we better understand the details of voiced speech patterns generated by individuals for

verification and stress-sensing applications. The results and accomplishments from this project are being presented to multiple programmatic areas to address Laboratory mission needs.

A New Approach for Reducing Uncertainty in Biospheric Carbon Dioxide Flux

Sonia Wharton (12-ERD-043)

Abstract

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

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

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) continued modeling work in collaboration with an industrial partner; (2) completed field campaigns at Tonzi Ranch, California, and Wind River, Washington, including over 500 hours of light detection and ranging measurements for determining boundary-layer profiles of wind speed, direction, and turbulence, and over 50 balloon-borne instrument platform launches for measuring profiles of air temperature, relative humidity, air pressure, and wind speed and direction up to the tropopause; (3) gathered and analyzed continuous CO_2, water, and energy fluxes at Wind River, Tonzi Ranch, and Site 300; (4) installed a soil respiration chamber at Tonzi Ranch and gathered and analyzed soil respiration fluxes at Wind River; (5) completed ACASA input files with meteorological-driving data and plant physiology parameters for Wind River and began modifying the code for Tonzi Ranch to include a seasonal leaf area index component and intensified drought season regime; and (6) made initial code runs for both sites.

Proposed Work for FY13

In FY13 we will (1) perform a second set of intensive field campaigns at the Tonzi Ranch and Wind River sites; (2) perform field campaigns at Site 300 and possibly at Sherman Island in which we deploy light detection and ranging instrumentation and launch balloon-borne instrument platforms to measure upper boundary-layer conditions, as well as continuously measuring carbon, water, and energy fluxes at each tower; (3) continue input forcing in ACASA, fully validate selected output variables (such as turbulence kinetic energy, wind speed, and soil CO_2 flux) using soil respiration and boundary-layer observations, and fill in gaps in the flux datasets; and (4) calculate errors associated with ACASA versus other methods to assess model performance.

Publications

Osuna, J. L., S. Wharton, and M. Falk, 2012. *A new approach for reducing uncertainty in biospheric CO_2 flux*. LLNL-PRES-582996.

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

Wharton, S., et al., 2012. *FY12 report: meteorological observations for renewable energy applications at Site 300*. LLNL-TR-590692.

Sub-Wavelength Plasmon Laser

Tiziana Bond (12-ERD-065)

Abstract

We propose to develop a nanometer-scale plasmon laser (a plasmon is a quasi-particle resulting from the quantization of plasma oscillations) that overcomes the diffraction limits of current laser diodes. Sub-wavelength size is achievable because of the strong field confinement of surface plasmon modes in metallic nanowires. The proposed

nanoscale vertical laser structure is amenable to high-density, large-area fabrication techniques and requires simpler processing, providing a cost-effective alternative to existing diode lasers. The plasmon waveguide configuration can potentially deliver a two-order-of-magnitude reduction in device size compared to current state-of-the-art vertical cavity surface-emitting lasers, a proportional increase in wafer device density, as well as reducing laser-array dimension beyond state of the art. Generating intense, nanoscale-localized optical-frequency fields opens many possibilities for prospective applications in nanoscience and nanotechnology—in particular, for near-field nonlinear-optical probing, high-speed imaging, intense heat harvesting and transfer, and nanoscale modifications useful in nanolithography techniques.

We expect to develop a nanoscale laser that is significantly smaller than the wavelength of its emitted light, which is possible because of strong field confinement of plasmon modes in metallic nanostructures. We intend to design a plasmon resonant cavity and perform characterizations with and without gain media. We will also conduct spectroscopic characterization of dyes or quantum dots—semiconductors whose electronic characteristics are closely related to the size and shape of the individual crystal. The output versus input power will be measured to determine lasing threshold and efficiency that, along with spontaneous versus stimulated recombination rates, will give a complete picture of device performance. We foresee applications for the plasmon laser in ultradense information storage, multichannel parallel data communication, multiplexed miniature spectroscopy, and enhanced metrology. Research in nanoscale optical fields also opens possibilities for prospective innovative applications such as near-field nonlinear-optical probing and imaging as well as the concept of nanometer-scale modifications.

Mission Relevance

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

FY12 Accomplishments and Results

We (1) designed tunable cavities to align with the emission and absorption features of the gain media, for which we used dye in ethanol; (2) fabricated the cavities from aluminum, gold, and silver; (3) tested the far-field optical properties of the cavities both with and without the gain media, achieving wideband tuning capability and aligned resonances; (4) developed a fully integrated setup for time-resolved photoluminescence of the dyes in absence or presence of the nanometer-scale arrays, along with optical characterization of the plasmonic resonances, achieving results indicating that plasmons enhance dye photoluminescence and decay rates—when plasmons were aligned, we observed a 36-fold reduction in lifetime and a 70% increase in photoluminescence relative to bulk properties; and (5) began infrared and visible near-field transmission complex measurements of nanometer-scale pillar arrays, using an adapted atomic force microscopy–near-field scanning optical microscopy setup at the University of California at Berkeley, obtaining near-field images with a planar configuration designed with the same properties as the vertical arrays to avoid perturbations of atomic force microscopy tips inside cavities and allow for more design flexibility.

Proposed Work for FY13

In FY13 we intend to (1) finalize the combined reflectance, photoluminescence, and lifetime setup to perform a test on spectral characterization of the laser dyes within our templates, using the flow cell we developed, to obtain information on absorption and emission enhancement as related to plasmon hotspot positions; (2) determine variation in emission patterns and directionality, spectral tuning, and broadband coverage; (3) introduce models of the quantum-based interactions between the gain media and plasmon in absorption and emission stages and validate them against collected data; and (4) continue our collaboration with the University of California at Berkeley and provide information on field localization over and within the pillars that we will use to validate our theoretical framework.

Publications

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

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

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

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

Stephen Libby (12-LW-009)

Abstract

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

California, to the study of possible modifications of Newtonian gravity at high accuracy at the centimeter-to-meter scale and to measure the gravitational constant at new levels of precision.

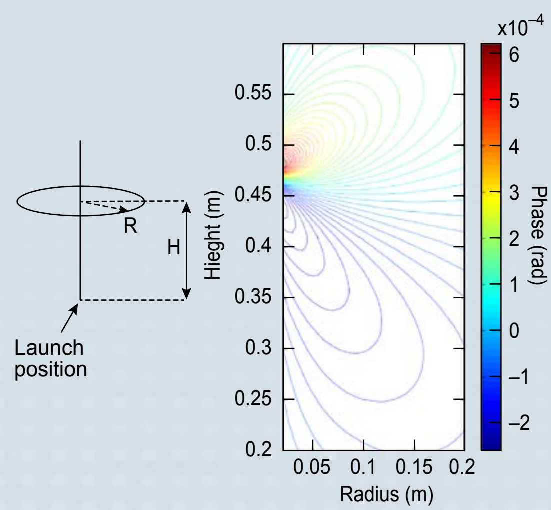
We expect, if successful, to make a significant contribution to fundamental physics by constraining grand unification models and the possible interplay of dark energy and particle physics by improving bounds on non-Newtonian mass-coupled forces over centimeter- to meter-length scales. In addition, this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

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) performed detailed signal sensitivity analysis of the error budget requirements for the AOSense gravity gradiometer for high-precision gravity measurements, discovering several novel cold-atom fountains—proof-mass arrangements that offer the potential for future ultrahigh-sensitivity measurements; (2) designed required mass-motion control for close-in phase-shift measurements (including geometrical proof-mass error budget constraints; couplings to the quadratic



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

level of shape, rotational noise, laser pointing, and atom cloud spatial variations; and full quantum corrections to one part in ten thousand of the cold-atom phases), which will lead to novel cold-atom measurements (such as the scalar Bohm–Aharonov effect for gravity); (3) performed a detailed retrofit design of the gravity gradiometer and the proof-mass translator and geometry and evaluated candidate proof-mass materials—tungsten, lead-tungstate crystal, and liquid mercury, finally selecting a nonmagnetic tungsten–copper–nickel heavy metal alloy; and (4) completed retrofit upgrades of the gradiometer laser and control system.

Proposed Work for FY13

In FY13 we will (1) implement the retrofit of the gravity gradiometer system—including a narrow line-width master laser and increased atom flux—and adapt the proof-mass translation system to accommodate our symmetric fundamental physics test geometry; (2) perform, after mounting proof masses, preliminary tests of the gravity gradiometer response and carry out any needed refinements of the gradiometer sensors; and (3) perform the first-ever direct measurement of quantum effects beyond the semiclassical in cold-atom gravity gradiometry.

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

Satinderpall Pannu (12-FS-010)

Abstract

In the past decade, there has been significant interest in developing in vitro human physiological platforms for medical research. However, there has been no significant effort to develop such a system that mimics the brain and central nervous system. Because a number of terrorist chemical agents such as Sarin affect the nervous system, an in vitro platform that recapitulates the brain and other neurological systems is required to develop effective medical countermeasures in a reasonable time. We propose to investigate the feasibility of a multi-cellular neuron tissue construct controlled by a simple micro-fluidic device to simulate the brain's function for use in such research. We will use direct ink deposition and traditional polymer micro-fabrication to develop the three-dimensional (3D) scaffolding required to house neuronal tissue. Utilizing in vitro platforms with primary human cells organized in a physiologically relevant manner will reduce preclinical testing and improve the relevance to clinical outcomes.

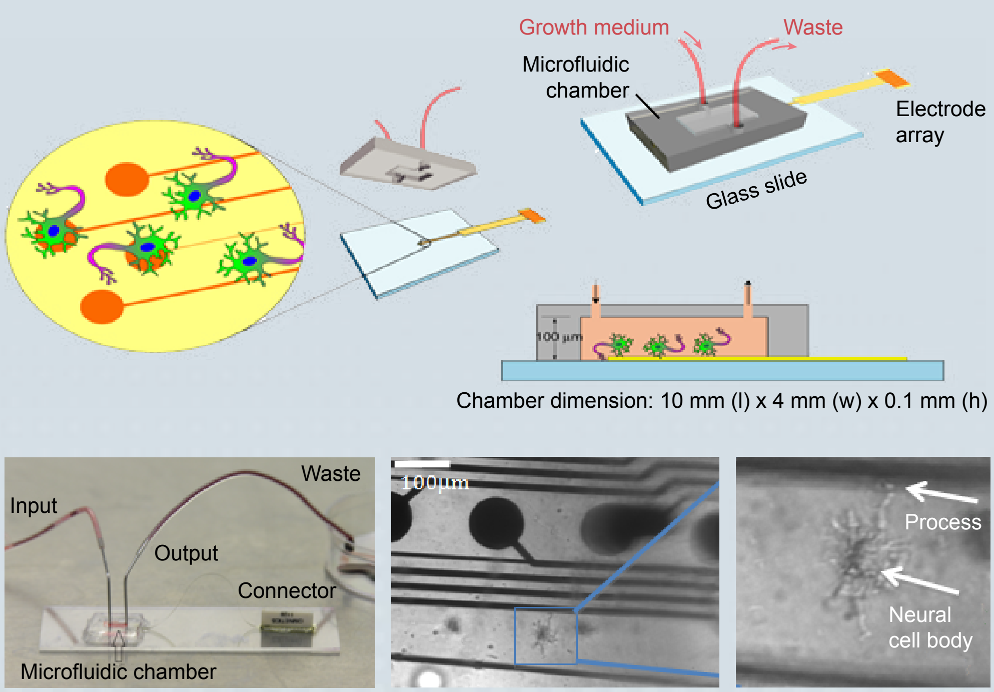
We are motivated by the need to rapidly develop medical countermeasures for chemical and biological attacks—at present, the timeline to develop such response is lengthy. The safety, efficacy, pharmacokinetics, and pharmacodynamics of medical countermeasures must be characterized in pre-clinical trials that include lengthy protocols involving animal testing. Frequently, the animal models do not accurately predict the clinical safety, efficacy, and tolerability in humans. We expect to leverage LLNL's expertise in micro-fluidics and electrode arrays to develop an in vitro platform demonstrating multi-cellular tissue survival in a 3D structure with a 3D array of electrodes. Our proposed physiological platform will enable stimulation and recording of output from neurons, which would yield valuable information about the cells and help maintain them for longer periods of time. Once this has been demonstrated, the Laboratory will be uniquely positioned to lead the field in neuron growth and research.

Mission Relevance

Our proposed project supports the Laboratory's strategic mission thrust in biosecurity for rapid mitigation of evolving and unknown threats by enabling timely research into medical countermeasures for response to a terrorist attack employing chemical or biological agents.

FY12 Accomplishments and Results

In FY12 we successfully fabricated a prototype microfluidic device with an embedded electrode array and tested it with dorsal root ganglion neuron cells from rats. Specifically, we (1) developed a microfluidic chamber comprised of a polymer chamber attached to a glass slide; (2) grew a neural cell body near an electrode array; (3) used optical microscopy to ascertain that the cells were healthy and were developing neural processes, and maintained the cells in these devices for seven days by periodically renewing the growth media; and (4) conducted initial electrophysiology experiments to determine our ability to stimulate the neurons and record a resulting action potential. Our research will be continued with an LDRD exploratory research project to create the first in vitro platform for the study of effects of human exposure to neurological toxins.



Overview of our in vitro chip-based human brain investigational (iCHIP) platform and optical microscopy showing electrophysical stimulation of rat neuron cells.

Energy Harvesting to Power Autonomous Sensors

Harry Radousky (12-FS-011)

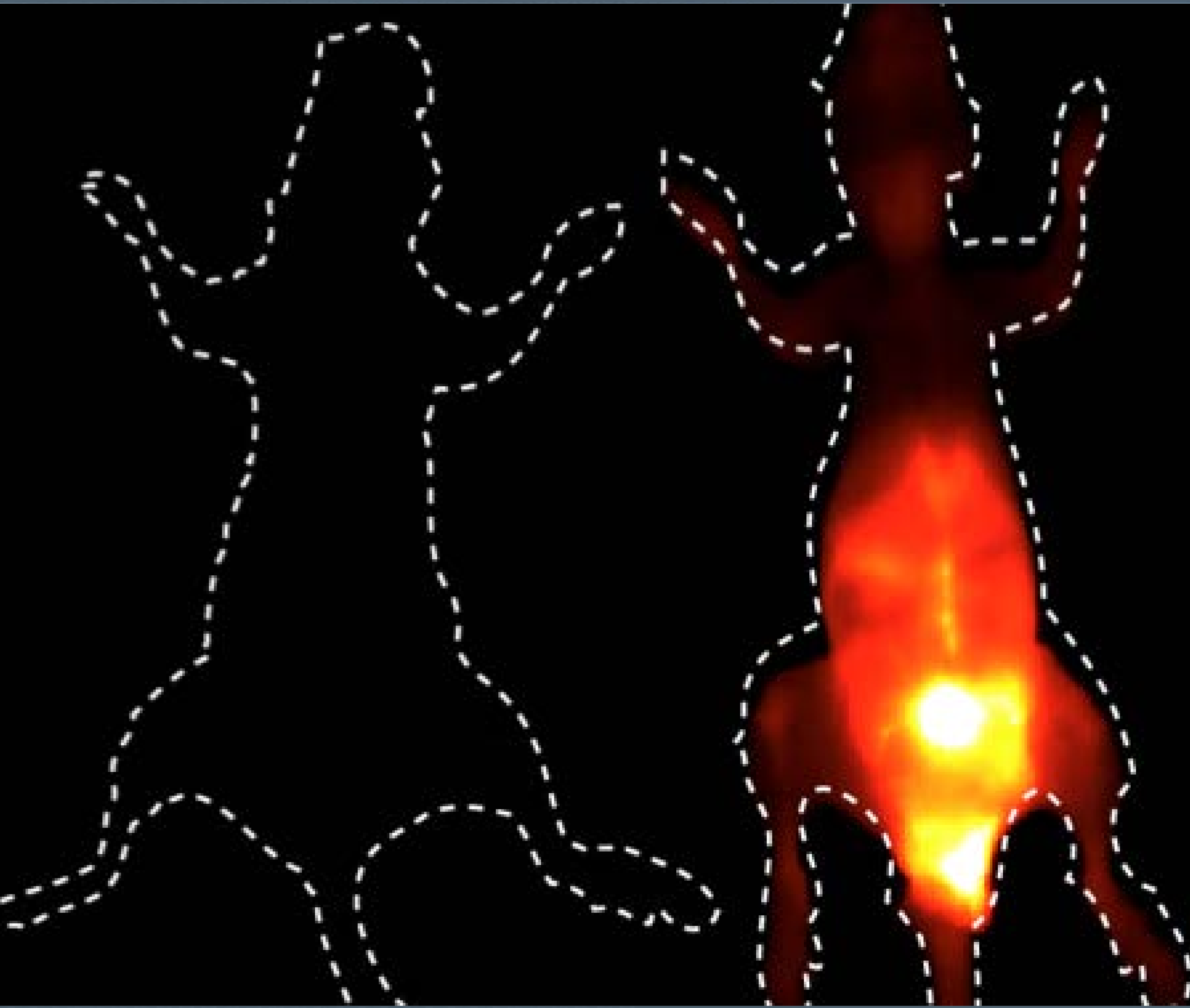
Abstract
We propose to explore the feasibility of alternative energy harvesting technologies to power autonomous sensors or other electronic devices deployed for monitoring the fuel cycle in a nuclear reactor to ensure safe operations and deter proliferated nuclear weapons capability. For autonomous sensors that perform their task without being connected to the interrogation unit, it is desirable to harvest the needed energy from the environment, living off the land, so to speak. We intend to focus on two adaptations of mechanical and thermoelectric harvesting technologies. The first technology we will explore involves mechanical and thermal energy harvesting based on nanometer-scale piezoelectric semiconductor wires and environmentally responsive polymers. This approach has the advantage of versatility—multiple energy sources can be targeted—along with high energy output. Our second approach will involve the harvest of low-quality thermal energy based on phase-change materials. This approach should not be confused with traditional thermoelectric devices, which are targeting thermal energy at relatively high temperatures.

We expect to determine the feasibility of producing power sources in the microwatt range for autonomous sensors relevant to monitoring the nuclear fuel cycle. We will scale up an energy-harvesting platform based on recent work to provide much higher power output and test additional energy-harvesting capability. The device relies on the response of a polymeric film to drive the piezoelectric effect in the nanowire array. Two key questions to answer from this feasibility study are whether multiple energy sources can be additive using the same platform, and the maximum power output of the devices. In addition, we intend to develop thermal harvesting from low-quality sources based on compound thermoelectric materials. This work will include device design and proof-of-principle demonstrations that will quantitatively evaluate the achievable power.

Mission Relevance
Autonomous sensors are vital for performing measurements in harsh environments characterized by high temperatures or corrosive atmospheres. Our proposal to develop alternative energy sources to power autonomous sensors for monitoring nuclear fuel cycles supports a core Laboratory mission to limit or prevent the spread of materials, technology, and expertise relating to weapons of mass destruction.

FY12 Accomplishments and Results
We demonstrated the feasibility of using zinc oxide nanowires to harvest both mechanical and low-quality thermal energy. First, we demonstrated mechanical harvesting with the periodic application of force, yielding an open-circuit voltage of 0.2 to 4.0 V and finding that voltage is enhanced by qualitatively increasing the bending curvature. To demonstrate thermal harvesting from low-quality heat sources, a nickel–

titanium alloy thin film—a phase-transition material with a transition temperature of approximately 50°C—was attached to the nanowire piezoelectric device, and the whole device assembly was bent at room temperature. Upon heating above 50°C, the alloy slip returned to its original flat shape, yielding an output voltage of nearly 1 V. In summary, this project has demonstrated the feasibility of our approach. The next step would be to develop and optimize a prototype by improving device performance to 5 to 10 V, demonstrating output power from a zinc oxide device, and investigating power levels from nanometer-scale wire devices based on lead zirconate titanate and using the gadolinium–silicon–germanium alloy $Gd_5Si_2Ge_2$ in place of nickel–titanium.



Laboratory Directed Research and Development

FY2012

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—the 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.

FY12 Accomplishments and Results

In FY12 we (1) completed our microarray analysis and identified a dramatic phenotype for co-cultured cells, demonstrating that osteoblasts lacking sclerostin protein promote cancer invasion by tenfold, while osteoblasts lacking the low-density lipoprotein protein 5 receptor inhibit cancer invasion; (2) continued to characterize the molecular events that contribute to this phenotype and identified several proteins such as matrix metalloproteinases and Dickkopf-related protein 1 that act as potential facilitators; (3) initiated in vivo animal work to test some of the findings we discovered in the cell–cell culture experiments; (4) identified 10 genetic loci, tested the evolutionary conserved elements, and identified several with enhancer activity; and (5) determined that the cancer invasion phenotype we observe when prostate cancer cells are co-cultured with sclerostin protein knock-out osteoblasts promotes a physical change in the prostate cancer cells characterized by filapodia, which are slender cytoplasmic projections. We hope to continue our research with support from the National Institutes of Health.

Publications

Hudson, B. D., and G. G. Loots, 2012. "Genomic profiling in bone and cartilage." *Genetics of Bone Biology and Skeletal Disease*, Academic Press, New York, NY. LLNL-BOOK-503531-DRAFT.

Hudson, B. D., et al., 2012. *SOST inhibits prostate cancer invasion*. ASBMR 2012 Ann. Mtg., Minneapolis, MN, Oct. 12–15, 2012. LLNL-ABS-567492.

Hudson, B. D., et al., 2012. *SOST inhibits prostate cancer invasion in bone*. LLNL-POST-564873-DRAFT.

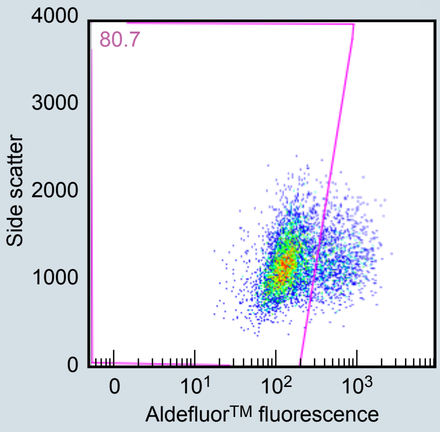
Establishing Cancer Stem Cell Longevity and Metastatic Potential

Bruce Buchholz (10-LW-033)

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 develop 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 the 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.



Aldefluor assay results for a bladder cancer cell line with resistance to a chemotherapy agent. A specific inhibitor of aldehyde dehydrogenase was used to set the gate for sorting the cells. Of the cells, 19.3% 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.

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.

FY12 Accomplishments and Results

We (1) identified cancer stem cell populations using an intrinsic marker and separated them from cell culture and human bladder tumors, finding that although the cultures contained high levels of cancer stem cells, the cells in excised tumors were too few to obtain reliable cell ages; (2) used the separated cancer stem cells in tumor xenograft studies in immunosuppressed mice; (3) identified a cancer stem cell marker that could be used in human breast cancer, in which tumors are larger and a cancer stem cell population could be large enough (that is, at least 500,000 cells) to obtain an age; and (4) published our results about the cancer stem cell marker in bladder tumor cells, the human bladder xenograft data, and breast cancer. In summary, this project established a marker of cancer stem cells in bladder cancer and separated populations from human tumors. Further work, including the extension of this technique to human breast cancer, is being supported by the National Institutes of Health.

Publications

Sarachine, M. J., and B. A. Buchholz, 2013. "Bomb pulse biology." *Nucl. Instrum. Meth. B*. **294**, 666. LLNL-JRNL-485203-DRAFT.

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

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. deVere White, 2012. "Stem-like cells in bladder cancer cell lines with differential sensitivity to Cisplatin." *Anticancer Res.* **32**(3), 733. LLNL-JRNL-522032.

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

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 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, such as treating overdoses of prescription, over-the-counter, or illegal drugs.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) incorporated a form of the CYP4503A4 protein and CYP450 reductase catalytic enzyme into a purified NLP construct, (2) co-expressed both the associated apolipoprotein and the target CYP450 and its reductase with membrane lipids and added surfactants and detergents for solubilization, (3) optimized reaction conditions for incorporation and expression of CYP450 and reductase into the NLP construct, (4) developed a colorimetric assay to assess CYP450 functionality, (5) investigated ways to assure CYP450 functionality during incorporation with NLPs, (6) developed ways to maintain CYP450 functionality using linear-dentritic polymer chemistry for formation of a CYP450 and nanometer-scale disk complex, and (7) demonstrated the ability of a CYP450 nanodisk construct to metabolize a substrate in vitro.

Proposed Work for FY13

In FY13 we will (1) optimize conditions to maintain CYP450 functionality using different detergents and reaction conditions; (2) investigate use of CYP4503A4, which has been shown to be more stable in the reactions in question; (3) test the metabolic functionality of the CYP450–NLP construct in vitro using caffeine as a T-2 surrogate; and (4) administer the CYP450–NLP construct to mice and determine the difference in metabolic and pharmacokinetic profiles between mice treated with CYP450–NLP and untreated controls.

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.

FY12 Accomplishments and Results

In FY12 we (1) characterized the in vivo distribution and pharmacokinetic profile of nanometer-scale lipoprotein (NLP) constructs following inoculation by intranasal, intraperitoneal, intravenous, and subcutaneous injection; (2) assessed inflammation induced by NLP and agonist constructs in vivo using a combination of fluorescence activated cell sorting analyses and an enzyme-linked immunosorbent assay;

(3) assessed inflammation induced by NLP and agonist constructs in primary human dendritic cells; (4) demonstrated, in collaboration with University of Rochester Medical Center, that NLP and agonist constructs protect mice against influenza-induced lethality; (5) demonstrated, in collaboration with the University of California Davis Cancer Center, enhanced activation of immune cells concomitant with a reduction in tumor burden in a mouse metastatic breast cancer model; and (6) demonstrated our ability to conjugate two additional agonists, muramyl dipeptide and flagellin, to the NLP platform.

Proposed Work for FY13

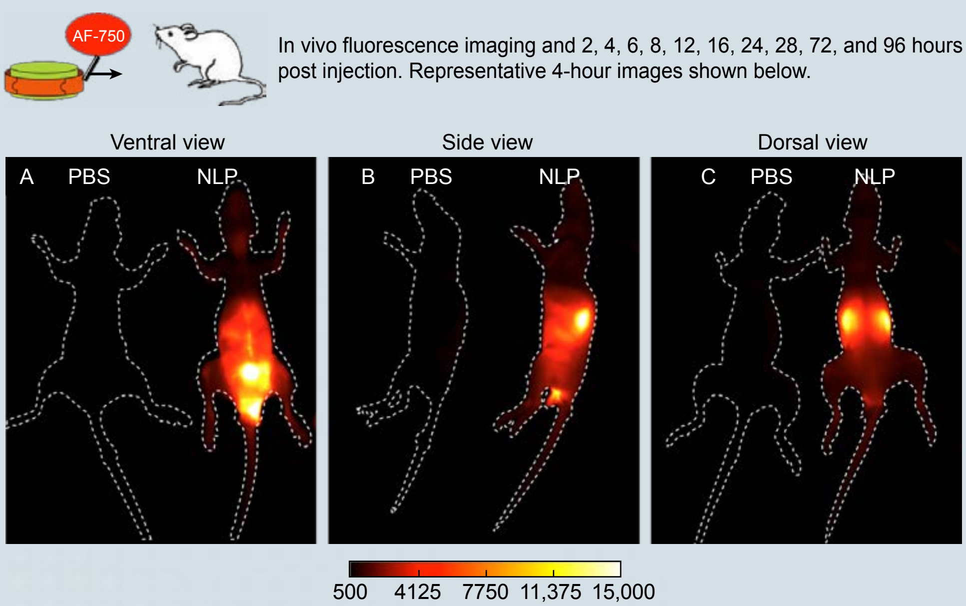
In FY13 we will (1) test our NLP and agonist constructs in vivo using relevant mouse models and taking advantage of LLNL's biosecurity level 3 facility, (2) continue to characterize NLP formulations containing multiple agonists in a single particle both in vitro and in vivo, and (3) test those constructs by demonstrating the ability to enhance innate immune responses in challenge models.

Publications

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

Rasley, A., et al., 2012. "Innate immune agonists conjugated to nanolipoproteins elicit robust inflammatory responses in mouse macrophages: Implications for host-based therapeutics." *J. Immunol*. **186**, 52.10. LLNL-POST-484037-DRAFT.

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



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

Unraveling of Assembly and Structure–Function Relationships of Poxviruses

Alexander Malkin (11-ERD-027)

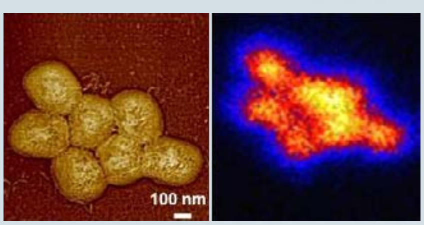
Abstract

Strong interest in poxviruses persists because of their unique replication cycle and assembly, the profound insights they provide into strategies to combat the host immune response, and the potential for deliberate release of poxviruses as a bioterrorist weapon. We aim to elucidate 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 (complete virus particle) and host–pathogen interactions, and determine forensic signatures using a multidisciplinary approach with a unique combination of nanometer-scale physico-chemical techniques and genetic and biochemical analysis. This improved knowledge will be pivotal for elucidating mechanisms of pathogenesis and developing countermeasures against viral agents.

We will identify and map target protein locations in vaccinia virion, and establish the relationships and links between viral assembly, chemistry, proteomic structure, and replication cycle as well as further develop molecular-scale models of 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 development of vaccines, therapeutics, viral detection, attribution, and bioforensics technologies.

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.



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

FY12 Accomplishments and Results

In FY12 we performed all proposed tasks. Specifically, we (1) completed atomic force microscopy and initiated nanometer-scale secondary-ion mass spectrometry characterization of subviral structures of the wild-type vaccinia virus, (2) initiated the same experimental characterization of virion and subviral structures of selected mutants representing phenotypic classes with selected defective replication properties, (3) completed characterization of formulation attributes of the vaccinia virus manufactured using selected processing procedures, and (4) developed methods to label DNA in virions and conducted analysis of labeled virions and substructures using atomic force microscopy and nanoscale secondary-ion mass spectrometry.

Proposed Work for FY13

In FY13 we will (1) complete characterization of virion and subviral structures of selected mutants, (2) complete correlation of topographical structures of vaccinia virion with selected specific viral gene products, (3) compile data on the functional repertoire of selected key vaccinia proteins, and (4) model the detailed architecture of vaccinia virion and the roles played by selected individual viral genes in the viral assembly and replication cycle.

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

Matthew Coleman (11-ERD-037)

Abstract

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

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

Mission Relevance

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.

FY12 Accomplishments and Results

We (1) collaborated with the University of California, Irvine to produce unique, single-chain antibodies against the secretion III protein LcrV; (2) selected the best LcrV-binding recombinant antibodies and characterized these single-chain antibodies with protein microarrays and fluorescence correlation spectroscopy analysis, unexpectedly finding preliminary binding affinities to be greater than 1 μM; (3) engineered and produced additional proteins, such as endolysin and phospholipase A, for additional screening and characterization; and (4) demonstrated the full functionality of the fluorescence correlation spectroscopy system we developed based on its ability to measure protein–protein interactions for the LcrV protein.

Proposed Work for FY13

Having demonstrated in FY12 the process for selecting, purifying, and analyzing single-chain antibodies, in FY13 we will extend this technology to characterize complex interactions. Specifically, we will (1) characterize protein interactions between LcrV and the *Yersinia pestis* outer membrane protein YopB, (2) test the ability of our synthetically generated antibodies to disrupt their interactions, (3) perform engineering and analysis of additional recombinant antibodies for proteins such as phospholipase A, and (4) perform single-molecule characterization of LcrV–phospholipase A interactions using labeled affinity reagents and fluorescence correlation spectroscopy.

Publications

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

Targeted Drug Delivery for Treating Traumatic Bone Injury

Nicole Collette (11-ERD-060)

Abstract

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

We will deliver a fully functional, tissue-specific drug system tested in vivo with synergistic bone-healing therapy that is ready for clinical settings. This treatment will utilize currently approved therapies in a novel way and will also enable development of novel methods for bone repair and regeneration. This therapy system will provide much-needed effective, nonsurgical treatments for traumatic bone injuries and other difficult fractures. In addition to defense applications, the resulting bone therapy

system will also have applications in biodefense and public medicine in general, especially in the areas of osteoporotic fractures and fractures complicated by diseases such as diabetes.

Mission Relevance

This work has applications that address Department of Defense research needs and supports LLNL's missions in national security and biodefense by providing a solid foundation—including materials design and novel approaches—for future research in the areas of infection treatment and prevention, to which this project's focus of healing fractures without infection can be extended.

FY12 Accomplishments and Results

In FY12 we (1) began in vivo work with a surgical mouse model of fracture repair; (2) determined, in our in vitro work, that nanolipid particles facilitate innate immune response and that the particles are rapidly taken up by target cells, are not cytotoxic, and have a short half-life in serum; (3) optimized our surgical technique without any animal loss or infection; (4) began analyzing several animal fracture data sets from experimentation with and without nanolipid particles; and (5) further developed a diabetic model of "difficult" fracture repair, because normal animals heal fractures well without intervention. This will allow us to test potential therapeutics in the context of a fracture that does not heal well. In addition, we have uncovered a mutant strain of mouse that does not heal fractures well, and we are in the process of characterizing this model, which may represent a novel model for uncovering the mechanisms of fracture repair.

Proposed Work for FY13

We will (1) continue to induce fractures and dose animals with optimized nanolipid particles to determine whether the particles play a role in enhancing innate immune response during the initial phases of fracture repair; (2) examine mutant mouse lines for novel targets for fracture repair, utilizing bone metabolism mutants and mouse genetic models of diabetes as the model of a difficult fracture; (3) use our in vivo studies to examine the efficacy of therapeutic particles by histology, molecular biology, and other techniques and thereby advance our analytical capabilities; and (4) publish our results.

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.

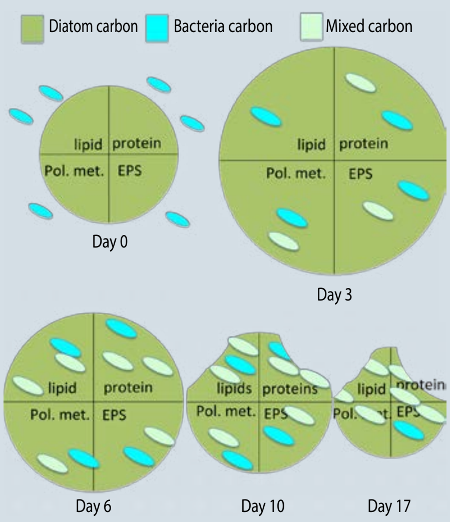
We will produce direct measurements of carbon fluxes into and out of degrading particles and identify the microorganisms responsible for these biogeochemical activities. In addition, we expect to elucidate the currently unknown effect of climate change on these processes. These results will increase 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.

FY12 Accomplishments and Results

In FY12 we (1) performed rRNA sequencing, probe design, and nanometer-scale secondary-ion mass spectrometry analysis of stable-isotope probing microarrays from our experiments on the microbial breakdown of marine particles with no climate effects; (2) incorporated the microarray data with isotope-ratio mass spectrometry data collected from particles labeled with stable isotopes; and (3) carried out a climate-effect experiment and collected samples for isotope-ratio mass spectrometry analysis at the University of California at Davis.



Conceptual understanding of marine particle degradation over time through mass spectrometric analysis of different molecular fractions (EPS = extracellular polysaccharides and Pol. met. = polar metabolites). The area of the particles corresponds roughly to particle mass over time.

Proposed Work for FY13

We will (1) conduct analyses of data from our additional climate-effect experiment conducted in FY12, including rRNA sequencing, probe design, and nanometer-scale secondary-ion mass spectrometry analysis of stable-isotope probing microarrays; (2) perform another climate-effect experiment for further analyses with isotope-ratio mass spectrometry; and (3) investigate the possibility of using probing microarrays for the direct analysis of marine particles labeled with stable isotopes, rather than using unlabeled particles and adding labeled substrates, with the goal of directly quantifying the carbon and nitrogen drawdown potential of individual bacterial species attached to particles.

Publications

Mayali, X., P. K. Weber, and J. Pett-Ridge, 2012. "Taxon-specific C:N relative use efficiency for amino acids in an estuarine community." *FEMS Microbiol. Ecol.* 2012 Aug 30, 10.1111/1574-6941.12000. LLNL-JRNL-553798.

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

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

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

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

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

Michael King (11-LW-009)

Abstract

Our objective is to explore the mechanisms by which explosive blasts cause traumatic brain injury (TBI). Unlike civilian injuries, which are usually caused by impacts and have been extensively studied, military TBI is 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.

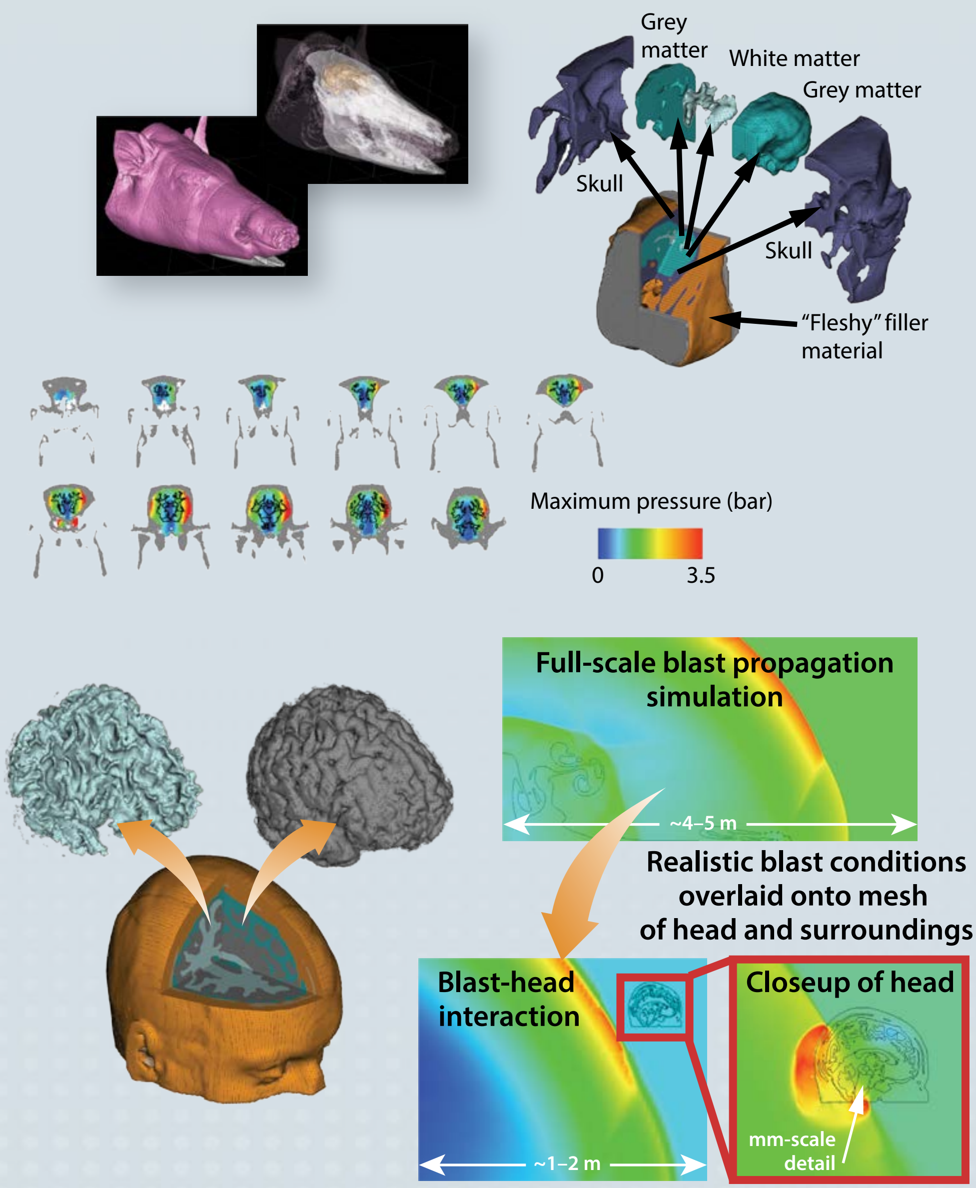
We expect to identify mechanisms by which explosive blasts damage the brain. This could allow development of more effective protective equipment for reducing traumatic brain injury and saving lives. Understanding damage mechanisms also has implications for rapid diagnosis in the field and could lead 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, multiscale 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.

FY12 Accomplishments and Results

We (1) compared our high-fidelity models of our pig experiments from FY11 to results from our University of Pennsylvania collaborators, determining that areas of the brain with damage generally correspond to shear strain from rotation; (2) expanded our



High-fidelity computational models used to study traumatic brain injury. Model of pig head along with images of the pressure that evolves during coronal rotation are shown at top. Model of a human brain with white and grey matter along with simulations of blast pressure acting upon it are shown on bottom.

simulation capabilities to a high-fidelity model of the human head in a realistic blast scenario; and (3) determined, in collaboration with a team led by Boston University Medical, that blast TBI may be caused by blast-wind-induced rotation of the head. In summary, this project conducted groundbreaking research into the causes of military TBI, especially from blasts, and produced modeling and simulation tools that are widely applicable to the future study of TBI and many other topics relevant to LLNL's missions. A next step would be to continue collaborating with Boston University Medical on the study of military TBI causes and prevention with support from the Department of Defense. Our work is poised to produce revolutionary new strategies for diagnosing, treating, and protecting against brain injury.

Proposed Work for FY13

Goldstein, L. E., et al., 2012. "Chronic traumatic encephalopathy in blast-exposed military veterans and a blast neurotrauma mouse model." *Sci. Transl. Med*. **4**(134), 16. LLNL-JRNL-531871.

In Vivo Modulation of MicroRNA for Cancer Therapy

Nicholas Fischer (11-LW-015)

Abstract

The goal of this proposal is to develop cancer therapies based on targeted modulation of microRNA (miRNA) in vivo. These are important regulatory molecules in living systems, and aberrant regulation of cellular miRNAs has been linked to cancer. In vitro modulation of miRNA in cancer cells has demonstrated their potential as therapeutic targets. However, targeted in vivo delivery of miRNA modulators is currently a major obstacle to using this modality. We will develop nanolipoprotein particles (NLPs) as universal, in vivo delivery platforms incorporating both miRNA modulators and tumor-targeting moieties. The delivery and efficacy of the miRNA 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 miRNA modulators to specific tissues using NLP 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 miRNA modulation as a possible therapy is precluded by the difficulty of its in vivo delivery. As such, the delivery of miRNA modulators using cancer-targeting nanolipoproteins is expected to alleviate this bottleneck, effectively accelerating the clinical application of this therapeutic technology.

Mission Relevance

In addition to the fact that the proposed technology and approach proposed in this study can have significant implications for cancer treatment, this project may contribute to national security and the Laboratory's strategic mission thrust in biosecurity by providing a potential therapeutic modality to address disease and illness caused by natural or engineered pathogens.

FY12 Accomplishments and Results

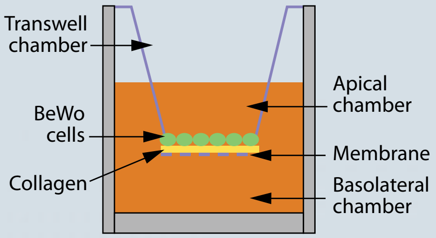
In FY12 we met two major milestones: demonstrating the enhanced internalization of targeted NLPs in cancer cells, using folate as the small-molecule-targeting agent, and successfully incorporating miRNA modulators into our NLP platform. These results highlight the versatility of the NLP platform to accommodate myriad cargo molecules of disparate chemistries. Although studies exploring the enhanced efficacy of targeted miRNA modulators in vitro were not successful—targeted NLPs did not decrease miRNA levels in treated cells—our detailed studies of the stability of NLPs in biological milieu demonstrated that NLPs are more ideally suited to in vivo applications requiring shorter therapeutic windows, rather than the long incubation times required for miRNA modulation. This was demonstrated by successful delivery of NLPs bearing immune therapeutics directly to tumors, resulting in a therapeutically relevant immune response. In summary, this project provided valuable insight into the preparation, characterization, and biological stability of the NLP platform and resulted in novel NLP constructs and innovative analytical techniques for NLP characterization and component quantification. The next step is to research the use of NLPs as therapeutic delivery platforms.

Publications

Fischer, N. O., 2012. *In vivo modulation of microRNA for cancer therapy*. LLNL-TR-604072.

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

Miranda Falso (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

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

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

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 to protect our water supply and public safety, in support of the Laboratory's mission in biosecurity.

FY12 Accomplishments and Results

In FY12 we (1) completed assays on the transfer of environmentally relevant levels of TCC labeled with carbon-14 in the BeWo b30 line of placental barrier cells—the sensitivity of accelerator mass spectrometry was needed to measure the low levels (<10%) that were transferred, (2) investigated the dosing of mice with TCC labeled with carbon-14 in their drinking water and determined that at least 37% and up to 76% of the dose was lost to adsorption by components in the bottle, and (3) generated customized water bottles to minimize adsorption of the TCC and allow for the animal experiments to be conducted.

Proposed Work for FY13

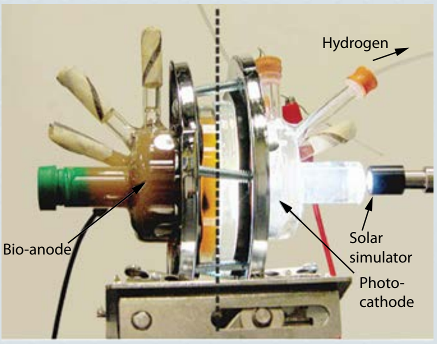
In FY13 we plan to (1) examine the transfer of three low doses of TCC from mother to offspring both in utero and through lactation, (2) examine the developmental outcomes in offspring of TCC-exposed mothers, and (3) examine the ability of exposed offspring to reproduce. We expect to see low levels of TCC transfer in utero and greater levels of TCC to transfer through lactation. These low levels will likely impact development and possibly interfere with reproduction.

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



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

promising of these fuels and is seen as an important future energy source. We propose to implement a simple and relatively straightforward strategy for hydrogen production by photosynthetic microorganisms using sunlight, sulfur- or iron-based inorganic substrates, and CO_2 as the feedstock. Carefully selected microorganisms with bioengineered beneficial traits will act as the biocatalysts 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 would be of great significance, not only eliminating the economic and agricultural burden of producing and growing the organic substrates, but also greatly reducing greenhouse gas emissions commonly associated with their production and usage. The use of sulfur- or iron-based inorganic substrates is likely to make hydrogen production more economically viable, because iron is both cheap and abundant and sulfur-containing iron minerals are moderately common. Our ultimate goal is to lead the way towards economically viable biological hydrogen production on a scale sufficient to supply a substantial portion of the hydrogen fuel market.

Mission Relevance

This project will enable a multidisciplinary group of scientists at LLNL to collaborate in achieving a basic understanding of critical microbial 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.

FY12 Accomplishments and Results

In FY12 we (1) experimentally demonstrated that hydrogen production by *Rhodopseudomonas palustris*, a bacteria efficient in the biodegradation of hydrocarbons, grown on thiosulfate is mainly through nitrogenases activity, with little or no contribution from hydrogenases; (2) verified that within the three nitrogenases present in *R. palustris*, molybdenum nitrogenase is expressed the most during growth on thiosulfate, suggesting its dominant role in hydrogen production; (3) continued curating the constraint-based model of *R. palustris* with new phenotypic empirical data, with a primary focus on balancing the redox environment of the cell, thus establishing the capability to identify mechanisms responsible for the differences between measured and model-predicted hydrogen production levels, because accurate implementation of these processes is essential for a thorough examination of cellular redox processes; and (4) began using our genome-scale model of *R. palustris* to examine the system's metabolic capabilities after consumption of different carbon sources coupled to various oxidizing agents.

Proposed Work for FY13

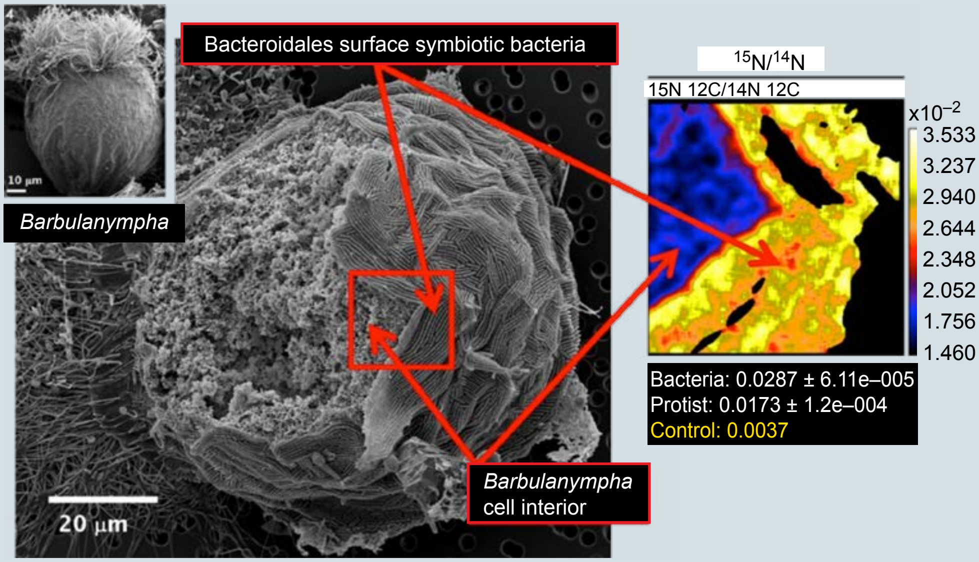
In FY13 we will test the contribution of individual hydrogenases and nitrogenases in hydrogen production in *R. palustris* when using thiosulfate as the electron donor, through direct mutation or activity measurement and with quantitative real-time polymerase chain reaction to amplify specific DNA sequences. Our goal is to complete the steps necessary to identify the most promising enzyme combination and electron-transfer pathways for efficiently producing hydrogen.

Protist Power: Deconstructing Termite Conversion of Wood to Biofuels

Peter Weber (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



The first direct isotopic evidence for nitrogen fixation by a termite hindgut microbe, in an intact Barbulanympha protist cell (from the Cryptocercus punctulatus hindgut) with a covering of the symbiotic bacteria Bacteroidales (left inset). Damage to the Barbulanympha cell during fixation reveals the cell interior and Bacteroidales surface symbiotic bacteria (center). The red box indicates the area in the nanoscale secondary ion mass spectrometry micrograph (right), which shows ratios of nitrogen-15 (^{15}N) to ^{14}N indicating higher ^{15}N enrichment in Bacteroidales surface bacteria than in the protist cell interior. The role of Bacteroidales in nitrogen fixation was corroborated by genomic evidence revealing several genes associated with nitrogen fixation. The control value of 0.0037 represents termites not exposed to ^{15}N-enriched air.

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 greatly increasing U.S. biofuels production capability. The goal of our research is to provide a scientific 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 the Livermore mission to reduce or counter threats to national and global security.

FY12 Accomplishments and Results

In FY12 we (1) completed isotopic imaging using nanometer-scale secondary ion mass spectrometry, finding evidence that in the gut of the lower termite *Paraneotermes simplicicornis*, the protist *Oxymonas dimorpha* was engulfing cellulose and providing the byproducts to its symbiotic bacteria; (2) found another protist–bacteria symbiotic pair in which the bacteria appear to be digesting the cellulose and sharing the byproducts with the protist—possibly the first observation of bacterial cellulose in "lower termites," a function solely performed by bacteria in "higher termites;" (3) demonstrated that in the wood-eating cockroach *Cryptocercus*, an insect closely related to lower termites, in situ nitrogen fixation is performed by a symbiotic bacterium, which shared the fixed nitrogen with its protist host; and (4) presented our results at international microbiology meetings. The work performed under this project has been selected for further support by the DOE Genome Sciences Program.

Publications

Carpenter, K. J., et al. 2011. *Protist–bacterial symbioses and carbon flux in the hindgut of a lower termite as inferred by stable isotopic labeling and high resolution SEM, TEM, and NanoSIMS analysis*. 6th European Congress of Protistology (ECOP 2011), Hesse, Germany, July 25–29, 2011. LLNL-ABS-557555.

Carpenter, K. J., et al. 2012. *Protist–bacterial symbioses and nitrogen fixation in the hindgut of a lower termite: Stable isotopic tracers and high resolution SEM, TEM, and NanoSIMS* *analysis*. 15th European Microscopy Congress (ECM 2012), Manchester, U.K., Sept. 16–21, 2012. LLNL-ABS-576272.

Carpenter, K. J., et al., 2011. *Protist power: Deconstructing termite microbial symbiotic conversion of wood to biofuels and H_2*. 6th European Congress of Protistology, Berlin, Germany, July 22–29, 2011. LLNL-PRES-491239.

Carpenter, K. J., et al., 2013. *SEM, TEM, and NanoSIMS imaging of lower termite hindgut microbes: Evidence for carbon transfer from an oxymonad protist to its bacterial symbionts*. LLNL-JRNL-557111-DRAFT.

Solar-Powered Microbial Electrolysis Cells for Hydrogen Production

Fang Qian (11-LW-054)

Abstract
The objective of this project is to develop a self-biased, solar-driven microbial electrolysis cell that uses solar energy to electro-hydrogenate organic waste into hydrogen gas. Compared to existing fermentation approaches based completely on microbial catalysis, our unique microbial electrolysis cell design will, for the first time, couple a semiconductor photoelectrode 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. Finally, we will scale up the solar microbial electrolysis cell system for integration into existing wastewater treatment pipelines for onsite waste-to-hydrogen conversion, in collaboration with the water reclamation plant in Livermore, California.

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 Laboratory's 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.

FY12 Accomplishments and Results

In FY12 we (1) used new semiconductor nanometer-scale materials that are chemically stable and have good solar-to-electron conversion capability, such as titanium dioxide and iron oxide nanorods, to serve as photo-electrodes in collaboration with the University of California, Santa Cruz; (2) established new, reconfigurable solar microbial electrolysis cell devices based on polydimethyl siloxane; (3) achieved hydrogen generation using a microbial fuel cell and photo-electrochemical assembly; and (4) achieved simultaneous wastewater treatment and electricity generation using microbial fuel cell technology in customized milliliter-scale devices. The successful completion of this project enabled a new potential technology using microbes, wastewater, and solar light to produce clean, renewable energy sources such as hydrogen.

Publications

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Wang, H., et al., 2012. *Self-biased and sustainable microbial electrohydrogenesis*. LLNL-JRNL-580052.

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Computational Advancements in Countermeasures for Emerging Bio-Threats

Felice Lightstone (12-SI-004)

Abstract

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

The successful outcome of this project will result in a new state-of-the-art capability at LLNL that will drastically accelerate medical countermeasure development and position LLNL as a world-class facility in computational pharmacology. This new capability will be the first of its kind to predict the pharmacokinetics and adverse side effects of a drug candidate from its chemical structure, using highly accurate physics-based methods coupled with informatics. Once accurate predictions are made, the drug development process can be shortened, and more drug candidates will succeed in clinical trials. Accurately predicting the pharmacokinetics of a drug candidate will dramatically reduce the time to Federal Drug Administration approval and have a profound impact on therapeutics for human health.

Mission Relevance

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

FY12 Accomplishments and Results

In the first year of our project we have met or exceeded the milestones to start developing each of the modules of a prototypic system that will ultimately predict pharmacokinetic properties and adverse side effects of drug candidates. Specifically, we (1) developed a multi-compartment physiologically based pharmacokinetic model for a drug, (2) created a model to predict relative probabilities of adverse drug reactions based on the chemical structure of a drug candidate and the biological pathways affected, (3) constructed 100% of the structurally available proteins for off-target interaction in all identified pathways, and (4) calculated reaction coordinates of drug compounds in cytochrome P450 enzymes and determined static membrane permeability parameters in homogeneous lipids.

Proposed Work for FY13

In FY13 we propose to continue developing each of the modules that will ultimately predict pharmacokinetic properties and adverse side effects of drug candidates. We will (1) identify parameters in the physiologically based pharmacokinetic model that could be determined through calculated methods, (2) create a network-based model that links known drug compounds to specific adverse side effects, (3) automate the protocol to better evaluate off-target interactions by drug-like molecules, and (4) compute kinetic parameters for small-molecule binding to proteins and membrane permeability.

Publications

Zhang, X. ,S. E. Wong, and F. C. Lightstone, 2012. "Message passing interface and multithreading hybrid for parallel molecular docking of large databases on petascale high performance computing machines." *J. Comput. Chem*. 2013 Jan 23. doi: 10.1002/jcc.23214. LLNL-JRNL-568309.

Molecular Movies with Dynamical Imaging of Biomolecular Interactions

Matthias Frank (12-ERD-031)

Abstract

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

A major bottleneck in structural biology is that many of the proteins performing critical cellular functions such as nutrient uptake, signal transduction, photosynthesis, and secretion are membrane proteins, whose structure cannot be determined by traditional x-ray crystallography. Consequently, most membrane protein structures remain unknown. We expect to help demonstrate the potential of coherent x-ray diffraction imaging to enable structure determination of membrane proteins and other macromolecules or complexes. If successful, this work will drastically expand proof-of-principle demonstration of this imaging technology, provide new sample preparation and delivery methods, and result in novel, high-impact structures and dynamics of protein complexes with near-

atomic resolution. This work would greatly aid our understanding of protein function applicable to a wide range of fields.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12, we (1) identified a number of protein and lipoprotein samples for coherent x-ray diffraction imaging, including several protein virulence factors from pathogenic bacteria and nanolipoprotein particles, then produced nanometer- and micrometer-scale crystals from these samples and conducted experiments at the Linac Coherent Light Source, Japan's Spring-8 Angstrom Compact Free Electron Laser, the Stanford Synchrotron Radiation Light Source, and the Advanced Photon Source at Argonne National Laboratory; (2) made progress in our novel sample delivery techniques—rather than developing a rapidly spinning sample disk prototype, we developed a somewhat simpler fixed-target approach that utilizes thin substrates moved by linear motors; and (3) demonstrated, in experiments at the Linac Coherent Light Source, that good, high-resolution diffraction patterns can be obtained from protein microcrystals and nanocrystals supported by such substrates at reasonably high data-acquisition rates, with our calculations showing a potential for a hundredfold reduction in sample volume required compared to the currently used continuous liquid jet injection.

Proposed Work for FY13

We will (1) continue our work on nanometer-scale crystallization of recombinant proteins and expressing membrane proteins in lipid cubic phase using co-translation, (2) produce and prepare low- and high-density lipoprotein particles and nanometer-scale crystals in solution for spraying with an aerodynamic lens, (3) continue to improve and demonstrate our fixed-target sample-delivery system and perform forward modeling of expected signal and noise from protein samples—such as nanometer-scale crystals or two-dimensional-arrays—embedded in lipid membranes held by this system, and (4) design pump–probe experiments that can be used to produce molecular movies.

Publications

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

Frank, M., 2012. *Study of the structure and function of a novel bacterial virulence factor isolated from* Francisella tularensis. LLNL-POST-568415-DRAFT.

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

Modulating Cellular Autophagy as an Alternative Line of Defense against Bacterial Pathogens

Catherine Lacayo (12-ERD-049)

Abstract

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

We expect to create a comprehensive and quantitative model of autophagy during bacterial infection and demonstrate that a pharmaco-modulatory approach can effect significant killing of invading bacteria by autophagy. In addition, we expect to determine the efficiency of autophagy in the degradation of invading bacteria by non-phagocytic cells—which are not involved in directly targeting and killing invading bacteria—and understand how autophagy affects survival of the host during infection. These questions have important implications when considering that all cells in our bodies have the potential of clearing infection using their inherent autophagic machinery and could use this mechanism to survive, instead of die, as a means to clear infection.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) developed a cutting-edge live-cell imaging system to spatiotemporally monitor infected living cells in vitro and visualize autophagy by microscopy; (2) cultured live host cells for days using a microfluidics platform; (3) fluorescently labeled bacteria and used them to infect host cells; (4) transfected the host cells to express fluorescently labeled proteins serving as autophagy markers; (5) visualized the process of infection in live cells with our new imaging system; (6) improved the efficiency and cost-effectiveness of fabricating microfluidic chambers to culture cells by successfully adopting a commercially available microfluidic perfusion system with environmental controls; and (7) began the process of integrating imaging of bacteria with autophagy markers to track and quantify the progression of autophagy during infection.

Proposed Work for FY13

In FY13 we will refine our analytical tools to generate a quantitative, dynamic view of autophagy during infection, modulate autophagy using pharmacological agents, and measure bacterial survival. Specifically, we will (1) quantify the progression and extent of autophagy by mapping the three-dimensional co-localization of bacteria and organelles involved in autophagy as function of time, (2) perform assay of intracellular bacterial survival and use electron microscopy to confirm involvement of autophagic machinery, and (3) pharmacologically modulate autophagy and measure any resulting changes in co-localization of autophagy markers and in bacteria–host cell survival.

Carbon Nanometer-Scale Membrane Channels

Aleksandr Noy (12-ERD-073)

Abstract

Living cells depend upon the flow of molecules across membranes for essential processes such as sensing, signaling, and energy production. Yet the cell membrane presents a formidable barrier to the transport of these molecules because they cannot cross the membrane unaided. As a result, living systems have evolved highly efficient trans-membrane protein channels that rapidly and selectively transport ions and molecules and play a key role in nutrient uptake, osmotic regulation, signal transduction, muscle contraction, and hormone secretion. We propose to create the first artificial inorganic ion channel using short barrels of carbon nanometer-scale tubes. The inner channel of a carbon nanotube is narrow, hydrophobic, and very smooth, which has a remarkable similarity to the properties of natural biological pores. We plan to cut carbon nanotubes in short pieces that match the thickness of a lipid bilayer, insert the nanotube barrel into the lipid bilayer membrane and form a pore in it that permits ion transport across the bilayer, and use chemical modification to alter channel selectivity. Creating a functional abiotic mimic for these protein channels can produce new therapeutic agents, biosensors, and pore-forming antibiotic agents, as well as a versatile model system for studying design rules for transport efficiency and selectivity in membrane channels.

We expect to demonstrate a functional scaffold of a membrane channel that replicates the membrane affinity and transport properties of biological ion channels. We propose to build a family of transporters that will be based on a common structural element—a carbon nanotube membrane channel. A short segment of a cut carbon nanotube will span a membrane and form a pore that mimics a biological ion channel. We will characterize transport efficiency and selectivity of these structures, as well as demonstrate specific targeting of these ion channels to bacterial membranes. The project also aims to characterize initial antibiotic activity of these structures using model bacterial systems.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) demonstrated our cutting protocols by producing small carbon nanotube dispersions and characterized them with atomic force microscopy, (2) demonstrated that our new procedure can produce carbon nanotube fragments in the size regime greater than 10 nm, (3) tested the extraction of carbon nanotube membrane channels using partitioning into a lipid vesicle and identified issues that will need to be addressed in FY13, and (4) started preliminary transport characterization measurements to demonstrate the passage of ions and protons through the nanotube pore, obtaining initial evidence of fast passage of protons and monovalent cations through the carbon nanotube pore in lipid bilayers, which positions us very well for performing transport measurements in FY13.

Proposed Work for FY13

In FY13 we will (1) continue work on reliable cutting protocols that produce dispersions with at least 10% of the carbon nanotubes smaller than 10 nm, (2) demonstrate high-yield separation of carbon nanotube membrane channels that isolate nanotube fragments greater than 10 nm, (3) refine extraction of carbon nanotube membrane channels using partitioning into a lipid vesicle and determine the efficiency of this procedure, and (4) perform initial transport measurements on the membrane channels.

Publications

Noy, A., 2012. *Kinetic model of gas transport in carbon nanotube channels*. LLNL-JRNL-574053.

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

Vanessa Tolosa (12-LW-008)

Abstract

In 2009, suicide was the 10th leading cause of death in the U.S., with veterans accounting for 20% of all such deaths each year. Suicide rates among active-duty military personnel reached record highs in 2010, and the rate among 18- to 29-year-old veterans increased by 26% between 2005 and 2007 alone. A majority of suicide cases are linked to major depressive disorder (MDD), the underlying causes of which are largely a mystery.

Consequently, many treatments remain ineffective or inefficient, and 20% of all MDD sufferers are deemed resistant to treatment. Deep brain stimulation has emerged as a promising tool to combat MDD, although its mechanisms are unknown and its parameters not optimal. We propose to develop a comprehensive method for treating and studying MDD and, in so doing, advance our understanding of both MDD and deep brain stimulation. We will combine a unique animal behavior model for depression with a novel multifunctional device to determine more-effective deep brain stimulation and pharmacological treatments as well as shed light on the pathology of MDD and other anxiety disorders.

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

Mission Relevance

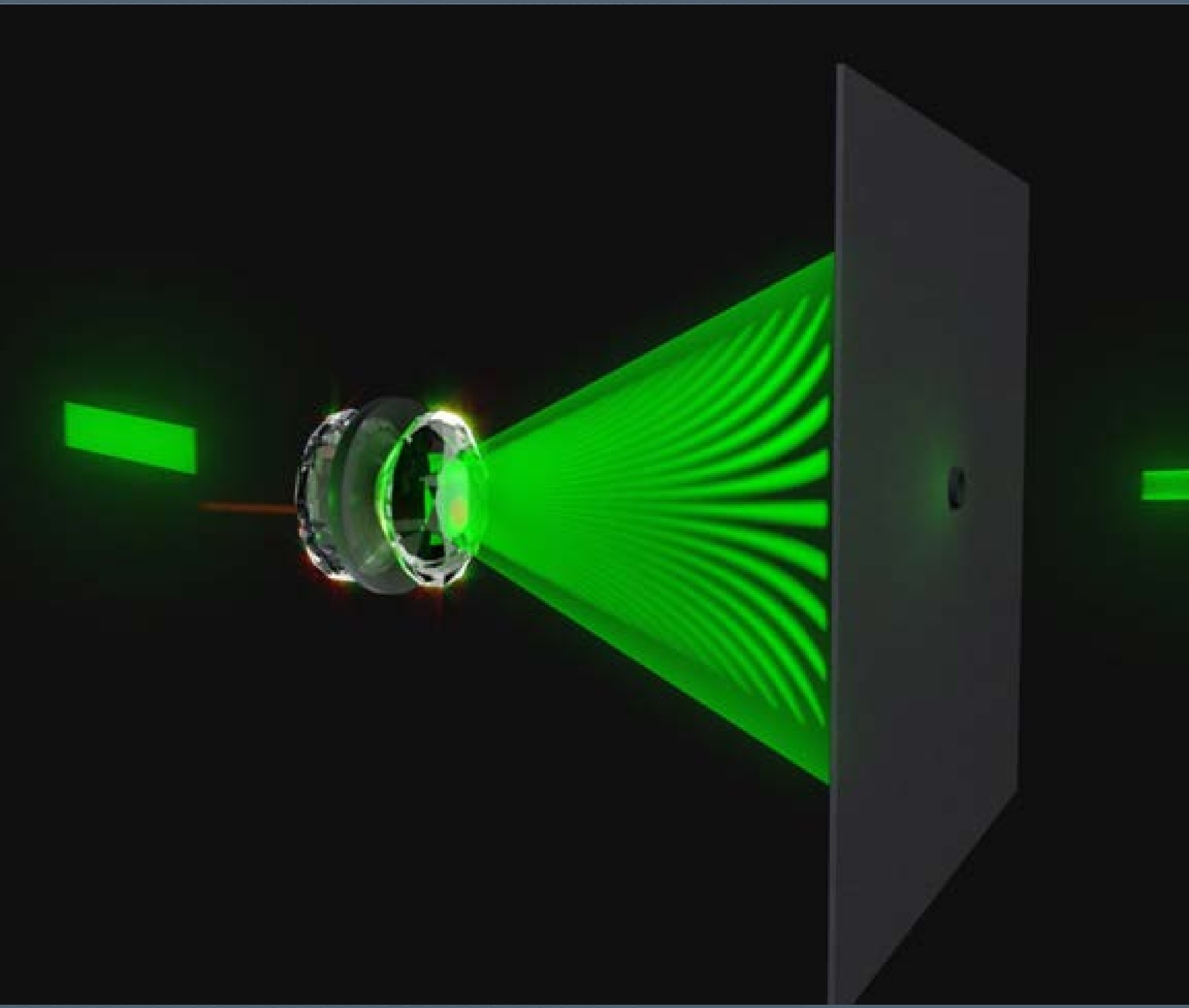
This project is directed toward improving military readiness in support of the Laboratory's national security mission. The Congressionally Directed Medical Research Program of the Department of Defense specifically identifies development of methods that will lead to improved prevention, detection, and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

FY12 Accomplishments and Results

In FY12 we (1) fabricated an interface, consisting of an electrode array and a micro-fluidic channel, to comprehensively study and treat MDD; (2) began developing and incorporating a chemical sensor to investigate the neurochemical basis of depression; and (3) tested and characterized each mode of our multifunctional array in preparation for eventual experimental use of such an array.

Proposed Work for FY13

In FY13 we will (1) test the electrical and chemical stimulation and recording functionality of the device in vitro, (2) begin in vivo studies using a behavioral rat model to investigate specific brain regions and receptors involved in deep brain stimulation for depression, and (3) modify our design and fabricate a new set of devices based on in vitro and in vivo data.



Laboratory Directed Research and Development

FY2012

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.

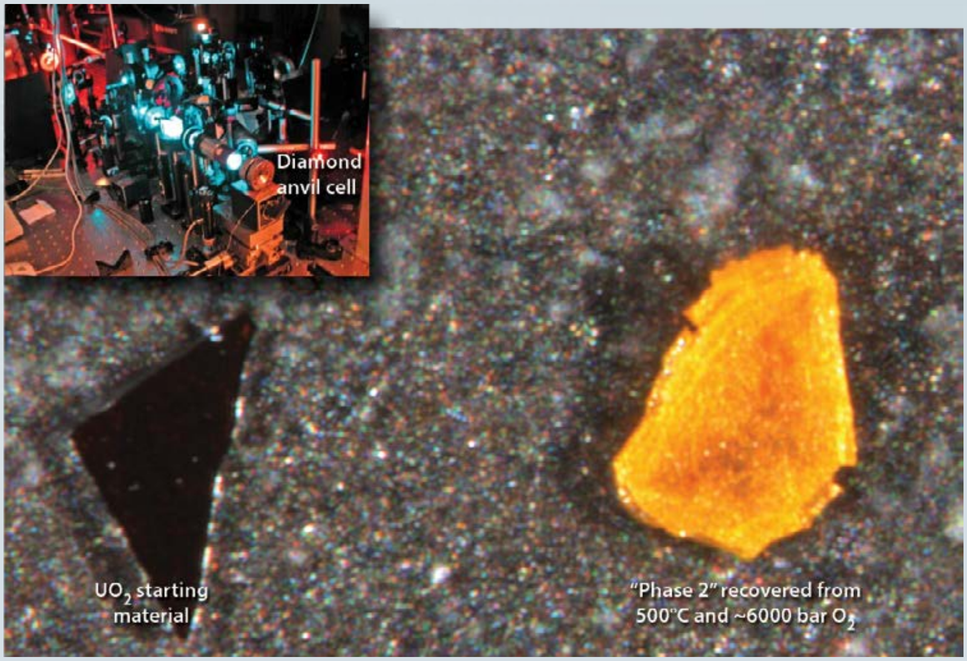
We expect to achieve breakthroughs in experimental science to increase the speed and fidelity of nuclear forensic analysis by (1) developing resonance ionization mass spectrometry to provide isotope-specific, definitive information on fuel composition in post-detonation scenarios within hours, without chemical separation; (2) developing a cavity ion source to increase mass spectrometry sensitivity by a factor of 10 to 25; (3) developing accelerator mass spectrometry to measure activation products at ultralow concentrations; (4) applying 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) characterizing nanometer-scale particles in fallout to provide the "ground truth" to evaluate new experimental techniques.

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.

FY12 Accomplishments and Results

In FY12 we (1) continued optimization of methods for rapid analysis of actinide isotope ratios in nuclear materials using resonance ionization mass spectrometry and demonstrated accuracy exceeding 0.3%, as well as the analysis of uranium at low (microgram/gram) concentrations in fallout glasses from U.S. nuclear tests; (2) developed apparatuses to conduct closed-system oxygen isotope exchange experiments with uranium oxide and produce miniaturized fuel pellets at the 0.5-g scale with commercial-grade qualities; (3) characterized approximately 50 uranium oxide samples synthesized with common and novel chemical processing techniques and demonstrated the use of thermogravimetric analysis to distinguish precipitates of uranium for forensic applications, as well as demonstrated the ability of morphology to distinguish precipitates and process history for otherwise chemically identical



We examined oxidation of uranium oxide in oxygen at pressures up to 0.9 GPa and temperatures of 450°C. The image shows material recovered from the reaction cell. The dramatic color change accompanies transformation to a previously unreported form of uranium trioxide, having far fewer crystalline defects and exhibiting a pseudo-tetragonal structure.

uranium oxides; (4) developed a novel spark-discharge apparatus capable of heating uranium samples to temperatures of approximately 1 eV to simulate melting and condensation processes occurring in fallout, and performed initial discharge and recovery experiments; (5) modified the ion source and immersion lens to improve ability to measure low levels of transition metal activation products using Livermore's Center for Mass Spectroscopy; and (6) analyzed melt glass in fallout debris from U.S. low-yield, near-surface nuclear tests and demonstrated an extreme range of uranium-235 contents, micro-scale uranium-isotope heterogeneity within individual melt spherules, and the presence of abundant krypton and xenon gas, produced by fission, from which cooling times can be estimated. This research has led to significant advances in nuclear forensic science by demonstrating the use of resonance ionization mass spectrometry for uranium isotope measurements as well as determining that uranium oxide is a Mott insulator—that is, an insulator despite predictions of conventional band theory. We have also determined that device signatures are significantly concentrated in fallout melt droplets compared to bulk soil, and demonstrated that process history of uranium oxides is reflected in morphological characteristics. This project has led to significant new funding for the Laboratory in pre- and post-detonation nuclear forensics from federal sponsors including NNSA NA-22 and NA-24 and the Defense Threat Reduction Agency.

Publications

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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 nanometer-scale 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.

FY12 Accomplishments and Results

In the final year of the project we (1) completed a sample-preparation scheme for thin-sectioning soils; (2) designed an ideal way to conduct combined scanning transmission x-ray microscopy and nanoscale secondary-ion mass spectrometry for soil analysis; (3) completed—with the help of a Lawrence scholar Ph.D. student who took on this research as his thesis work—our third multifactorial synthetic rhizosphere incubation, which combines carbon-13 labeled Zinnia elegans plant cells, diffusible manganese(III) ligand complexes, and a native soil minerals matrix in a highly controlled experimental mimic of a plant root system; and (4) published our results, including an invited book chapter. The success of this project has enabled new understanding of the mechanisms and persistence of interactions between organic molecules and minerals, as well as a critical new scientific approach: combined scanning transmission x-ray microscopy and secondary-ion mass spectrometry imaging, which has received high praise in the scientific community. Support through a Genomic Science Program project on carbon cycling renewal is pending.

Publications

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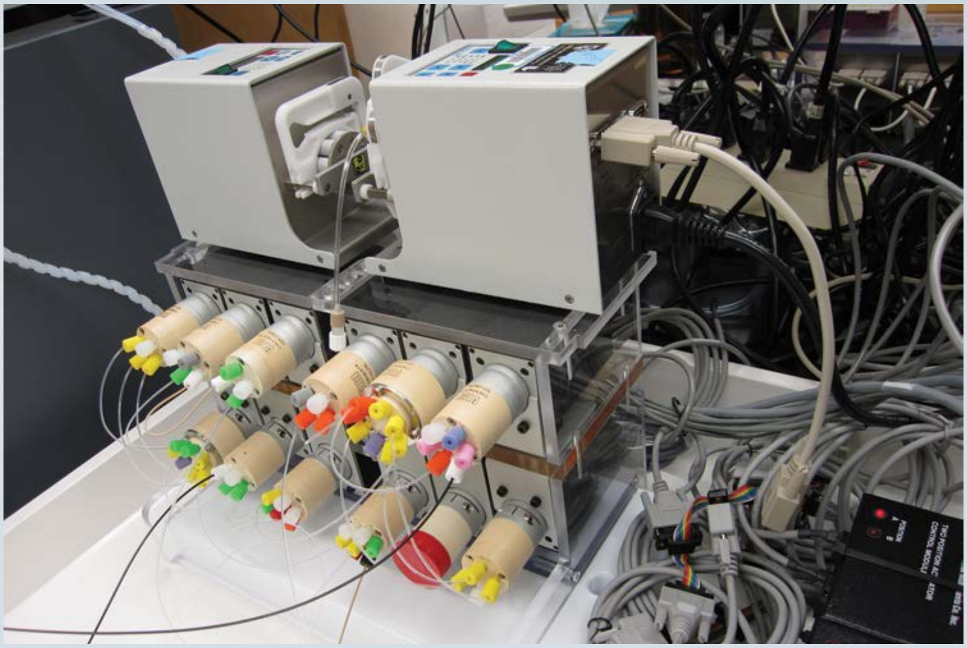
Fundamental Chemical Behavior of Superheavy Elements through Applications of Online Isotope Production and Automated Chemical Systems

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



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.

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

FY12 Accomplishments and Results

In FY12 we (1) began developing model chemical systems for group-14 elements in collaboration with the University of Nevada, Las Vegas; (2) identified modifications to our automated chemical system for chemistry at CAMS and behind the recoil transfer chamber at Texas A&M University; (3) designed and fabricated a chemistry interface chamber; (4) performed tests of our CAMS target chamber, performed required modifications, and tested it with and without beam on target foils; and (5) collaborated with Texas A&M University to design a chemistry experimental facility at their Cyclotron Institute, including developing chemical systems for initial element 104 experiments using the Texas A&M cyclotron for isotope production and Livermore's automated chemical system for analysis.

Proposed Work for FY13

In FY13 we propose to (1) work with our collaborators at GSI on the search for element 119; (2) work with Texas A&M to set up the new facilities, the recoil transfer chamber, and online chemistry capability; (3) perform initial chemistry experiments at Texas A&M using our automated chemistry system with heavy-ion irradiation; (4) continue developing chemistry for element 114 with the University of Nevada, Las Vegas; and (5) complete the setup of online isotope production capability at CAMS using the newly fabricated target and chemical interface chambers.

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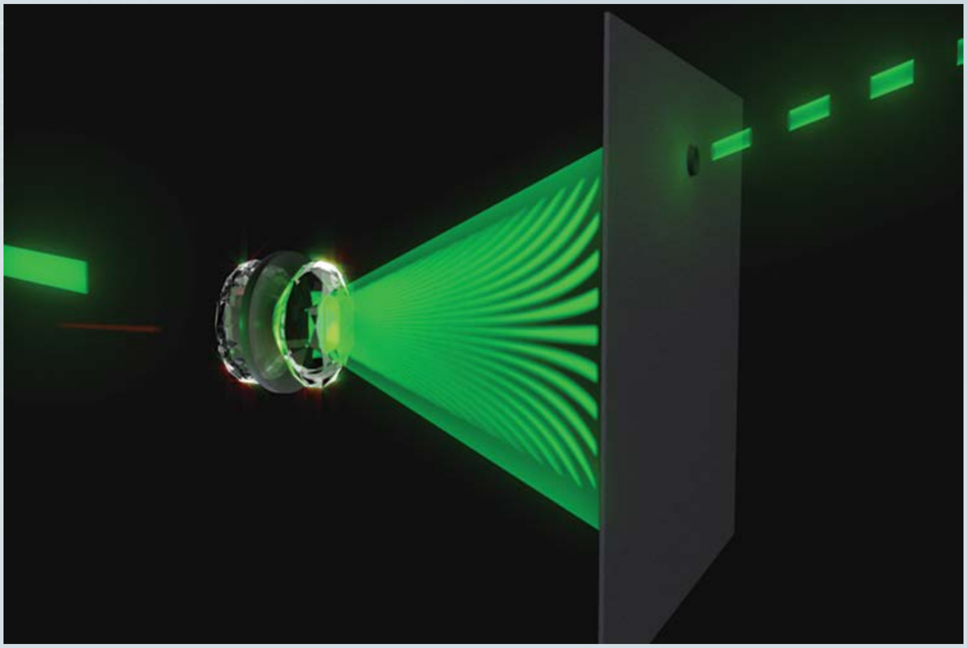
Detonation Performance of Improvised Explosives via Reactive Flow Simulations and Diamond Anvil Experiments

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



A schematic representation of sound speed measurements at high pressures in a diamond anvil cell using a photo-acoustic light-scattering technique.

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.

FY12 Accomplishments and Results

In FY12 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 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 FY13

In FY13 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

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

Tadashi Ogitsu (11-ERD-073)

Abstract

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

Successful completion of the project will provide detailed information on microscopic properties of the surfaces of semiconductor elements in groups III through V of the periodic table as well as general structural and dynamical properties of water–semiconductor interfaces. In addition, we aim to gain 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

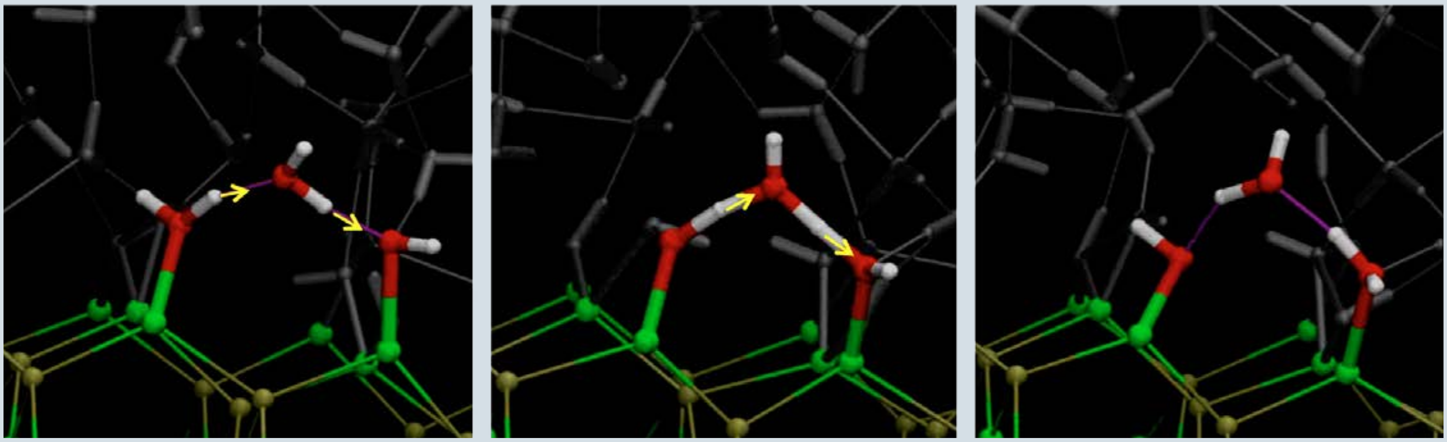
Development of efficient, environmentally responsible solar-to-chemical energy conversion has been identified as a strategic DOE goal for a secure, sustainable energy future. This project directly addresses this, as well as the Laboratory's mission to enhance the energy and environmental security of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

FY12 Accomplishments and Results

In FY12 we continued FY11 investigations on the effect of surface morphologies on surface proton transfer and how that might lead to water formation. Specifically, we (1) completed our study of the morphologies of surface oxygen and hydroxyl on indium phosphide and gallium phosphide, (2) performed simulations of nitrogen doping and the nitrogen K-edge x-ray emission spectrum of n-doped group III–V compounds, and (3) found evidence that the morphologies of gallium and indium phosphide surfaces can affect the hydrogen-bond network properties of interfacial water, which in turn govern proton transport parallel to the surface.

Proposed Work for FY13

In FY13 we will focus on studying the effect of nitrogen doping on group III–V elements using the surface and interface models we developed in FY12. We will then compare the results with experimental characterization of the electrode to understand



Water molecules adsorbed on an indium phosphide (001) surface were observed forming an ice-like hydrogen-bonding network, with frequent proton exchanges. The implications of this behavior in hydrogen evolution reactions and corrosion resistance are discussed in a recent paper published in the *Journal of Chemical Physics.*

the underlying mechanisms linking surface morphology, solar-to-hydrogen conversion efficiency, and corrosion resistance.

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., T. Ogitsu, and E. Schwegler, 2012. "Local structural models of complex oxygen- and hydroxyl-rich GaP/InP(001) surfaces." *J. Chem. Phys.* **136**, 064705. LLNL-JRNL-511456.

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Predicting Weapon Headspace Gas Atmosphere for Modeling Component Compatibility and Aging

Elizabeth Glascoe (12-ERD-046)

Abstract

As the nuclear stockpile ages and undergoes life-extension measures, concerns about material compatibility and aging arise. In a warhead, material incompatibilities and degradation can result in a loss of functionality. Moreover, these problems may appear after decades of apparent compatibility or after a change made as part of a life-extension program. We currently lack a science-based understanding of what constitutes the gas-phase signatures of such undesirable material changes that are detectable in systems surveillance. Our goal is to develop a novel capability for assessing material compatibilities with age—specifically, a model that simulates the reactive transport of volatile species through materials. The model will be based on fundamental physical and chemical properties of the materials and be versatile enough to apply to different geometries, sizes, and arrangements. Our novel approach of using a reactive transport code to predict material compatibilities could also have wide-ranging applications in military, aerospace, medicine, and other fields.

We will develop a novel methodology and capability to predict material compatibilities. Our computational model will simulate the transport and chemical reactions of volatile species and thereby predict the resulting constituents of headspace gas. To this end, we will characterize and quantify the fundamental physics of transport, sorption, and chemical-reaction kinetics and mechanisms, creating for the Stockpile Stewardship Program a novel method for assessing the long-term compatibility of warhead materials.

Mission Relevance

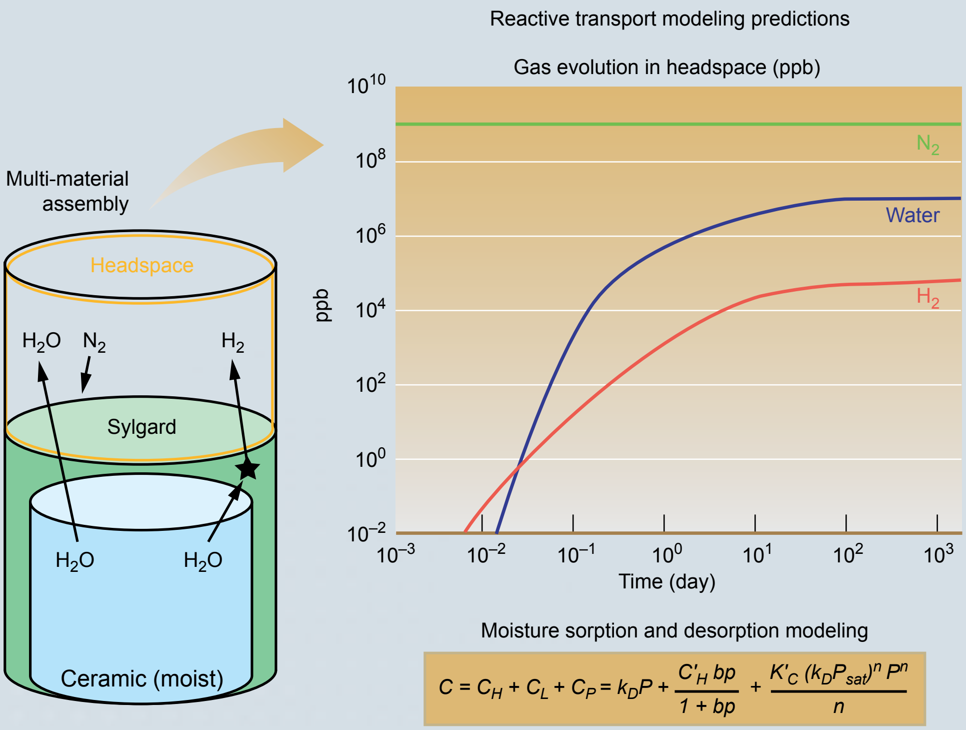
This project supports the Laboratory's stockpile stewardship mission by providing the capability to predict the chemical compatibility of materials over the long term, thereby enabling greater predictive foresight in selecting replacement materials for stockpile life-extension programs.

FY12 Accomplishments and Results

In FY12 we (1) generated data for a compatibility model for the diffusion and sorption of moisture through selected materials, (2) began development of the compatibility model using the reactive transport codes NUFT and ToughReact, (3) used sensitivity analysis and uncertainty quantification techniques to provide quantitative confidence in our predictions and prioritize experiments for validation and parameterization, (4) developed a small experiment for model validation, and (5) began to quantify the chemical kinetics necessary for our reactive transport modeling.

Proposed Work for FY13

In FY13 we plan to (1) continue characterizing the diffusion and sorption parameters for vapors in our selected materials, (2) continue development of our compatibility model in conjunction with validation experiments of various scales and complexities, (3) characterize the chemical kinetics of our selected materials with relevant volatile species, and (4) prepare and publish manuscripts on our material characterization and compatibility model in peer-reviewed journals and conference proceedings.



Using fundamental physics and chemistry parameters and reactive transport modeling, we are developing a capability to predict aging and chemical compatibility of multiple-material assemblies with possible military, aerospace, and national security applications.

Publications

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Lu, C., et al., 2012. *Modeling gas transport and reactions in polydimethysiloxane.* TOUGH Symp., Berkeley, CA, Sept. 17–19, 2012. LLNL-CONF-565272.

Equation of State of Polymers Under Extreme Conditions with Quantum Accuracy

Nir Goldman (12-ERD-052)

Abstract

Accurate modeling of materials containing the bonded elements carbon, hydrogen, and oxygen (C–H–O), such as polymers and low-density foams, is essential for National Ignition Facility (NIF) target capsule design and laser-driven ramp wave studies in which chemical reactivity can lead to unexpected relationships of pressure to density, or Hugoniot states, for a single shocked state. We will create a predictive capability for materials containing C–H–O that are accurate at temperatures greater than 1 eV and pressures greater than 10 Mbar for use in interpreting and designing experiments on NIF. This capability will include density-functional tight-binding parameters that will increase computational efficiency by several orders of magnitude over standard quantum codes, while achieving comparable accuracy. Our models will help to fully elucidate the equations of state and chemical processes that occur in these experiments.

We will develop a high-efficiency quantum model for simulating C–H–O materials at high temperatures and pressures to circumvent the critical issues of timescale and system length presented by standard quantum codes, while also achieving comparable accuracy. Our ultimate goal is to develop and constrain hydrodynamics-code simulations by efficiently and accurately estimating equation-of-state data and the chemical kinetic effects of shock compression and release behavior in bonded C–H–O materials. Our simulations will address critical needs for experiments on NIF by helping to interpret current and proposed ramp compression experiments using carbon-based foams, and by making predictions for future NIF capsule experimental design.

Mission Relevance

This project supports the Laboratory's missions in national and energy security by creating a predictive capability for essential materials in fusion ignition capsules used in stockpile stewardship and fusion energy research. This capability could also be extended to other fusion target materials such as diamond and beryllium, to strongly shocked energetic materials used in national security efforts, and to planetary fluids such as hydrogen and water, in support of LLNL's basic science mission.

FY12 Accomplishments and Results

In FY12 we (1) conducted quantum simulations of a polystyrene-like composite at conditions relevant to NIF capsule implosion—results exhibit excellent agreement with both previous experimental and quantum simulation results; (2) observed extremely fast chemical reactivity in our simulations, contrary to what has been inferred from recent experiments; and (3) began creation of new density-functional tight-binding interaction potentials that include new expressions for repulsive energy as well as new and larger basis sets for the carbon atoms. Our initial results so far have yielded good agreement with previous quantum simulations and experiments.

Proposed Work for FY13

In FY13 we will (1) continue development of our density-functional tight-binding parameter set through comparison to quantum molecular dynamics simulations of different C–H–O bonded systems under dynamic strain—this will include further determination of repulsion energy terms and basis sets for our systems and conditions of interest, (2) begin to extend our density-functional tight-binding simulations of shock-compressed C–H–O materials to nanosecond timescales—we will validate our simulations through comparison to available experimental results, and (3) begin to use our density-functional tight-binding models to predict equation-of-state data and chemical kinetics during isentropic expansion of materials after shock compression. Methods that include quantum nuclear vibrational effects in temperature calculations will be developed to provide more accurate comparison to experiments, as needed.

Publications

Goldman, N., and L. E. Fried, 2012. "Extending the density functional tight binding method to carbon under extreme conditions." *J. Phys. Chem. C* **116**(3), 2198. LLNL-JRNL-491027.

Hamel, S., et al., 2012. "The equation of state of CH1.36: First-principles molecular dynamics simulations and shock-and-release wave speed measurements." *Phys. Rev. B* **86**(9), 094113. LLNL-JRNL-555819.

New Energetic Materials

Philip Pagoria (12-ERD-066)

Abstract

With few investigators in the U.S. developing new energetic compounds, we propose to identify methods that will provide two such compounds. Together, they would improve current and future weapon systems, while at the same time having fewer deleterious environmental and health effects. The two are highly oxidized energetic compounds for use as replacements for ammonium perchlorate in rocket propellants and liquid energetic plasticizers for both propellant and explosives applications. We envision discovering new reaction pathways that will be important for the scientific community because of the structural similarities to biologically active compounds.

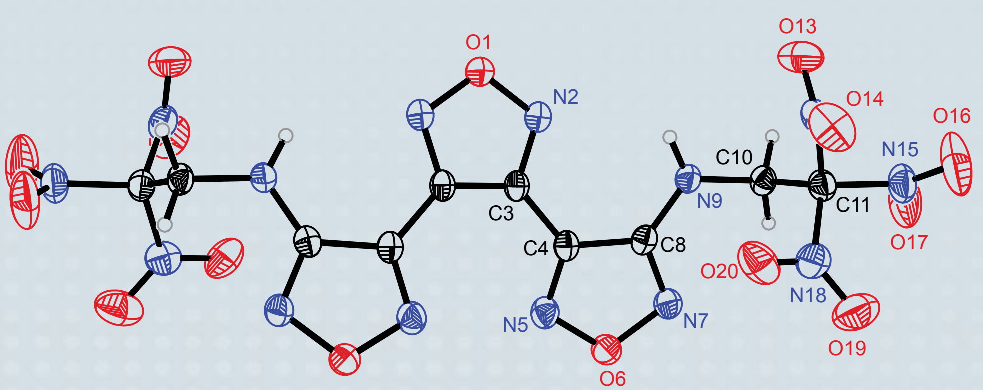
If successful, we will provide new energetic compounds of utility to current and future weapon designers, along with increased understanding and expertise in energetic materials synthesis. Our new compounds will be fully characterized with respect to sensitivity, equation of state, and thermal stability—important parameters for weapon designers and modelers. The new compounds and intermediates may have pharmaceutical applications because they are structurally similar to known biologically active compounds. This research will help to fill the gap in basic research in energetic materials synthesis in the U.S.

Mission Relevance

This project overlaps with Laboratory missions in biochemistry and stockpile stewardship. The development of these new compounds will give weapon designers new materials to achieve enhanced performance and reduced sensitivity, leading to a safer and more secure stockpile. This effort also will lead to a better understanding of synthesis efforts in foreign countries and of the performance, synthesis, and sensitivity of homemade explosives, in support of national security.

FY12 Accomplishments and Results

In FY12 we prepared four new energetic materials while determining their chemistries and reactivity. Specifically, we (1) found that the precursor LLM-201 for one our target



X-ray crystallographic analysis of LLM-204, a new energetic compound with a density of 1.755 g/cm³, which meets our criteria for thermal stability and has been characterized with respect to small-scale safety tests.

compounds has desirable physical and sensitivity properties for a melt-pour TNT replacement and has been scaled up to 25 g; (2) determined valuable information about the chemical reactivity of amino substituted 1,2,4- and 1,2,5-oxadiazoles, trinitroethanol, and nitroform; and (3) prepared three new energetic materials, 4LLM-199, LLM-204, and LLM-206, comprised of trinitroethylamino-functionalized-heterocycles—the precursors to these compounds have been scaled up to 10 to 50 g. Both LLM-199 and LLM-204 meet our criteria for thermal stability and have been characterized with respect to small-scale safety tests.

Proposed Work for FY13

In FY13 we will continue to investigate new energetic compounds consisting of highly oxidized, nitro-substituted heterocycles containing pendant dinitromethyl or trinitromethyl moieties as possible ammonium perchlorate replacement compounds. Specifically, we will continue to synthesize a variety of heterocyclic precursor compounds with functional groups that may be converted to the dinitromethyl and trinitromethyl moiety because the heterocyclic backbone affects the stability of our target energetic compounds. In addition, we will continue to characterize our target compounds with respect to thermal stability, sensitivity, and performance.

Publications

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

DeHope, A., and P. F. Pagoria, 2012. *Synthetic efforts towards high-nitrogen content 1,2,4-oxadiazoles*. LLNL-PRES-547779.

DeHope, A., and P. F. Pagoria, 2012. *Synthetic efforts towards high-nitrogen content 1,2,4-oxadiazoles*. Joint Army-Navy-NASA-Air Force (JANNAF) 45th Combustion/33rd Airbreathing Propulsion/33rd Exhaust Plume and Signatures/27th Propulsion Systems Hazards Joint Subcommittee Mtg., Monterey, CA, Dec. 3–7, 2012. LLNL-CONF-543611.

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

Determining the Coulomb Potential in Electronic Structure Calculations

Antonios Gonis (12-ERD-072)

Abstract

We propose to develop a code based on an analytic solution of the longstanding problem of material self-interaction to determine the potential from Coulomb interaction—that is, the potential acting on an electron because of Coulomb interaction between electrons in a material—whether at a level of atoms, molecules, or bulk solid. We will conduct the research within the Kohn–Sham formulation of density functional

theory and develop a single generic coding subroutine for the case of periodic materials, then finalize its development with respect to finite systems such as atoms. We will provide a unique, analytic, and computationally rigorous method for electronic structure calculations that is consistent with strict requirements of quantum mechanics and that is no more difficult computationally than other methods currently in use, which are mostly numerical and invariably approximate.

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

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) implemented a formalism within a conventional Kohn–Sham scheme to calculate the ground-state energies of atoms in the periodic table, carrying out the initial set of calculations on the basis of Cartesian coordinates, which allowed us to calculate the energies of atoms from helium to krypton in the so-called "exchange only" mode (i.e., ignoring the correlation energy functional); (2) developed the method within spherical coordinates, using the speed gained to complete calculations for the entire set of atoms in the periodic table from helium to livermorium; and (3) compared our calculations with those of the optimized effective potential method, the method closest in spirit to ours. Our comparisons demonstrated the uniqueness of our method and also exposed the limitations of the alternative method most closely in formalism to ours.

Proposed Work for FY13

Our goal in FY13 is to extend the formalism of calculating exchange potential derived from the Coulomb repulsion in the Kohn–Sham implementation of density functional theory to periodic systems, in particular to solids, based on a Green function formalism. In addition, we will implement the exchange potential formalism in two simple metals (copper and nickel), as well as in semiconductors and oxides.

Publications

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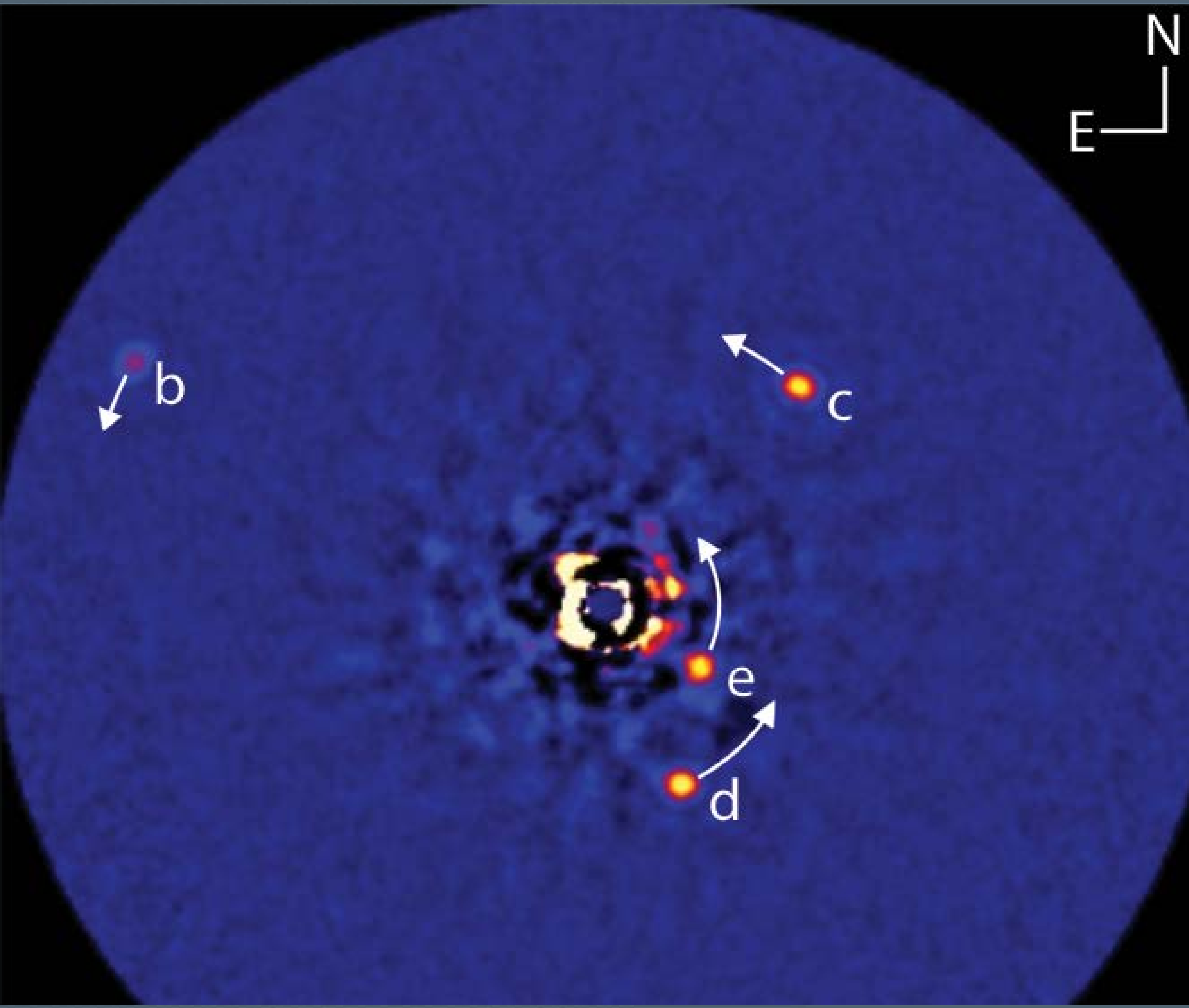
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Enhancing Climate Model Diagnosis and Intercomparison

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 being 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 to 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.

FY12 Accomplishments and Results

In FY12 we (1) processed data from the Aquarius satellite mission for use in the evaluation of climate models, (2) developed new metrics characterizing trends in ocean salinity, and (3) developed an innovative approach to replicating data stored in an evolving distributed archive to keep that data up to date locally—a portion of the data was re-processed to produce results that could be directly compared to satellite instrument measurements of atmospheric temperatures. In summary, our enhancement of both access to and development of climate model data tools and our development of new higher-level data products (notably for ocean salinity) has led to a number of research papers and positions us to make further advances under renewed outside funding in future years, specifically for DOE's Office of Basic Energy

Research. This project has positioned us to perform further innovative research in the analysis of ocean salinity and heat content trends, as well as in the application of metrics to evaluate recent climate model results.

Publications

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Creating Optimal Fracture Networks for Energy Extraction

Frederick Ryerson (11-SI-006)

Abstract

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

A major deliverable of this project will be a computational hydraulic fracturing simulation capability, GEOS, that allows the design of subsurface fracture networks in a variety of geologic settings to support the extraction of deep geothermal energy and natural gas from shales. This capability will describe the optimal fracture network, how to

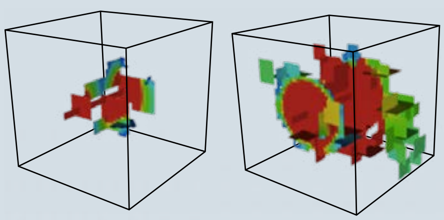
create this network, how to determine what has been created, and how this perturbed system evolves over time. This high-fidelity code will describe both hydraulic and explosive fracturing in the subsurface, and will be linked to a wave propagation code for predicting seismicity associated with the hydraulic 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.

FY12 Accomplishments and Results

In FY12 we (1) developed support for hydraulic fracture simulations by adding the ability to dynamically change the mesh topology (i.e., propagate a fracture along any face-constrained path), suitable fracture criteria at the fine-scale (resolved crack tip) as well as the coarse-scale (unresolved crack tip), and special quasi-two-dimensional finite-volume elements at separated faces (i.e., couple changes in fluid pressure with mechanical response) within the GEOS code framework for both three-dimensional and massively parallel cases—these capabilities enable simulation of flow through the fracture network as well as fracture network propagation; (2) completed work for an explicitly integrated case, appropriate for dynamic loading rates; (3) worked to develop an implicit solver, which will provide support for the slower pressure increases in conventionally stimulated reservoirs; (4) implemented a model for a sub-fracture resolution of permeability alongside the Livermore distinct element code for joint dilation, enabling simulation of dilating, rough fractures; (5) developed a simple seismic source-term generation model within the GEOS framework by incorporating an explicit, adjustable fidelity representation of the fractures in a Lagrangian framework; (6) completed development of capabilities to accommodate multiple, interacting fractures, which include the effects of induced stress fields in the near-field of the fracture process zone as well as the coalescence of fractures; (7) successfully tested source inversion using adjoint wave tomography on synthetic cases and on real data for seismic events at The Geysers geothermal field in northern California to constrain depth, location, and source parameters—the results compare well with those in the literature; and (8) maintained a seven-station broadband seismic array to monitor stimulation of AltaRock's enhanced geothermal system at the Newberry Caldera in Oregon.



Our preliminary three-dimensional GEOS computational hydraulic fracturing simulation capability. The initial rectilinear fracture network contained within an initial elastic domain (left) has a surface area of 12,000 m^2. Following stimulation at a fluid pressure 1 MPa above the minimum principal compressive stress, the network surface area increases to 85,000 m^2 (right). Colors indicate fluid pressure, with hot colors representing the highest values.

Proposed Work for FY13

We will (1) develop a mesh-splitting approach in which fractures pass through elements rather than along element boundaries; (2) run coupled problems using both time-implicit and time-explicit solvers for efficient modeling of fracture events and longer-term events, such as loading and flow; (3) validate a mechanics model of multiple fractures against available experimental data; (4) develop rate-dependent models for fracturing relevant to explosively driven methods; (5) analyze the Newberry and Salton Sea data sets using adjoint wave tomography and a Bayesloc algorithm tuned to improve locations in the micro-seismic regime; (6) create a synthetic micro-seismic three-dimensional dataset using a fourth-order finite-wave propagation code

(SW) to test the detection and location algorithms; and (7) integrate a simplified model of fracture growth.

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Ezzedine, S., et al., 2012. *Enhancing heat extraction and minimizing water losses by deploying silica gel in enhanced geothermal systems*. ARMA Intl. Conf. and Expo., Chicago, IL, Sept. 23–25, 2012. LLNL-PROC-539191-DRAFT.

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Ezzedine, S., et al., 2012. *Uncertainty quantification for hydromechanical coupling in three dimensional discrete fracture network application to enhanced geothermal systems*. 46th US Rock Mechanics/Geomechanics Symp., Chicago, IL, June 22–28, 2012. LLNL-POST-544953-DRAFT.

Ezzedine, S. M., et al., 2012. *Uncertainty quantification of 3D flow, heat and mass transport in stochastic discrete fracture networks*. AGU Fall Mtg. San Francisco, CA, Dec. 3–7, 2012. LLNL-PRES-563996-DRAFT.

Fu, P., S. M. Johnson, and C. R. Carrigan, 2012. "An explicitly coupled hydro-geomechanical model for simulating hydraulic fracturing in complex discrete fracture networks." *Int. J. Numer. Anal. Meth. Geomech*. 23 Aug. 2012, 10.1002/nag.2135. LLNL-JRNL-519272.

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Detection and Attribution of Regional Climate Change with a Focus on the Precursors of Droughts

Celine Bonfils (11-ERD-006)

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 detect-and-attribute 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 changes in atmospheric circulation are widely recognized as key factors triggering droughts. We will determine whether an emerging human signal exists in the frequency and spatial structure of drought precursors and how relative human and natural contributions to drought evolve over time.

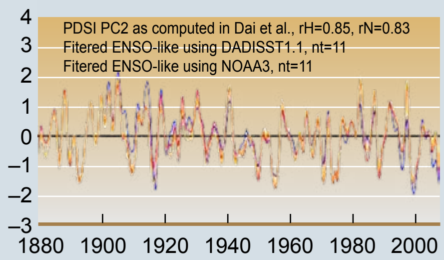
This project will improve our scientific understanding of drought mechanisms. We expect to investigate new avenues in detection and attribution, including new variables and regional-scale tools. Ultimately we will generate knowledge that can inform policy decisions on adapting to drought in a changing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

FY12 Accomplishments and Results

In FY12 we (1) characterized drought behavior in observations and in various climate runs of the 20th and 21st century using two widely used drought indices; (2) examined whether the temporal statistics of a specific drought-conducive mode of variability found in FY11 evolve in response to future simulated global warming; (3) began to incorporate new simulations of the historical and future climate change from the Coupled Model Intercomparison Project Phase 5; and (4) computed a metric of drought-related atmospheric circulation change suitable for both observations and model runs. Our project helped to explore the feasibility of this type of drought research and led to an Early Career Research Program proposal that successfully received funding through the DOE Office of Science. With this award, we will continue to improve the scientific understanding of the nature and causes of past droughts. This will be done through investigation of the naturally driven and externally forced components of presumed large-scale drought precursors. Particular attention will be given to the sensitivity of the results to specific uncertainties.



We found that the second mode of variability of the observed Palmer Drought Severity Index (PDSI PC2) (blue line) responds with a three-month lag to the temporal component of our drought-conducive mode of variability (ENSO-like PC) (red and orange lines), computed with two different observational data sets of historical sea-surface temperatures. Negative values in the ENSO-like time-series are associated with a La Niña-type pattern and more subtropical droughts. All time-series are low-pass-filtered.

Publications

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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)

Abstract

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

We expect to produce infrared spectra of the atmospheres of the planets HR8799b and HR8799c—making these the lowest-temperature exoplanets ever spectroscopically characterized—and use the results to adapt models of the planets' complex atmospheres. We will use precision adaptive optics astrometry and computational simulation to find orbital parameters that are consistent with observations and with formation and evolution scenarios. Next, through our large-scale GPI project to survey 600 stars, which is the most sensitive and complete imaging search for planets to date, we will produce detailed characterizations of the atmospheric structure and composition of previously inaccessible classes of exoplanets.

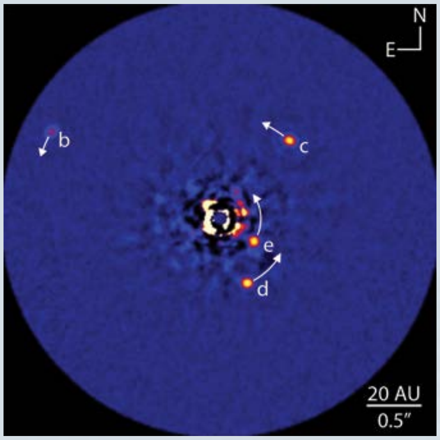

Image of the HR8799 system showing the newly discovered fourth planet, HR8799e. Arrows show the direction and speed of orbital motion.

Mission Relevance

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

FY12 Accomplishments and Results

During FY12 we carried out spectroscopic analysis of the HR8799 system. Specifically, we (1) obtained, with specialized filtering of high signal-to-noise spectra, the best-ever spectrum of an extrasolar planet—this shows clear evidence for molecular species such as water and carbon monoxide at concentrations that require deep vertical circulation in the atmosphere, as well as an enhancement in heavy elements consistent with the accretion process that formed our own solar system; (2) performed observations at higher spectral resolution that may directly measure rotation rate of the planet; (3) began orbit fitting of HR8799 with new 2012 data; (4) developed an advanced wavefront control technique to cancel diffraction speckle artifacts and tested it on GPI; and (5) began detailed science planning for the international 900-hour GPI campaign that we will lead, including developing simulation and image-analysis tools and calibration algorithms.

Proposed Work for FY13

We will (1) conduct high-spectral-resolution observations of HR8799, which may allow us to directly measure the planets' rotation rates—the first such measurement of an exoplanet; (2) test new algorithms for extracting planetary signals from correlated spectral noise, using both past data on HR8799 and new data from GPI; and (3) complete planning for the GPI science campaign and lead the "first light" science and commissioning observations, studying known planetary and disk systems with the most advanced adaptive optics instrument ever constructed.

Publications

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

Karis Mcfarlane (11-ERD-053)

Abstract

Our goal is to determine the short-term effects of land use and management on soil respiration and total ecosystem respiration fluxes in a mature northern hardwood forest, and to attribute changes in the regional biosphere signal of atmospheric carbon-14 dioxide ($^{14}CO_2$) to disturbances from logging operations using radiocarbon as an isotopic tracer. We will compare observations of carbon-14 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 determining 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.

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.

FY12 Accomplishments and Results

In FY12 we (1) continued pre-harvest $^{14}CO_2$ measurements from air sampled at the Willow Creek eddy covariance tower and the WLEF-TV regional tall tower in northern Wisconsin; (2) began data analysis of the $^{14}CO_2$ measured at the two towers; (3) continued pre-harvest soil respiration flux measurements at soil surface and from depth profiles and conducted intensive sampling for $^{14}CO_2$ of soil respiration from depth profiles in the field; (4) conducted soil incubation experiments and soil density fractionation to determine isotopic end members for bulk soil respiration and characterize soil carbon stocks, distribution, and long-timescale dynamics; (5) began data analysis on approach and findings for pre-harvest soil and ecosystem respiration fluxes and $^{14}CO_2$ and $^{13}CO_2$ levels; and (6) presented our work at domestic and international conferences.

Proposed Work for FY13

In FY13 we will (1) complete pre- and post-harvest $^{14}CO_2$ measurements from air sampled at the Willow Creek eddy covariance tower and the WLEF-TV regional tall tower, (2) complete data analysis of $^{14}CO_2$ measured at the two towers, (3) complete pre- and post-harvest soil respiration flux measurements and begin more intensive sampling for $^{14}CO_2$ of soil respiration in the field, (4) complete soil incubation experiments and soil density fractionation to determine isotopic end members for bulk soil respiration, and (5) publish our results on post-harvest soil and ecosystem CO_2 fluxes and our WLEF-TV tower $^{14}CO_2$ results.

Publications

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The Feasibility of Using Aerosols to Discriminate and Quantify Greenhouse Gas Emissions by Source

Eric Gard (11-FS-010)

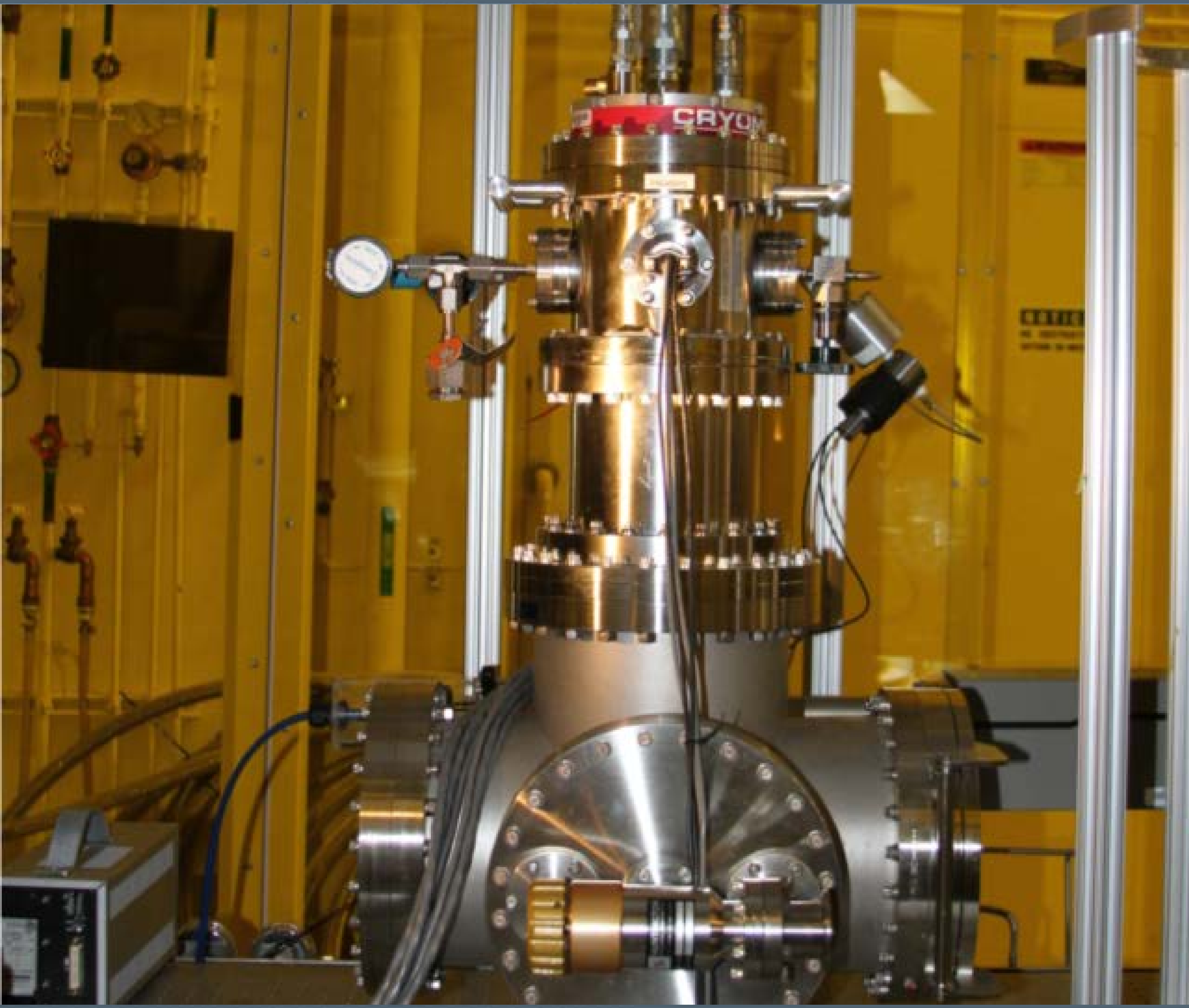
Abstract
We propose to assess the feasibility of using aerosol (particulate) composition, concentration, and size distribution information to improve the discrimination and quantification of greenhouse gas emissions, and assess how these analyses might help determine the emission source. An independent 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 terms of the 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.

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.

FY12 Accomplishments and Results
In FY12 we detailed the design and performance of a conceptual system for worldwide greenhouse gas emissions characterization, which would provide the accuracy and precision in both carbon dioxide emissions measurements and inverse transport modeling to quantify emissions at the country level with weekly and monthly time resolution. We determined that adding aerosol particle measurements to the minimum suite of gas-phase measurements would result in a very small improvement in overall uncertainty, which is dominated at this point by large errors introduced by the inverse modeling steps that are essential for greenhouse gas source apportionment. We found that while the proposed aerosol measurements would provide valuable information on greenhouse gas emissions, the cost-to-benefit ratio of deploying them within the network is not feasible.

Energy Supply and Use



Laboratory Directed Research and Development

FY2012

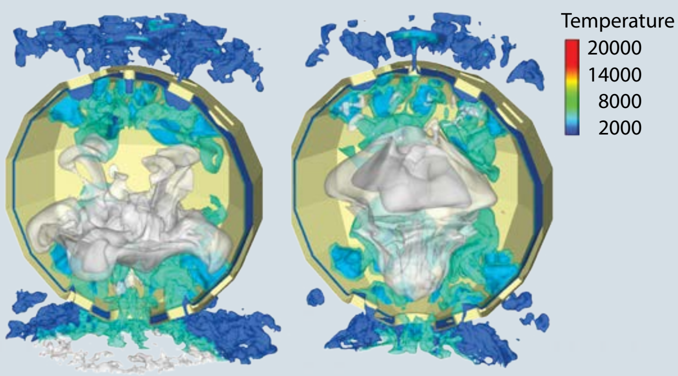
Dynamic Chamber Processes for LIFE: Simulations and Experiments on Beam Propagation and Chamber Clearing

Jeffery Latkowski (10-SI-009)

Abstract

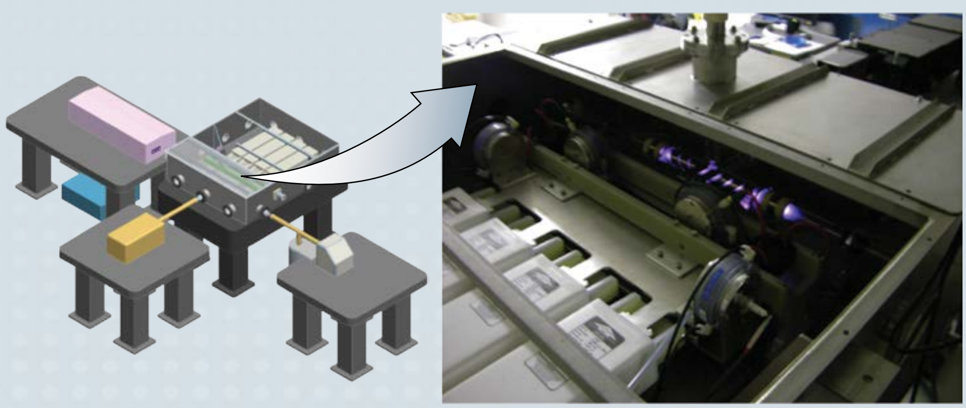
To mitigate the challenges of nuclear energy and to advance the timetable of the availability of fusion sources, the Laboratory envisions a novel once-through, closed nuclear-fuel cycle based on the laser inertial fusion energy (LIFE) concept. We propose to couple modeling and experiments to explore 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.

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,



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 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 laser inertial fusion energy.

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.

FY12 Accomplishments and Results

In FY12 we (1) studied the radiative cooling of xenon using the theta-pinch facility and measured the xenon stall temperature, although reduction in funding prevented planned testing of laser propagation; (2) optimized the vent-and-fill protocol using many-shot simulations in two- and three-dimensions to estimate quasi-steady-state densities and temperatures using integrated radiation-hydrodynamics calculations; and (3) improved our understanding of the early-time gas response to target explosion using various radiative and hydrodynamic simulations and integrated simulations to explore hydrodynamic motions under the influence of the blast wave induced by target explosion, including the effects of radiation, turbulence, and shock propagation in the fusion chamber. In summary, this project explored and developed new fusion chamber designs and simulation capabilities critically required for enabling the LIFE concept. The next steps are to experimentally validate these design codes and our LIFE chamber designs.

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Design of Novel Catalysts to Capture Carbon Dioxide

Roger Aines (10-ERD-035)

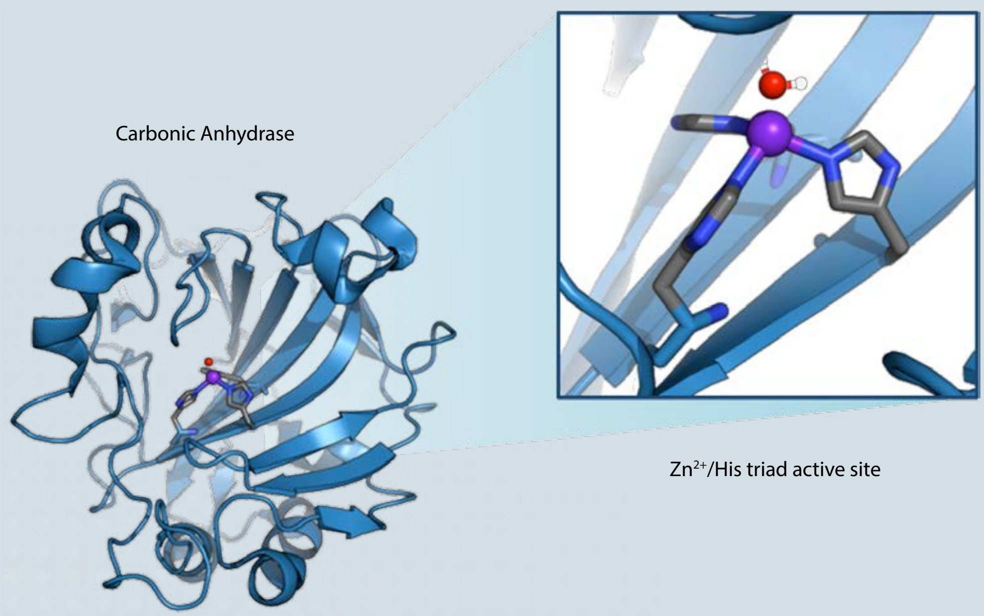
Abstract

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 1000 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. To enable feasible application, the chemistry of the capture process must be more efficient than current methods to keep the capture device size



We have created simple versions of the carbonic anhydrase enzyme by considering only the functions of the active site in the enzyme and then building small molecules that perform those functions without a protein structure.

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.

FY12 Accomplishments and Results

We created a number of catalysts that mimic the behavior of carbonic anhydrase, using computational methods to examine specific functions, particularly release of the product (i.e., bicarbonate) from the reactive complex. Maximizing this release resulted in a new set of noncyclic catalysts with optimized behavior in the presence of high concentrations of bicarbonate. We also created a series of computational tools and approaches to rapidly screen and design new catalysts. In summary, this project has created a series of functional mimics for carbonic anhydrase and established methods for mimicking the active site of other enzymes using quantum mechanical modeling. The DOE Office of Fossil Energy and the Advanced Research Projects Agency–Energy has begun supporting the use of these carbonic anydrase mimics in follow-on research to enhance CO_2 capture.

Publications

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

David Camp (10-ERD-055)

Abstract

Underground coal gasification (UCG) is a critical emergent technology to boost 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 as well as in computational geosciences.

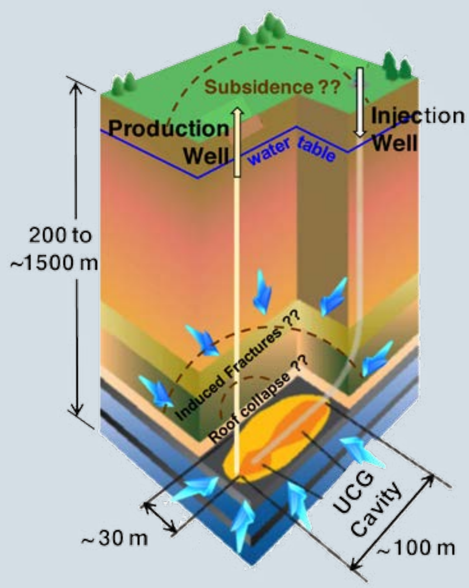
We expect to provide a far stronger understanding and predictive capability for UCG operations than is currently available. Our coupled simulation and geophysics monitoring approach will allow improved siting, design, permitting, operation, monitoring, and environmental performance of pilot and commercial 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. It will help spur the deployment of UCG to reduce the cost and accelerate delivery of potentially low-carbon, secure energy to the nation. In addition, this project capitalizes on high-performance computing to demonstrate leadership in unclassified computing applications.

FY12 Accomplishments and Results

In FY12 we finished creating and validating the world's best computational model for UCG, a three-dimensional, full-physics model that provides geometric and operational flexibility to handle a wide range of UCG configurations. The simulator includes domain models for cavity gas, coal wall, rock wall, and rubble zones in the near field; non-isothermal unsaturated transport and spalling (collapse) in the mid-field; and hydrology and rock and soil mechanics in the far field. Specifically, we (1) finished developing and validating the wall-zone model, three-dimensional boundary tracker, cavity gas model, and rubble zone model; (2) ran rock and soil mechanics and hydrology models for actual UCG field-test conditions; (3) interfaced all domain models to create an integrated simulation capability, including manually coding the rock and soil mechanics interfacing; (4) simulated a difficult but important UCG field test with results matching the data well; and (5) presented our results at a key conference attended by most UCG practitioners, modelers, and potential sponsors. In summary, we have created the world's best computational simulator for UCG, making it possible to analyze construction and operation alternatives in complex geology. The simulator has been well received by the UCG community, and discussions have already begun about collaborating with companies and institutions seeking to use the simulator.



Underground coal gasification (UCG) produces energy-rich gas from deep coal seams without mining. Lawrence Livermore created the world's most advanced computational simulator for UCG, making it possible to analyze construction and operation alternatives in complex geology. The figure shows a typical UCG well layout, with assessment of the risk of subsidence and roof collapse.

Publications

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Reid, C. M., et al., 2012. *Application of integrated multiphysics simulator to underground coal gasification*. 29th Intl. Pittsburgh Coal Conf., Pittsburgh, PA, Oct. 15–18, 2012. LLNL-ABS-553931.

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

Robin Miles (11-SI-004)

Abstract

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

Our research will lead to a fundamental understanding of material attributes required to design a structurally sound injected laser fusion target, based upon both measurements and models of 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, 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.

FY12 Accomplishments and Results

We (1) performed extensive measurements of the phase-change temperatures and vapor pressures for the liquid-to-solid transitions of hydrogen and deuterium frozen in ultralow-density aerogels (~30 mg/cm^3)—results show depressed freezing temperatures of over 1°K; (2) built an apparatus to measure the permeability of low-density foams to liquids for input into structural models of liquid hydrogen moving in foams during acceleration—results showed little permeability and that a "poroelastic" model (porous media whose solid matrix is elastic and fluid is viscous) could be used to model hydrogen in foam; (3) modeled response of the capsule-support membrane to various axial and transverse loads with consideration of the stress and friction of the membranes—results showed dampening of motion with added pre-stress; (4) performed static material tests on new carbon nanometer-scale tube composites; (5) built an apparatus to measure the displacement of variable-thickness membranes under an acceleration load; (6) demonstrated new fast-growth chemical-vapor-deposition diamond materials, with a growth rate of 4.5 µm/h; and (7) measured relevant optical properties of candidate membrane materials—results showed graphene (single-atomic-layer carbon sheets) to be a good infrared absorber.

Cryostat used to measure depressed freezing temperature of hydrogen isotopes in ultralow-density foams used in laser fusion targets.

Proposed Work for FY13

We will (1) move the calorimeter cryostat to a tritium area or finish building the system for measuring mechanical properties of solid DT ice, depending on the success of vapor-pressure measurements of deuterium in lower-density aerogels; (2) continue developing new carbon-composite films and complete dynamic tests of candidate membrane materials; (3) perform system-level thermal tests to verify our thermal analysis; and (4) test thermal and mechanical properties of the new fast-growth-rate chemical-vapor-deposition diamond materials.

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.

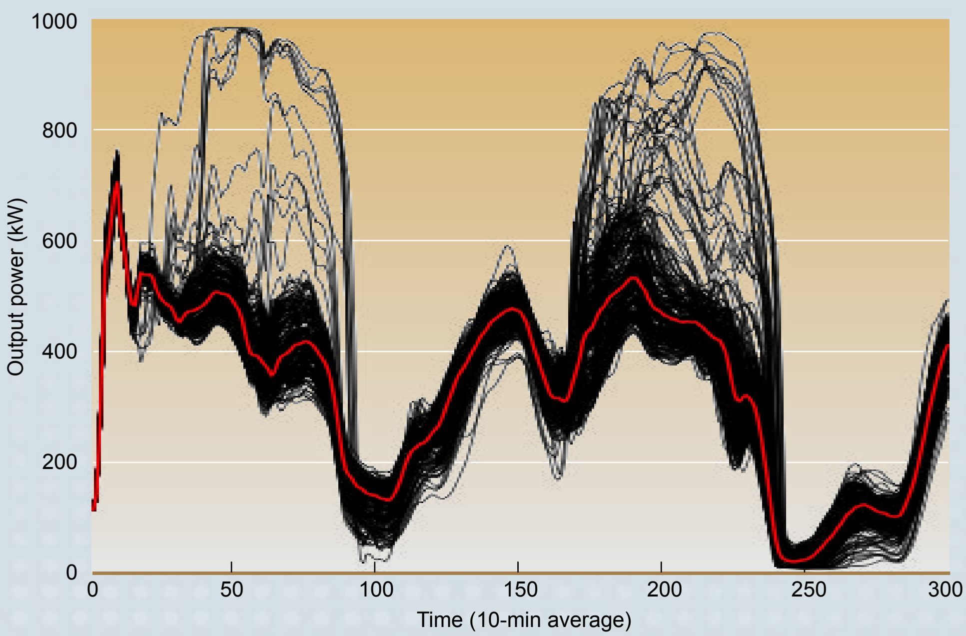
Forecasting and Uncertainty Quantification of Power from Intermittent Renewable Energy Sources

Wayne Miller (12-ERD-069)

Abstract

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

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



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

line power. Deliverables will be the best-practice process for performing validated forecasts, and the suite of tools needed to perform the forecasts.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) acquired meteorological data at Site 300 and an adjacent wind farm using the existing meteorological tower and a mobile light detection and ranging system at Site 300, and a mobile light detection and ranging system and a wind farm supervisory control and data acquisition system at the wind farm; (2) tested various Weather Research and Forecasting (WRF) Model ensemble options and chose ensembles based on varying WRF Model physics packages and parameters because of better error control; (3) ran a large ensemble forecast over the wind farm and identified forecast errors based on the variations; (4) began developing a reduced-order model for turbine power that improves the standard power curves, which have errors up to 100%, and correlated the actual inflow characteristics to the measured turbine power output; (5) prototyped the uncertainty qualification process for evaluating ensemble errors on turbine power output, evaluating an ensemble for power production; and (6) collaborated with the University of Wyoming to couple WRF Model ensembles with the HELiOS model, achieving the ability to simulate realistic turbulent atmospheric inflows in a state-of-the-art rotor computational fluid-dynamics code to determine loads and power.

Proposed Work for FY13

In FY13 we will (1) perform additional field data campaigns to obtain verification and validation statistics; (2) implement and test several forecasting approaches for comparative error analysis; (3) couple, for high-resolution modeling of wind farm terrain, the WRF Model code and the CgWind large-eddy simulation code for wind engineering applications; (4) further refine and implement the forecasting uncertainty process; and (5) begin efforts to compare forecast methods for short-, medium-, and long-term forecast windows.

Increasing the Lifetime of Rechargeable Batteries

Christine Orme (12-LW-030)

Abstract

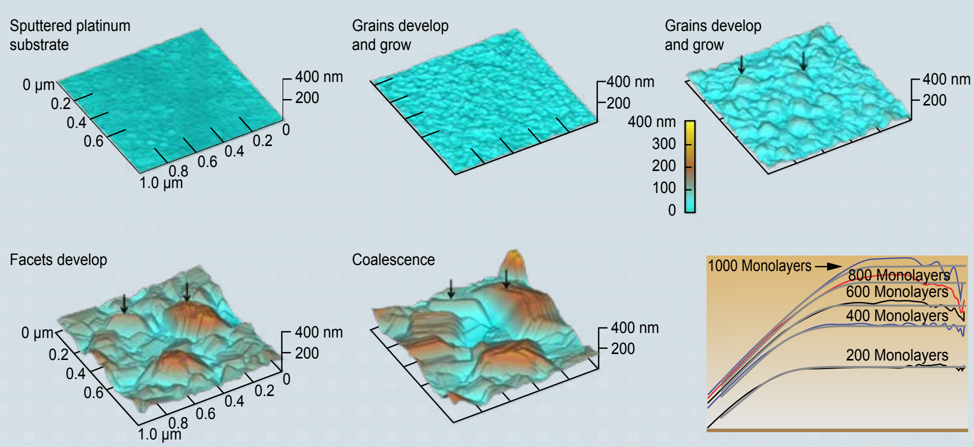
Our overarching goal is to increase the lifetime of rechargeable batteries by preventing failures from dendrite formation and nonreversible processes. It is a major challenge to design the chemistry, geometry, and charge cycle such that a battery is truly reversible and returns to its original configuration after charging. One of the biggest problems occurs as

the oxide converts back to a metal, re-plating onto the anode during recharging. We propose to directly image the cyclic charging and discharging of anode materials using in situ atomic force microscopy, transmission electron microscopy, optical microscopy, and Raman microscopy. All techniques will monitor dynamical changes under electrochemical and temperature control. Together, these techniques will give us the ability to image at higher spatial and temporal resolution than has been reported to date, allowing us to develop an understanding of the parameters and chemistries that control zinc dendrite nucleation and kinetics in alkaline chemistries and ionic liquids. We will also investigate several additives that we believe have the potential to stabilize the zinc interface. We hypothesize that quantifying the dendritic morphological evolution will provide new mechanistic details of how electrolytes and additives impact the stability of battery interfaces.

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

Mission Relevance

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



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

FY12 Accomplishments and Results

In FY12 we met our milestones by directly imaging charging and discharging phenomena of zinc anodes using the three complementary in situ techniques of atomic force microscopy, optical microscopy, and ultra-small-angle x-ray scattering. Specifically, we completed a comparison between atomic force microscopy and ultra-small-angle x-ray scattering in an alkaline battery electrolyte that measured surface kinetics and quantified roughness evolution as a function of current density. This baseline study measured scaling exponents, shape anisotropy, and correlation lengths that are metrics for evaluating additives and other electrolytes. Results suggest a transition in the smoothing mechanism as a function of charging rate. We also completed an atomic force microscopy and optical investigation of zinc recharging in an ionic liquid. These results indicate that both the roughness and correlation length saturate, eventually evolving to a steady-state morphology, which differs from fast-growing instability observed in alkaline electrolyte.

Proposed Work for FY13

In FY13 we propose to evaluate how additives alter important crystal growth parameters (such as nucleation density, growth rates, and diffusion rates) during charging and discharging. In addition, we will evaluate commercial and proprietary ionic liquids. Specific milestones include (1) optimizing concentration of the additive bismuth and determining its mechanism of interaction, (2) screening several other additives (bromide and thiol-based), and (3) determining molecular mechanisms of dendrite formation in commercial and proprietary ionic liquids.

Publications

Keist, J., et al., 2012. *Coupling in-situ techniques to analyze zinc deposition and dissolution for energy storage applications*. 2012 MRS Fall Mtg. and Exhibit, Boston, MA, Nov. 25–30, 2012. LLNL-ABS-562111.

Keist, J., et al., 2012. *In situ techniques to analyze electrodeposition of zinc within ionic liquid electrolytes*. 2012 Gordon Conf. Electrodeposition: Electrochemical Materials Synthesis and Applications, Biddeford, ME, July 29–Aug. 3, 2012. LLNL-POST-574015.

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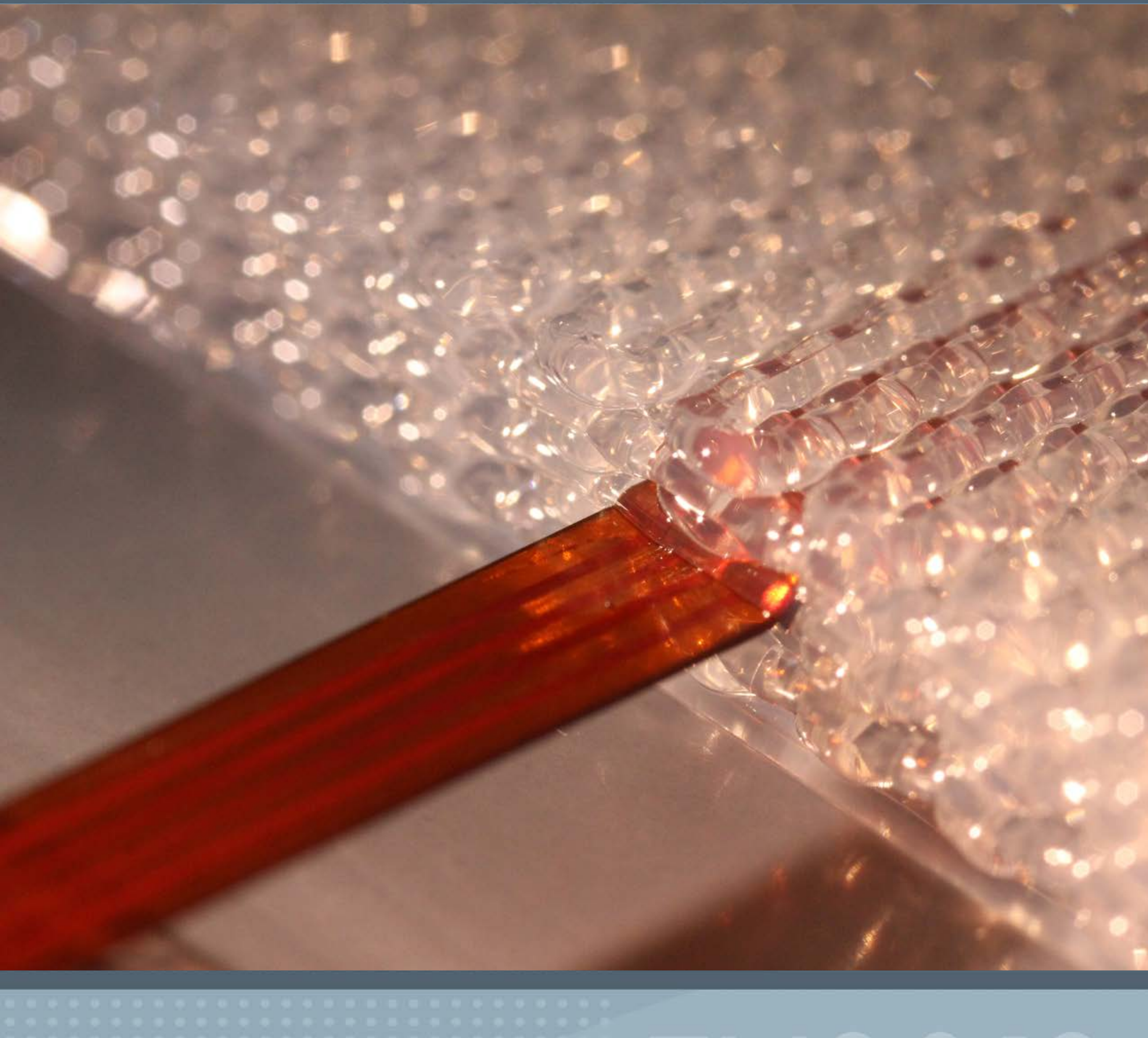
Engineering and Manufacturing Processes



Laboratory Directed Research and Development

FY2012

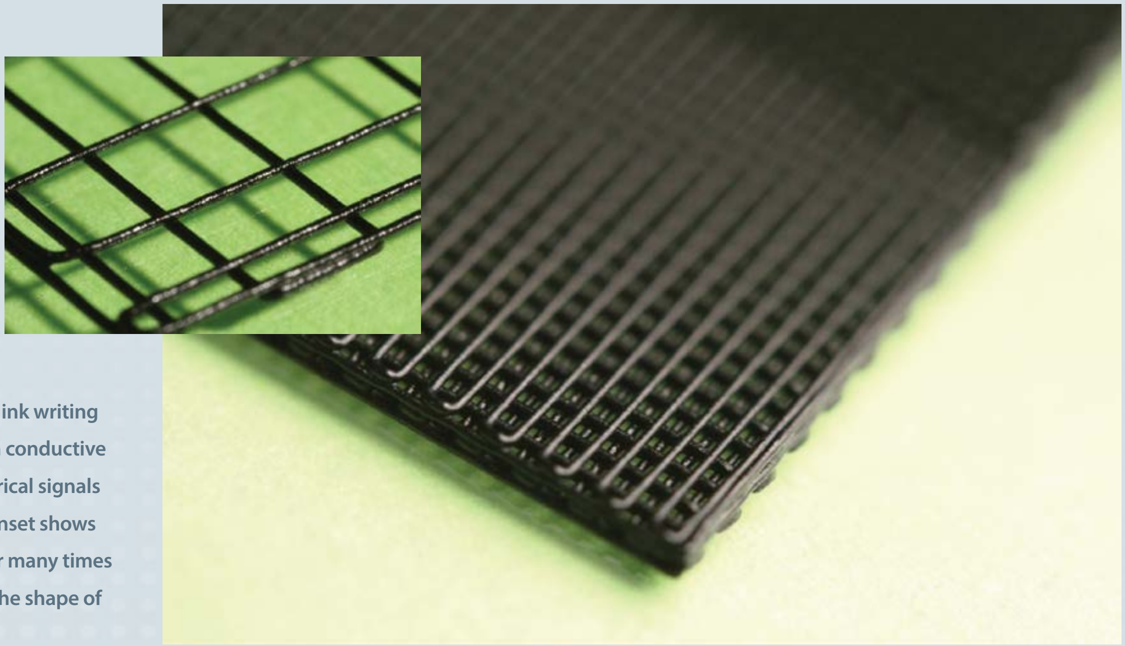
Scalable High-Volume Micro-Manufacturing Techniques for Three-Dimensional Mesoscale Components

Christopher Spadaccini (11-SI-005)

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 production of three-dimensional 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 three-dimensional 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 the ability to scale to large numbers of components. We intend to design and fabricate a "new" material with



A structure printed using the direct ink writing technique. The ink, formulated with conductive carbon and silicone, conducts electrical signals even as it flows and is printed. The inset shows how the filaments are spanned over many times their diameter while still retaining the shape of the extrusion nozzle.

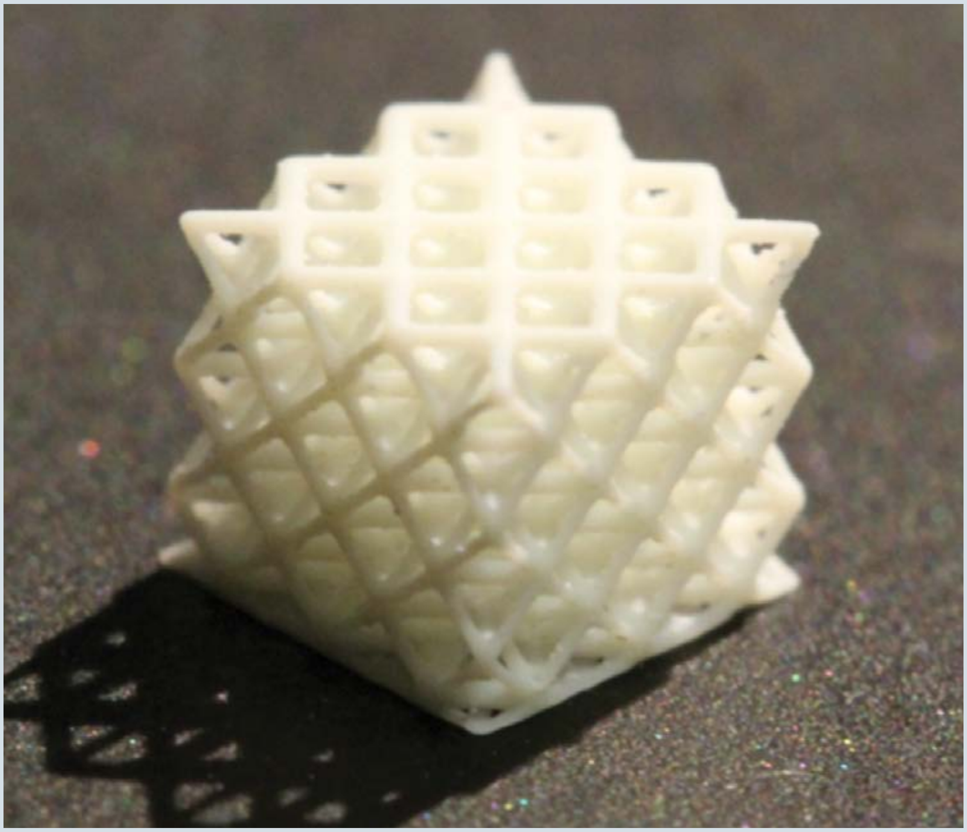
specified properties such as thermal expansion versus stiffness outside the bounds of those attainable with bulk materials processed via traditional synthesis methods. Success in meeting our proposed objectives will both illustrate capabilities of the process and demonstrate novel materials and structures 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" three-dimensional 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 and reducing or countering threats to national and global security.

FY12 Accomplishments and Results

In FY12 we improved and expanded upon our micro-manufacturing technologies, and our efforts in microcellular materials demonstrated previously unachievable material property combinations. Specifically, we (1) fabricated and characterized micro-lattice architectures with ultrahigh stiffness-to-density ratios and lattices with negative Poisson's ratio, (2) fabricated and characterized binary thermite materials energy



This cellular alumina micro-lattice architecture, which exhibits high stiffness and low weight, was fabricated using projection micro-stereolithography. The feedstock material was photopolymer binder loaded with alumina nanoparticles, and subsequent thermal post-processing removed the polymer binder, leaving only denser alumina.

release rates spanning two orders of magnitude, (3) produced a three-dimensional mesoscale component with complex geometry, and (4) demonstrated novel structures by combining our micro-manufacturing techniques.

Proposed Work for FY13

In FY13 we will continue to advance our micro-manufacturing techniques by scaling up to produce larger parts, fully incorporating multiple-material capability into several of our systems and designing additional materials with unique microstructures, providing previously unachievable properties. Specifically, we will (1) design a micro-architecture for a negative Poisson's ratio, which will produce a structure that will contract in one direction when compressed in another; (2) fabricate and characterize that structure; (3) establish the capability to fabricate heterogeneous structures with our projection micro-stereolithography system; (4) fabricate and characterize thermite materials with a fully three-dimensional design; and (5) fabricate additional unique components by combining micro-manufacturing technologies.

Publications

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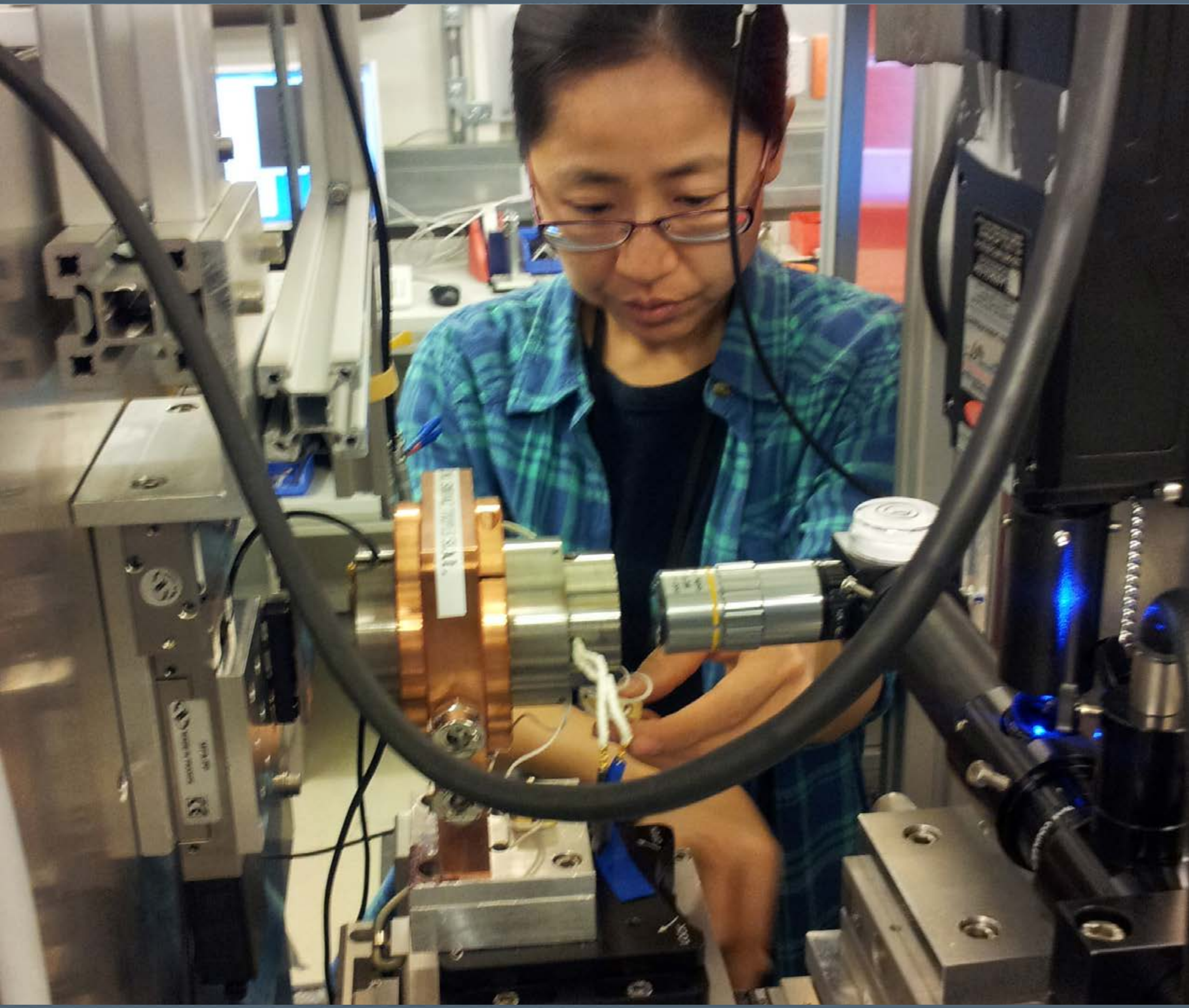
Materials Science and Technology



Laboratory Directed Research and Development

FY2012

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 the transformation from the body-centered cubic phase to the hexagonal closely packed phase in iron by leveraging recent advances in synchrotron x-ray diffraction, and characterize the high-pressure hexagonal closely packed 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 thermo-mechanical loading in situ using a diamond anvil cell. This will provide direct variant selection, stress state, and growth measurements for the transformation from body-centered cubic to hexagonal closely packed phase in individual grains embedded within an iron polycrystal; observations of mechanical twinning in hexagonal closely packed iron (if any); and material properties for the high-pressure hexagonal closely packed 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 multiphysics 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.

FY12 Accomplishments and Results

In FY12, we (1) demonstrated the feasibility of combining far-field high-energy diffraction microscopy with a near-field technique in a single instrument, including successfully testing the instrument; (2) performed a final series of experiments showing no orientation relationship associated with the alpha–gamma phase transition in iron and illustrating direct observations of two mechanisms for the alpha–omega phase transition in zirconium; and (3) completed and released our open-source analysis suite heXRD, which is being implemented and further developed at the Advanced Photon Source, the Advanced Light Source, and the Cornell High-Energy Synchrotron Source. Experiments on cerium were deemed redundant for now but could be performed in follow-on work. In summary, this project has provided a measurement technique—and the necessary analysis software—that enabled first-of-kind observations into the mechanisms underlying displacive phase transitions in materials subject to extremes of pressure and temperature in situ. On the basis of our results, we secured a partnership with the Air Force Research Laboratory and Carnegie Mellon University to conduct follow-on work at the Advanced Photon Source to develop a dedicated high-energy diffraction microscopy instrument. We have also secured support from the Department of Defense under the Joint Munitions Program.

Publications

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Material–Coolant Interactions in Fusion Reactors

Bassem El-Dasher (10-ERD-056)

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.

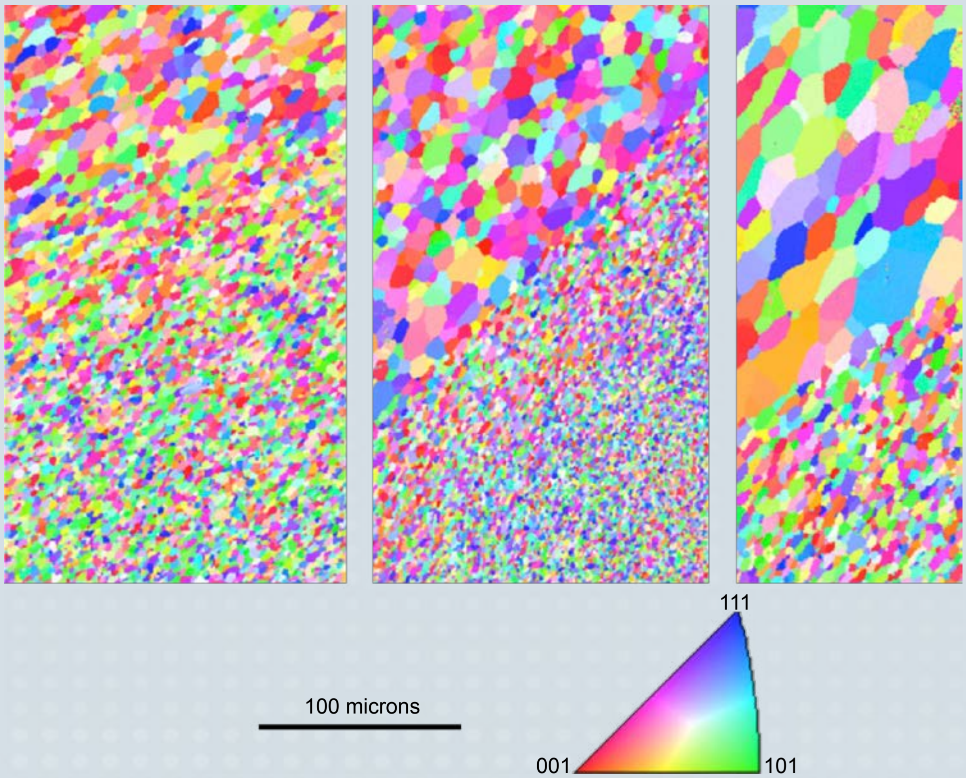
We expect to (1) assess the threat of corrosion, stress corrosion cracking, and hydrogen embrittlement in candidate reactor materials; (2) elucidate the mechanisms involved in degradation of these materials; (3) develop predictive models to explain observed phenomena, thereby demonstrating scientific 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 in nuclear waste disposal, safety, carbon sequestration, or proliferation.

FY12 Accomplishments and Results

In FY12 we (1) studied the effect of adding both stainless steel and beryllium electrodes and found that the secondary steel reduced the corrosion rate by a factor of two, while the addition of a beryllium electrode reduced the corrosion of the primary steel



Zone interfaces on the advancing side of a friction-stir weld, a solid-state joining process in which the metal is not melted but mechanically intermixed with another metal at the place of the join, then softened so the pieces can be fused using mechanical pressure. Welding conditions of 400 revolutions per minute and 4 inches per minute are shown on the left, 300 revolutions per minute and 2 inches per minute (middle), and 500 revolutions per minute and 1 inch per minute (right), demonstrating the increase in microstructural gradient as a function of heat input in MA956 oxide-dispersion-strengthened steel.

electrode to virtually zero—this was unfortunately at the expense of the beryllium electrode, which dissolved within ten seconds of introduction into the molten salt; (2) performed testing with lithium on both ODS and reduced-activation ferritic and martensitic steels and found virtually no penetration of the lithium within the steels—however, when the same testing was conducted on specimens where stress was present, measurable infiltration of lithium was observed; (3) conducted multiple experiments in situ at Argonne National Laboratory's Advanced Photon Source to compare hydrogen uptake in a variety of steels—the kinetics of hydrogen bubble formation were found to be lower in ODS steels than in reduced activation ferritic and martensitic steels by a factor of three, and perhaps more interesting was that the presence of helium in the steels from prior ion-beam irradiation seemed to retard the hydrogen bubble formation, contrary to conventional wisdom; and (4) determined the appropriate heat input required for successful friction-stir welding of ODS steels, a solid-state joining process in which the metal is mixed, not melted. The successful conclusion of this project has yielded valuable insight into how the steels of interest to inertial-confinement fusion interact with structural materials as well as the development of novel electrochemical in situ techniques that have been adopted by other groups at the Laboratory. The information gleaned from this work has been used to inform materials decisions in the Laser Inertial Fusion Program at Livermore, and inspired a follow-on LDRD project focused on understanding the accelerative effects of stress on corrosion in environments relevant to inertial-confinement fusion.

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

This project is designed to apply a suite of in situ x-ray spectroscopy and electron microscopy techniques to investigate the performance of advanced materials for energy storage, specifically electrodes based on group IV element nanometer-scale crystals and capacitors based on nanoporous carbon. The development of 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 the project will yield information regarding the mechanisms of energy storage in nanostructured electrodes and their degradation 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. Moreover, it is aligned with research needs that were defined by a recent DOE Office of Basic Energy Sciences study on electrical energy storage—specifically, the need to implement new in situ techniques to investigate chemical, electrochemical, and physical processes at solid–liquid and solid–gas interfaces.

FY12 Accomplishments and Results

In FY12 we successfully conducted a series of soft x-ray spectroscopy measurements of materials for a rechargeable lithium ion battery (metal oxide cathodes), along with associated lithium-containing control samples, focusing on the electronic structure and geometrical environment of the lithium ions in these materials. These measurements enabled the assignment of spectroscopic signatures for specific electronic configurations of lithium—signatures that are invaluable for distinguishing the local environment and bonding of lithium ions in different electrodes for lithium ion batteries and establishing how the bonding environment of the lithium changes as the electrodes degrade during operation. We also conducted successful in situ x-ray spectroscopy studies of the transition metal elements and oxygen in the metal oxide cathodes, although faulty equipment at the experimental beam-line limited the amount of valuable information obtained from these studies. In summary, this project developed novel in situ capabilities, using soft x-ray spectroscopy and transmission electron microscopy, to investigate electrical-energy-storage materials under operating conditions, and used those capabilities to identify the modes and mechanisms of charge storage and actuation in these materials. The in situ soft x-ray spectroscopy and transmission electron microscopy capabilities developed in this project have already been used in research supported by the DOE Office of Basic Energy Sciences.

Research team members Jonathan Lee (left) and Michael Bagge-Hansen (right) prepare to test the vacuum integrity of a cell for in situ x-ray spectroscopy measurements of electrical energy storage materials.

Publications

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

Jaime Marian (11-ERD-023)

Abstract

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

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

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) finished a first version of the kinetic Monte Carlo code for a type of displacement known as screw dislocation motion and continued to make the code more robust and efficient; (2) followed a different approach for the selected alloy of study than initially planned—rather than developing a full tungsten–titanium potential for semiempirical atomistic calculations, we formulated a solute interaction model within the kinetic Monte Carlo model of dislocation motion; (3) parametrized the solute model with density functional theory calculations for the tungsten–rhenium system and computed vacancy formation as well as migration and solute migration energies, all as a function of several components of the stress tensor; (4) continued to work on characterization of tungsten potentials, particularly what controls screw dislocation core structure; and (5) performed calculations of damage accumulation in pure tungsten for different temperatures with and without simultaneous helium production.

Proposed Work for FY13

For FY13 we propose to (1) improve the kinetic Monte Carlo model of thermally activated dislocation motion and add solute interactions to the calculations to obtain solution hardening effects, (2) link the kinetic Monte Carlo results to a crystal plasticity model of tungsten and tungsten–rhenium to achieve experimentally relevant strains, (3) calculate the temperature dependence of void hardening in tungsten, and (4) propose conditions (e.g., temperature, stress, and irradiation dose) under which tungsten is most ductile to maximize usefulness and lifetime.

Publications

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

Manyalibo Matthews (11-ERD-026)

Abstract

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

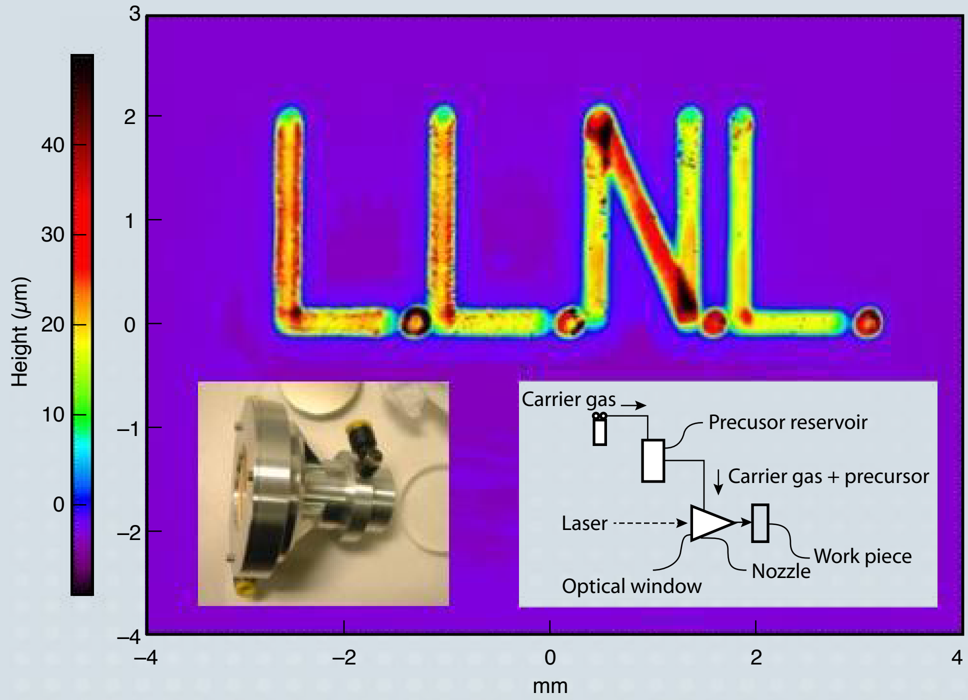
Using optimized short-pulse laser light, we expect to achieve more precise control of laser-machined morphologies by limiting laser-parameter-dependent energy absorption. We will establish a method for introducing controlled silica-rich vapor at lower temperatures to fill damage cavities and eventually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

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) developed Schlieren and infrared absorption imaging to study gas flow dynamics and chemistry in situ during laser exposures and demonstrated nanosecond-resolved laser plume spectroscopy using a gated intensified charge-coupled device camera; (2) conducted finite-element analysis of our laser-based surface-shaping experiments including hydrogen-assisted material removal, a laser-based chemical-vapor-deposition process, and variable wavelength infrared laser treatments—our models are capable of accurately predicting surface shape under a variety of laser and chemical conditions; (3) installed and tested an infrared laser capable of tuning between 9 and 11 µm and producing up to 188 W of power—additional laser and electrical safety controls were deemed necessary and implemented; (4) demonstrated controlled, local deposition of high-damage-threshold silicon dioxide onto silica optics and developed in situ diagnostics to guide re-planarization; and (5) demonstrated controlled, direct-write deposition over millimeter-length scales using a motorized stage—modeling of quantitative deposition rates are in agreement with experimental results and reveal competition between mass transport and reaction kinetics.



Height map of direct-write, laser chemical-vapor deposition of silicon dioxide using an optical nozzle assembly (inset) for precursor gas delivery.

Proposed Work for FY13

We will (1) continue experimenting with, developing, and modeling our laser chemical-vapor-deposition system as a means of damage mitigation by planarization; (2) explore the use of in situ and ex situ diagnostics, including sum frequency generation and photo-acoustic spectroscopy to probe laser-induced tetraethylorthosilicate polymerization to silica; and (3) study the use of 9- to 11-µm infrared laser light for surface-shaping damage mitigation of fused silica optics in both ambient and chemically active environments (such as tetraethylorthosilicate, dihydrogen, ozone, or dioxygen.

Publications

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Understanding the Stochastic Nature of Laser-Induced Damage

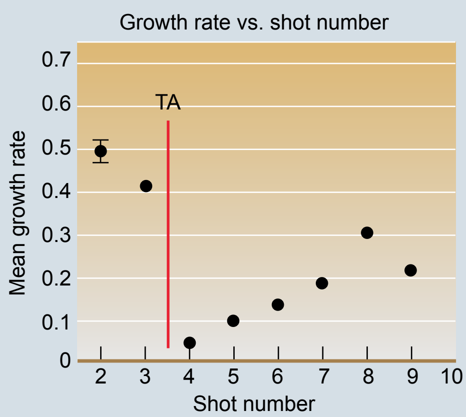
Christopher Carr (11-ERD-030)

Abstract
Laser optic surface damage is of particular concern for laser facilities because even small damage sites only a few micrometers in size can quickly grow large enough to destroy the optic, thereby resulting in damage to downstream optics as well. In fact, laser-induced optics damage will remain a key constraint on the operation of inertial-confinement fusion laser facilities for the foreseeable future. Damage growth is widely believed to be statistical in nature, but the internal features of damage sites are 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 facilities to operate at energy densities high enough to fully enable research in inertial-confinement fusion and laser ignition fusion energy, in support of the Laboratory's missions in stockpile stewardship and advanced lasers and applications.

FY12 Accomplishments and Results
We have (1) quantified a number of physical attributes of damage sites that affect growth rate and probability of growth; (2) began a study of how shot and sample history affects the evolution of damage sites, during which we confirmed expected effects of fluence and pulse duration and shape; (3) discovered unexpected effects of growing sites in materials with different bulk properties and of exposing growing sites to hydrocarbons; (4) demonstrated trends of sites that have very different growth constants; and (5) demonstrated the technique of using environmental and material differences to isolate and examine physical features that affect the rate at which sites grow.



The average growth rate of over 100 damage sites is plotted as a function of shot number. The sample on which the damage sites reside was thermally annealed (TA) between shots three and four. The dramatic drop in growth rate caused by thermal annealing has been attributed to closure of subsurface fractures surrounding the damage sites.

Proposed Work for FY13

We will (1) build on our FY12 experiments to develop empirical rules to describe both the probability of growth and the growth rate of damage sites based on internal features of damage sites—current models for these phenomena take into account only damage site size, (2) combine the description of internal site evolution and our mature description of site growth as a function of laser parameters and diameter with advanced predictive models using Monte Carlo and classification techniques, and (3) complete experiments to help understand the fundamental nature of damage site growth.

Publications

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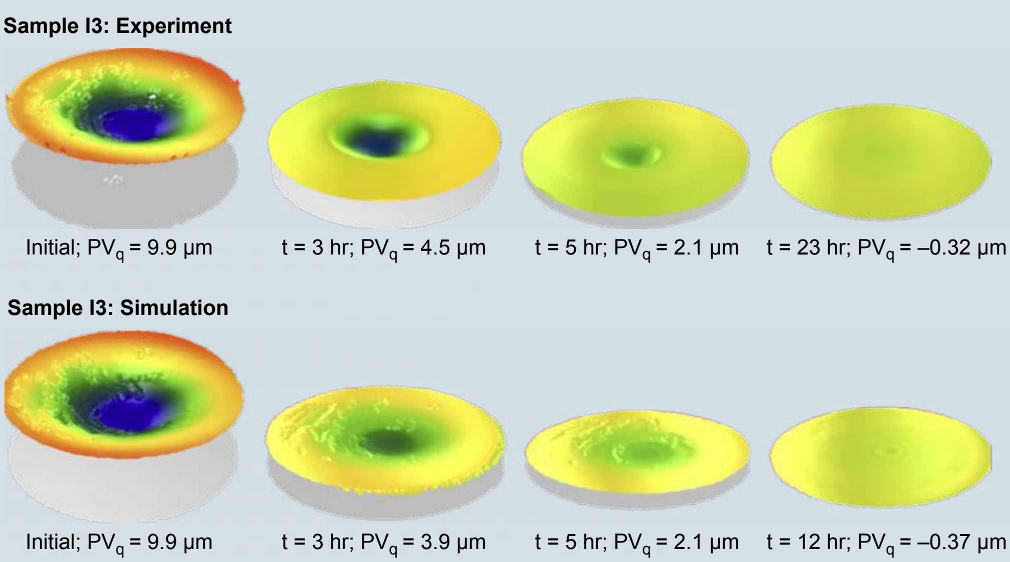
Fundamentals of Figure Control and Fracture-Free Finishing for High-Aspect-Ratio Laser Optics

Tayyab Suratwala (11-ERD-036)

Abstract

With this project, we will scientifically investigate critical phenomena affecting the full-aperture finishing of high-aspect-ratio optical components, conduct full-aperture optical polishing in environments free of rogue particles by understanding and preventing key sources of the particles, and then develop methods to enhance the lifetime and reduce the cost of inertial-confinement fusion and laser inertial fusion optics. To this end, we will perform specifically designed polishing experiments and use models to understand the effects of pad wear, mechanical bending, residual stress, and thermal effects on nonuniform removal. In addition, we will determine methods to control rogue particles, such as using 100% humidity to control particle size distribution. The fusion laser optics that we will target include main debris shields, continuous phase plate substrates, disposable debris shields, and blue and red blockers for use in high-peak-power laser systems such as the National Ignition Facility at Livermore and amplifier glass for high-average-power laser systems.

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



Surface of a fused silica (100-mm-diameter) optical workpiece during polishing using our new convergent method (top row), and simulation of the same polishing conditions using the SurF (sped-up robust feature) simulation code (bottom).

Mission Relevance

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

FY12 Accomplishments and Results

In F12 we (1) studied nonuniform pad wear, workpiece bending, and residual stress as well as developed methods to mitigate their effects on workpiece removal; (2) demonstrated, for the first time, the convergent polishing of 100-mm round, square, and high-aspect-ratio workpieces; (3) developed, modeled, and optimized "pitch button blocking" to hold high-aspect-ratio workpieces during polishing; (4) developed new, novel chemical and filtration methods for improving particle size distributions of cerium dioxide polishing slurries; and (5) measured and modeled the influence of temperature distribution during polishing and its influence on nonuniform material removal.

Proposed Work for FY13

In F13 we will (1) develop methods to improve the long-term (many hundreds of hours) convergence point of workpieces; (2) develop a chemical model to explain the behavior of surfactants in stabilizing particle size distributions without altering particle activity; (3) measure the temperature dependence of the cerium dioxide polishing reaction with silica; (4) expand our understanding of figure control to smaller lengths, including the relationship between size distribution of small slurry particles and measured roughness on the polished workpiece; (5) expand the capability of convergent polishing to large (12-in.) spherical workpieces and other glass types; and (6) investigate our hypothesis of the role of 100% humidity on reducing scratch densities.

Publications

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Hydrogen Melting and Metallization at High Density

Michael Armstrong (11-ERD-039)

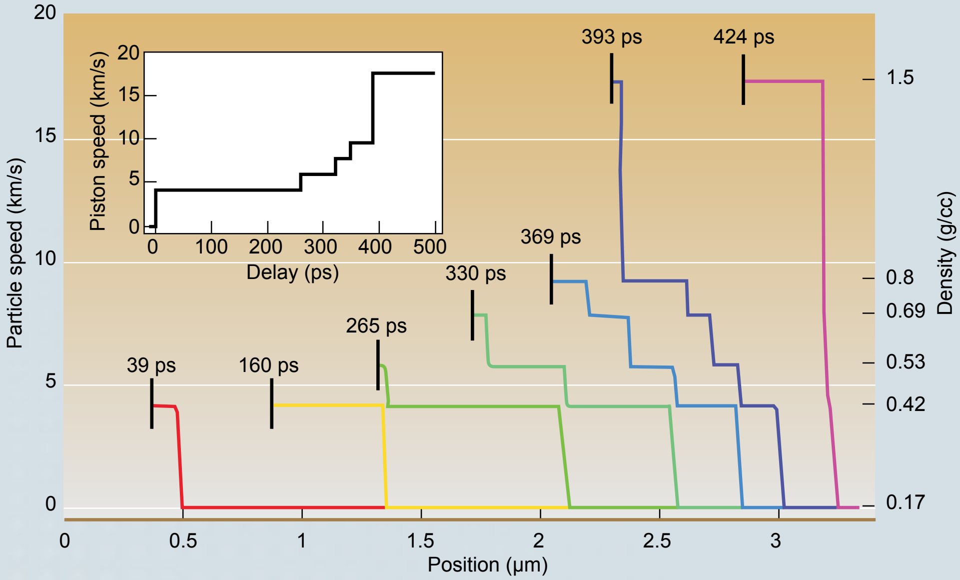
Abstract

The objective of our project is to experimentally determine phase boundaries in hydrogen–deuterium melts and to examine metallization phase transitions at high density (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.

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.



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

FY12 Accomplishments and Results

In FY12 we (1) achieved the quasi-isentropic compression of deuterium in less than 100 ps, which is more than 10 times faster than previously observed—a significant, unanticipated discovery that may enable the compression of hydrogen and isotopes to very high density with substantially less energy than used in conventional experiments using tabletop laser systems; (2) observed a rapid transition suggesting ramp-to-shock transition—results with deuterium pre-compressed to 36 GPa suggest an initial elastic compression and a ramp-to-shock transition with no melting, while results obtained with deuterium pre-compressed beyond 50 GPa indicate an elastic shock compression but no ramp-to-shock transition over the 100-ps time scale of the experiment; and (3) conducted hydrodynamics simulations of compression in cryogenic liquid deuterium, obtaining results that suggest that extreme density can be achieved on ultrafast time scales with a low-energy laser.

Proposed Work for FY13

We will ramp-compress liquid deuterium on a timescale more than 10 times shorter than in FY12, achieving a density as high as 10 times that of initial cryogenic density with a tabletop laser system. At this density range, metallization is expected. We will also simulate the ramp compression of deuterium on an ultrafast time scale to determine the optimum compression profile and experimentally vary the compression profile to adjust the temperature range over which metallization occurs.

Publications

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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 polycrystalline, amorphous, or other microstructure. Our experiments will combine the dynamic diamond-anvil cell with time-resolved x-ray scattering and imaging to study the dynamics of pressure-induced transitions in the dinitrogen molecule.

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

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we used the dynamic diamond anvil cell to study compression-rate-dependent physical processes in a variety of materials using synchrotron x-ray scattering. We studied bismuth, gallium, barium, iron, and tin at the Advanced Photo Source at Argonne National Laboratory and DESY in Germany. Specifically, we (1) performed time-resolved x-ray diffraction at compression rates up to 120 GPa/s—for polycrystalline samples, the minimum time resolution was approximately 2 ms, which is a convolution of the x-ray intensity, scattering cross section, and detector efficiency; (2) determined this limit can be lowered using higher-intensity fourth-generation sources such as the Linac Coherent Light Source at Stanford University or European X-Ray Free Electron Laser in Germany; (3) collected data demonstrating clear compression-rate dependence of the crystal structure and transition in bismuth and analyzed results of experiments on gallium, barium, iron, and tin; and (4) performed time-resolved imaging studies using white-beam x rays with time resolution down to 500 μs to observe pressure-induced crystal morphology and growth, and determined that the limited contrast between solid and liquid phases is a challenge to this approach—monochromatic or pink (narrow-bandwidth) x rays will improve the contrast.

The Livermore dynamic diamond-anvil cell is used to study the influence of compression rate on the pressure-induced freezing, growth, and morphology of liquid metals. Here, researcher Jing-Yin Chen is setting up the anvil cell for operation at the extreme conditions beam line located at the Petra-III third-generation synchrotron x-ray source at DESY in Germany.

Proposed Work for FY13

In FY13 we will continue our studies of time-resolved dynamics with an emphasis on high temperatures, higher compression rates, and imaging to study liquid–solid transitions in mercury, bismuth, and gallium. Specifically, we will (1) use diffraction to measure the kinetics of these materials' solidification, possible metastability, and novel compression-induced phases; (2) use imaging to identify the growth morphology (uniform or dendritic) of the solidification; and (3) conduct time-resolved studies at the Linac Coherent Light Source using the dynamic diamond-anvil cell and the techniques we have refined at third-generation x-ray sources.

Publications

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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, especially for next-generation computing. 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. We propose a unique approach to creating a perfect spin polarization of electrons on the surfaces of nonmagnetic materials by using an electric field, not a magnetic field. We propose to demonstrate the perfect spin polarization of electrons by x-ray magnetic circular dichroism. By closely analyzing the x-ray magnetic circular dichroism spectrum, information can be obtained on the magnetic properties of the atom, such as its spin and orbital magnetic moment.

In this project, we will attempt to develop a new class of nonmagnetic 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 for quantum computing or communications. The novel idea of using an electric field on a nonmagnetic material to create a perfect spin polarization of electrons would have a great impact in the field. Such a nonmagnetic 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.

FY12 Accomplishments and Results

In FY12 we performed x-ray magnetic circular dichroism measurements on the topological insulator bismuth selenide with circularly polarized light. The measurements were performed both without voltage and with a voltage of 5 mV, which showed very different characteristics for the 2P3/2 spin-orbital of selenium. This is because of the spin alignment (magnetic moment) on the surface caused by spin-momentum locking of the moving electrons by the applied voltage. Our data proves that the surface of a topological insulator could be potentially used for highly spin-polarized nonmagnetic materials in spintronics.

Publications

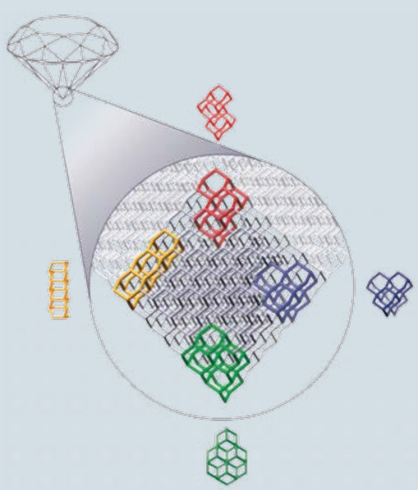
Yu, S. W., and T. W. Barbee, 2012. *Demonstration of electron spin imbalance in two dimensional electronic states*. LLNL-TR-602592.

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. We intend to explore the fundamental properties of diamondoid self-assembled monolayers and their potential to produce clean, thin, or templated 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.



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.

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.

FY12 Accomplishments and Results
In FY12 we (1) successfully fabricated diamondoid monolayers using siloxane and alkene chemistries—a butadiene-functionalized tetramantane formed orientationally ordered thin films; (2) determined that dihydrogen wetting was not significantly different on diamondoids compared to alkanethiols; (3) determined that these diamondoid layers are stable to about 450°C, which should be sufficient for lower-temperature diamond chemical vapor deposition; (4) worked to develop diamond deposition on these diamondoid substrates; and (5) further characterized diamond deposited on these monolayer substrates. Over the course of this project, we submitted a record-of-invention on new methods for forming orientationally ordered and chemisorbed diamondoid monolayers on silicon substrates. In addition, we intend to submit scientific manuscripts for publication and work with collaborators on further development of diamond deposition technology.

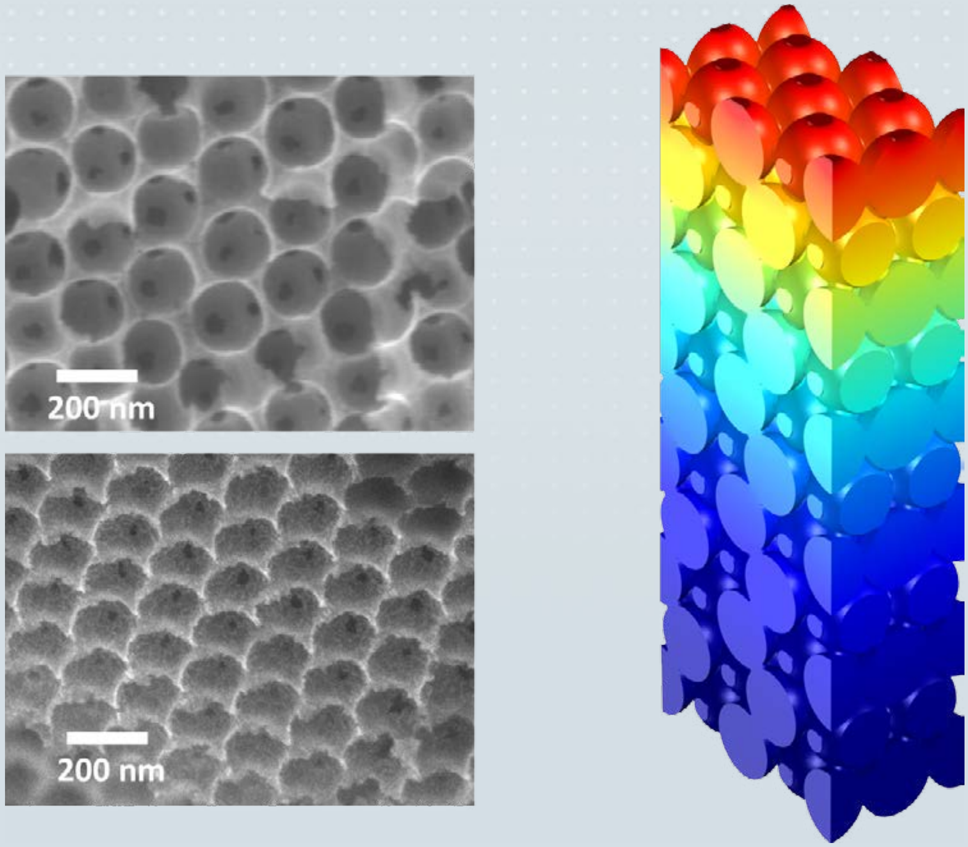
Proposed Work for FY13
In FY13 we plan to (1) examine the transfer of three low doses of TCC from mother to offspring both in utero and through lactation, (2) examine the developmental outcomes in offspring of TCC-exposed mothers, and (3) examine the ability of exposed offspring to reproduce. We expect to see low levels of TCC transfer in utero and greater levels of TCC to transfer through lactation. These low levels will likely impact development and possibly interfere with reproduction.

Mass Transport through Porous Materials

Michael Stadermann (11-LW-037)

Abstract
Porous systems are found in a wide variety of surface-reaction energy applications, from storage to catalysis. The porous structures are used to increase available surface area and thereby enhance the reaction rate. However, mass transport limitations caused by pore size distribution and shape 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.

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



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. Additionally, 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.

FY12 Accomplishments and Results

In FY12 we (1) attempted to deposit nickel oxide into a carbon template, although the carbon material proved to be unstable at the deposition voltages—an unexpected result, because the depositions on surrogate carbon surfaces worked without problems; (2) performed several different treatments on the carbon electrodes to stabilize them, but none worked, and so the template material was switched to nickel, which existing literature suggested would perform well; (3) began deposition with nickel, bringing a different set of problems compared to carbon, a primary one being the high aspect ratio of the nickel-coated template, resulting in sealed pores; (4) successfully fabricated multiple nickel templates suitable for experiments, and determined the deposition conditions for depositing nickel oxide onto these structures, although no transport measurements were completed; and (5) modeled the impact of structure on transport behavior for all proposed geometries, although measurements were outside the scope of this project. A record of invention was filed for our nickel oxide deposition method. The next step would be to find an industrial partner to license the technology and continue developing it under a cooperative research and development agreement. The designer pore structures have also drawn the attention of potential collaborators at Stanford University who are interested in modeling turbulent flow in porous structures.

Publications

Merrill, M. D., M. A. Worsley, and M. Stadermann, in press. "Determination of the NiOOH charge and discharge mechanisms at ideal activity." *Electrochim. Acta*. LLNL-JRNL-539131.

Materials for Inertial Fusion Energy Reactors

Michael Fluss (12-SI-002)

Abstract

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

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

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) identified a set of three ferritic–martensitic steel materials (HT9 modified for reduced activation, EUROFER97, and modified F82H) for multiple simultaneous ion-beam simulations of radiation damage accumulation in a fusion neutron environment; (2) began characterization work and developed techniques for performing micro-mechanical testing as a function of depth in the specimen in collaboration with the University of California at Berkeley; and (3) completed irradiation of EUROFER97 using a multiple simultaneous beam of hydrogen, helium, and iron ions.

Proposed Work for FY13

In FY13 we will focus on a comparison of pulsed ion radiation with multiple ion beams (hydrogen, helium, and iron) to steady-state radiation. We intend to use the neutron-irradiated reduced activation EUROFER 97 for the experiment, which is a highly ranked candidate material for fusion energy first-wall and blanket structural components. We will conduct post-irradiation examination using transmission electron microscopy and micro-mechanical testing using focused ion-beam extracted specimens obtained from the irradiated material. In addition, we hope to develop specimens for spallation neutron irradiations at the Paul Scherrer Institute in Switzerland or at Los Alamos National Laboratory in New Mexico.

Ultrahigh Burn-Up Nuclear Fuels

Patrice Erne Turchi (12-SI-008)

Abstract

One of the key questions for the U.S. as it seeks to incorporate nuclear energy in its clean-energy strategy is how to more completely burn nuclear fuel in its power plants. We propose to make significant advances in the basic science for development and qualification of advanced, ultrahigh burn-up nuclear fuel. To achieve this goal, we will couple modern computational materials modeling, fabrication, and characterization capabilities and targeted performance-testing experiments using ion-beam facilities. This project will establish the scientific foundation for selecting the optimum fuel type for advanced reactor concepts.

Our work combines a robust experimental program with validated modeling. We will experimentally quantify phase stability and kinetics of phase transformations, inter-diffusion, microstructural evolution, micro-mechanical properties, and the influence of severe radiation environments on fuel performance. Ultimately, we will have a validated model for advanced nuclear energy materials under extreme conditions of radiation,

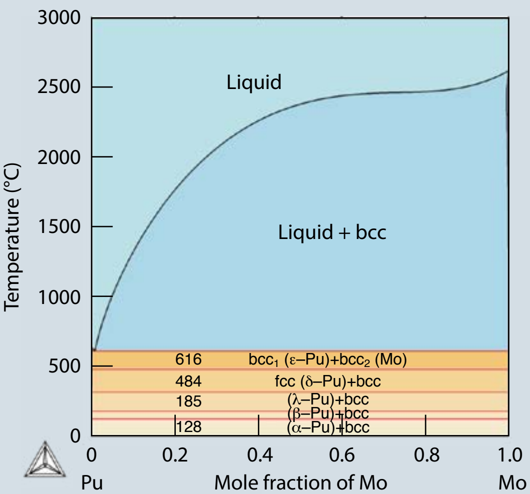
temperature, and evolving chemistry. We will also have a science-based path forward to an optimized inert matrix fuel, while contributing to the development of a validated nuclear-fuel database. Furthermore, the successful accomplishment of this project will augment LLNL's credibility within the nuclear energy community.

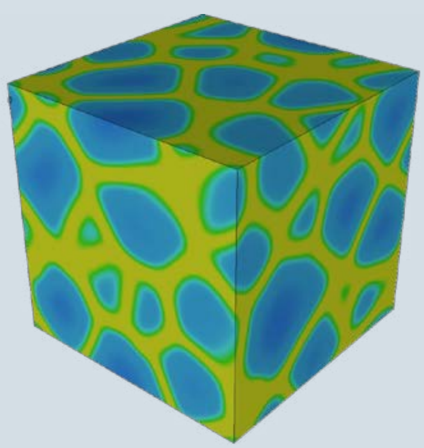
Mission Relevance

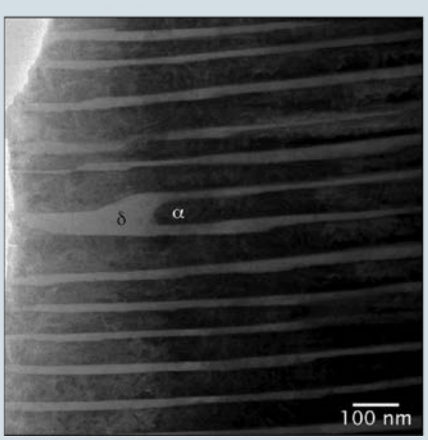
Our approach to developing the science of advanced nuclear energy fuels aligns well with the Laboratory's energy and national security missions. Development of both advanced fuel cycles and hybrid fusion–fission concepts face the same scientific challenges. This research will extend LLNL capabilities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

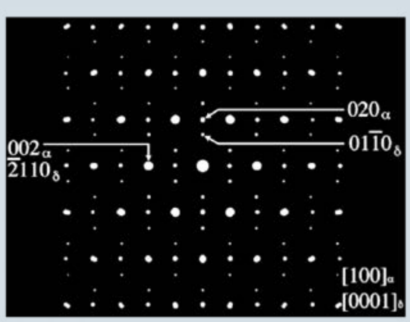
FY12 Accomplishments and Results

In FY12 we extended our ab initio work to include various alloys of americium, neptunium, and uranium, studying whether phase separation or ordering were modified because of irradiation. Specifically, we (1) completed a complete thermodynamic assessment of the plutonium–molybdenum phase diagram with input from ab initio









Thermodynamically assessed plutonium–molybdenum phase diagram with input energetics from ab initio calculations is shown top left. Snapshot of a phase-field simulation of coring effect in gold–nickel is shown top right (green is the liquid phase and blue is the face-centered cubic solid phase, with dark blue areas associated with higher composition of nickel). Transmission electron microscopy analysis of a low-zirconium-content as-cast bulk uranium–zirconium alloy, fabricated at Texas A&M University, shows the existence of the alpha and delta phase microstructure (bottom left). Verification by transmission electron microscopy diffraction analysis of the two-phase microstructure in uranium–zirconium alloys is shown bottom right.

energetics; (2) predicted a new phase in uranium–molybdenum at a three-to-one composition based on ab initio calculations, and developed a preliminary thermodynamic database for molybdenum–plutonium–uranium; (3) applied kinetic modeling to uranium–zirconium diffusion couples to optimize geometry for future ion-beam experiments; (4) upgraded our phase-field code to include thermal gradient effect and non-periodic boundary conditions, and applied it to the effect of coring in alloys and site redistribution in uranium–zirconium in a gradient of temperature; (5) prepared and characterized more samples of uranium–molybdenum, uranium–zirconium, and the metal matrix alloy zirconium–iron–copper at Texas A&M University, and carried out further characterization using x-ray diffraction and transmission electron microscopy as a function of annealing time—our studies on actinide alloys showed that transmission electron microscopy is indispensable to reveal features that were not seen before; (6) observed and characterized, using transmission electron microscopy, as-cast uranium–zirconium bulk alloys and determined the lamellar structure of the decomposition in alpha and delta phases (even at low zirconium composition); (7) observed short-range order in the quenched body-centered cubic uranium–molybdenum alloy—further analysis may reveal the existence of a new phase; and (8) conducted the very first ion-beam experiment on bulk uranium–zirconium and performed post-irradiation examinations to study microstructural changes caused by irradiation effects.

Proposed Work for FY13

In FY13 we propose to (1) synthesize and characterize more uranium–molybdenum, uranium–zirconium, zirconium–iron–copper bulk samples (at other compositions), and uranium–molybdenum and uranium–zirconium diffusion couples; (2) conduct irradiation experiments at Livermore's Center for Accelerator Mass Spectrometry and use the post-irradiation examination facility to study phase formation and diffusion as functions of temperature, dose, and dose rate; (3) continue ab initio work on complex actinide- and zirconium-based coating mixtures and further develop the search engine to predict optimum materials properties; (4) develop, implement, and apply diffusion kinetic modeling to guide and support the experimental effort, with initial focus on uranium–zirconium; and (5) continue extending our phase-field code to non-periodic boundary conditions, study microstructure evolution in bulk and diffusion couple materials in a temperature gradient, and compare our code and studies to experimental results.

Publications

Landa, A., et al., 2012. *Ab initio study of advanced metallic nuclear fuels for fast breeder reactors*. 2012 MRS Spring Mtg. and Exhibit, San Francisco, CA, Apr. 9–13, 2012. LLNL-CONF-552336.

Landa, A., et al., 2012. *Ab initio study of advanced metallic nuclear fuels for fast breeder reactors*. TMS 2012 Ann. Mtg. and Exhibition: Linking Science and Technology for Global Solutions, Orlando, FL, Mar. 11–15, 2012. LLNL-ABS-490670.

Landa, A., et al., 2012. *Ab initio study of advanced metallic nuclear fuels for fast breeder reactors*. CALPHAD XLI, Berkeley, CA, June 3–8, 2012. LLNL-ABS-541312.

Lordi, V., and P. E. A. Turchi, 2012. *Alloy optimization for metallic inert matrix nuclear fuels.* CALPHAD XLI, Berkeley, CA, June 3–8, 2012. LLNL-ABS-558416.

Lordi, V., et al., 2012. *Interdiffusion kinetics in U–Zr*. TMS 2012 Ann. Mtg. and Exhibition: Linking Science and Technology for Global Solutions, Orlando, FL, Mar. 11–15, 2012. LLNL-ABS-491639.

McKeown, J., et al., 2012. *Report on characterization of U-10 wt.% Zr alloy*. LLNL-TR-534973.

Turchi, P., 2012. *Acquisition of diffusion from ab initio: Comments on a good usage of the pseudo-potential method*. 2012 NIST Diffusion Workshop, Gaithersburg, MD, May 3–4, 2012. LLNL-PRES-553992.

Turchi, P., and A. Landa, 2012. *Thermodynamic database, lower length scale-thermodynamic assessment of the ternary alloy system Mo–Pu–U*. LLNL-TR-553775.

Turchi, P., A. Landa, and P. Soderlind, 2012. *Thermodynamic properties of complex actinide alloys*. TMS 2012 Ann. Mtg. and Exhibition: Linking Science and Technology for Global Solutions, Orlando, FL, Mar. 11–15, 2012. LLNL-PRES-535971.

Turchi, P., V. Drchal, and J. Kurdnovsky, 2012. *Ab initio phase diagrams of BCC-based transition metals—Consequences on properties*. TMS 2012 Ann. Mtg. and Exhibition: Linking Science and Technology for Global Solutions, Orlando, FL, Mar. 11–15, 2012. LLNL-PRES-535891.

Turchi, P. E. A., et al., 2012. *Electronic structure and thermodynamics of actinide-based alloys.* Intl. Symp. and Workshop Electron Correlations and Materials Properties of Compounds and Alloys, Porto Heli, Greece, July 9–13, 2012. LLNL-PRES-562907.

Novel Rare-Earth-Based Permanent Magnets

Scott McCall (12-ERD-013)

Abstract
Recent restrictions by China on the export of rare earth elements have prompted concern about the impact a shortage would have on advanced world economies, which is a significant national security concern. The physics of 4f electrons found in rare earth elements make them peerless with respect to potential magnetic properties. Rare earth elements are essential components of the strong permanent magnets

necessary for all technologies requiring a passive magnetic field such as regenerative braking systems in hybrid automobiles, lightweight motor systems for compact hard-disk drives, and advanced megawatt windmills. We propose to create new rare-earth-element permanent magnets by developing a high-temperature synthesis capability and coupling it closely with the world-class capabilities already present at Lawrence Livermore in materials characterization, high-pressure physics, and quantum simulations to rapidly and systematically develop new materials.

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

Mission Relevance

Our development of new high-strength magnets that use fewer (or cheaper) rare earth elements supports the Laboratory mission in energy and environmental security by providing a critical component for a clean, renewable energy source and fuel-efficient hybrid automobiles. In addition, research on rare earth elements can provide insight into the properties of actinide elements, which are important for stockpile stewardship science research.

FY12 Accomplishments and Results

In FY12 we (1) established a high-temperature synthesis laboratory, including building a quartz bench for sealing samples under controlled atmospheres with a hydrogen torch and installing an inert-atmosphere glove box; (2) installed a tetra arc furnace to grow high-quality single-crystal samples to centimeter lengths by heating constituent material to temperatures over 3,000°C, which is an ideal technique for high-melting-point materials; (3) finished installing a furnace for removing and recycling an almost 30-year-old ion implanter; (4) began developing techniques to grow samarium alloy single crystals, including the use of antimony and tin fluxes; (5) mapped the structure versus pressure of polycrystalline samples using diamond anvil cells; and (6) succeeded in modeling the effects of pressure on crystal structure and began a direct comparison of our models with experimental results.

Proposed Work for FY13

In FY 13 we will (1) employ our new sample-growing capabilities to produce single-crystal samples with a 2:17 chemical structure, including nitride or hydride versions; (2) explore related intermetallic compounds and employ chemical doping with guidance from our developing theoretical models; (3) finish characterizing the physical properties of these compounds as a function of pressure, and converge experimental and theoretical measurements of the magnetic moments; and (4) perform spectroscopy as a function of pressure at the Advanced Photon Source at Argonne National Laboratory on our single-crystal samples and related systems to determine site-specific orbital and spin contributions to the overall magnetic moment.

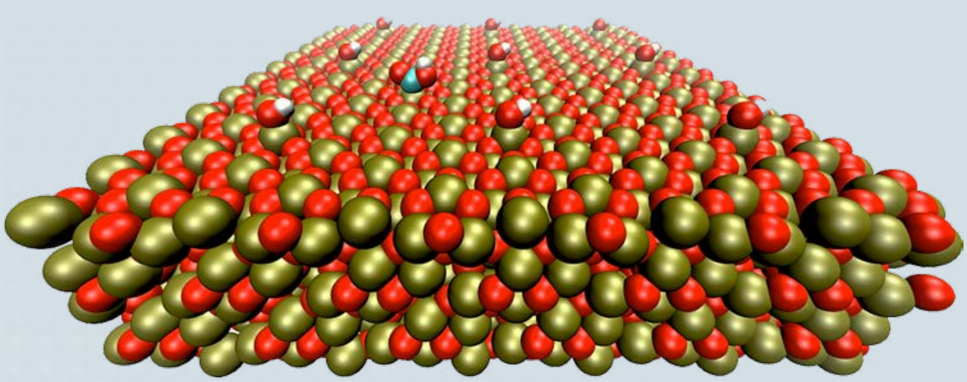
First-Principles Materials Characterization and Optimization for Ultralow-Noise Superconducting Qubits

Vincenzo Lordi (12-ERD-020)

Abstract
The application of superconducting quantum bits (qubits) in quantum information processing is currently limited by unidentified noise sources, which reduce decoherence time to impractical levels. This project aims to use first-principles atomic-scale simulations to develop a quantitative understanding of the microscopic origins of noise in superconducting qubits and devise strategies for reducing the concentration or impact of the sources identified. We will characterize the sources of paramagnetic noise by developing atomistic models of defects in constituent materials and at the interfaces between them, focusing on niobium, rhenium, and aluminum superconductors; aluminum oxide tunnel junctions; and silicon–silicon dioxide substrates, based on the results of experimental superconducting qubit fabrication.

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

Mission Relevance
This project supports LLNL's cyber security mission by furthering the realization of solid-state quantum information processing systems that could play an important role



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

in future cryptography technologies. By developing the science for fundamental computational materials, this project also bolsters the Laboratory's science and technology foundations in materials on demand, high-performance computing and simulation, and measurement science.

FY12 Accomplishments and Results

The goals for FY12 were modified early when experimental data showed that substrate effects were more significant in introducing paramagnetic noise than the metal–substrate interface, because measured noise scaled much more with the area enclosed than the width of the conductor. Consequently, new FY12 goals focused on identifying surface features on sapphire substrates that introduce paramagnetic noise. Accomplishments included (1) generating atomistic models of sapphire surfaces with different terminations, including the bare surface with and without intrinsic vacancies, termination with hydrogen and hydroxyl and a representative moisture-exposed substrate, and termination with two dozen different adsorbates; (2) constraining spin density-functional calculations to determine energy difference from the ground state to the first magnetic excited state for each model; and (3) determining that certain surface terminations, including incomplete hydrogen and hydroxyl passivation (reduction of reactivity), possess low-energy excited magnetic states that can cause flux noise in a superconducting qubit—we are pursuing a series of surface treatment experiments to validate the calculations and test predicted mitigation strategies.

Proposed Work for FY13

In FY13 we will focus on using the results from FY12 to develop passivation strategies for sapphire to minimize localized paramagnetic noise sources on the surface. In particular, we will (1) investigate how exposure to humidity can change surface hydroxyl termination to create a stable surface without localized paramagnetic defects; (2) explore alternate passivation strategies to substitute surface hydroxyls, closely comparing these calculations with experimental results; (3) construct and explore models of the amorphous and heterogeneous surfaces of silica substrates; and (4) implement an analytical tool to link microscopic properties to macroscopic device characteristics.

Publications

Adelstein, N., J. L. DuBois, and V. Lordi, 2012. *First-principles simulation of magnetic defects on the alpha quartz substrate for superconducting qubits*. 2013 MRS Spring Mtg. and Exhibit, San Francisco, CA, Apr. 1–5, 2013. LLNL-ABS-598072.

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

High-Fluence, Multipulse Laser Surface Damage: Absorbers, Mechanisms, and Mitigation

Jeffrey Bude (12-ERD-023)

Abstract

The lifetime and performance of optical systems designed to guide high-photon fluxes are limited by degradation and damage to key optical components at high-photon fluences. Even high-quality optical surfaces without flaws can degrade through extensive multipulse optical stress and can suffer damage from absorption by damage precursors. The mechanisms of this degradation and the nature of these precursors are unknown. We will employ a suite of integrated tasks that closely link processing, characterization, and modeling to develop a scientific understanding of the mechanisms that govern high-fluence optical damage and degradation, and develop techniques to improve the high-fluence lifetime for optical glasses and other related optical materials.

We expect to determine the physical mechanisms of high-fluence damage initiation in optical materials, including the links between absorption and damage and the nature of damage-precursor absorption, and identify the physical origin of high-fluence surface damage precursors on optical glasses and the processes that introduce them onto surfaces during fabrication. We will develop processes to reduce whole-optic fluence damage by reducing precursors and modulation from etched flaws. We will also develop accelerated multipulse optical stress protocols to characterize degradation from billions of pulses and to clarify degradation mechanisms. This work will advance understanding of laser–matter interactions and extend operational lifetime and performance of high-fluence laser systems.

Mission Relevance

This work directly addresses ignition and stockpile stewardship challenges by optimizing the use of large inertial-confinement fusion laser systems and supports inertial fusion energy missions. Reduction of high-fluence damage on optical glasses will allow fusion-class lasers to operate at higher fluences and with reduced sensitivity to contrast. Understanding how optical surfaces degrade under extreme multipulse stress will guide design and use of optics for fusion energy systems, and understanding how absorption leads to damage can clarify the mechanisms of laser damage from contamination, damage in coatings, and the role of radiation-induced defects on damage in optics. More broadly, understanding laser–matter interactions is a frontier problem in condensed matter physics.

FY12 Accomplishments and Results

In FY12 we (1) began tests to measure absorption-front formation thresholds; (2) constructed a high-resolution transmission imaging system that allows high-resolution (<1-μm), high-sensitivity (<0.1%) transmission mapping for wavelengths down to 357 nm; (3) built a high-fluence damage threshold system and successfully mapped precursor densities over ranges previously untested—25 to 150 J/cm^2;

(4) monitored process-induced changes to high-fluence damage precursor densities, which led to process modifications that reduced damage initiation by over twentyfold on small-scale parts; (5) demonstrated that wide-gap salts can be high-fluence precursors, which helped to identify and physically locate classes of precursors—that is, nanometer-scale precipitates; (6) completed and tested the hydrofluoric acid vapor etching system and successfully etched silica samples; and (7) established a small-spot, high-average-fluence test setup, with which we performed initial stress experiments at 355 nm out to 20^6 J/cm^2—equivalent to 7 million inertial fusion energy laser pulses—and measured transmission changes with the transmission imaging system.

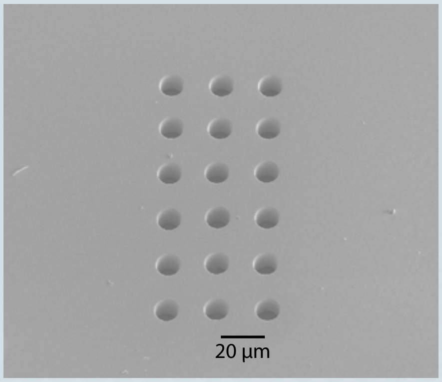
Proposed Work for FY13

In FY13 we will (1) perform tests to measure absorption-front formation thresholds for artificial precursor layers, (2) complete the high-resolution transmission system for high-repetition-rate testing and perform initial tests equivalent to up to 10^9 shots, (3) conduct experiments to identify the mechanisms of nanoscale precipitate formation during liquid-phase silica processing and continue to test methods to reduce these precipitates and correlate them to damage, (4) test etching rates and surface quality achieved with gas-phase etching, and (5) develop and test initiation models based on surface absorption from nanoscale precipitates.

Publications

Laurence, T. A., et al., 2012. "Extracting the distribution of laser damage precursors on fused silica surfaces for 351-nm, 3-ns laser pulses at high fluences (20–150 J/cm^2)." *Opic. Express* **20**(10), 11561. LLNL-JRNL-541972.

Shen, N., et al., 2012. "Thermal annealing of laser damage precursors on fused silica surfaces." *Optic. Eng.* **51**(12), 121817. LLNL-JRNL-539671.



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

A Scalable Topological Quantum Device

George Chapline (12-ERD-027)

Abstract

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

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

Mission Relevance

Lawrence Livermore has a long history of using its superior computer facilities for national security applications, and it is consistent with this mission to develop novel approaches to data processing for national security needs. If successful, our approach to quantum information storage and processing could have a profound impact on this mission. Using three-dimensional topological insulators could be an enabling technology for intelligence and surveillance applications, where because of massive volumes of data, it is difficult or impossible to analyze data in real time.

FY12 Accomplishments and Results

In FY12 we (1) fabricated millimeter-sized bismuth telluride samples doped with antimony and pure bismuth antimony samples using the Bridgman method; (2) measured resistance versus voltage characteristics of the samples at temperatures from room temperature to that of liquid helium, adjusting the antimony doping to maximize low-temperature resistance; (3) measured the transport properties of our samples in a high magnetic field at the National High Magnetic Field Laboratory at Florida State University, demonstrating, in the case of bismuth antimony, the existence of surface conduction states; (4) collaborated with Lawrence Berkeley National Laboratory to develop a method for using focused ion beams to produce 10-µm holes in samples and demonstrated the ability to produce hundreds of holes in our samples; and (5) designed and built an optical cryostat to study the effect of microwave and infrared irradiation on the surface conductivity our samples, so that the cryostat can be used for FY13 experiments, including measuring the effect of holes on the surface resistance.

Proposed Work for FY13

In FY13 we plan to study the transport properties of our sample after exposure to a far-infrared pulse. Our hope is that by tuning the chemical potential to lie in the valence band, we can selectively excite topological surface currents. This work will differ from that originally proposed in that we have decided to use a laser operating at a wavelength of about 8 to 10 µm rather than a terahertz source to excite our samples.

Publications

Qu, D., Y. Hor, and R. J. Cava, in press. "Quantum oscillations in magnetothermopower measurements of topological insulator Bi_2Te_3." *Phys. Rev. Lett*. LLNL-JRNL-594632-DRAFT.

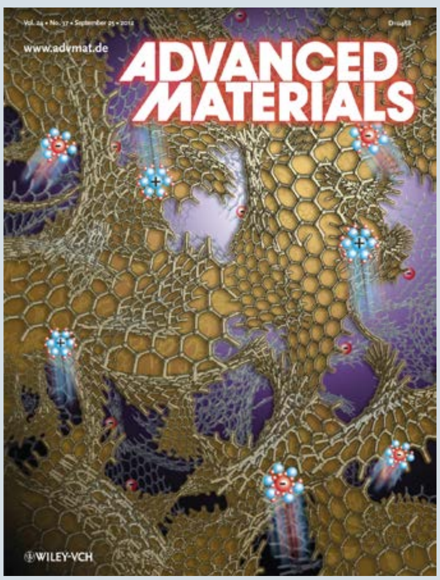
Dynamically Tunable Nanometer-Scale Materials: From Atomic-Scale Processes to Macroscopic Properties

Juergen Biener (12-ERD-035)

Abstract

The future of sustainable energy strongly depends on scientific advances in materials for energy storage and conversion. Material interfaces are of particular importance, because all processes relevant to energy storage, whether physical or chemical, occur here. In classical bulk materials, only a negligible fraction of atoms are surface atoms. Consequently, the interfacial area in these materials is quite small. By contrast, nanometer-scale porous materials—materials with pores smaller than 100 nm—have surface areas so large that the majority of atoms are part of a surface. Because of their high accessible interfacial area, the properties of these materials are thus determined by surface interactions. We propose to establish a fundamental understanding of interfacial charge-transfer phenomena on interface-controlled materials by using a combination of experimental and theoretical tools, and use this information to develop a new class of dynamically tunable three-dimensional (3D) nanometer-scale materials for the next generation of energy-storage technologies.

We expect to develop a fundamental understanding of interfacial phenomena related to electrical energy storage that is needed to develop the next generation of devices for energy storage and harvesting. In addition, we will develop novel 3D graphene-based materials (a honeycomb crystal film of graphitic carbon) with improved electrical energy-storage performance. Ultimately, the project will lead to the development of dynamically tunable bulk materials whose mechanical, chemical, and physical properties can be controlled by interfacial electric fields.



The journal cover that featured our article "Macroscopic 3D nanometer-graphene with dynamically tunable bulk properties." The cover art represents the three-dimensional (3D) structure of a new polymer-derived nanometer-scale graphene bulk material that consists of a 3D network of single-layer graphene nanometer-scale platelets. The material is mechanically robust and combines a graphene-like surface area with a porosity that allows macroscopic properties to be dynamically controlled through ion-induced interfacial electric fields.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) prepared freestanding graphene samples and performed initial electrochemical measurements and simulations to establish a set of design rules for the next generation of 3D graphene electrodes, (2) performed advanced fabrication of 3D graphene monoliths and fabricated the first oxygen-doped 3D graphene materials, and (3) characterized the structural properties and electrochemical performance of 3D graphene, demonstrating that it is a promising actuator material. The proposed boron and nitrogen doping experiments were deferred to FY13.

Proposed Work for FY13

In FY13 we will (1) begin in situ Raman studies of freestanding graphene electrodes; (2) fabricate the second generation of 3D graphene monoliths based on the design rules we established in FY12, (3) develop methods for nitrogen and boron doping and continue oxygen doping, (4) investigate the electronic structure and electrochemical performance of doped graphene materials and explore the concept of electrochemically gated bulk transistor materials, (5) expand atomic-scale modeling by explicit inclusion of applied potential and electrolyte composition, (6) start mesoscale modeling of charge transport in 3D graphene electrodes, (7) evaluate non-aqueous electrolytes, and (8) explore surface engineering for performance improvements.

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Understanding Absorbing Dopant-Ion Oxidation States in Glass Using Redox Chemistry and Dopant Pairs for High-Fluence Optical Filters

Kathleen Schaffers (12-ERD-041)

Abstract

Our objective is to explore and extend the current methods and fundamental understanding of how to achieve precise control of the oxidation state of colored dopant ions (particularly copper and iron ion pairs) in a glass host. With this understanding, we will develop an optic with the appropriate absorption characteristics for a suitable red blocker (high transmission at 351 nm, low transmission at 1,053 nm) for high-peak-power lasers. This will be accomplished through experimental glass melting, doping with metallic ions and ion pairs, material and spectroscopic characterization, and developing a predictive model for choosing laser optical glasses.

The scientific understanding developed from this study will provide the tools to formulate a model for predicting the reduced-to-oxidized ratio of a dopant ion in specific glass hosts as well as the spectral positioning and width of the absorption bands within a glass. We will also develop a capability to fabricate robust optical filters with a controllable quantity of the desired oxidation state of copper and iron dopant ions and the appropriate spectral characteristics. In addition, we will have chosen a suitable glass that also meets the

solarization, damage resistance, and manufacturability requirements for high-power laser applications.

Mission Relevance

The basic knowledge gained from this study can improve upon LLNL's leadership in producing optics with extreme requirements for specific optical applications, in support of the Laboratory's strategic mission thrust in advanced laser optical systems and applications. In particular, the development of a red blocker optic for use in high-peak-power laser systems such as the National Ignition Facility will be critical to improving efficiency for ignition.

FY12 Accomplishments and Results

In FY12 we (1) characterized three base-glass systems for suitability as a red blocker filter for high-fluence lasers; (2) conducted single-dopant-ion studies in both the base-glass systems and in solution, leading to the determination that single dopants are not sufficient to control the oxidation state of the dopant ion; (3) began oxidation–reduction ion-pair studies in solutions and in representative base-glass systems, identifying a dopant pair that meets the spectroscopic requirements, after which we began work to better understand properties in various hosts, including fused silica; (4) set up a laser system to study the high-fluence behavior of glasses and characterized all of the doped systems for spectroscopic properties, including the solutions and doped glasses produced, and the high-fluence behavior for many of the systems; and (5) began developing a suitable model to simulate and predict spectroscopic properties, including performing benchmarking solution experiments with a model to begin learning about oxidation state and coordination environments.

Proposed Work for FY13

In FY13 we will define the best host material for an iron–tin–carbon dopant system, focusing on fused silica as a good candidate. Specifically, we will (1) continue dopant-ion-pair studies, with an emphasis on understanding dopant concentrations and effects on the oxidation–reduction reaction; (2) perform solution studies to support both the host material and modeling efforts; (3) explore the ability to dope iron–tin–carbon into a fused silica host; (4) complete a working model to simulate the optical and chemical properties of the dopant ions and host materials studied; and (5) fully characterize the high-fluence behavior of the most promising materials.

Multiscale Capabilities for Exploring Transport Phenomena in Batteries

Ming Tang (12-ERD-053)

Abstract

We propose to build state-of-the-art multiscale capabilities for modeling transport phenomena in batteries. Such capabilities will overcome limitations of the traditional

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

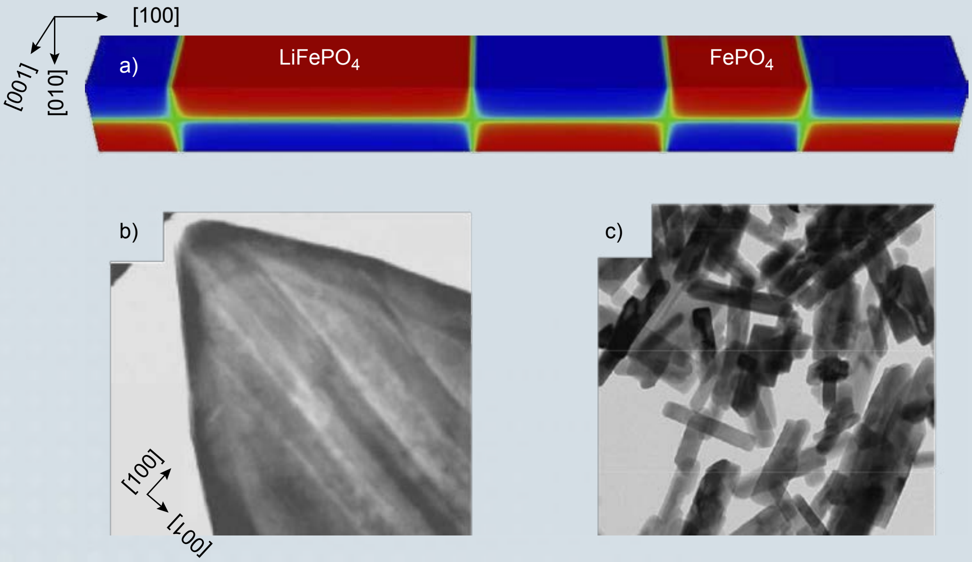
If successful, we expect to produce comprehensive multiscale capabilities for modeling charge-transport processes in batteries that will position LLNL at the frontier of high-performance computing relevant to energy storage technology. We will create a high-performance code for efficiently performing large-scale battery cell-level simulations, and a mesoscale model that quantitatively explains and predicts the important coupling phenomena in nanoscale structured electrodes and their influence on battery performance. This research will produce an improved scientific understanding of the charge-transfer kinetics at the electrolyte and electrode interfaces and implications for electrode microstructure optimization.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we made progress in both the modeling and experimental study of cathode materials for lithium-ion batteries. Specifically, we (1) used theory and phase-field



Simulated $LiFePO_4$ and $FePO_4$ domain structure in an olivine particle from phase-field modeling (top) compared with the experimental observation (bottom left). The $LiFePO_4$ nanometer-scale rod particles synthesized by the hydrothermal approach are shown bottom right.

modeling to shed light on the intriguing domain structure experimentally observed in olivine ($LiFePO_4$) cathode particles, discovering that the ordered domain structure originates from a unique surface mode of spinodal decomposition upon particle lithium loss; (2) used modeling predictions to help determine electrochemical conditions for domain formation and its implications for battery performance; (3) successfully synthesized and characterized a series of nanoscale $LiFePO_4$ particles with tunable sizes and morphologies using the hydrothermal method—the electrochemical performance of these particles will be subsequently tested to validate model predictions; and (4) completed the apparatus setup for the in situ synchrotron soft x-ray spectroscopic measurement of battery electrode materials on a new beam line at the Advanced Light Source at Lawrence Berkeley National Laboratory.

Proposed Work for FY13

For FY13 we will (1) develop a phase-field model for simulating charge transport in electrolytes and integrate it with the model for solid electrodes; (2) perform first-principles calculations of lithium transport properties in nanoscale electrodes and provide parameter input to the phase-field model; (3) develop synthesis routes for nanoscale lithium insertion cathodes such as lithium iron phosphate and anodes such as silicon and carbon, with tunable morphology; (4) collect in situ synchrotron x-ray spectra of nanoscale electrodes in electrochemical cycling and analyze the results; and (5) conduct transmission electron microscopy observations of the charge and discharge processes in nanoscale cathodes.

Publications

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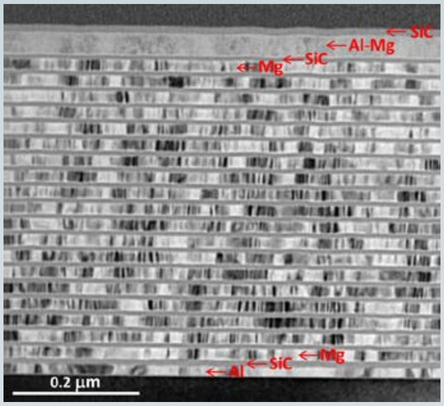
Multilayer Thin-Film Science for Core Missions

Regina Soufli (12-ERD-055)

Abstract

Unique scientific facilities and strategic missions are emerging that require beyond-state-of-the-art multilayer thin-film coatings to produce efficient optical systems for laser fusion systems. Such advanced multilayer coatings do not exist today. We propose new research on fundamental multilayer thin-film science topics including roughness and microstructure manipulation, corrosion mitigation, defect reduction, and smoothing. We will develop high-performance, ultrashort-period, corrosion-resistant x-ray multilayer coatings and defect-free dielectric multilayer coatings for applications to inertial-confinement fusion and inertial fusion energy programs.

We expect our advanced ultrashort-period and corrosion-resistant multilayer coatings will enable x-ray imaging at the Linac Coherent Light Source free-electron laser at Stanford University, and significant improvements in National Ignition Facility diagnostics at Livermore. The new coatings will also provide enhancements to nuclear radiation-detection systems, as well as increase scientific capabilities for optics relevant to next-generation solar physics and astrophysics missions. Our defect-free dielectric multilayer coatings will be resistant to laser damage and would be an enabling technology for successful operation of fusion energy programs at Lawrence Livermore.



Transmission electron microscopy cross-sectional image of a magnesium–silicon carbide (Mg–SiC) multilayer coating with an aluminum–magnesium (Al–Mg) corrosion barrier layer located underneath the top SiC layer. The Al–Mg layer was created by depositing a magnesium layer followed by an aluminum layer. The two layers intermix spontaneously to create the Al–Mg layer.

Mission Relevance

The proposed research is well aligned with Livermore's strategic mission focus area of advanced laser optical systems and applications, through development of multilayer coatings for wavelengths near and below 6.8 nm—a key technical need for x-ray astronomy, radiation detection, photolithography, microscopy, National Ignition Facility diagnostics, and free-electron laser experiments. The project also supports the Laboratory's core competency in materials on demand and measurement science and technology.

FY12 Accomplishments and Results

In FY12 we met all of our deliverables. Specifically, we (1) elucidated the origins and propagation of atmospheric corrosion in magnesium–silicon carbide (Mg–SiC) multilayer reflective coatings; (2) demonstrated efficient aluminum–magnesium (Al–Mg) corrosion barrier layers for Mg–SiC; (3) discovered the process of spontaneous intermixing of crystalline, nanometer-scale aluminum and magnesium layers, which results in the Al–Mg corrosion barrier; (4) demonstrated corrosion-resistant Mg–SiC coatings with high reflectance in up to three narrow bands in the wavelength region of 25 to 80 nm; (5) achieved the highest reported narrowband peak reflectance (40.6%) at a wavelength of 76.9 nm and near-normal incidence angles; and (6) achieved a breakthrough in micrometer-scale coatings, using defect planarization to successfully demonstrate greater than 90% defect height reduction—exceeding our FY14 goal of 75% for 1- and 2-µm features—and created mirror coatings with a laser resistance greater than 100 J/cm^2 (at 10 ns and 1064 nm), a more than tenfold improvement over control coatings not created with planarization.

Proposed Work for FY13

In FY13 we propose to (1) continue studying the corrosion and aging of Mg–SiC multilayers; (2) perform further studies (including molecular dynamics modeling) of the physics of the Al–Mg corrosion barrier structures we developed to elucidate the intermixing and phase-change properties crucial to corrosion resistance; (3) further optimize performance of the corrosion-resistant Mg–SiC multilayers; (4) optimize the silicon oxide and hafnium oxide multilayer defect-planarization process (for inertial-confinement fusion and inertial fusion energy applications) to reduce the processing time (currently at 26 hours)—which includes exploring simultaneous deposition and etching; and (5) conduct electric-field modeling (using finite-element analysis) and laser damage testing of the multilayer to determine the magnitude of planarization needed to realize high-fluence mirror structures over embedded defects.

Publications

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Super-Strained Three-Dimensional Semiconductors

Lars Voss (12-LW-043)

Abstract

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

We expect to enable a new class of highly tunable semiconductor devices that do not rely on heterogeneous structures of differing materials for control over the electronic properties. In addition, the external strain achieved through our approach will allow for greater volumetric effect and will decouple the choice of materials from the achievable strain. Thus, the approach will impact devices that rely on both single semiconductors and multiple semiconductor heterogeneous structures, enabling performance beyond what is currently achievable.

Mission Relevance

This work furthers NNSA and LLNL missions by impacting a broad array of semiconductor devices. These include both laser and light-emitting diodes in support of Livermore's strategic mission thrust in inertial confinement fusion energy; gas, radiation, and other types of sensors in support of stockpile stewardship and nuclear threat elimination; and energy-harvesting technologies such as photovoltaics and thermoelectrics in support of energy security.

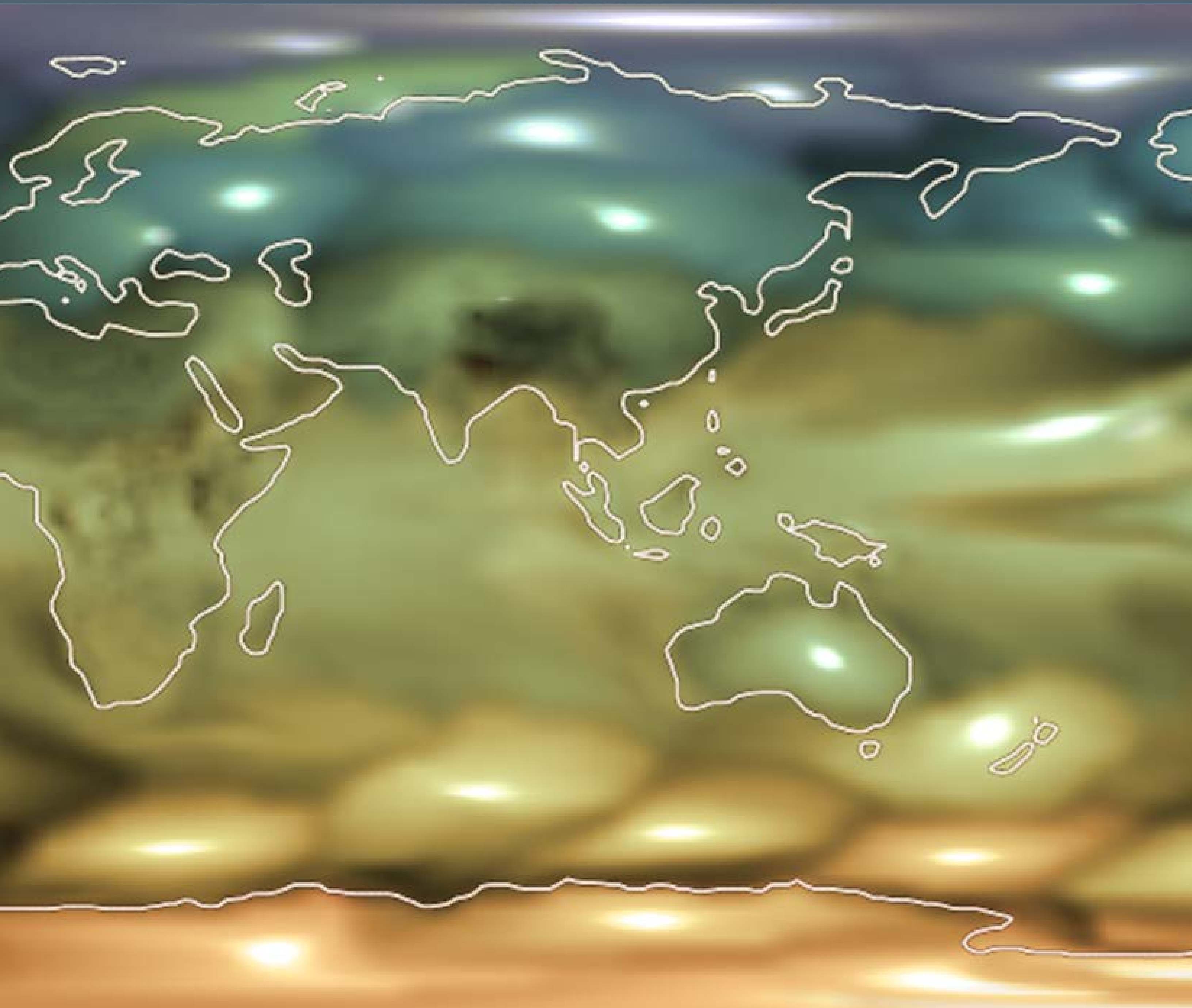
FY12 Accomplishments and Results

In FY12 we (1) fabricated three-dimensional microstructures using dry etching on both silicon and gallium arsenide substrates, creating microstructures ranging in aspect ratio from 2:1 to 25:1 and thereby providing the necessary platform for further work; (2) demonstrated controllable strain on silicon microstructures, using Raman spectroscopy to measure the tensile and compressive strain of silicon microstructures made with silicon dioxide and silicon nitride coatings; (3) developed characterization methods for measuring band gap shifts in our structures using electrical testing, including creating a reliable temperature-monitoring setup enabling measurement at temperatures lower and higher than room temperature and examining several different methods of extracting the band gap from the resulting data; (4) conducted preliminary work on using first-principles simulations with density functional theory; and (5) established a collaboration with the Molecular Foundry at Lawrence Berkeley National Laboratory to grow nanometer-scale wires for our experiments in FY13.

Proposed Work for FY13

In FY13 our work will focus on applying what was learned in FY12 to actual semiconductor devices. Specifically, we will apply compressive and tensile strains to various geometries on devices such as p–n diodes (a p-type semiconducting layer joined to an n-type semiconducting layer) and light-emitting diodes to achieve control over the electronic properties of these devices, and continue work on modeling these structures.

Mathematics and Computing Science



Laboratory Directed Research and Development

FY2012

The Advance of Uncertainty Quantification Science

Richard Klein (10-SI-013)

Abstract

Uncertainty quantification deals with the propagation of uncertainty in multiphysics 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 multiphysics and multiscale 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, quantification and reduction of uncertainty in the inertial-confinement fusion design process will optimize target designs so that they are more likely to perform as intended and less damaging to laser operations.

FY12 Accomplishments and Results

We (1) developed, implemented, and assessed methods for both adjoint error estimates and direct error estimate evolution for a series of increasingly difficult partial-differential equations, including both linear and nonlinear hyperbolic systems of equations, parabolic systems of equations, advection-diffusion equations, Burgers' equation, time-split equations, shallow-water equations, and equations describing a porous medium; (2) developed multiple adaptive sample refinement methods that combined feedback from multiple response models with different adaptive sample criteria for use in high-dimensional sample spaces; (3) tested the methodologies for a series of problems with increasing high dimensional complexity, quantitatively assessing how well these various adaptive sample refinement methods did in competition with each other, and began preliminary work on applying these methodologies to the climate model; (4) developed a novel aggregation methodology that combines several adaptive sample refinement approaches to balance several often-competing criteria, rather than optimizing any individual one, to produce the best sample for a given candidate set of new sample training points and thereby act as a decision-analysis engine in the uncertainty qualification pipeline; (5) began implementing the decision-analysis engine into the pipeline and testing the new aggregation methodology, including increasingly complex high-dimensional functions; (6) extended the aggregation methodology to use a learning-improvement approach that gives further intelligence to the decision-making capability; and (7) developed a new set of topological approaches, including Morse–Smale complexes, topological spines, and neighborhood-based adaptive sampling, to more accurately describe the structure of high-dimensional uncertainty spaces, and applied these techniques to a climate model problem with a 28-dimensional uncertainty space. In summary, this project developed a powerful set of uncertainty qualification methodologies; a powerful, self-guided uncertainty qualification pipeline incorporating these methodologies; and applications using these methodologies to problems important to stockpile stewardship, global climate change, and inertial-confinement fusion. The NNSA Office of Advanced Simulation and Computing has provided support to continue this research to develop advanced uncertainty qualification approaches associated with numerical error in codes, topological adaptive sampling, sparse and scalable emulators, aggregation in the context of adaptive sampling using regression models with multivariate response, and application of these uncertainty qualification methods to predictions accompanied by uncertainty in stockpile devices.

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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 (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 compute 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. This will be accomplished 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.

FY12 Accomplishments and Results

In FY12 we (1) developed a new version of AMG (the algebraic multi-grid algorithm), a benchmark of multicore-aware, sparse linear solvers; (2) demonstrated the automated root-cause analysis of coding errors in message-passing interface programs at scale on BlueGene/P supercomputing systems as applied to several programs including ddcMD (domain decomposition plus molecular dynamics); (3) developed a prototype implementation of techniques that asynchronously redistribute computational load for our test bed applications; and (4) developed and implemented an asynchronous tree-based checkpoint offload engine. The successful conclusion of this project enabled new scalable algorithms for multicore systems, new programming methodologies to use those systems effectively, and highly scalable and novel techniques for debugging on extremely large-scale systems and to improve the resilience of programs running on them. Nearly all work of the project will continue through a variety of sources, including NNSA's Advanced Simulation and Computing program and the Office of Science's Scientific Discovery through Advanced Computing program.

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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, and a powerful 2009 attack that targeted dozens of government and private sites underscores how unevenly prepared the U.S. government is to block such multipronged assaults. We propose to develop a prototype discrete-event simulation using the QWarp platform (formerly named NetWarp) 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 QWarp 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, faster, 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.

FY12 Accomplishments and Results

In FY12 we further advanced the capabilities of QWarp and BackStroke. Specifically, we (1) demonstrated the ability of QWarp to simulate a virtual network comprising a thousand nodes or more in a scalable, parallel manner in a network simulation based on the NS-3 network simulator 3, including the unique capability of QWarp to both maintain proper synchronization among all of the Virtual Machine nodes and NS-3 nodes and accurate timing; (2) showed that QWarp supports nodes that use Intel x86 processors or ARM processors and that are running either Linux or Windows operating systems—or, presumably, any other system using a processor supported by the Quick Emulator standard for hardware simulation—as well as any client, server, or routing applications; (3) demonstrated the ability of BackStroke to completely automatically generate reverse code for a significant subset of the C++ language, including on at least one realistic, optimistic simulation model of airports and air traffic using the Rensselaer Optimistic Simulation System, with a runtime almost as good as that of hand-generated code; and (4) published papers about our work. In summary, this project has demonstrated that QWarp enables a kind of "multiscale" parallel network simulation in which some parts of the model are composed of ordinary NS-3 nodes and some are full-blown virtual machines running real operating systems and application software, and does so in a way that all parts of the model are properly synchronized and the timing is accurate. The next step is to demonstrate the use of QWarp in real cyber security scenarios and to further improve the integration with

NS-3. We also demonstrated BackStroke as a means of enhancing optimistic discrete event simulation by making it easier to use reverse computation, by automatically generating reverse code for functions and methods written in C++, avoiding the writing and debugging by hand of this code. The next step is to extend BackStroke to cover more parts of the C++ language, including arrays, pointers, and standard libraries and create a library of test cases and benchmarks to demonstrate its utility.

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" multigrid 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 will bridge the gap between the larger scale atmospheric modeling being performed at LLNL and turbine-scale flows, and therefore 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 multigrid 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 the 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 multigrid algorithms and moving-grid-generation algorithms developed under this proposal will impact various other simulation fields.

FY12 Accomplishments and Results

In FY12 we (1) improved the performance and robustness of the approximate factored compact scheme—showing the new scheme to be two orders of magnitude faster than the baseline scheme while using an order of magnitude less memory—and performed verification and validation of a number of configurations with moving geometry; (2) made important improvements to the fourth-order-accurate parallel multigrid solver, which was demonstrated to be hundreds of times faster than state-of-the-art Krylov solvers, while also requiring an order of magnitude less memory; (3) demonstrated robust and efficient parallel, moving-grid simulations on wind turbine and other configurations; (4) evaluated the large-eddy simulation turbulence model for use with the new approximate factored compact scheme; and (5) presented our results in publications and at talks at conferences and universities and completed a number of publications. In this project, we successfully developed a new, extremely efficient, parallel algorithm for simulating three-dimensional incompressible flows with moving geometries using high-order-accurate numerical approximations that will have an impact on a wide class of problems of importance to the DOE, including—but not limited to—the modeling of wind turbines and wind farms. The capabilities developed under this project are already slated to be used in upcoming projects, such as wind park modeling to be conducted with an industrial partner and evaluating new large-eddy turbulence models and near-field turbine models in collaboration with several universities.

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, Netherlands, July 10–15, 2011. LLNL-CONF-483351.

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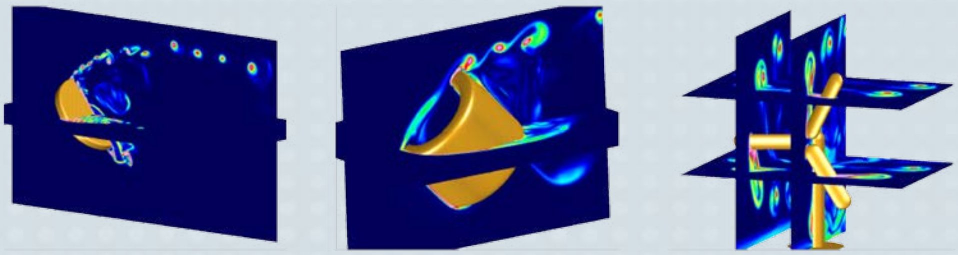

An example of using the new high-performance simulation capability created in this project to accurately and efficiently solve difficult problems involving turbulent fluid flows and moving structures.

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.

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Binary Analysis

Daniel Quinlan (10-ERD-039)

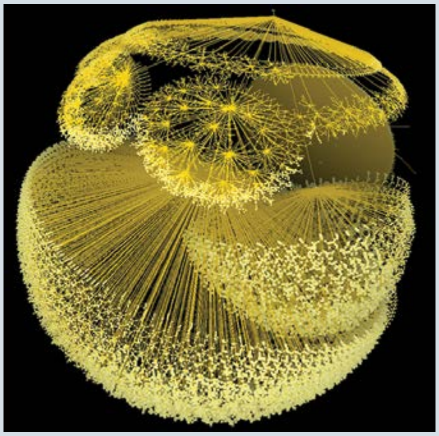
Abstract

We propose to develop new binary analysis techniques required for the analysis of software and, in particular, for the evaluation of specific behavioral properties that are critical for cyber security classification software. Our work will also address the use of parallel machines for analysis of binaries and 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 widely used software in 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.



This is an image of the structure of a binary executable and shows that the different parts have separately complex substructures that are key to reading and interpreting the binary executable.

FY12 Accomplishments and Results

In FY12 we worked with Carnegie Mellon University's Computer Emergency Response Team (CERT) to analyze many new difficult forms of binaries and develop the formal-methods proofs associated with understanding them and supporting custom forms of analysis of them. We released our results—expertise and infrastructure—through the ROSE framework for access by new collaborators attracted by this project, including CERT and the DOE Office of Electricity Delivery and Energy Reliability. In summary, we developed infrastructure and tools for binary analysis that led directly to new cyber security capabilities at LLNL. Our work led to support from the DOE Office of Electricity Delivery and Energy Reliability to focus on analyzing binaries as part of supply chain integrity. We also expect interest directly from electric utilities.

Publications

Quinlan, D., and C. Cohen, 2010. "Discovery of C++ data structures from binaries." *Proc. 6th Ann. Workshop on Cyber Security and Information Intelligence Research*. LLNL-ABS-428049.

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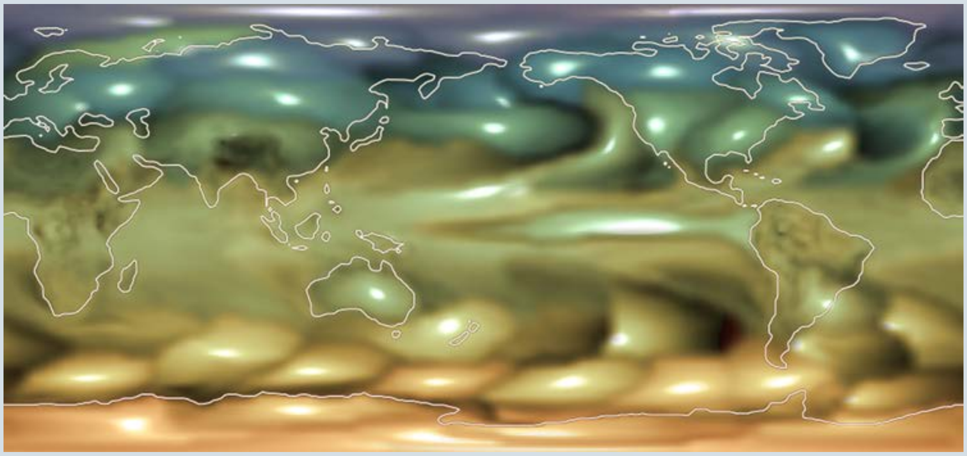
Uncertainty Visualization

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 computational 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 the exploration of high-dimensional merit



Automatic detection of climate zones via nonlinear correlation of temperature distributions. Well-known climate patterns such as the El Niño Southern Oscillation and the Pacific Decadal Oscillation appear as salient features, with bright highlights indicating the most central climate tendency for each region.

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 as well as 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 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.

FY12 Accomplishments and Results

In FY12 we (1) developed a technique for visualizing, analyzing, and clustering spatially varying distributions in random fields; (2) devised a new affinity measure for spectral clustering based on beta skeletons and bilateral filtering; (3) proved the optimality of Delaunay triangulations, a method for representing a set of sample points, for gradient-based topological segmentation; (4) developed a randomized algorithm for computing approximate Delaunay graphs with statistically important edges; (5) made a comprehensive study of bases and optimization schemes for compressive sensing of climate data; (6) devised an asymptotically faster algorithm for computing empty region graphs; (7) defined several novel weighted proximity graphs, including natural neighbor, spectral neighbor, and perturbed empty region graphs; and (8) invented a new nonlinear correlation measure for random variates based on sets of points (manifoldness). This project successfully achieved its two primary goals of developing visual analysis techniques for depicting high-dimensional and often irregularly and sparsely sampled functions arising in uncertainty quantification parameter studies, as well as visually representing the uncertainty in random fields and the statistical dependence between variables. These techniques have been adopted in data analysis applications across several Lawrence Livermore programs that have committed to follow-on funding. Our results are more broadly impacting LLNL programs and the science community through integration with the Laboratory's uncertainty quantification pipeline and through the release of independent software libraries.

Publications

Correa, C. D., and P. Lindstrom, 2012. *Locally-scaled spectral clustering using empty region graphs*. ACM SIGKDD Conf. Knowledge Discovery and Data Mining, Beijing, China, Aug. 12–16, 2012. LLNL-CONF-513768.

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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 network 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 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 has 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 developed in this project can be transferred and developed for a host of related applications in data mining and analysis, such as 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.

FY12 Accomplishments and Results

We have met all our objectives for FY12. Specifically, we (1) performed multilevel research that enabled creation of a parallel multilevel eigensolver and tested it successfully on up to 25,600 processors; (2) discovered that low-rank approximations can be used for seed-set expansions, an important application of interest to DOE and external sponsors; (3) discovered a fast algorithm using the pseudo-inverse of the Laplacian matrix of a graph, and showed it to be equivalent to filtering global effects, giving accurate rankings at a cost lower than spectral decompositions—however, this method must be recomputed for every new query; (4) developed prototype low-rank update approximations as an alternative to costly standard updating methods—fully developing these new methods are beyond the scope of this project, but the work we did forms a basis for further investigation; and (5) delivered our codes to both the internal and external community, who began integrating it into their own research. We have shed considerable light on the use of eigensystem decompositions for clustering, community identification, and seed-set expansions in the power-law graphs common to data mining, cybersecurity, and intelligence applications. Moreover, we have integrated our algorithms into efficient parallel codes for processing extremely large graphs on massively parallel supercomputers. We have delivered our research discoveries and our codes to Livermore's Sequoia supercomputer project and its external sponsor, and both are integrating the research and codes into their continuing efforts. We intend to perform follow-on research that builds upon the work performed for this project.

Publications

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Yoo, A., at al. 2011. *Graph analytics on HPC*. SC11—Connecting Communities through HPC, Seattle, WA, Nov. 12–18, 2011. LLNL-PRES-501071.

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 a thousand, 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 server 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–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. The approach will also allow increased system reliability and greatly reduce the burden of inserting fault tolerance into the computing system, as well as enable 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.

FY12 Accomplishments and Results
In FY12 we (1) improved and further validated the persistent-memory simulator and transitioned the code base into a memory-mapped fault handler serving pages from a mapped file in the flash file system—the handler was released to open source, and flash array vendors have expressed interest in adopting it; (2) developed a dynamic-memory allocator and demonstrated its use in three simulation codes—the LULESH shock hydrodynamics code, the LAMMPS parallel molecular dynamics simulator, and the ParaDiS dislocation dynamics simulation code (additionally, the allocator was used by

others to construct a bioinformatics database); (3) demonstrated a data stream filter running on our storage controller emulator; (4) demonstrated our graph algorithm on the BlueGene/Q supercomputing system with 131,000 cores; and (5) designed a priority queue manager for use in the controller.

Proposed Work for FY13

In FY13 we will (1) complete the pages that serve the memory-mapped fault handler, implement it in persistent memory, and evaluate its performance; (2) design, implement, and evaluate the application programming interface for active storage controllers for host-to-storage processing communication; (3) implement multiple persistent heaps (large pools of memory) and demonstrate them in an application; (4) implement a library mechanism to bundle methods that access persistent heap objects with the specific heap; and (5) extend the checkpoint scheme to an in situ analysis usage case.

Publications

Ames, S., M. B. Gokhale, and C. Maltzahn, 2011. *A searchable file system metadata service based on a graph data model*. 2011 IEEE 6th Intl. Conf. Networking, Architecture, and Storage (NAS), Dalian, China, July 28–30, 2011. LLNL-CONF-454941.

Ames, S., M. B. Gokhale, and C. Maltzahn, 2011. "QMDS: A file system metadata management service supporting a graph data model-based query language." *NAS '11—Proc. 2011 IEE 6th Intl. Conf. Networking, Architecture, and Storage*, p. 268. IEEE Computer Society, Washington, DC. LLNL-JRNL-518611.

Essen, B. V., and M. Gokhale, 2011. *Unifying memory and storage with persistent–random access hardware*. LLNL-PRES-506891-DRAFT.

Pearce R., M. Gokhale, and N. Amato, 2011. *A memory mapped approach to checkpointing*. 6th Parallel Data Storage Workshop, SC11—Connecting Communities through HPC, Seattle, WA, Nov. 13, 2011. LLNL-CONF-499091-DRAFT.

Pearce R., M. Gokhale, and N. Amato, 2010. *Multithreaded asynchronous graph traversal for in-memory and semi-external memory*. SC10—The Future of Discovery, New Orleans, LA, Nov. 13–19, 2010. LLNL-CONF-390163.

Pearce, R., M. Gokhale, and N. Amato, 2011. *Traversing massive graphs in NAND flash*. LLNL-POST-507571-DRAFT.

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Wong, D., and M. Gokhale, 2011. *A memory mapped approach to checkpointing*. 6th Parallel Data Storage Workshop, SC11—Connecting Communities through HPC, Seattle, WA, Nov. 13, 2011. LLNL-CONF-499091-DRAFT.

Compressive Sensing for Wide-Area Surveillance

Paul Kidwell (11-ERD-022)

Abstract

This project will, for the first time, 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 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

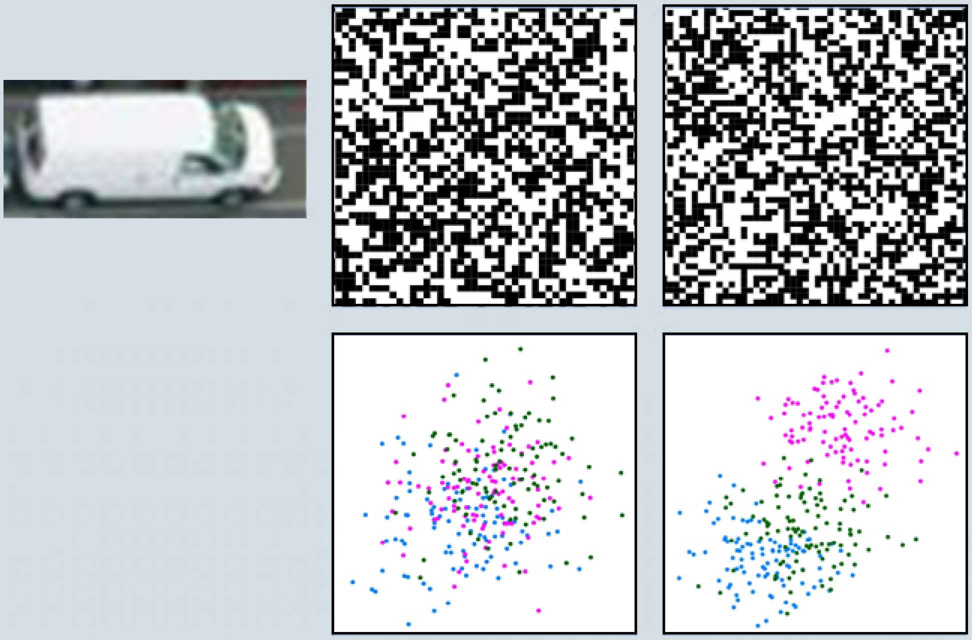
The innovations achieved in this project will further the Laboratory's mission in cyber security, space security, and intelligence by developing advanced data-management capabilities that will yield higher-quality intelligence in wide-area aerial surveillance.

FY12 Accomplishments and Results

In FY12 we (1) developed a classifier capable of operating on lower-dimensional signals; (2) designed a linear projection matrix to minimize data collection while maximizing separation between classes in the compressed domain; (3) compared the classifier's performance on real datasets to that using linear discriminant analysis and information discriminant analysis codes, achieving favorable results; (4) generated background models directly from the compressed signal—it was shown that under this operational paradigm when using a literal background subtraction, a minimum of an 80% reduction in foreground sampling requirements is achievable on operational wide-area motion imagery data; and (5) developed an automated tracking algorithm capable of operating directly on compressed signals and estimating location directly, without the need to reconstruct each image, and used it on a small-scale video sequence to produce results comparable to those obtained by traditional algorithms.

Proposed Work for FY13

In FY13 we will focus on the continued development of algorithms operating in the compressive domain, with an emphasis on scalability. Specifically, we will create capabilities for (1) change detection—that is, identifying wide-area change with a hierarchical search strategy that uses a sequence of hypothesis tests at progressively finer scales to identify the source of variation; (2) dictionary-based classification—that is, a Bayesian nonparametric approach offering a potentially better representation of the underlying geometry, yielding a soft classification with a basis learned directly from data at hand and with basis vectors associated with hidden-class variables; and (3) scalable, automated tracking, which we will achieve through extensions such as performance characterization of wide-area motion imagery.



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

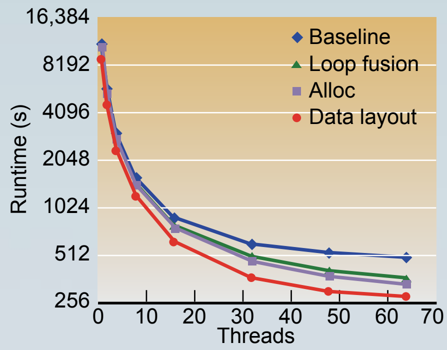
Data Abstractions for Portable High-Performance Computing

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 transferred to newer supercomputers without requiring major rewriting and while maintaining performance.

This research will result in C++ code data abstractions relevant to Livermore's Advanced Simulation and Computing codes that, when implemented with the ROSE compiler, automatically rewrite the code for selected high-performance computing systems. The primary metric for success will be performance of the ROSE-generated specialized codes on diverse computer platforms. We will demonstrate the ease of writing and maintaining the data abstraction version of a code, the breadth of machines on which that code can achieve high performance, and the level of performance achieved on each of those machines. We will also assess how semi-automated processes using the ROSE system could expedite transforming an existing code to one that effectively uses the data-abstraction approach.



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

Mission Relevance

By enabling high-performance computing codes to be quickly and efficiently moved to new computing systems as they come on line, 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.

FY12 Accomplishments and Results

In FY12 we (1) determined we could explore multiple data layout schemes without using data abstractions; (2) implemented optimization techniques for diverse data layouts; (3) updated the TALC multi-architecture compiler (previously developed at LLNL) for using ROSE to generate new data layouts from existing codes; (4) generated multiple versions of LULESH (Livermore Unstructured Lagrangian Explicit Shock Hydrodynamics) with different layouts and demonstrated that capability on a more complex code, xALE (Arbitrary Langerian–Eulerian); (5) measured performance gains for various data layouts using a mini-application LULESH by first optimizing LULESH to establish a fair baseline for studying data layout improvements—these optimizations yielded performance resulting in 62% fewer memory reads, a 19% smaller memory footprint, and 770% more floating point operations; (6) manually created 13 data layouts for LULESH and tested performance on 4 different computers, which showed data layout options could add 15 to 20% performance gains, depending on the layout and machine; and (7) confirmed performance gains for all versions of LULESH produced via our updated TALC tool.

Proposed Work for FY13

In FY13 we will (1) define and implement optimization techniques and directives in ROSE to demonstrate the range of data layouts that can be supported with current data abstractions; (2) evaluate our LULESH codes on the Advanced Simulation and Computing Purple and BlueGene/L supercomputing platforms, and possibly the BlueGene/Q platform, to assess performance; and (3) analyze a second mini-application code for similar data-layout performance studies using Livermore's Mulard high-order diffusion code or Los Alamos National Laboratory's CLAMR code, a cell-based adaptive mesh refinement mini-application developed as a test bed for hybrid algorithm development.

Publications

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

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

Sharma, K., et al., 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 the 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.

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 strategic focus. Our methodology will form a key and crucial component for a variety of analytic surveillance systems, and will help establish the Laboratory's reputation as a leader in analysis of cyber security and intelligence data.

FY12 Accomplishments and Results

In FY12 we developed new adaptive particle filters that automatically adjust to fit time-evolving data. Specifically, we (1) investigated the possibility of a hybrid system combing full and smart particles, but had to abandon the smart-particle approach because of unsatisfactory modeling performance; (2) developed dynamic particle filters capable of analyzing time-evolving, concept-drift data by combining two approaches—a "forgetting" factor that differentially weights recent data and mixture components that are split and merged into the model; and (3) evaluated and benchmarked the performance of particle filters trained with decreasing amounts of data.

Proposed Work for FY13

Our goal in FY13 is to pursue architectural and algorithmic approaches that significantly increase the data analysis rates of our tools to handle the high data throughput rates in streaming applications. As for architecture, we will implement our particle filters in Storm, a massively parallel, high-throughput framework for processing enormous amounts of data on the fly. For algorithms, we will explore adaptive sampling criteria to increase scalability of our particle filters by selectively choosing observations that arrive in the stream, so that updates to the model are restricted primarily to those that maximize information gain. These two complementary approaches should enable us to increase data throughput by orders of magnitude.

Publications

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

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

The Role of Plasma Electromagnetic Fields in Anomalous Mass Diffusion: Applications to High-Energy-Density Science

Peter Amendt (11-ERD-075)

Abstract

Our objective is to conduct a comprehensive study of anomalous diffusive effects in plasmas relevant to LLNL's core missions in stockpile stewardship and inertial-confinement fusion. This work takes advantage of the Laboratory's expertise in high-energy-density science to explore a number of existing anomalies in the growing inertial-confinement fusion and high-energy-density science database that are not explained by conventional methods such as hydrodynamic mix. We propose to develop tools for including ion diffusive phenomena in our physical and computational descriptions of various laboratory phenomena. Analytical methods will be developed and particle-in-cell simulations performed to arrive at a detailed understanding of barodiffusion (diffusion of species brought about by pressure gradients) and thermal diffusion, with eventual incorporation into LLNL's suite of radiation-hydrodynamics production codes.

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 (MIT), will explore the physics of low-Mach-number, collisional plasma shocks and their role in anomalous, nonclassical mass diffusion. A comprehensive understanding of non-fluid (plasma) shock behavior is central to several core missions of the Laboratory, including stockpile stewardship and the pursuit of fusion at the National Ignition Facility.

FY12 Accomplishments and Results

In FY12 we (1) tested the hybrid particle-in-cell simulation code LSP (large-scale plasma) in assessing shock morphology in a spherically converging geometry; (2) studied multi-ion species distributions in LSP to assess the strength of mass diffusion in high-energy-density plasmas in the presence of electric fields; (3) continued analysis and proposals for shots on the OMEGA laser at the University of Rochester in collaboration with MIT, finding that the amount of entropy and heat generated by deuterium and tritium separation during multiple shock passage of the encapsulated fuel significantly affects ignition performance margins; (4) conducted further work that showed the importance of species separation between the helium and carbon in National Ignition Facility ablators in degrading the rocket efficiency; and (5) published a paper in Physical Review Letters highlighting a new source of energy transport from binary species diffusion in capsules that is not currently described in our baseline radiation-hydrodynamics fluid codes.

Proposed Work for FY13

In FY13 we propose to (1) design and conduct implosion experiments on the OMEGA laser in collaboration with MIT to test the phenomenon of species separation across a shock front, (2) finish LSP simulations of recent MIT-led experiments of exploding pushers with various fuel mixtures to test fuel stratification as an explanation for several published nuclear product anomalies, (3) continue LSP simulations of National Ignition Facility implosions to assess the role of species separation on hotspot formation, (4) finish analyzing dissipation energy in binary species plasmas by high-Mach-number shock traversal, (5) begin analytically and computationally studying magnetic field generation in imploding capsules, and (6) begin LSP simulations of the hohlraum target capsule gas and wall interface for interpenetrating plasmas.

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.

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

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, even 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 are behind 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 Livermore's high-performance computing resources to support the Laboratory's mission in energy security.

FY12 Accomplishments and Results

In FY12 we focused our grid modeling and uncertainty work on supporting the development of stochastic optimization methods, with the Plexos grid-modeling software as the core. Specifically, we (1) developed code to stitch together multiple days and to analyze multiple timescales for decisions, with assistance from collaborators—IBM provided the CPLEX solver, Energy Exemplar provided the Plexos grid-modeling software, and the California Independent System Operator Corporation provided models of the western U.S. grid; (2) hired a postdoctoral researcher to implement semi-definite programming algorithms to solve the optimal AC power-flow problem; and (3) deployed codes from our collaborators on Livermore's high-performance computing resources.

Proposed Work for FY13

In FY13 we will use the parallel integer and combinatorial optimization framework developed at Sandia National Laboratories to develop algorithms for the parallel search of branch-and-bound algorithm trees and to develop checkpoint methods to track incumbent bounds that can be used to "prune" portions of the tree. In this work, we will use randomly generated test problems, models of the western U.S. grid provided by the California Independent System Operator Corporation, and models of the eastern U.S. grid obtained by our collaborators at Princeton. Finally, we plan to implement heuristics and algorithms for processor scheduling and load balancing that will improve the performance of optimization algorithms in high-performance computing environments.

Publications

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

Secure Virtual Network Enclaves

Domingo Colon (12-ERD-016)

Abstract

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

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

Mission Relevance

By providing military and other organizations with the capability to dynamically create secure network enclaves and thereby enable the management of production-level network applications in potentially compromised network infrastructures, this project supports the Laboratory's mission in cyber security.

FY12 Accomplishments and Results

In FY12 we (1) developed techniques for systematically defining a virtual enclave environment from the perspective of a network focused on security, (2) developed the capability to automatically build a virtual enclave environment from a security-focused intermediate representation, (3) explored and identified characteristics that are unique to purely virtualized environments and that form the basis for novel security measurement approaches, and (4) began the process of defining and implementing custom measurement techniques that are capable of enforcing mission-specific security policies at a high level of granularity, with measurement components provisioned on a custom per-enclave basis in support of mission-defined security controls.

Proposed Work for FY13

In FY13 we will explore the notion of a distributed, secure virtual enclave environment. Specifically, we will (1) develop algorithms and technical approaches for measuring security across distributed enclave environments, (2) define security properties that are capable of dictating how disparate enclave environments can interoperate in a secure manner, (3) define novel measurement techniques and controls to enforce security guarantees across geographically distributed environments, (4) define techniques for measuring the remotely observable characteristics of enclave environments, and (5) investigate how to characterize (i.e., measure) the public profile exhibited by an ephemeral virtual enclave environment as it is created and operated.

A Network Simulator and Its Applications

Peter Barnes (12-ERD-024)

Abstract

We propose a simulation capability that will establish LLNL as the national leader in large-scale network simulations, with demonstrated applications in cyber security, global network situational awareness, performance modeling, and prediction. Predictive analysis of cyber mission risk and performance is one of the major gaps in our national cyber capability. What we propose is groundbreaking on all fronts. We will simulate realistic networks, derived from real and synthetic network maps, that incorporate real hardware and geographic constraints at the enterprise scale (at least 103 nodes), incorporate near-real-time updates from the global Internet, and generate traffic from realistic traffic models matched to observed data.

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

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) developed an automated process to generate network simulation models directly from network mapper data, generated a model network and simulated modest trivial traffic, and implemented a basic active mapping technique within the simulation; (2) developed performance metrics for mission data-flow problems and began parameter studies for this application in models representative of Department of Defense scenarios; (3) completed porting the Quagga BGP router implementation to our simulator, which would enable simulation of the routing of multiple autonomous systems covering a portion of the worldwide Internet; (4) installed the simulator on LLNL high-performance computing clusters; (5) began a private-sector partnership to couple the Internetwork Operating System routing software emulator in our simulation; and (6) developed several improvements to our network simulator framework.

Proposed Work for FY13

In FY13 we will (1) scale up our laboratory model to a complete level of activity, (2) finish our studies of parameters of the mission execution application and develop plans for realistic and relevant models in collaboration with the Department of Defense, (3) develop performance improvements for the parallel implementation of our network simulator with a Department of Defense laboratory, and (4) develop working models utilizing our private-sector partner's Internetwork Operating System emulation.

Publications

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

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

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

Chunhua Liao (12-ERD-026)

Abstract

High-performance computing at the exascale will require compute node architectures that have thousands of cores, a deep memory hierarchy, and heterogeneous components. This will significantly increase the complexity of designing and adopting programming models that map applications to these architectures. Coupled with the fact that standardized node-level programming models often lag several years behind their target architectures, there is a significant risk that no model will be available for programming exascale architectures when the machines are finally deployed. Our objective is to develop an open framework to assist users—both programming-model researchers and application developers—in building node-level programming models to explore essential exascale issues. This project will evaluate and demonstrate a framework to support the construction of various programming models for heterogeneous architectures tailored to different application requirements.

Our primary deliverable is a framework that assists users in creating various node-level programming models targeting exascale architectures. The framework will be written in

the C++ programming language and iteratively released under a Unix-like open-source license, providing maximum freedom for users from both research and commercial communities. Users will be able to contribute new components, thereby continually increasing the functionality provided by our framework. If successful, this project will revolutionize the high-performance computing software stack, permitting software teams to write applications and design programming models tailored to their applications.

Mission Relevance

Ensuring that applications work well with current and future high-performance computing architectures is essential for every mission at the Laboratory. As new architectures become available, programming models will need to be updated or even overhauled to better map applications to these new architectures. Our project will develop in-house expertise with new programming models that will help design and use high-performance computers, in support of LLNL's strategic foundation in high-performance computing and simulation.

FY12 Accomplishments and Results

In FY12 we focused on defining programming-model building blocks to address the issues of baseline parallelism and data locality. Specifically, we investigated representative applications and kernels and manually translated them to leverage graphics processing units, developing three levels of building blocks: (1) language-level directives to express parallelism and data transfer for graphics processing units; (2) compiler-level building blocks for directive parsing, kernel extraction, and instrumentation support; and (3) runtime support to facilitate programming tasks for graphics processing units, including memory allocation and data transfer. Working with students, we also explored compiler optimizations for power efficiency and a software-based redundancy transformation to address the resilience challenge for exascale computing.

Proposed Work for FY13

In FY13 we will explore advanced analysis and optimization building blocks to improve the performance and power efficiency of programming models built on our framework. In particular, we will focus on approaches most likely to both improve performance and save power, including (1) implementing a vector program for important computation kernels, (2) optimizing data locality for better use of hierarchical memory in graphics processing units, and (3) optimizing Laboratory applications for specific domains using stencil operations on structured arrays.

Publications

Lidman, J., et al., 2012. *ROSE::FTTransform—A source-to-source translation framework for exascale fault-tolerance research*. 2nd Intl. Workshop Fault-Tolerance for HPC at Extreme Scale (FTXS 2012), Boston, MA, June 25–28, 2012. LLNL-CONF-541631.

Rahman, S., et al., 2012. *Studying the impact of application-level optimizations on the power consumption of multi-core architectures*. ACM Intl. Conf. Computing Frontiers, Cagliari, Italy, May 15–17, 2012. LLNL-CONF-599780.

Adaptive Model Reduction for High-Fidelity Simulations

Michael Singer (12-ERD-029)

Abstract

We propose to develop, implement, and apply adaptive model-reduction techniques to enable the fast and accurate predictions of high-fidelity simulations and to enhance the design and control of simulations based on partial differential equations. Our new projection-based methods will automate the extraction of essential physical processes in a complex, multiphysics simulation, which in turn will enable computational effort to be focused on resolving the underlying important phenomena that drive the system. Our techniques will minimize the preprocessing effort that is required to construct the models because the reduced models evolve locally and change dynamically with the underlying physics. Our techniques will be broadly applicable to a multitude of LLNL mission-critical domains. Upon successful completion of the project, we will have designed, implemented, and tested state-of-the-art model-reduction algorithms that are applicable to numerous modeling and simulation problems. Our methods will enable fast and accurate simulation of complex science and engineering systems, especially in the contexts of prediction, design, and control. These new simulation capabilities will facilitate further developments in such domains as uncertainty quantification and design optimization. Our methods will be specifically applied to assist the fast and accurate power prediction of energy systems, which is relevant to Livermore's proposed Energy Research Park.

Mission Relevance

The computer simulation and modeling techniques developed in this work will advance the state of the art in computational science and enable prediction, design, and control of multiscale systems on high-performance computers. In particular, our work will enhance energy security by providing computational tools to better predict and harvest wind energy, and will contribute to the mitigation of climate change by enabling and enhancing the development of clean, renewable, and sustainable energy systems.

FY12 Accomplishments and Results

In FY12 we began studying the development, implementation, and application of adaptive model-reduction techniques to enable fast and accurate predictions of high-fidelity simulations and to enhance the design and control of partial-differential-equation-based simulations. The project was concluded early, however, after the principal investigator left the Laboratory.

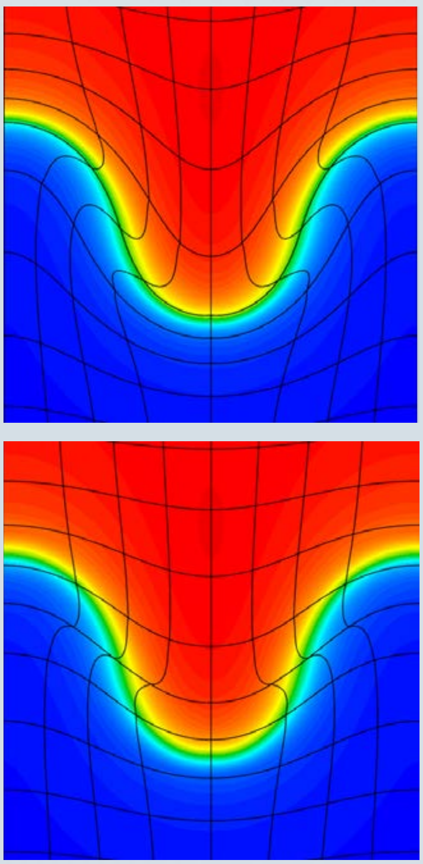
High-Order Curvilinear Arbitrary Lagrangian–Eulerian Hydrodynamics

Tzanio Kolev (12-ERD-030)

Abstract

The framework of arbitrary Lagrangian–Eulerian (ALE) codes, used to model fluid and material response on an unstructured grid (or mesh), form the core of large-scale hydrodynamics codes used at LLNL for stockpile stewardship and other mission-relevant work. Current ALE schemes are an improvement over pure Lagrangian methods, but also introduce numerical problems such as lack of energy conservation and artificial material breakup. Recent advances in high-order curvilinear finite elements have shown significant benefits in the Lagrange phase of ALE, and we propose to apply this curvilinear technology to the ALE advection phase to develop new and more robust high-order ALE algorithms, while preserving the accuracy of the high-order Lagrange step. To this end, we will research and develop new methods for optimization of curvilinear mesh geometry representations, conservative monotonic high-order field remapping, and the handling of multiple-material curvilinear zones. This work will leverage previous research at LLNL on high-order curvilinear finite elements for the Lagrange phase.

This project will produce the first high-order curvilinear method for ALE hydrodynamics, enabling higher-quality simulations of multi-material ALE hydrodynamics. These algorithms can potentially eliminate the need for adjusting mesh-motion parameters and for manual intervention by users, minimize diffusive errors by running longer in a Lagrange mode, improve accuracy by diminishing mesh imprinting and improving symmetry preservation, and more effectively utilize future multi-core and graphics processing unit architectures because of the algorithms' local intensity in floating-point operations.



Our newly developed high-order discontinuous Galerkin advection algorithm enables the conservative and accurate remapping of functions with steep gradients on highly distorted curved meshes for hydrodynamic simulations. High-order density field on a distorted Lagrangian mesh (left) is remapped to a new, relaxed curvilinear mesh (right). The two density fields are virtually identical, illustrating the high accuracy of the remapping algorithm.

Mission Relevance

Hydrodynamics simulations are of critical importance in numerous Laboratory applications, including stockpile stewardship, inertial-confinement fusion, and other mission-relevant efforts. The proposed research will develop new high-order curvilinear, 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.

FY12 Accomplishments and Results

We (1) developed and investigated different approaches for curvilinear mesh optimization, including high-order extensions of currently existing methods such as harmonic smoothing based on novel high-order mesh Laplacians, Winslow–Crowley nonlinear mesh optimization, polynomial filtering, and the Mesquite software toolkit for mesh optimization; (2) proposed a family of new high-order advection-based discontinuous Galerkin remap schemes that generalize the classical "swept volumes" technique and avoid the expensive computation of curved-element intersections—we achieved very encouraging preliminary results; and (3) implemented and extensively tested the above ideas in our finite-element research code.

Proposed Work for FY13

In FY13 we will design, implement, and analyze novel high-order curvilinear, finite-element numerical algorithms to enable more accurate, robust, and reliable ALE hydrodynamics simulations. Specifically, we will (1) continue investigating and analyzing new conservative, monotonic, and accurate interpolation methods for high-order field remapping on curvilinear meshes; (2) demonstrate the advantage of the new remapping methods in simplified settings such as simple ALE calculations in which material interfaces are kept Lagrangian, and in single-material problems; (3) begin researching high-order multi-material zone treatment; and (4) implement and test the new algorithms in a parallel research code.

Publications

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

Dobrev, V. A., T. V. Kolev, and R. N. Rieben, 2012. "High order curvilinear finite elements for Lagrangian hydrodynamics." *SIAM J. Sci. Comput*. **34**(5), B606. LLNL-JRNL-516394.

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

Efficient and Accurate Metagenomics Search Using a k-mer Index Stored in Persistent Memory

Jonathan Allen (12-ERD-033)

Abstract

Developing the capability to detect and diagnose engineered and emerging diseases across a global network is a national biosecurity research priority. "Metagenomics" sequencing (the study of genetic material recovered from environmental samples) has emerged as a powerful genetic survey tool for research to generate an unbiased and detailed description of a biological sample. We will develop novel, massively parallel algorithms to detect and identify pathogens in biological samples by searching pathogen genome databases indexed by their constituent k-mers—specific amounts of nucleic acid or amino acid sequences that can be used to identify regions within biomolecules such as DNA. This approach requires shorter laboratory preparation time prior to sequencing and no prior knowledge of the contents required for analysis. The ability to efficiently search

emerging metagenomics databases presents a powerful new tool that could be used for pathogen detection and characterization.

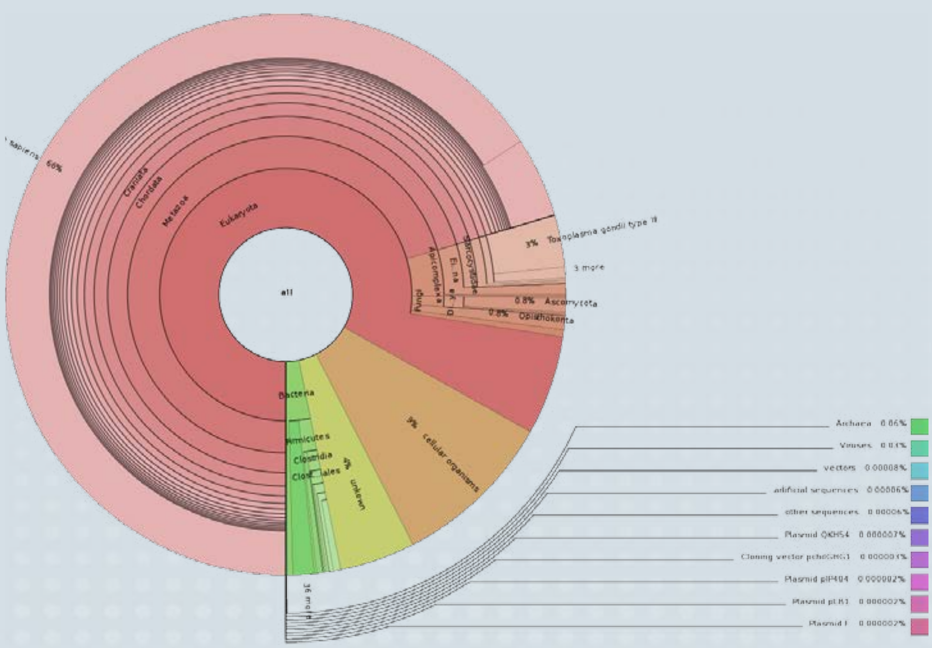
We expect that the new software tools we create will enable orders-of-magnitude improvement in turnaround time from the submission of a biological sample's DNA to its taxonomic and functional characterization. Further, the analysis can be performed on commodity hardware—that is, computer systems manufactured by multiple vendors, incorporating components based on open standards—and utilizing multiple and many-core processors combined with high-performance flash storage, making this analysis potentially deployable to field sites worldwide. The project results would thus demonstrate technical pathways by which challenging computational hurdles in metagenomics analysis can be overcome to transition current research tools into technology that can be exploited by government agencies tasked with pathogen surveillance, diagnostics, and characterization.

Mission Relevance

Our approach could advance the forefront of metagenomic analysis for the biodefense needs of the nation, positioning us to partner with others to tackle challenging analyses of large-scale clinical, environmental, and forensic metagenomic samples. This effort therefore supports LLNL's mission focus area of biosecurity research, as well as bolstering the Laboratory's core competency in high-performance computing by using low-cost, multicore computing nodes augmented by persistent flash computer memory.

FY12 Accomplishments and Results

In FY12 we (1) implemented tools in software for testing our k-mer indexing strategy on a pathogen genome database and created the largest—to the best of our knowledge—k-mer database of its kind; (2) implemented the metagenome search



Taxonomic classification of 2.3 billion genetic sequencer reads taken from the Tyrolean Iceman (also known as Ötzi). Analysis was completed in less than 20 hours on a single large-memory computer using commodity hardware.

algorithm to exploit flash memory for classification on the genetic-strain-level for the input of metagenomic samples; (3) applied benchmarking measures for algorithm run time and classification accuracy under varying computational resource requirements; and (4) expanded our database to include all bacterial, fungal, and viral genomes—not just pathogenic variants—which allows our sample classification tool to be more widely applicable than previously anticipated.

Proposed Work for FY13

We propose to extend the methods and benchmarks developed for strain-level classification to address the problem of classifying samples containing organisms not previously sequenced. Specifically, we will evaluate and test extended indexing strategies that tolerate greater sequence mismatches between the query sample and reference database, investigating the use of multiple k-mers of shorter length and variable length k-mers. In addition, we will modify accuracy performance metrics to measure the amount of sequence differences from the reference database that can be introduced and still support sample classification. This approach should enable us to identify novel organisms that previous methods were not able to classify.

Publications

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

Automatic Complexity Reduction for Simulations of Electromagnetic Effects

Daniel White (12-ERD-038)

Abstract

Our goal is to develop a new method for reducing the complexity of simulating electromagnetic effects on circuits. While documented experimental evidence shows that an electromagnetic wave of modest power can temporarily shut down an electronic circuit, not all circuits are affected and not under all conditions. Simulation is required to better understand the electromagnetic effect. LLNL has sophisticated massively parallel finite-element and boundary-element codes for solving Maxwell's equations, but on the order of trillions of simulations are required to understand how electromagnetic effects vary with circuit layout, frequency, and location of the circuit. Based on a combination of reduced-order models, radial basis functions for interpolating matrix triple products, and parameters adaptivity, we propose to develop an automatic complexity-reduction algorithm for simulating electromagnetic effects, test it on supercomputers, and validate the results using experimental electromagnetic effects data.

If successful, the project will result in an automatic complexity-reduction algorithm for predicting electromagnetic effects. This new method for reducing the complexity of exploring parameters via simulation has many applications, including heat transfer, elasticity, and related partial differential equations. We expect to publish the results in peer-reviewed journals and to license software.

Mission Relevance

Electromagnetic effects can disrupt any device that contains an electronic circuit, including improvised explosive devices in war zones, cell phones, information technology equipment, the electrical grid, and industrial and military control systems. By creating the capability to predict electromagnetic effects, this research has the potential to support the entire range of Lawrence Livermore strategic missions, including national and international security and energy security missions.

FY12 Accomplishments and Results

We successfully implemented a prototype general-purpose reduced-order model algorithm, which consists of three steps: (1) sampling relevant parameters; (2) performing a change of basis, thereby reducing the problem dimension; and (3) interpolating for efficiency. We tested this algorithm on several boundary-element electromagnetics problems. One problem consisted of a printed circuit board within a metal enclosure for which we created a reduced-order model for the induced current on the printed circuit board traces for every location of the circuit within the enclosure. A second problem consisted of the same printed circuit board in a metal enclosure, with an aperture whose position was varied. The reduced-order model gives the induced current on the circuit traces for every location of the aperture. For a third test problem, our reduced-order model algorithm resulted in ill-conditioned matrices, and we will investigate the cause of this.

Proposed Work for FY13

In FY13 we propose to (1) identify the root cause of ill conditioning of the reduced-order model discovered in FY12, and to develop a more robust algorithm; (2) develop an error estimator to terminate the process for our automatic, adaptive reduced-order model—a conservative error estimator leads to inefficiencies (too large a model), whereas an optimistic estimator leads to an inaccurate model; and (3) apply our algorithm to some large-scale test problems, which will require significant computational resources to generate the true solution. This will provide important information for FY14.

Publications

Lange, K. J., and D. A. White, 2012. *A comparison of radial basis functions for MIROM of a boundary element electromagnetics simulation*. LLNL-POST-558893.

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

A Linearly Scalable Algorithm for First-Principles Molecular Dynamics at Exascale

Jean-Luc Fattebert (12-ERD-048)

Abstract

Current molecular dynamics algorithms with $O(N^3)$ complexity—that is, requiring computational resources of calculations that increase with the cube of *N*, the number of atoms in the system—will not be able to take full advantage of the orders-of-magnitude increase in computational power expected by the end of the decade. We will therefore develop and implement a first-principles molecular dynamics simulation technology with reduced complexity—$O(N)$ (linearly scalable) instead of $O(N^3)$—to simulate molecular systems. We will focus on making the capability truly scalable and reliable for routine use in applications involving thousands of atoms simulated with many thousands of processors. To this end, we will also develop a faster convergence solver for the sparse representation of solutions, implement the $O(N)$ algorithm needed for the parallel linear algebra, develop and implement an algorithm for constant-pressure simulations, and use the first-principles molecular dynamics computer code MGmol to implement new algorithms based on real-space finite differences on a uniform grid, or mesh.

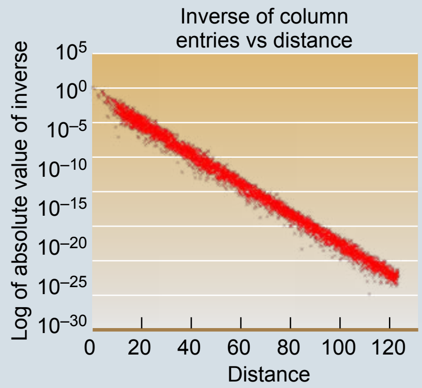
The reduced-complexity $O(N)$ algorithm we develop should be able to simulate hundreds of thousands of atoms from first principles on exascale computers. This capability will enable research with more realistic models of matter than we use today, involving, for instance, more realistic defects and more complicated molecular structures for the study of nucleation in materials or calculation of the equation of state of polymers with realistic molecular structures.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we hired a new postdoctoral researcher and developed parallel linear algebra solvers to calculate selected elements of the inverse of the Gram matrix coupling localized molecular orbitals to density functional theory. Specifically, we (1) developed and implemented a new parallel and scalable algorithm that eliminates long-range coupling when computing selected elements of the inverse of relevant matrices; (2) coupled, after testing its parallel scaling and accuracy on matrices generated from real density functional theory applications, the new algorithm to the density functional theory code MGmol and started the debugging, testing, and evaluation phase; and (3) instituted substantial design changes to MGmol to accommodate the new algorithm, which improved the code in general.



Decay properties of matrix elements (inverse Gram matrix) as a function of spatial distance between corresponding molecular orbitals for a polymer density functional theory calculation. Our new algorithm makes use of that exponential decay in insulating systems.

Proposed Work for FY13

We propose to (1) fully couple our newly developed algorithm to the MGmol code; (2) perform weak-scaling studies of density functional theory applications for up to 50,000 atoms and identify bottlenecks; (3) demonstrate *O(N)* scaling for up to 50,000-atom systems and publish our results in a high-impact peer-reviewed journal; and (4) develop new algorithms to speed up solvers for the MGmol code.

Laser Lethality Experimentation, Modeling, and Simulation Capability

W. Howard Lowdermilk (12-ERD-050)

Abstract

Existing experimental data and models are inadequate to develop systems to counter the threat of modern ballistic missiles. The need therefore exists for an experimentally validated, predictive modeling and simulation capability to optimize the design and performance of anti-ballistic-missile laser weapon systems and to reduce the need for costly full-scale testing. Our goal is an experimentally validated model for the laser-induced fracture and fragmentation of thin metal plates and pressure vessels in single- and multiple-layer configurations. We will conduct laser interaction experiments and measure thermo-physical properties using the National Ignition Facility to enable and guide the development of a new ALE3D (arbitrary Lagrangian–Eulerian three-dimensional) code and to validate the resulting laser–target interaction models.

We will produce measurements of the physical properties of select materials in relevant regimes of temperature and stress loading, characterizations of laser-induced fracture and fragmentation of thin metal plates and pressure vessels in single- and multiple-layer arrangement, and ALE3D capabilities for modeling laser¬–target interaction, culminating in an experimentally validated model. We will also demonstrate our new model's capability and practicality for countering laser lethality problems important to the military. This capability will enable the timely and cost-effective design and optimization of anti-missile laser weapon systems needed to defend against modern ballistic missiles.

Mission Relevance

This project directly supports LLNL's national security mission by meeting the currently unfilled need for a validated, predictive modeling capability to evaluate laser lethality and missile vulnerability for laser-based anti-missile systems. In addition, the capability to be developed will also be applicable to similar fracture and fragmentation problems relevant to National Ignition Facility target experiments, in support of stockpile stewardship and the Laboratory's energy security mission.

FY12 Accomplishments and Results

In FY12 we (1) developed a method to accurately measure laser absorptivity by metals that is sensitive to surface conditions; (2) measured absorption versus temperature of 0.8-μm wavelength diode laser radiation by the aerospace metals aluminum, steel, and titanium from room temperature to near-softening points; (3) developed and implemented a numerical algorithm that accounts for convective and radiative loss and improves experimental data-processing accuracy; (4) began measurement of temporal and spatially resolved two- and three-dimensional strain and deformation under controlled mechanical and thermal stress in the same metals to support modeling of deformation fields in metal plates at temperatures up to but not including fracture; (5) surveyed aluminum alloy properties and model parameters; (6) enhanced heat transfer, surface contact, and element erosion components of the ALE3D material model and simulations, and completed an initial ALE3D model of laser-induced metal fracture; and (7) began preparation for experiments to characterize laser-driven buckling, fracture, and fragmentation of stressed and thermally loaded thin metal plates.

Proposed Work for FY13

In FY13 we propose to (1) complete strain deformation measurements and use data in ALE3D to identify appropriate constitutive models for relevant materials, heating, and strain rates; (2) complete laser interaction measurements on stressed and thermally loaded plates and begin measurements on pressurized vessels, and plan for dual-layer plate and plate and vessel measurements; (3) enhance ALE3D for integration of heat transfer, thermal contact, and element erosion, as well as develop ablative pressure and traction boundary conditions for free surfaces; and (4) develop a fracture model with laser-driven fragment ablation and begin model validation.

Publications

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

Rubenchik, A. M., et al., 2012. *Laser–material interaction experiments using a diode array to simulate a DE weapon-class source*. LLNL-PRES-512874.

Theory and Simulation of Large-Amplitude Electron Plasma and Ion Acoustic Waves with an Innovative Vlasov Code

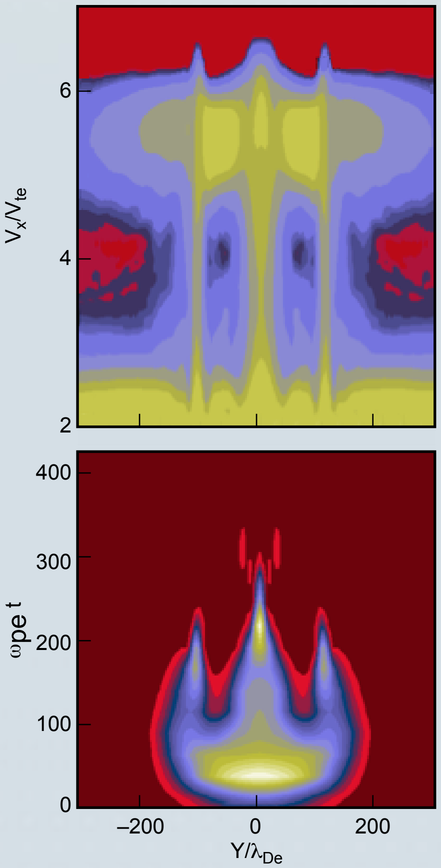
Richard Berger (12-ERD-061)

Abstract

The goal of the National Ignition Campaign is to implode capsules and ignite the deuterium–tritium fuel with x rays converted from laser light in a high-atomic-number plasma. If successful, this is a potential path to fusion energy, a safe and carbon-free source of energy. Experiments at the National Ignition Facility to realize this goal depend on predictable propagation of the 192 laser beams through hot dense plasma. We propose to examine processes that affect laser light propagation directly and the effects on the distribution of electrons from plasma waves excited by laser light. Specifically, we intend

to improve understanding of the kinetic processes, described by the Vlasov equation, that determine the nonlinear state of large-amplitude electron plasma waves and ion acoustic waves and the self-consistent distribution of electrons and ions associated with these waves in two dimensions. We will use Livermore-developed 2D + 2V Vlasov codes (two dimensions in space and velocity) to study nonlinear ion acoustic and electron plasma waves in hot dense plasmas.

We expect to establish the dependence of transverse and longitudinal modulation instability of an electron plasma wave-on-wave amplitude and wavelength over the Debye length. We will also study the nonlinear evolution of two-species ion acoustic waves, specifically the slow mode of carbon–hydrogen plasmas. We will create a multiple-ion species Vlasov code by generalizing the choice of boundary conditions in the VALHALLA code as well as the input options, and consider collisions in VALHALLA for inclusion, if feasible. Nonlinear ion acoustic waves will be studied for single- and multiple-ion species with VALHALLA with the same methods developed for the study of electron plasma waves. The results will be applicable not only to stimulated Raman and Brillouin scattering but also to other effects such as two-plasmon decay and ion acoustic waves driven by an inter-penetrating plasma.



The tail of electron distribution in a plasma shows the trapped electrons in a wave with filaments and a beam of de-trapped electrons at time $t = 200$ in units of the plasma frequency (top). The electric-field energy density averaged over the wave propagation direction is graphed against time and the transverse direction (bottom), showing how the wave focuses and filaments over time.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) conducted simulations of one-dimensional (1D) multispecies ion waves using the Eulerian Vlasov code, SAPRISTI; (2) conducted simulations with our 2D Vlasov code, LOKI on the modulation instabilities of electron plasma waves and presented the results at two conferences; (3) compared the results of the 2D LOKI simulations to 2D particle-in-cell simulations by collaborators at the University of California, Los Angeles; (4) implemented absorbing boundary conditions in LOKI in collaboration with Switzerland's Laboratory of Intelligent Systems and the University of California, Los Angeles; and (5) evaluated approaches to adding particle collisions to LOKI and developed theory to interpret the simulations. In addition, we hired a postdoctoral researcher to help with the Vlasov simulations.

Proposed Work for FY13

In FY13 we will (1) continue work on electron plasma wave modulation instabilities in 2D, (2) begin studies to determine the thresholds for and the nonlinear state of the Langmuir decay instability, (3) finish implementing multiple species in the Vlasov code, and (4) begin 2D LOKI simulations of ion acoustic waves.

Publications

Banks, J. W., et al., 2012. *Vlasov simulations of electron plasma and ion acoustic waves: Self-focusing and harmonics*. 54th Ann. Mtg. APS Division of Plasma Physics, Providence, RI, Oct. 29–Nov. 2, 2012. LLNL-ABS-490292.

Banks, J. W., et al., 2012. *Vlasov simulations of electron plasma waves: Self-focusing and modulational instability*. APS March Mtg. 2012, Boston, MA, Feb. 27–Mar. 2, 2012. LLNL-ABS-523173.

Banks, J. W., et al., 2012. *Vlasov simulations of the filamentation and trapped electron sideband instability*. 42nd Ann. Anomalous Absorption Conf., Key West, FL, June 25–29, 2012. LLNL-ABS-552655.

Berger, R. L., et al., in press. "Electron and ion kinetic effects on nonlinearly driven electron plasma and ion acoustic waves." *Phys. Plasmas*. LLNL-JRNL-480183.

Berger, R. L., et al., 2012. *Kinetic simulations of electron plasma waves: Trapped electron filamentation and sideband instabilities*. 42nd Ann. Anomalous Absorption Conf., Key West, FL, June 25–29, 2012. LLNL-ABS-552676.

Chapman, T. D., et al., 2012. *Kinetic simulations of electron plasma waves: Trapped electron filamentation and sideband instabilities*. 54th Ann. Mtg. APS Division of Plasma Physics, Providence, RI, Oct. 29–Nov. 2, 2012. LLNL-ABS-564171.

Chapman, T., et al., 2012. *Nonlinear waves in multi-ion species plasmas at low electron-to-ion-temperature ratios*. 42nd Ann. Anomalous Absorption Conf., Key West, FL, June 25–29, 2012. LLNL-ABS-552319.

Strozzi, D. J., et al., in press. "Characterizing electron trapping nonlinearity in Langmuir waves." *Phys. Plasmas*. LLNL-JRNL-574812.

Winjum, B. J., et al., 2012. *PIC simulations of the trapped electron filamentation instability in finite-width electron plasma waves*. 42nd Ann. Anomalous Absorption Conf., Key West, FL, June 25–29, 2012. LLNL-ABS-552677.

Predictive Models for Target Response During Penetration

Tarabay Antoun (12-ERD-064)

Abstract

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

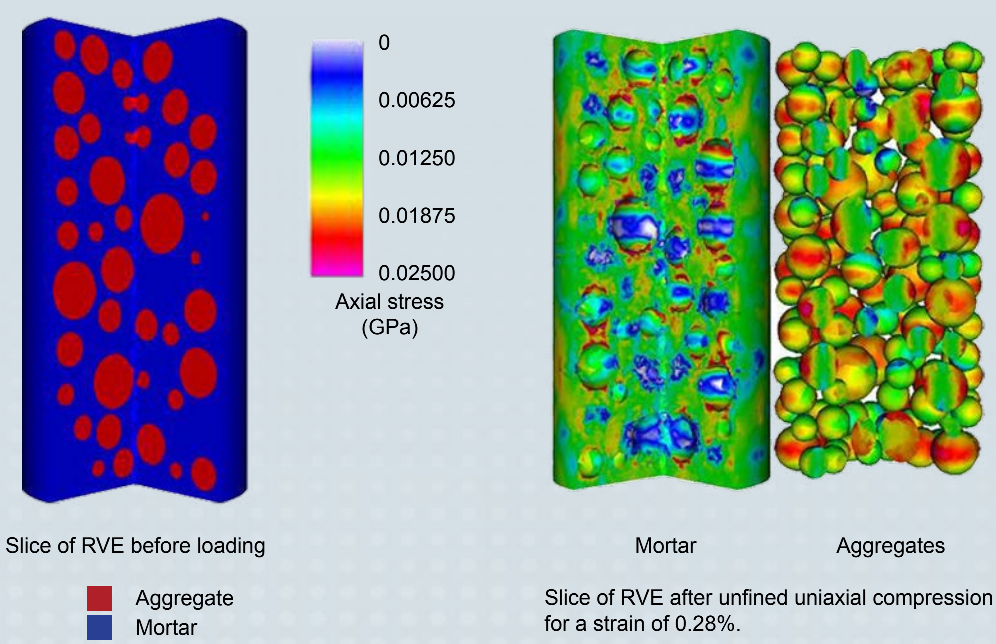
Successful execution of this project will result in a new capability with unprecedented fidelity for modeling the response of frictional materials to extreme dynamic loading environments such as those encountered during the interaction of an earth penetrator with a geologic target or the interaction of a bullet or a shaped charge with ceramic armor. This modeling framework will support the design of advanced penetrating weapons that are smaller, lighter, faster, and more effective against hardened and deeply buried targets. Also, this work will make it possible to design more efficient transparent ceramic armor capable of providing superior protection against a wide range of threats, including shaped charges and improvised explosive devices. We expect that this novel modeling capability will be applicable to the programmatic needs of various sponsors, including the Defense Advanced Research Projects Agency, Army Research Laboratory, Defense Threat Reduction Agency, and various branches of the armed services.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we achieved all our proposed milestones. Specifically, we (1) developed a continuum model, implemented a number of model features, and performed parametric studies to determine which features are most important for penetration mechanics; (2) developed a robust computational model of a representative volume that allows discrete fracture and insertion of contact physics along the newly fractured surfaces; (3) applied a penetration-relevant loading path to the representative volume and performed mesoscale simulations to generate a synthetic database for developing the continuum model; (4) documented baseline continuum simulation capabilities and



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

provided boundary conditions for mesoscale simulations; and (5) assessed and documented model sensitivities, mesh sensitivities, and scalability of the model using the arbitrary Lagrangian–Eulerian multiphysics ALE3D code.

Proposed Work for FY13

For FY13 we propose to (1) complete implementation of our micromechanically based continuum model and begin linking the evolution of model parameters to mesoscale simulation results; (2) investigate mesh dependence during softening and strain localization, and investigate different methods for mitigating this effect; (3) improve the fracture modeling algorithm and develop an adaptive fragment meshing strategy to improve the efficiency of mesoscale simulations; (4) validate the mesoscale modeling approach by comparing it to data on the dynamic behavior of sand (a relevant granular material); and (5) publish preliminary validation with available experimental results and observed trends to reduce the statistical space of sensitivity study.

Proposed Work for FY13

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

Applying High-Performance Computing and Simulation to Future Energy Challenges

Clara Smith (12-ERD-074)

Abstract

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

We expect to provide fundamental progress in the basic scientific challenges of applying high-performance computing and simulation to energy issues. Partnering with the private

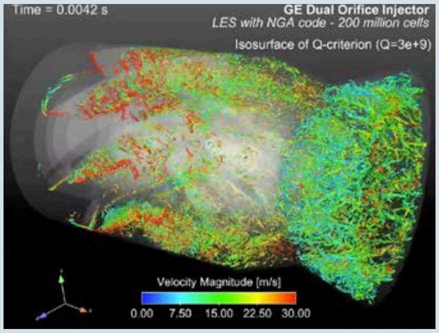
sector, we have identified representative near-term research topics and goals. Specifically, we intend to demonstrate application of uncertainty quantification and contingency analysis to the solution of large energy-grid problems such as electricity production and distribution. We will also provide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

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we initiated collaborations with several industrial partners by applying high-performance computing to challenging problems in the energy sector. Specifically, we (1) performed scaling tests and calculations in collaboration with industrial partner GE Global Research and Bosch using large-eddy and direct numerical simulation codes to improve the speed of combustion modeling; (2) completed 7,000 simulations in 2 dimensions and 30 simulations in 3 dimensions providing valuable insight into actual mechanisms for spall formation in collaboration with Potter Drilling using the Lawrence Livermore PSUADE uncertainty and GEODYN geological drilling simulation codes; (3) scaled up the PSLF (Positive Sequence Load Flow) software computer code with GE Energy and performed a test case of over 4,000 contingency analyses, reducing time requirements from 23.5 days estimated on a single computer core to 23 minutes on over 4,000 nodes; (4) reduced time requirements in completing 400 solutions with ISO New England from 200 hours to 2 hours in preparation to intelligently evaluate robust unit commitment for the electricity grid; and (5) completed energy simulations with United Technologies Research Center to better understand which of 917 parameters affect building energy use most significantly.



Fuel flow velocities in a dual-orifice injector calculated on Livermore's Sierra supercomputer using the NGA large-eddy and direct numerical simulation code as part of the GE Global Research incubator project.

Proposed Work for FY13

In FY13 the results from our simulations will be compared to results obtained with the conventional design and development cycle. Specifically, we will (1) determine the benefits and disadvantages of the two software codes developed in collaboration with GE Global Research in describing fuel spray in a combustion chamber as compared with experimental data, (2) compare the Bosch simulations of an internal combustion engine transitioning from spark ignition to homogeneous-charge compression ignition in four scenarios to experimental data and explain the observed phenomena, (3) conduct parametric and sensitivity analyses to determine the most influential parameters in optimizing thermal spallation drilling, (4) run test cases with the parallelized code to demonstrate and quantify the increase in solution convergence for representations of large electricity grids, (5) work with ISO New England to run simulations to identify the appropriate levels under which the algorithm of robust unit commitment exhibits economic advantages and compare the results to the current industry-accepted deterministic method for selecting the least-cost dispatch of available generation sources to the electricity grid, and (6) work with United Technologies Research Center to use building energy models to determine the first-level parameters and second-level interaction parameters that determine the energy efficiency of buildings.

Publications

Smith, C. A., 2012. *hpc4energy incubator busy growing energy technologies*. LLNL-ABS-546312.

Smith, C. A., and J. Grosh, 2012. *SC12 panel proposal: Applying high-performance computing at a national laboratory to the needs of industry; case study: hpc4energy*. SC12—The Intl. Conf. High Performance Computing, Networking, Storage and Analysis, Salt Lake City, UT, Nov. 10–16, 2012. LLNL-ABS-556131.

Min, L., 2012. *Perspectives on advanced computing methods and technologies for real-time control center operations*. LLNL-ABS-585193.

Walsh, S. D. C., 2012. *Grain-scale thermal spallation simulation*. LLNL-PRES-581314.

Evaluating the Feasibility of Co-Evolving Network Simulations

James Brase (12-FS-006)

Abstract

We propose to evaluate the feasibility of using agent-based simulation to model human social networks, which connote complex sets of relationships between members of social systems at all scales from interpersonal to international. These, in turn, influence the behavior of complex technological networks such as the Internet or large-scale power grids. Integrated simulation of the co- evolution of technological and social networks is required to accurately model observed network behaviors, and developing a predictive simulation capability for complex information networks is important to many national security applications. We will evaluate this feasibility by using an agent-based simulation to drive a discrete-event network simulation tool and comparing the resulting network behaviors to real data sets. Our principal deliverables will be a working prototype simulation code for the defined scenario and a report on feasibility.

We expect to demonstrate that we can use simulations of co-evolving technological and social networks to more accurately model observed behaviors of the technological network. If successful, we will demonstrate the causal relationship between activities of humans in the social network and activity in the information network, and adaptation of social network activity based on technical network behaviors. This overall co-evolution model then gives us the needed tool to model integrated activities including the actions of attackers and defenders in computer networks as well as those of background users.

Mission Relevance

The ability to predict behaviors of complex networked systems is critical to understanding their performance and to controlling their behavior in applications such as cyber security as well as power grid control, in support of the Laboratory's strategic mission focus areas in cyber security and energy and climate. Cyber security will benefit from the ability to quantitatively evaluate cyber defense methods and to evaluate cyber mission risk. Energy programs will benefit from understanding performance of complex power systems that combine energy grid networks with information networks.

FY12 Accomplishments and Results

In FY12 we (1) defined a botnet evolution scenario to test coupled simulations (botnets are loose networks of computers that are controlled by a third party for malicious intent); (2) developed a framework for agent-based activity simulations in the NS-3 network simulator, which was then employed by a summer student in his CyberDefenders project on botnets; (3) evaluated the simulation performance and network activity statistics for varying levels of botnet activity and defensive responses; and (4) determined a simulation framework and botnet performance showing approaches for scaling to million-entity simulation sizes. Our feasibility study results are the basis for a new simulation being developed for sponsor application and have contributed to proposals for large-scale network activity simulations to the Department of Defense and others. The agent-based simulation framework is continuing to be used to support doctoral research at the University of California, Santa Cruz.

Reduced-Order Modeling

Kyle Chand (12-FS-007)

Abstract

Reduced-order models provide efficient approximations for the essential features of complex, large-scale, mathematical models. Reduced-order models are designed to be orders of magnitude more efficient than the corresponding high-fidelity models they approximate. In wind energy, for example, reduced-order models generated from large eddy simulations have great potential in optimization problems such as turbine placement, as well as enhancing models for turbine wake aerodynamics. For building efficiency, reduced-order models derived from high-fidelity simulations can be used in the design of control systems and for optimal sensor placement. We propose to explore current state-of-the-art reduced-order models, with the goal of assessing the feasibility of applying such methods to strategic Lawrence Livermore initiatives. Our approach will include application of reduced-order models to model problems that represent LLNL simulation tools based on partial differential equations. Model systems such as the convection–diffusion equation provide a realistic framework to evaluate different reduced-order modeling techniques while remaining amenable to mathematical analysis of the efficiency, accuracy, and stability of reduced-order model approximations.

We expect to focus on two fundamental components of reduced-order models: the automated distillation of high-fidelity simulation data into lower-dimensional, "reduced," approximations and the application of these approximations to enable rapid, reduced-order simulations. Issues of interest for the first component include dynamic selection of optimal basis vectors that maximize the amount of information included in the reduced-order model while minimizing the size of the lower-dimensional subspace. Research topics for the second component, solution of the reduced model, include stability of the projected system of equations and producing error bounds on the entire process. In addition, we will suggest fruitful avenues of further research and application of reduced-order models.

Mission Relevance

Combining LLNL's existing simulation expertise based on partial differential equations with a reduced-order model infrastructure positions the Laboratory to expand into new areas critical to its missions in energy and national security.

FY12 Accomplishments and Results

We (1) investigated the current state of the art for constructing reduced-order models from simulations based on dynamic partial-differential equations; (2) partitioned the time domain of a model partial-differential equation and tuned a fixed-size reduced-order model to each sub-domain, thus creating an effective method for achieving an efficient, accurate, and small-sized time-partitioned, reduced-order model for dynamic problems; and (3) investigated the construction of reduced-order models for simulations that use overlapping grids, finding that weighting the discrete inner product in a manner that approximates the continuous inner product yields a significantly more accurate reduced-order model. In summary, this project demonstrated the feasibility of applying reduced-order models to simulation tools based on complex partial-differential equations, and explored the benefits of adaptive reduced-order modeling techniques. The results of this project led to further work focusing on developing composite, adaptive reduced-order models suitable for various mission-relevant LLNL applications.

Publications

Chand, K. K., et al., 2012. *Reduced-order modelling for dynamic simulations: LDRD feasibility study final report*. LLNL-TR-605014.

Visualization of Computer Networks at Arbitrary Scales

Carlos Correa (12-FS-008)

Abstract

We propose to study algorithmic techniques for visual analytics of computer networks at arbitrary scales. Despite current efforts in extracting useful statistics of networks, visualization of large networks remains a challenge because of computational cost and visual complexity. In this feasibility study we will test two hypotheses. First, we will determine

whether factorization of networks can be used not only to scale current visual metaphors and tools, but also to provide more insightful views of network data. Second, we will determine whether visual tools are useful at arbitrary scales thanks to the predictive nature of matrix factorization techniques. We will explore benchmarks of information and social networks used in previous research efforts and that are representative of networks used within LLNL.

We expect our study will provide the foundation to develop visualizations of networks at an arbitrary scale and enable algorithms that support different operations and visual analytic tools available within this new framework. We will focus on a single method for matrix factorization (stochastic Kronecker graphs generally proposed as a generative model for large-scale real-world networks). We believe that our project will lead to a more complete research effort that merges visualization and analytics relevant to efficient matrix decomposition methods, general visualization algorithms, and generative models of networks. In addition, we expect our research has application to system-building efforts aimed at developing tools that can be incorporated into the network analytics systems within the Laboratory.

Mission Relevance

This study is an important step towards scalable visual analytics as a support tool for global intelligence and cyber-security. Network data is at the core of performance analysis of simulations running on large clusters in cyber-security, biosecurity, global intelligence, and counterterrorism and nonproliferation efforts. Scalable visualization tools will be crucial in providing analysts the means for faster decision making, while coping with the growing complexity, scale, and heterogeneity of network data.

FY12 Accomplishments and Results

This feasibility study was expected to provide the foundation to develop visualizations of networks at an arbitrary scale and enable algorithms that support different operations and visual analytic tools available within this new framework. During this work, we explored Kronecker product factorization of adjacency matrices arising from graphs. Preliminary results were promising with more work needed to validate the method on a broader set of graphs.

Feasibility of Predicting Protein Antigenicity over Multiple Resolutions of Function

Eithon Cadag (12-FS-009)

Abstract

The computational identification of antigens—pathogen proteins that elicit a potentially detectable immune response in the host—has wide-reaching implications for efforts in biodefense, including assessment of immune history (previous exposure to pathogens), diagnostics development, disease staging, vaccine design, and elucidation of disease mechanisms. Detection and treatment of emerging and engineered infectious diseases will

require new, rapid methods of antigen prediction to not only expedite research but also direct attention to potentially neglected targets and handle the evolving landscape of biological threats. We propose to assess the utility of applying functional information from protein sequence, structure, and molecular interactions to predicting antigenicity. Current methods for predicting antigens or their epitopes (antibody binding sites) rely primarily on sequence, physicochemical characteristics, or, infrequently, protein structure. We will test the feasibility of using predicted function as a discriminating feature for antigenicity. In contrast to examining antigenicity across broad functional categories, we will use existing Laboratory bioinformatics capabilities to examine function across a spectrum of sources and at varying levels of detail.

We expect to assess the feasibility of using computational methods to screen for probable antigens against pathogens at a proteome scale using functional data at varying degrees of granularity. In addition, we will use functional, structural, and systems biology information to identify patterns of antigenicity as a way of informing experimental research. Using data from various molecular contexts, predictive functions, or features, we believe we can identify proteins likely to provoke an immune system response, and thereby provide insight into the interactions between host and pathogen. Antigen prediction used in this way layers explanation onto computational results, which can guide basic research into mechanisms of pathogenicity, further suggesting avenues for therapeutic intervention.

Mission Relevance

This study will explore the feasibility of devising novel immuno-informatics algorithms and expanding the Laboratory's capabilities in protein informatics to include tools for rapidly and accurately predicting pathogen immune biomarkers in support of LLNL's mission focus area in biosecurity for the rapid mitigation of evolving and unknown biothreats.

FY12 Accomplishments and Results

In FY12 we conducted a study to assess the feasibility of using functional data to predict the immunogenicity of proteins. We employed a number of statistical methods and codes and original protein microarray data provided by a collaborator to build a method for associating various functional annotations with diagnostic outcomes in healthy and sick individuals. Our preliminary results suggest that certain functional annotations may be predictive of immunological reactivity of pathogen proteins. The next step would be further work to confirm whether functional annotations would be useful in selecting antigens for diagnostic protein microarrays.

Optimizing Application Cache Performance

Kathryn Mohror (12-FS-013)

Abstract

Tools for understanding computer application cache performance are critically lacking. Although programmers understand the benefits of temporal and spatial locality, no

tool exists for measuring how close any given piece of code approaches optimal performance. As we move to ever-larger computer clusters, the penalty for suboptimal performance continues to increase. Recent NNSA exascale planning working groups have identified efficient cache layout by applications and the availability of tools for memory efficiency analysis as specific technical challenges for exascale computing capabilities extending beyond the currently existing petascale. With Intel's introduction of several new classes of hardware performance counters, we are now able to revisit this problem. We propose to explore and evaluate the effectiveness of these new counters with regard to understanding and improving cache behavior. We will deliver a robust low-level library for data collection and show how the data it generates is useful for expert analysis.

The expected outcome of this work will be initially limited to developing code and expertise in using novel hardware performance counters to diagnose the effects of data layouts on application performance. Following this six-month study, we can then map out what would be required to translate this work into production-quality tools. Ultimately, we expect a standard tool along the lines of the PAPI (performance application programming interface) library to access performance counters that are able to automatically provide the user with insight as to how computer code may be improved for greater performance.

Mission Relevance

Our tool will provide users with a deeper understanding of program behavior across the memory hierarchy. While this is important on today's machines, it promises to be critical as we move towards exascale computing. Ensuring the readiness of our codes for exascale is integral to the Laboratory's strategic roadmap, and their continued performance contributes to LLNL's core competency in high-performance computing and simulation.

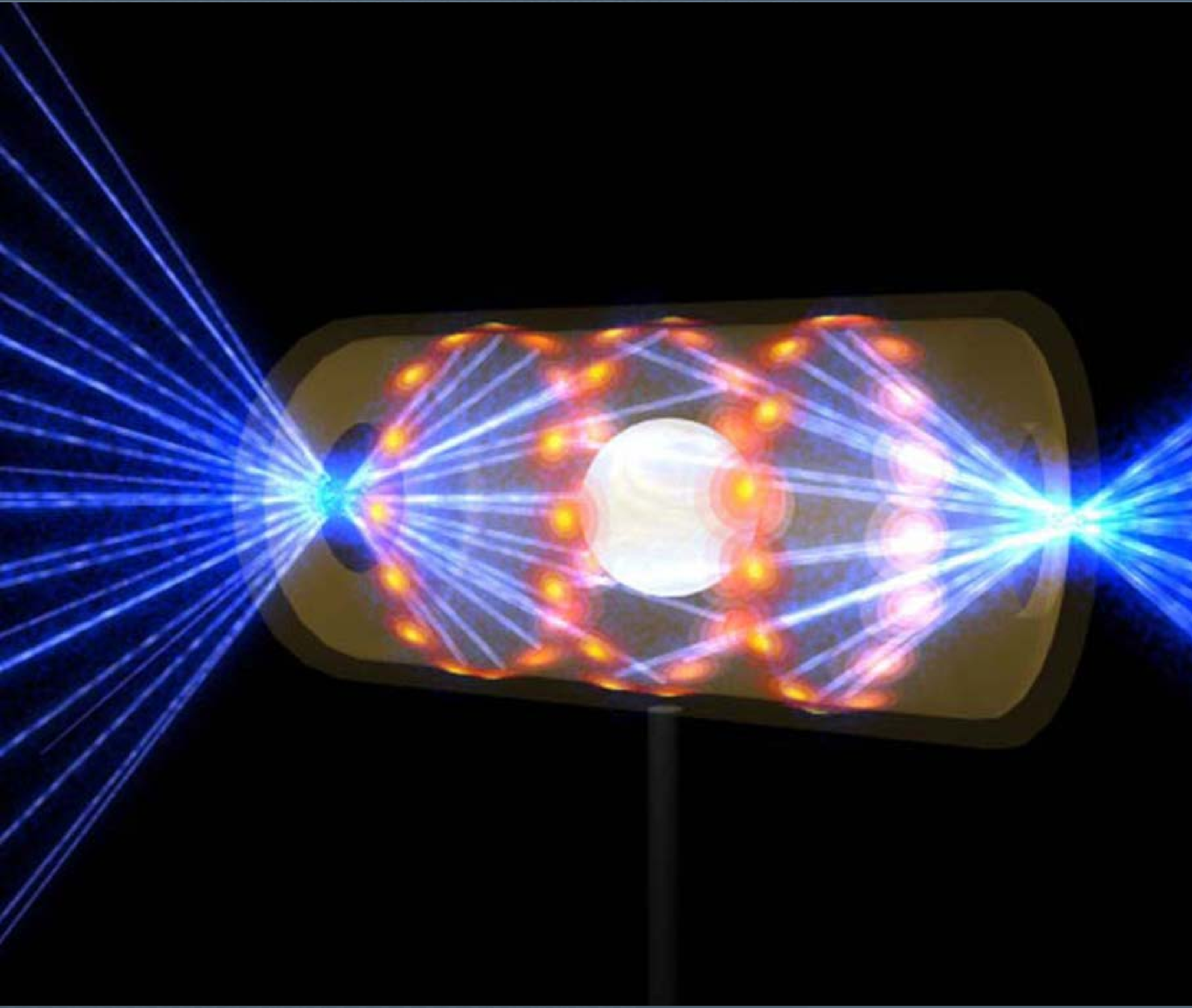
FY12 Accomplishments and Results

In FY12 we (1) created a runtime library to access data from the new memory performance counters on Intel's latest processors; (2) used our library to evaluate the potential and limitations of these new counters; (3) validated our tool for correctness and found that the latency values and memory reference service locations reported were sound, even varying the instruction-sampling rate and finding that our tool gives accurate results even at low rates; and (4) demonstrated that our tool is useful for understanding application memory performance with different versions of a benchmark that had different memory access patterns. In summary, this project created the capability to extract the memory performance characteristics of applications and identify problematic data structures for further analysis and optimization. The next step would be to collaborate with an industrial partner, which we are seeking to do through a Small Business Technology Transfer grant to enable us to commercialize the results of this project for the computing community as a whole.

Publications

Mohror, K., and B. Rountree, 2012. *Cache performance analysis and optimization*. LLNL-TR-604112.

Nuclear Science and Engineering



Laboratory Directed Research and Development

FY2012

Advanced Inertial-Fusion Target Designs and Experiments for Transformative Energy Applications

Peter Amendt (11-SI-002)

Abstract
Inertial-fusion energy research is gaining momentum in the scientific world and the energy arena. A key challenge for realizing fusion energy production is to develop a viable and experimentally validated "point design," which is an integrated simulation used to assess overall performance for the laser target within the larger framework of a fusion energy system. This project will develop central hot-spot ignition and fast-ignition target designs for the laser inertial fusion energy (LIFE) concept, validated with experiments on the OMEGA laser at the University of Rochester and at the National Ignition Facility (NIF) at Livermore. This planned research, founded upon the extensive experience of Laboratory scientists and engineers who developed the point design for NIF, will deliver a suite of 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 energy gain (less than a hundredfold) through innovative measures—such as low drive temperature, rugby-ball-shaped hohlraum target capsules, and radiation shields—will be applied and tested at NIF. In parallel, we will strive to develop a suite of point designs for high-gain (greater than a hundredfold) fast ignition using the high-efficiency hohlraums developed for central hot-spot ignition. Experiments at NIF to test the key physics uncertainties in the fast-ignition concept are also planned.

Mission Relevance
The proposed research could have a groundbreaking, transformative impact on LIFE, fusion energy in general, and high-energy-density matter research. The research will thus help enhance America's national security by developing proliferation-resistant advanced energy technologies, economic security by improving energy efficiency, and energy security by reducing energy imports and greenhouse gases.

FY12 Accomplishments and Results
In FY12 we conducted the first high-density-carbon ablator implosion campaign on the OMEGA laser. Specifically, we (1) demonstrated high-performance plastic carbon–hydrogen baseline ablators to compare with the current LIFE point design of high-density carbon ablators; (2) demonstrated LIFE-relevant fuel-filling methodologies using the drill, fill, and plug technique—excellent core symmetry was demonstrated and the observed neutron yields were 30 to 50% of "clean" predictions; (3) modified the LIFE hohlraum point design to reduce the risk of instability caused by laser–plasma interaction by effecting a nearly equal partitioning of the inner and outer cone peak

powers and energy compared with the FY11 design, which used nearly threefold more energy in the inner cones; and (4) delivered a rugby hohlraum design (without radiation shield pair) for testing on NIF in FY13 to validate the predicted efficiency improvements.

Proposed Work for FY13

In FY13 we propose to (1) conduct an experimental study on the OMEGA laser of cylindrical and rugby-ball-shaped gold and lead hohlraums with laser entrance-hole shields, our goal being to baseline their performance relative to current LIFE target point designs; (2) finalize the LIFE hohlraum point design with high-density carbon and aluminum ablators, using lead hohlraums and aerogel-supported liquid deuterium–tritium fuel loading; (3) complete a laser–plasma interaction assessment of the LIFE point design with high-density carbon ablators; (4) complete a second-generation fast-ignition point design; (5) assess past rugby-ball-shaped hohlraum performance on the OMEGA laser; and (6) analyze the results from high-density carbon implosions on OMEGA in FY12.

Publications

Amendt, P., et al., 2011. "LIFE pure fusion target designs: Status and prospects." *Fusion Sci. Tech*. **60**(1), 49. LLNL-PROC-463711.

Shay, H. D., et al., 2012. "Implosion and burn of fast ignition capsules—Calculations with HYDRA." *Phys. Plasma*. **19**(9), 092706. LLNL-JRNL-550171.

Development of an Approach to Imaging for Use in a Verification Regime

Mark Cunningham (11-ERD-051)

Abstract

Current U.S. national security policies clearly call for moving past Cold War arms control towards verifiable stockpile reductions. The focus is likely to shift to determining whether or not an item is a nuclear warhead, and if so, whether it is most probably in an operationally deployed configuration. Imaging may play an important role in these verifications. Our objective is to establish the utility and optimization of a new concept for a gamma-ray imaging system appropriate for nuclear treaty verification and on-site inspections 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 expect a successful project will establish the utility of a new type of gamma-ray imaging system for future arms-reduction treaty applications. This will be performed by

determining the efficacy of a set of metrics derived from gamma-ray images, such as the density and isotopic profile of materials that are used in warhead construction, which is a new concept that has not been realized for objects such as a nuclear warhead. In addition, we will develop a prototype gamma-ray imager design that can be used for building a proof-of-concept prototype with follow-on funding.

Mission Relevance

This research supports the central Laboratory strategic missions of ensuring national security—specifically, reducing the threat of nuclear attack by a rogue state or terrorists through technologies that support nuclear nonproliferation treaties.

FY12 Accomplishments and Results

In FY12 we (1) successfully validated our Monte Carlo simulations of radiation transport in three-dimensional objects and in a gamma-ray imager by comparing the images from the LLNL-built gamma-ray imaging spectrometer system to our Monte Carlo simulations, (2) finished a study of a range of direct and indirect attributes, and (3) began designing an optimized gamma-ray imaging system (the building and testing of the optimized system is no longer part of this project).

Proposed Work for FY13

In FY13 we propose to (1) optimize the filter for two-dimensional coded-aperture gamma-ray images, (2) modify the three-dimensional reconstruction algorithm to accommodate noncontiguous radiation sources, and (3) design a prototype coded-aperture gamma ray that is optimized for arms-control applications.

The High-Energy-Density Science of Inertial-Confinement Fusion Ignition

Gilbert Collins (12-ERD-076)

Abstract

Achieving controlled thermonuclear burn at the National Ignition Facility using inertial-confinement fusion is a major goal for the Laboratory. Optimizing implosions for igniting and propagating thermonuclear burn requires understanding high-energy-density states over an unprecedented range of conditions. However, to date, none of the physical models integral to ignition simulations has benchmarked data for most of the ignition implosion regime. Using results from the National Ignition Campaign as a guide, we will assess, benchmark, and improve key physical models used for inertial-confinement fusion calculations. These models include equation-of-state, opacity, nuclear reactions, kinetics, and electric- and magnetic-field generation. Models will also incorporate transport quantities such as thermal conduction, electron–ion equilibration, ion-stopping power, viscosity, and the interplay between these areas of physics in a high-energy-density environment.

We expect to develop advanced, benchmarked models of the underlying physical processes that control inertial-confinement fusion ignition in concert with validation measurements and data from integrated ignition experiments. Versions and representations that can be used directly in target design calculations will be developed and qualified. The resulting simulations will include quantifiable uncertainties associated with underlying physical processes. This approach should significantly improve the predictive power of target design calculations and agreement with ignition experiments. Our research will also allow identification of potential new science in the extreme physical regimes where inertial-confinement fusion targets are ignited.

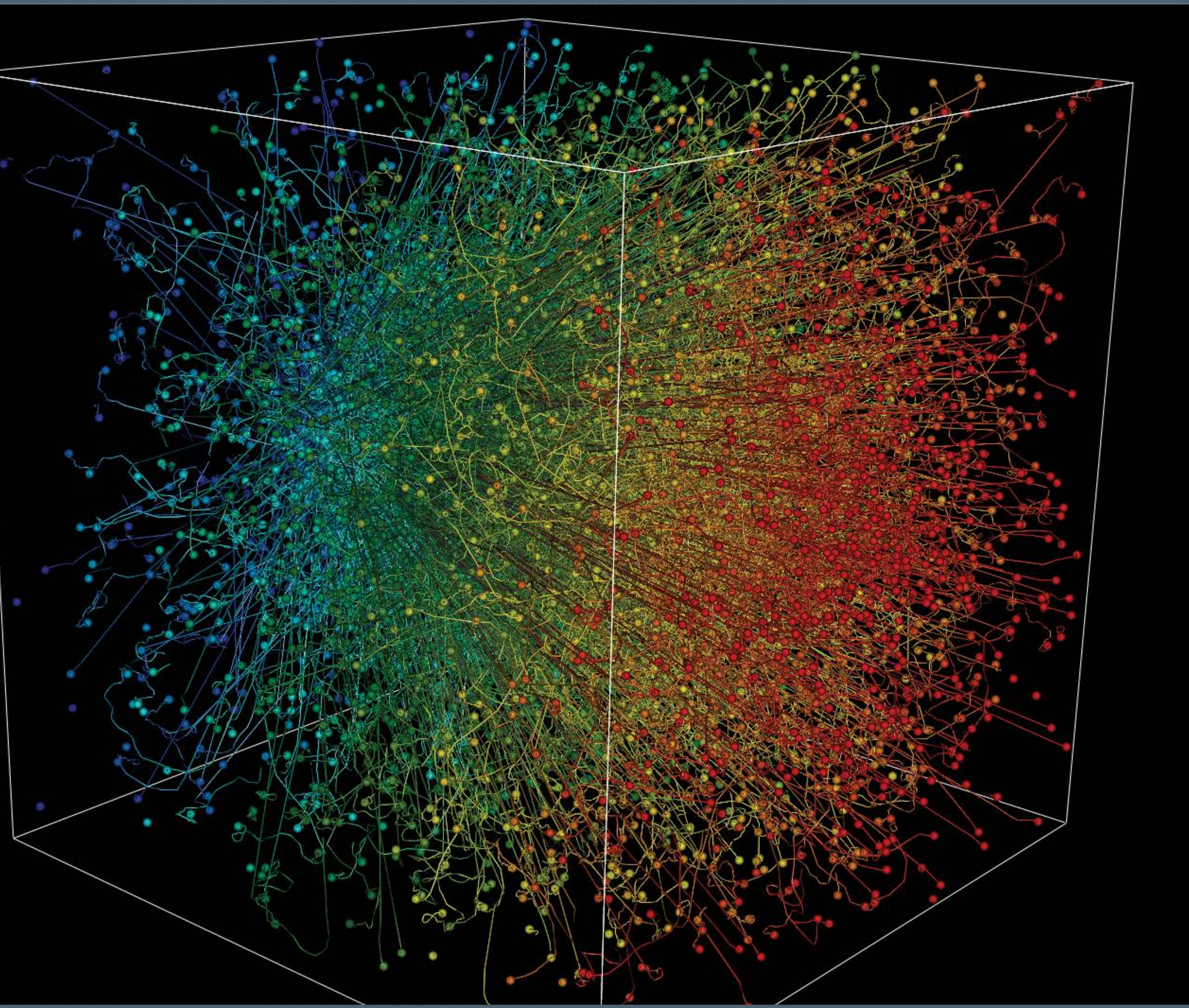
Mission Relevance

This work is closely aligned with LLNL missions in stockpile stewardship science and basic research in high-energy-density science. Project experiments are important for optimizing the robust ignition platform that is a critical goal of the National Ignition Facility. The fundamental high-energy-density physics being researched and modeled under this project is common to inertial-confinement fusion targets and nuclear explosions.

FY12 Accomplishments and Results

We started work on the project in late FY12 and (1) began to design equation-of-state experiments to guide theory for regimes that are important to improving ignition implosion performance, (2) outlined key areas of radiation production and transport to improve ignition implosion performance, and (3) began to design experiments for hohlraum target capsules and ablators to address opacity and emissivity questions. We deferred the onset of calculations and modeling because of funding constraints. The project gave us a start in defining an experimental campaign to understand the underlying physical processes in target capsules. A great deal of design work was completed in the relatively short period of research and the team will transition to ongoing programmatic work in support of ignition target physics.

Physics



Laboratory Directed Research and Development

FY2012

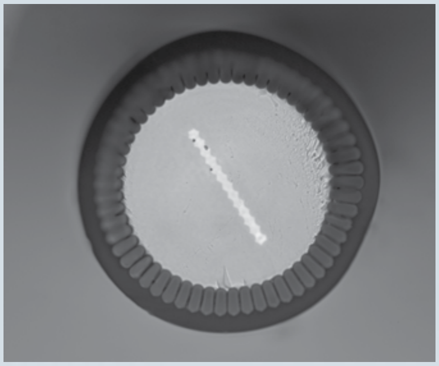
Science and Technology of Unconventional Fiber Waveguides for Emerging Laser Missions

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 new 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..



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

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.

FY12 Accomplishments and Results

Our program has been highly successful at meeting its goals. In FY12 we (1) fabricated both passive and active (rare-earth laser ion-doped) versions of the ribbon fiber using LLNL's new optical fiber production capability; (2) demonstrated clearly that we can launch high-purity, higher-order modes into these fibers and convert them back to a Gaussian beam via a variety of techniques including diffractive optical elements, mode coupling by application of controlled pressure to the fiber waveguide, off-axis launch, and launch through a phase plate; (3) shown our active ribbon fiber can stably amplify a higher-order mode by greater than a hundredfold or be operated as an oscillator in a single higher-order mode; and (4) developed new waveguide concepts in the area of flattened higher-order modes, which resulted in three patents in progress. We believe these new ideas will be important in a number of applications in telecommunications, transport of high-power laser light, and possibly advanced dielectric laser accelerators. Demonstration of the ribbon fiber technology has led to outside interest in continued development, and we will be submitting a proposal with an industry collaborator relevant to solid-state lasers. Further, a number of outside collaborators have been impressed with the demonstration of our capability to fabricate complex photonic crystal fiber structures. We believe the results of our project will generate interest in LLNL's fiber production and fiber laser research for years to come.

Publications

Bullington, A., et al., 2012. "Mode conversion in rectangular-core optical fibers." *Appl. Optic*. **51**(1), 84. LLNL-JRNL-493311.

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Sridharan, A., et al., 2012. "Mode converters for rectangular-core fiber amplifiers to achieve diffraction-limited power scaling." *Proc. SPIE, Laser Technology for Defense and Security VIII*, vol. 8381, p. 838103. SPIE, Bellingham, WA. LLNL-ABS-567412.

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 from hundreds of kilometers away 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 the technology for standoff

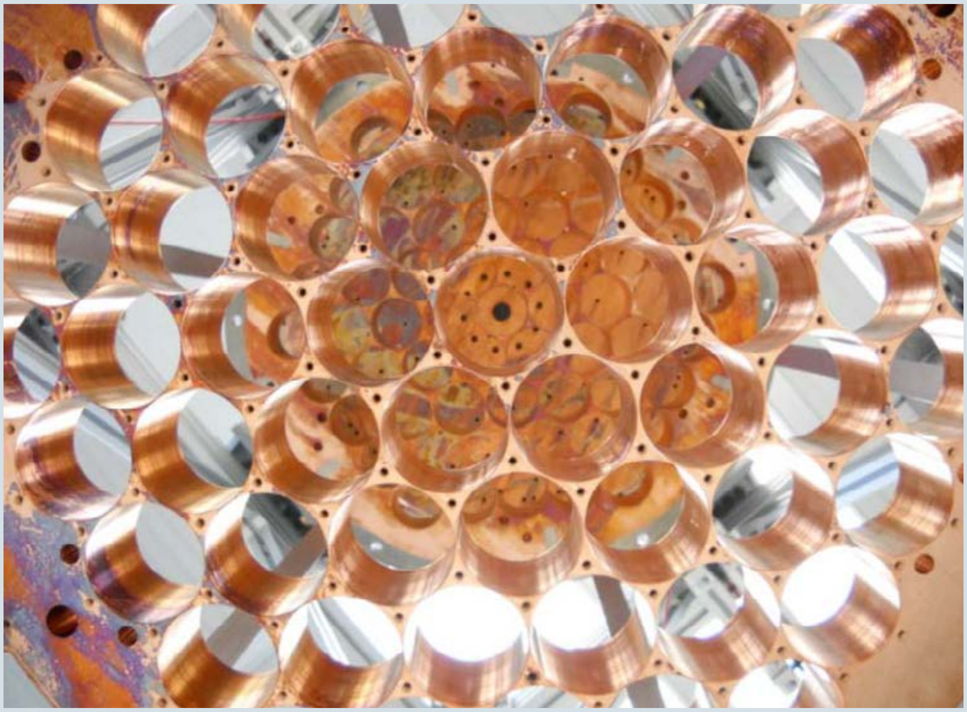
detection of nuclear reactors with antineutrino detectors, which will enable the discovery of operating reactors across borders, as well as provide reactor safeguards. In addition, we will develop technologies for fissile material search and monitoring by addressing essential needs for practical deployed systems, including neutron and gamma-particle identification; directionality; nanosecond-scale timing resolution; low-cost, large-solid-angle coverage; and spectroscopy.

Mission Relevance

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 Laboratory access to cutting-edge technology, expertise, and potential hires.

FY12 Accomplishments and Results

We (1) published the world's most precise measurement of the neutrino mixing angle theta-13; (2) helped deploy the Large Underground Xenon Detector underground on schedule, although the dark matter search was delayed by problems at the hosting facility—LLNL will remain as an author on the world-record dark matter search results likely to be announced in FY13; and (3) continued 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. In summary, this project developed novel rare-event techniques for use in a range of nonproliferation applications, including reactor monitoring with antineutrinos and efficient, low-energy neutron spectroscopy



A key component of the ultrasensitive Large Underground Xenon Detector, which will search for dark matter, is the copper photomultiplier tube mounting structure designed and built by Lawrence Livermore National Laboratory.

for fissile material monitoring. The technology we developed for remote reactor discovery has led to a $4 million new project from NNSA, and the Office of Science is supporting follow-on work on short-baseline neutrino oscillations and the Large Underground Xenon Detector. Furthermore, NNSA and the Office of Science are, based on ideas developed in the project, considering for the first time jointly funding a $100 million nonproliferation and science fusion exercise. The project also helped recruit a world-class low-temperature physicist and five postdoctoral researchers, one of whom was named Laboratory Postdoc of the Year in 2012 and two of whom were converted to staff; led to one master's degree and four doctorate degrees; and generated numerous peer-reviewed publications.

Publications

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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.

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Sorensen, P., 2012. *From dark matter to neutrino–nucleus scattering in liquid noble gas detectors*. LLNL-PRES-607022.

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Mix at the Atomic Scale

Paul Miller (10-ERD-004)

Abstract

The need to compute the effects of mixing 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 will develop a transport theory with the aid of molecular dynamics and continuum simulations. Large-scale simulation problems will be used for discovery and exploration,

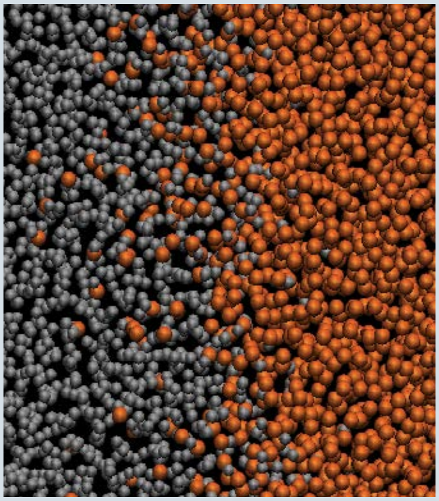
to gauge the adequacy of the methods, and to assess model improvements. Through an analysis of these simulations, we will begin developing a more complete, atomically informed model of mixing for use in continuum calculations. By the end of this project, we expect to have made significant improvements 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.

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.

FY12 Accomplishments and Results

In FY12 we (1) used our Yukawa capability of electrical potential between ions to simulate diffusivity and viscosity in a hot, dense mixed-plasma case; (2) completed our work on the equation of state derived from the molecular dynamics code; (3) completed the comparison of our molecular dynamics and continuum results; and (4) provided assessment of dilute hard-sphere and dilute Lennard–Jones models using our simulated self-diffusivities (including finite-size effects) and derived an empirical analytic formulation, assessed cross-correlation effects, and compared our results to experimental transport coefficients. The results of this project provide new insights into the modeling of self- diffusion, inter-diffusion, and transport coefficients of liquid-metal and dense-plasma systems over a range of temperatures, pressures, and concentrations. Follow-on activities from our research were incorporated into the ongoing Cimarron Project, a multi-institution collaboration dedicated to the prediction and measurement of properties of dense, highly collisional, and strongly-coupled plasmas.



Molecular dynamics simulation of diffusion broadening of an interface between hydrogen (gray) and argon (orange) ions. Atomic-scale diffusion plays a central role in reactions of non-premixed systems as well as other physical processes.

Publications

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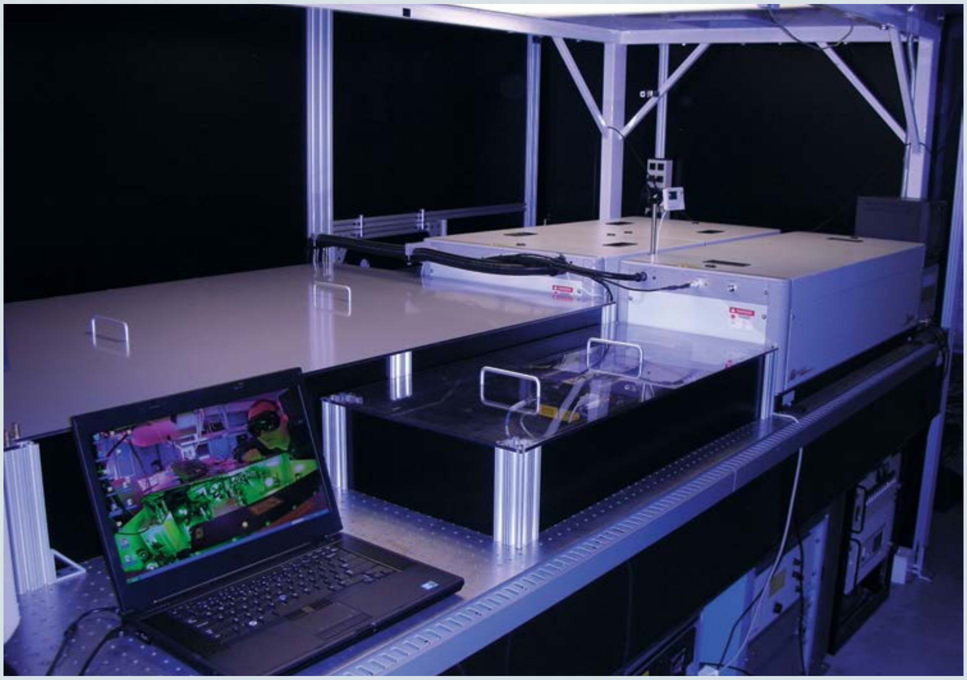
High-Gradient Inverse Free-Electron Laser Accelerator

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. The technology has the potential to be 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 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 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.



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.

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.

FY12 Accomplishments and Results

In FY12 we (1) completed the high-power laser system—design specifications of 500 mJ, 100 fs, and 10 Hz repetition rate were demonstrated; (2) completed construction of the vacuum transport system and successfully propagated the laser to the IFEL undulator; and (3) conducted IFEL acceleration experiments—spatial and temporal overlap was achieved as well as beam size and focal position optimization. Our project has enabled the capability to generate multi-terawatt laser pulses and high-brightness electron beams synchronized to the 100-fs level for a variety of radiation production and advanced acceleration applications, in addition to demonstrating the IFEL as a viable accelerator technology for compact applications requiring hundreds of megaelectronvolt beams. In addition, the project will provide Ph.D. thesis data for a University of California, Los Angeles graduate student. Continuing research will be pursued through Defense Threat Reduction Agency funding of compact gamma-ray source development and through other agencies as a novel x-ray free-electron laser driver.

Publications

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Modeling and Measuring Quark–Gluon Plasma Shock Waves

Ron Soltz (10-ERD-029)

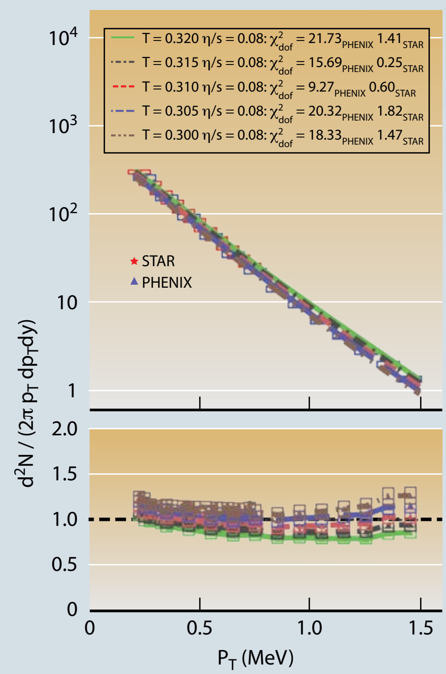
Abstract

The possibility of developing shock waves in heavy-ion collisions was first suggested as a consequence of the compression of nuclear matter during 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 if 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.



STAR and PHENIX experimental data at the Large Hadron Collider in Switzerland compared with results obtained with the CHIMERA (comprehensive heavy-ion model evaluation and reporting algorithm) simulation package developed under this project.

FY12 Accomplishments and Results

In FY12 we (1) used our CHIMERA (comprehensive heavy ion model evaluation and reporting algorithm) framework, which runs multistage hydrodynamic models coupled to the microscopic transport of hadronic collisions, to compare modeling results to experimental signatures measured in heavy ion collisions at the Large Hadron Collider and the Relativistic Heavy Ion Collider at Brookhaven National Laboratory; (2) used these initial comparisons to place statistically rigorous constraints on the initial temperature- and viscosity-to-entropy ratios; and (3) began implementing these tools in a new hydro model based on the Chombo partial differential equation solver, which will enable us to accurately model jet energy loss and the initial state geometry fluctuations needed to describe higher moments, thereby providing a complete description of a heavy-ion collision. In summary, this project produced the most sophisticated comparisons ever achieved between the models of quark–gluon plasma evolution and experimental data collected in relativistic heavy ion collisions at the Relativistic Heavy Ion Collider and the Large Hadron Collider. The DOE Nuclear Physics Program has expressed interest in supporting the continuation of this work.

Publications

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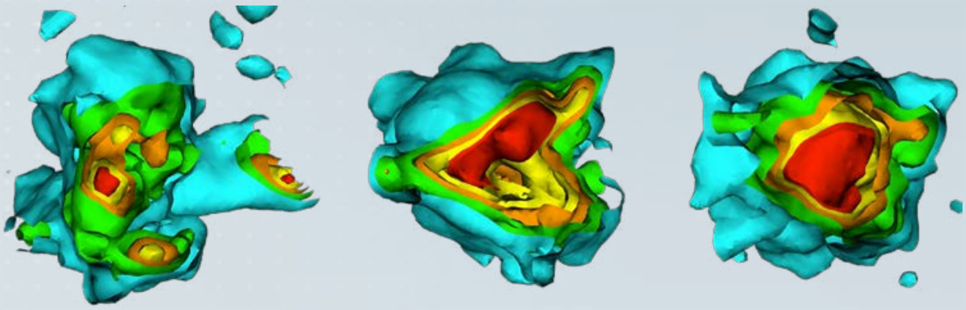
Unlocking the Universe with High-Performance Computing

Pavlos Vranas (10-ERD-033)

Abstract

We propose to use large-scale numerical simulations on LLNL's BlueGene supercomputers—in particular Sequoia, which has ranked as the world's fastest—to unravel the inner workings of the universe. We will explore 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 (LHC) 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



Cutaway views of three-dimensional contour plots of the neutron electric charge-density magnitude in strongly interacting theories with 2-, 6-, and 10-flavor particle species (left to right, respectively) from simulations on Livermore's Sequoia supercomputer. Larger flavor numbers may be discovered at the Large Hadron Collider near Geneva, Switzerland.

help understand and guide the largest and highest-energy experiments of our time—those performed at the LHC. No other groups in the world are pursuing this type of research at this scale.

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.

FY12 Accomplishments and Results

We (1) conducted special unitary group-2 Yang–Mills theory large-lattice simulations, obtaining the measurements of the running coupling and establishing intricate details of the theory and how it relates to discovery of the Higgs particle at the LHC; (2) completed numerical simulations with physical 140-MeV pions and an equation-of-state simulation with staggered fermions; and (3) completed our study on clover lattices of the lattice QCD nuclear parity violation and the S-wave scattering of strangeness-3 baryons. The successful conclusion of this project demonstrated that strongly coupled theories are viable candidates to explain experimental results at LHC, and has calculated finite- and zero-temperature QCD properties that are directly relevant to corresponding experiments. Follow-on research on lattice strong dynamics in the LHC post-Higgs discovery era, with the central goal of predicting and explaining the new experimental tera-electronvolt physics results, will be supported by DOE's Office of High-Energy Science. DOE's Office of Nuclear Physics and the Scientific Discovery through Advanced Computing Program will support follow-on research on finite- and zero-temperature lattice QCD studies relevant to the Relativistic Heavy Ion Collider experiments and nuclear physics experiments at Brookhaven National Laboratory.

Publications

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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 in the 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.

FY12 Accomplishments and Results

In FY12 we (1) carried out crystalline searches for nitrogen-based high-energy-density materials based on liquid molecular dynamics simulations and identified nitrogen–carbon and nitrogen–silicon compounds as best candidates, which were investigated thoroughly, resulting in the prediction of novel, metastable structures—we additionally fine-tuned the nitrogen–silicon structures and determined the energy that they can store; (2) performed simulations of carbon dioxide liquid mixtures, as well as various carbon–oxygen stoichiometry ratios—because new experimental data on carbon dioxide appeared during the year, we turned our attention back to pure carbon dioxide to better determine its high-pressure and temperature stability; and (3) developed efficient methods for finite-temperature phase stability that included free-energy calculations of compositionally complex systems and phase-search techniques—our focus remained mostly on the former because the importance of this development has become evident. We have formulated a general methodology for free-energy calculations and performed a comprehensive test on a diverse set of systems. This project has resulted in the discovery of novel nitrogen-based high-energy-density materials, new approaches for finite-temperature phase searches, and a powerful method for free-energy calculations of liquids, which makes possible the study of complex systems that were previously not feasible and has already been applied to important problems in high-pressure physics. Our free-energy calculation capabilities are now being used in the equation-of-state program at Livermore, and the crystalline search techniques will be implemented and further developed for programmatic Laboratory work.

Publications

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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.

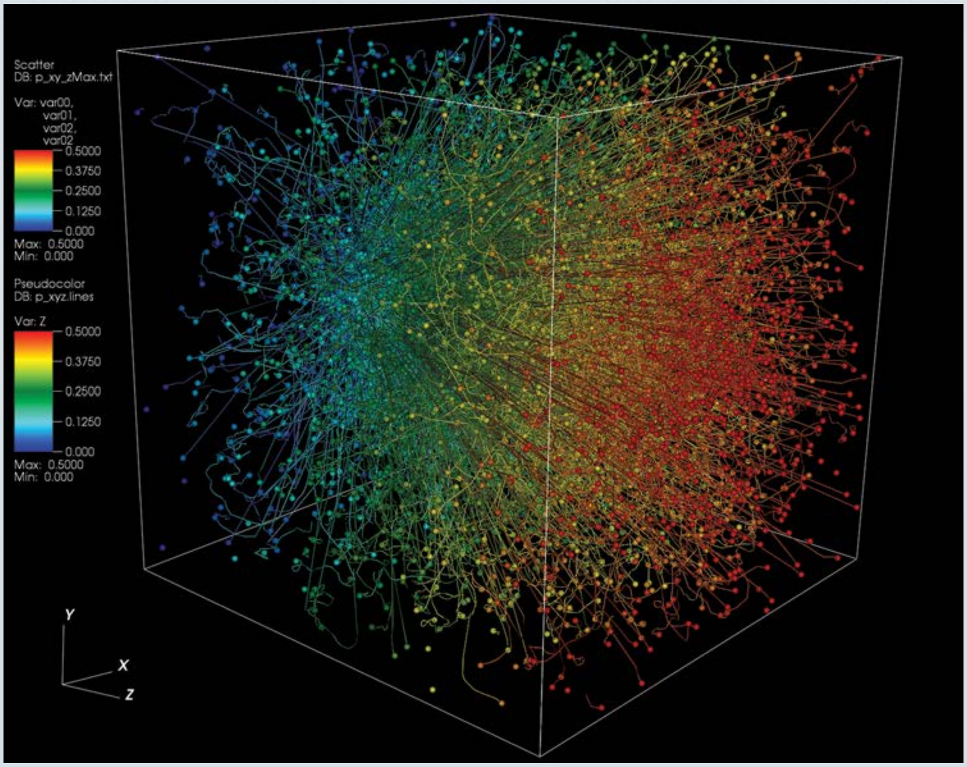
We expect to lay the scientific foundation for a new class of intense positron sources. In particular, we will (1) develop an understanding of the detailed physics behind laser-to-positron coupling, (2) investigate physics of the sheath electric field behind the target and its effect on average positron 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 scientific 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.

FY12 Accomplishments and Results

In FY12 we (1) completed the experiments on positron emittance and annihilation radiation; (2) completed several new campaigns on positrons—in particular, new positron data was taken on a high-intensity laser in Osaka, Japan as well as in the United Kingdom; and (3) completed the most comprehensive simulations of laser-generated



Simulation of the positrons created as an ultra-intense laser hits a millimeter-thick slab of solid gold. The dark blue positrons exit back towards the laser, and the red positrons make it out of the rear of the target. These positrons are detected in our unique electron–positron spectrometer that has been used on the Titan laser at Lawrence Livermore and the OMEGA Extended Performance laser at the Laboratory for Laser Energetics in Rutherford, New York, and more recently on the Firex laser in Osaka, Japan.

positrons to date. Our research has led to a novel method of one-laser-pulse positron acceleration to high energies, and also a novel neutron source, for which we are obtaining a patent. We intend to pursue positron shots on the National Ignition Facility once the Advanced Radiography Capability ultra-short pulse laser is completed.

Publications

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Wilks, S. C., 2011. "Electron-positron jets created by ultra-intense lasers." *Bull. Am. Phys. Soc*. **56**(16), UI2.00001. LLNL-ABS-491095.

Multiscale Polymer Flows and Drag Reduction

Todd Weisgraber (10-ERD-057)

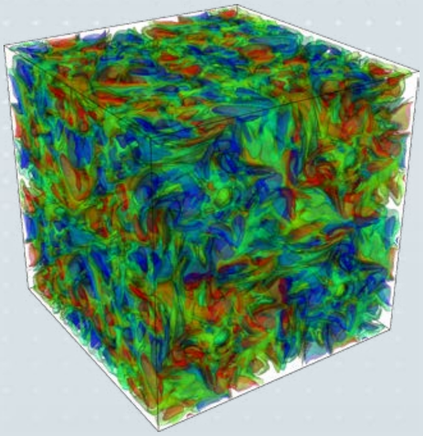
Abstract

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, multiscale simulations of drag reduction that will increase our understanding of the underlying physics.

We expect to perform a series of large-scale simulations to provide valuable insight into a fundamental hydrodynamics problem, including a more detailed understanding of the complex dynamics of polymers and turbulence. Additional knowledge of drag reduction could conceivably make it possible to optimize this effect in more practical applications. A modest decrease in drag would dramatically improve the deployment efficiency of naval vessels and reduce expenditures in the civilian sector. In addition to the fundamental science, we will also develop a high-performance computing capability applicable to biosecurity and Rayleigh–Taylor instability research.

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,



A simulation, conducted with 1,500 processors, of a vortex structure in a turbulent flow as part of our research into the fundamental mechanisms of drag reduction.

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.

FY12 Accomplishments and Results

In FY12 we focused on the role of distributed wall roughness in the onset of turbulence in channels. Specifically, we (1) demonstrated for the first time with our simulations that small-amplitude wall topology can initiate turbulence without any other flow perturbations, (2) showed that the wavelength of flow structures induced by the wall is a function of the Reynolds number used to characterize different flow regimes, and (3) completed development of our lattice-Boltzmann adaptive-mesh refinement algorithm.

Proposed Work for FY13

In FY13 we will continue simulations of roughness-induced transition to turbulence. Specifically, we will (1) investigate the role of smaller amplitude roughness elements and generate a neutral stability curve for transition, (2) analyze evolution of the frequency and structural content of the flow to elucidate how roughness disturbances are amplified, and (3) implement hydrodynamic fluctuations in our adaptive-mesh framework to understand their contribution to the onset of turbulence.

Publications

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Weisgraber, T., and B. J. 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

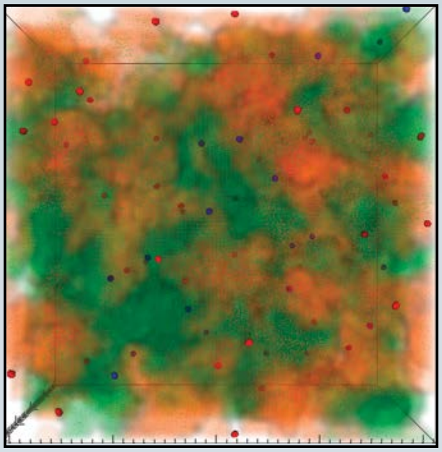
Jonathan DuBois (10-ERD-058)

Abstract

First-principles simulation of materials properties has played an essential role in 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. This will increase the accuracy of state-of-the art calculations by two orders of magnitude, so that the typical error lies within the chemical accuracy bound. We will also develop a constant-pressure diffusion Monte Carlo algorithm in which the proton and electron degrees of freedom, along with the cell degrees of freedom, can adjust. Finally, we will implement several novel enhancements to the standard path-integral quantum Monte Carlo method using the knowledge garnered in ground-state quantum Monte Carlo calculations to extend the path-integral quantum method to lower temperatures.



Snapshot of a full many-body path-integral quantum Monte Carlo simulation of a strongly coupled degenerate plasma mixture of lithium (red spheres) and hydrogen (blue spheres). The instantaneous delocalized electron distributions are represented as orange (spin up) and green (spin down) point clouds. This calculation is representative of a new first-principles simulation capability.

Mission Relevance

Accurate first-principles simulations of materials are essential for current and future 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.

FY12 Accomplishments and Results

In FY12 we (1) completed the exact evaluation of low-temperature helium-3, (2) performed a comprehensive path-integral Monte Carlo study of a finite-temperature homogeneous electron gas, (3) laid the theoretical and algorithmic framework for use of our unbiased Monte Carlo method for determining thermal conductivity of fermionic systems and began development of an exact fermion path-integral Monte Carlo approach for inhomogeneous systems, (4) performed extensive studies of candidate structures for solid hydrogen at high pressure with high-fidelity first-principles simulations in this regime, and (5) conducted, using our newly developed constant-pressure ensemble Monte Carlo code, studies of phase transitions in quantum hard-sphere models.

Proposed Work for FY13

During the two-month continuation of this project in FY13, we will complete current work on the finite-temperature homogeneous electron gas. We will also continue to work on the formulation of a general simulation approach to exact finite-temperature many-body fermion systems capable of efficiently treating inhomogeneous systems.

Publications

Brown, E. W., et al., 2012. *Path integral Monte Carlo simulation of the warm-dense homogeneous electron gas*. LLNL-JRNL-604175-DRAFT.

DuBois, J. L., 2011. *Algorithms for exact quantum simulations at the exascale*. LLNL-PROP-517051.

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Tubman, N., J. L. Dubois, and B. Alder, 2012. "Recent results in the exact treatment of fermions at zero and finite temperature." *Advances in quantum Monte Carlo*. American Chemical Society, Washington, DC, p. 41. LLNL-BOOK-493754.

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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.

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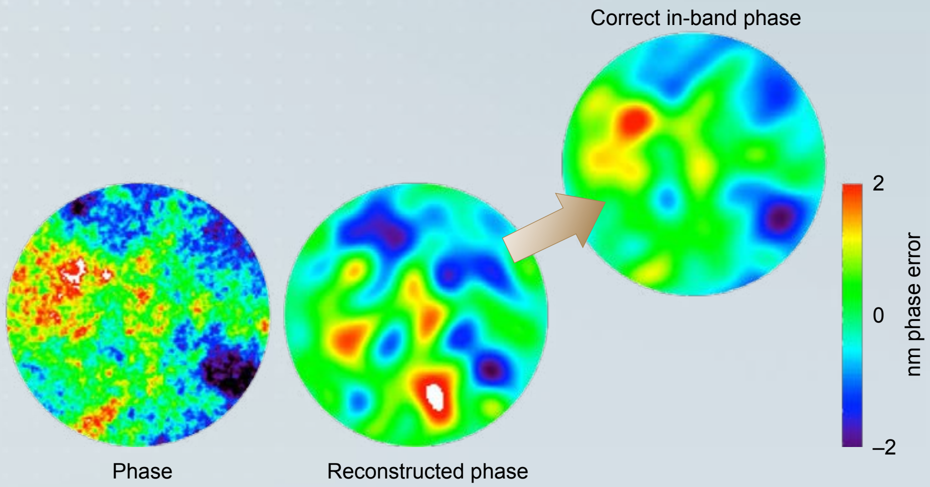
Whitley, H. D., J. L. DuBois, and K. B. Whaley, 2011. "Theoretical analysis of the anomalous spectral splitting of tetracene in ^{4}He droplets." *J. Phys. Chem. A* **115**(25), 7220. LLNL-JRNL-464446.

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 Stanford's SLAC Linac Coherent Light Source and the National Synchrotron Light Source II at Brookhaven National Laboratory. This new class of x-ray optics will



A simulation illustrating how aliasing can complicate the measurement of phase and that the resultant error may equal the in-band aberration to be measured.

unleash the full scientific potential of these new national facilities. Our research builds on recent LLNL efforts to develop the primary x-ray mirror systems for the Linac Coherent Light Source and the adaptive-optic-based Gemini Planet Imager to be initially deployed on the Gemini South telescope in the Chilean Andes. Livermore's acknowledged world leadership in extreme adaptive optics with state-of-the-art design, coupled with fabrication expertise developed for the Linac Coherent Light Source, provides the foundation necessary for our novel research in adaptive x-ray optics.

We expect to produce a prototype system, including the x-ray adaptive optic, a wave-front sensor, control and sensing algorithms, and a detailed performance model. It will be developed and tested at an existing light source. This research will establish a national capability that all DOE facilities and labs can draw upon to build new beam lines or end stations. By enabling delivery of coherent or nanometer-scale focused x-rays, this research will enable advances in physics, chemistry, and biology. If prototype systems can be developed and proven effective, this pioneering research could serve as the basis for many instruments at future facilities that require x-ray imaging.

Mission Relevance

This research supports the Laboratory's foundations in energy manipulation and advanced measurements, enabling fundamental scientific discoveries in diverse areas relevant to LLNL and DOE. In particular, 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.

FY12 Accomplishments and Results

In FY12 we (1) completed detailed evaluation of three mirror actuation concepts and selected a single design for construction—a major milestone for the project; (2) finished developing a comprehensive suite of modeling and simulation tools to evaluate the actuation, wave-front sensing, and propagation of coherent radiation through these optical components; and (3) incorporated strain gages into the mirror design in lieu of the prototype wave-front sensor.

Proposed Work for FY13

In FY13 we propose to (1) finish fabricating the mirror system, including several tens of actuators to correct the most performance-limiting figure errors, positional feedback to measure and control actuation, and electronic drivers and software control to adjust actuation on appropriate timescales; (2) calibrate the mirror using visible wavelength techniques at LLNL and then test the mirror at wave length at a DOE x-ray light source, possibly the Stanford Synchrotron Radiation Lightsource or the National Synchrotron Light Source at Brookhaven National Laboratory; and (3) compare measured performance to predictions derived from modeling and simulation tools developed earlier in the project.

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, U.K., Apr. 4–5, 2011. LLNL-ABS-510933.

Pardini, T., et al., 2012. "Aliasing in a Hartmann wavefront sensor at x-ray wavelengths." *Proc. SPIE* **8503**, 85030H. LLNL-CONF-569272-DRAFT.

Poyneer, L. A., B. Bauman, and B. A. Macintosh, 2012. "Simulating wavefront correction via deformable mirrors at x-ray beamlines." *Proc. SPIE* **8503**, 85030I. LLNL-PROC-568453-DRAFT.

Advanced Algorithm Technology for Exascale Multiphysics Simulation

Charles Still (11-ERD-017)

Abstract

We propose to develop computational algorithms that will enable multiphysics simulation codes to efficiently utilize exascale computers. Within the next decade, NNSA plans to site an exascale computer at LLNL with a thousandfold performance increase and only a tenfold electricity requirement. The system architecture is a dramatic departure from hardware of today, and codes will have to change drastically to utilize it. We will characterize which multiphysics 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 an exascale-tuned hydrodynamics and diffusion reference code as proof of principle on exascale surrogate hardware.

Current multiphysics codes were designed to meet criteria that are very different from the demands of exascale systems. Power constraints and performance targets are driving those future systems to have more than a thousand cores per node, leading to restricted memory and message-passing bandwidth per node. Algorithms must exhibit significant intra-node concurrency while limiting data motion to take advantage of these systems. Using a coupled hydrodynamics–diffusion reference code as an example, we will develop computational algorithms that will enable multiphysics simulation codes to efficiently utilize exascale machines.

Mission Relevance

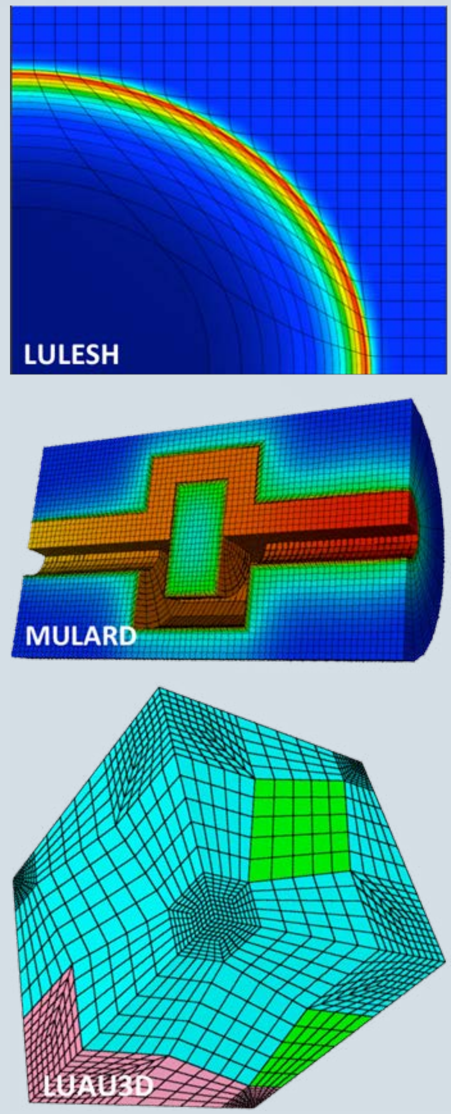
There is a pressing need to ensure that the current Livermore suite of advanced simulation and computation codes for defense applications can successfully migrate to the exascale computers coming to LLNL. Our research will provide the performance metrics and methodology needed for transitioning multiphysics codes to exascale systems, along with a coupled arbitrary Lagrangian–Eulerian (ALE) hydrodynamics and diffusion reference code. This work furthers the Laboratory's stockpile stewardship science mission and will bolster Livermore's scientific and technological foundation in high-performance computing and simulation.

FY12 Accomplishments and Results

In FY12 we (1) developed multiple-group diffusion and advection proxy applications representative of our research code xALE; (2) explored memory-motion and parallelism metrics; (3) developed code and data transformations to optimize proxies for the given metrics; (4) began work on integrating those transforms into xALE; (5) performed initial investigations of massive multithreading and computational transforms in xALE; (6) delivered an optimized Lagrangian hydrodynamics proxy LULESH, which shows a twofold to threefold performance improvement on exascale surrogate architectures; (7) explored, in collaboration with university and private-sector partners, programming models for future architectures; and (8) began knowledge transfer to Livermore's Advanced Simulation and Computing platforms.

Proposed Work for FY13

In FY13 we will (1) explore metrics based on memory and parallelism, rather than floating-point performance; (2) create proxy applications for radiation diffusion and hydrodynamic advection; (3) produce code and data transformations that improve the metrics on the proxy applications; and (4) complete our analysis on whether the individual node-focused optimizations can be combined into an efficient, scalable code on Livermore's Sequoia supercomputing system.



In 2012 we delivered an optimized Lagrangian hydrodynamics proxy application (LULESH) and new proxy applications for multigroup radiation diffusion (Mulard) and advection (LUAU3D) advanced simulation and computation codes for defense applications.

Publications

Brunner, T. A., 2012. *A new multigroup thermal radiation diffusion mini-application*. SIAM Conf. Parallel Processing for Scientific Computing (PP12), Savannah, GA, Feb. 15–17, 2012. LLNL-ABS-494432.

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Brunner, T. A., 2012. *Mulard: A multigroup radiation diffusion compact application*. LLNL-PRES-581073.

Brunner, T. A., 2012. *Mulard: A multigroup thermal radiation diffusion mini-application*. Exascale Research Conf. Mtg., Arlington, VA, Oct. 1–3, 2012. LLNL-PROC-537572.

Karlin, I., et al., 2012. *Exploring traditional and emerging parallel programming models using a proxy application*. 27th IEEE Intl. Parallel and Distributed Processing Symp., Boston, MA, May 20–24, 2003. LLNL-CONF-586774.

Karlin, I., et al., 2012. *Memory and parallelism tuning exploration using the LULESH proxy application*. 2012 Intl. Conf. High-Performance Computing, Networking, Storage, and Analysis (SC12), Salt Lake City, UT, Nov. 10–16, 2012. LLNL-ABS-567120.

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Karlin, I., et al., 2012. *Programming model support necessary for adapting high-performance code to differing platforms*. 2012 SIAM Conf. Computational Science and Engineering, Paphos, Cyprus, Dec. 5–7, 2012. LLNL-ABS-580414.

Karlin, I., et al., 2012. *Transformations to ALE miniapps for current and arriving architectures*. LLNL-PRES-580614.

Karlin, I., et al., 2012. *Tuning the LULESH mini-app for current and future hardware*. 2012 Nuclear Explosive Code Development Conf., Livermore, CA, Oct. 10–26, 2012. LLNL-ABS-564179.

Miller, B. J., and N. A. Gentile, 2012. *Monte Carlo Benchmark (MCB)*. LLNL-PRES-557033.

Still, C. H., 2011. *Challenges beyond the software pipeline*. Salishan Conf. High-Speed Computing, Gleneden Beach, OR, Apr. 25–28, 2011. LLNL-PRES-481274.

Still, C. H., 2012. *High-level abstractions for portable performance using LULESH*. 2012 Intl. Conf. High-Performance Computing, Networking, Storage, and Analysis (SC12), Salt Lake City, UT, Nov. 10–16, 2012. LLNL-ABS-580432.

Still, C. H., 2012. *Using mini-applications to explore multiphysics codes at exascale*. LLNL-PRES-528656.

Still, C. H., et al., 2012. *AATEMPS: Advanced algorithm technology for exascale multiphysics simulation*. LLNL-PRES-541772.

Temperature-Dependent Lattice Dynamics and Stabilization of High-Temperature Phases from First-Principles Theory

Per Söderlind (11-ERD-033)

Abstract

We propose to extend our current capability in quantum-mechanical equation-of-state and materials investigations by developing and implementing a new finite-temperature self-consistent phonon calculation scheme for large-scale, first-principles electronic structure calculations on massively parallel computer platforms. By combining state-of-the-art electronic structure theory with the proposed self-consistent phonon approach, we aim to dramatically improve our capability for predicting high-temperature phase diagrams of materials and provide a complementary approach to the more time-consuming quantum molecular dynamics simulation method. Specifically, we will focus on the temperature-induced body-centered-cubic phases all actinide elements adopt prior to melt.

Our results will predict and explain the occurrence of the body-centered-cubic crystal structure as the prevalent melting phase in many metallic systems, including the actinides. This will be of significance because, for the first time, the high-temperature portion of the phase diagrams for actinides (and other metals) can be computed from first principles with the reliability that already exists for the low-temperature region. 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 current theory or experiment. Hence, our capability to address an essential region of many metals phase diagrams can be improved dramatically with this research.

Mission Relevance

The properties of actinides and other metals must be well understood and predictable to ensure reliability of the nation's nuclear stockpile. Here we present a new idea that, together with our current expertise and capabilities, can substantially improve the reliability of our actinide phase models—particularly at high temperatures—relevant to LLNL's mission in stockpile stewardship science.

FY12 Accomplishments and Results

In FY12 we (1) computed stable phonon dispersion for body-centered-cubic uranium metal—a major milestone—finding the phase to be very unstable at lower temperatures and setting the stage to develop, with our self-consistent lattice dynamics scheme, an accurate, fully relativistic electronic structure to assist with stabilizing this phase at higher temperatures; (2) found that, for d-transition metals, alloying pure titanium with small amounts of vanadium lowered the temperature at which the alloy is mechanically stable; (3) studied two titanium–vanadium alloys, finding that the temperature dependence of stabilization as a function of alloying agree very well with the experimental phase diagram; and (4) studied vanadium metal and determined that it has a pressure-induced phase transition to a rhombohedral phase at around 70 GPa and demonstrated that this instability cannot be removed—that is, the body-centered-cubic phase cannot be stabilized—by increasing the temperature, resolving prior speculation about the material.

Proposed Work for FY13

We propose to apply the lattice dynamics scheme for technologically important alloys, first focusing on the titanium–vanadium system. Alloy phases stabilized in phonon–phonon interaction have not been computed—previous studies have focused on elemental metals. We will therefore (1) make our calculations and compare them with existing experimental phase diagrams for this system, (2) make modifications to our electronic-structure codes to properly describe the titanium–vanadium alloy system, and (3) apply the phonon approach to study the instability of vanadium metal under pressure, possibly determining that temperature effects eliminate the well-known instability of vanadium at about 70 GPa.

Publications

Söderlind, P., et al., 2012. "High-temperature phonon stabilization of γ-uranium from relativistic first-principles theory." Phys. Rev. B 85(6), 060301. LLNL-JRNL-522133.

Söderlind, P., et al., 2012. *Temperature-dependent lattice dynamics and stabilization of high-temperature phases from first-principles theory*. 2012 MRS Fall Mtg. and Exhibit (MRS 2012), Boston, MA, Nov. 25–30, 2012. LLNL-ABS-509674.

Söderlind, P., et al., 2012. *Temperature-dependent lattice dynamics for bcc uranium from first-principles theory*. 141st Ann. Mtg. and Exhibition of the Minerals, Metals, and Materials Society (TMS 2012), Orlando, FL, Mar. 11–15, 2012. LLNL-ABS-491592.

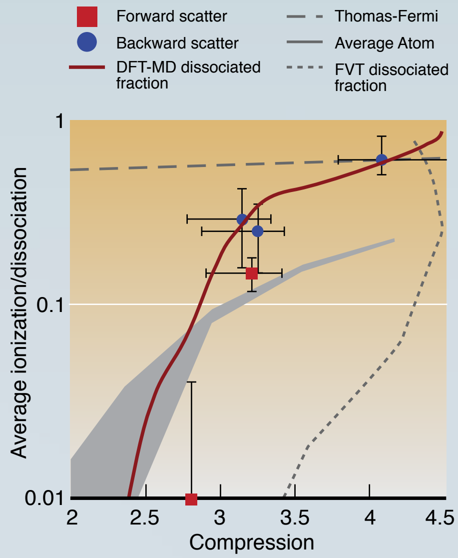
Dynamics of Ultrafast Heated Matter

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.



Experimental ionization data of deuterium and comparison with simulations as a function of compression along the Hugoniot (the relationship of pressure to density) for a single shocked state. The comparison with density functional theory (DFT) molecular dynamics (MD) simulations shown as a dark red line indicates that the conditions for metallization coincide with molecular dissociation without passing through an extended intermediate atomic state.

Mission Relevance

Ultrafast heating of matter is an important phenomenon for understanding physical processes relevant to laboratory astrophysics and high-energy-density science laboratory programs. This newly proposed application of x-ray scattering would provide definite data important to several Laboratory missions. For instance, equation of state and collision processes are important for successful modeling of experiments at NIF, in support of the Laboratory's missions in stockpile stewardship, climate and energy challenges, and laser ignition fusion energy.

FY12 Accomplishments and Results

In FY12 we (1) performed precision measurements of the ultrafast melting process involved in x-ray scattering on free-electron lasers, which showed melting on a 100-fs timescale; (2) assessed the material properties of shocked deuterium and beryllium, including determining their conductivity and equation of state—the molecular dynamics simulations indicate Drude-like behavior of mobile electrons; (3) incorporated the influence of detailed balance on proton-heated matter into a temperature diagnostic that is independent of the velocity distribution function; and (4) designed a new experiment on NIF to measure the properties of highly compressed matter. We have found that conditions for metallization in dynamically compressed deuterium coincide with molecular dissociation without passing through an intermediate atomic state.

Proposed Work for FY13

In FY13 we will conduct a series of x-ray Thomson scattering experiments to measure ultrafast phenomena. The experiments will use NIF fusion capsules, carbon phase transitions, and compressed deuterium and will be conducted on NIF, the Linac Coherent Light Source at the Stanford Linear Accelerator Center, the OMEGA laser at the University of Rochester, and Livermore's Titan laser. After experimentation, we will begin analyzing the scattering data.

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.

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Astrophysical Collisionless Shock Generation by Laser-Driven Laboratory Experiments

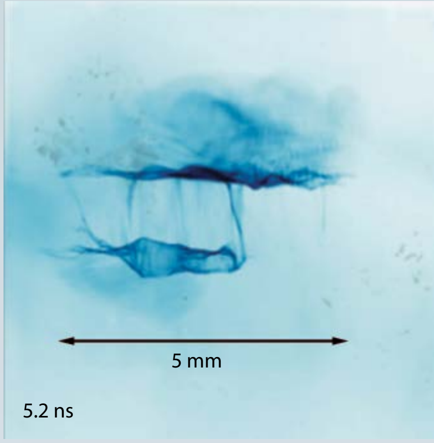
Hye-Sook Park (11-ERD-054)

Abstract

The exact mechanism of ultrahigh-energy cosmic-ray creation is a longstanding mystery. Collisionless shocks are a possible mechanism for creating, through Weibel instability, magnetic fields that may reveal shock acceleration dynamics that amplify cosmic-ray

particle energies beyond 1015 eV. We seek to observe and measure the creation and amplification of magnetic fields from collisionless shocks—specifically, high-Mach-number shocks created by the counter-streaming laser-produced plasmas associated with dynamic astrophysics processes. Using the OMEGA laser 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 collisionless shocks and generation of magnetic fields.

If successful, this project will generate a wealth of knowledge about the physics of collisionless shock dynamics. Our observations and experimental results will be important for understanding plasma processes occurring in astrophysical collisionless shocks, such as generation of magnetic fields and acceleration of cosmic-ray particles, which remain open questions of great interest in modern astronomy and astrophysics. The experiments we develop will also add new diagnostic capabilities for measuring magnetic fields, electron spectra, and the density and structure of plasmas capabilities that will be applicable to later NIF experiments. This project will also attract the world's foremost experts in this field to participate in those NIF experiments.



Self-organized electromagnetic fields are created from high-velocity counter-streaming plasmas as viewed by proton beams. Our experiments will study astrophysical shock-generation mechanisms using laser-created high-velocity plasma flows.

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.

FY12 Accomplishments and Results

In FY12 we performed experiments on the OMEGA laser and Livermore's Titan laser facilities to understand the interactions of high-velocity counter-streaming plasma flows that could produce collisionless shocks. Specifically, we (1) discovered, using a proton probe beam, unexpected stable field structures from small-scale plasma processes; (2) studied possible mechanisms of generation of these structures as well as plasma conditions when the two flows intersect; (3) successfully installed a gated optical imager on OMEGA and utilized it to image shock formation; (4) performed several collaborative experiments with the international Astrophysical Collisionless Shock Experiments with Lasers effort; and (5) initiated a NIF experiment design utilizing our OMEGA results.

Proposed Work for FY13

In FY13 we will (1) continue to work on understanding collisionless shock physics and related phenomenon, such as ion heating from intra-jet hydrodynamics; (2) refine and enhance our diagnostic probes to measure magnetic fields and distinguish their generation mechanisms; (3) utilize the OMEGA Extended Performance laser's schlieren imager for flow measurement and other 4-omega optical diagnostics to capture images of collisionless shock formation; (4) perform an experiment on Livermore's Jupiter or Titan laser to create collisionless shocks in expanding plasmas in a gas-filled chamber; (5) compare our results with radiation-hydrodynamics and particle-in-cell simulations; and (6) design further experiments to be conducted on NIF.

Publications

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Control of Impulsive Heat Loads in Tokamaks: Measurements and Modeling

Max Fenstermacher (11-ERD-058)

Abstract

Hot, magnetically confined fusion plasmas in tokamak nuclear reactors generally exhibit periodic impulsive edge instabilities called edge-localized modes (ELMs). The impulsive nature of the ELM plasma heat load on wall materials in such devices will lead to unacceptably high erosion and damage 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 will 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 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 southern France, in support of the energy security mission. The project also supports Livermore's science and technology foundations in fusion and high-energy-density science, as well as high-performance computing and simulation.

FY12 Accomplishments and Results

In FY12 we (1) employed new motional Stark effect edge data in the ELM model; (2) compared BOUT++ ELM simulations to low-collision experimental data; (3) implemented synthetic diagnostics for comparison to fast-camera images; (4) implemented four- and five-field models and flux-limited parallel conduction to predict particle and energy transport; (5) upgraded the resonant magnetic perturbation penetration models to multi-machine coil geometries, including a more accurate calculation of external magnetic coil fields; (6) verified linear ideal and non-linear island solutions, including a study of both resistive and hyper-resistive reconnection in slab geometry; (7) analyzed fluid transport generated by reconnection in the three-field drift-magento-fluid dynamics ELM model and its impact on turbulence; and (8) compared interpretive and predictive UEDGE fluid transport code simulations versus data for transport by resonant magnetic perturbations at high collision rates.

Proposed Work for FY13

We will (1) calculate penetration of resonant magnetic perturbations in a rotating plasma in realistic x-point and divertor geometry; (2) calculate magnetic flutter transport induced by resonant magnetic perturbation using BOUT++; (3) determine, at the DIII-D reactor, the self-consistent perturbed magnetic field that allows plasma shielding; (4) continue comparing DIII-D data and simulations with regard to ELM dynamics, size, and heat-flux distribution for low-collision regimes; (5) measure simultaneous changes in heat and particle fluxes induced by resonant magnetic perturbations using a new periscope diagnostic and injected impurities, if necessary, to enhance imaging resolution; and (6) compare our best simulations of ELM suppression by resonant magnetic perturbations in low-collision plasmas with complete DIII-D diagnostic measurements.

Publications

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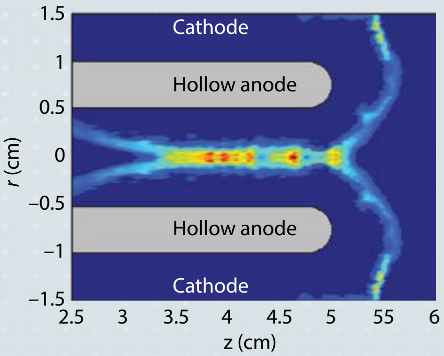
Investigation of Fast Z-Pinches for Scalable, Large-Current, High-Gradient Particle Accelerators

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, in which an electrical current generates a magnetic field that compresses, or pinches, the plasma. We will perform unique, first-ever probe-beam experiments that will measure the gradients



Deuterium ion density is indicated in the first fully kinetic simulations of a dense plasma focus Z-pinch, in which an electrical current generates a magnetic field that compresses, or pinches, the plasma. These simulations are being used to unravel the mechanisms behind instability-driven beam acceleration in this classic plasma configuration.

directly and develop state-of-the-art, fully kinetic plasma simulations. Our motivation is twofold. First, at the mega-volt level, dense plasma focus (DPF) devices optimized for beam production and acceleration could serve as the basis for compact, intense radiological sources such as directional neutron sources, and also as unique high-current ion injectors. Second, the DPF device could, if scaled to higher levels, lead to multistage, very-high-gradient plasma-based accelerators notably simpler than current laser or electron-beam systems, thus revolutionizing accelerator technology and applications.

We expect to produce a fundamental understanding of the acceleration gradients in hot, dense Z-pinch plasmas, and use that knowledge to examine how these plasmas can be systematically exploited for accelerator applications such as a new class of high-current injectors and a fundamentally new kind of high-gradient plasma accelerator using multiple staged Z-pinch devices. We will also experimentally demonstrate, for the first time, use of a DPF device as a very-large-current injector source for an induction linear accelerator. Success in this project could potentially be a revolutionary breakthrough in accelerator technology that would lay the groundwork for enabling mission-relevant applications such as portable devices for the very-large-standoff detection of special nuclear material using giga-electronvolt protons.

Mission Relevance

The work supports LLNL's national security missions—specifically homeland security, nonproliferation, and accelerator science—by providing the technological and scientific basis for compact alternative radiological sources and next-generation accelerator technology for active interrogation and radiography. Successful resolution of the physics behind the Z-pinch plasmas formed in a DPF device would resolve longstanding questions in plasma physics, a core Livermore competency.

FY12 Accomplishments and Results

In FY12 we (1) produced and characterized record sub-kilojoule and greater than 1-kilojoule DPF plasmas with ion-beam energies greater than 400 keV and gradients of approximately 50 MV/m; (2) assembled the 4-MeV radio-frequency quadrupole ion probe beam and the beam transport required for it, and conducted the first-ever experiments of a beam into a DPF plasma; (3) completed the first-ever simulations of DPF using both fully kinetic particle-in-cell and hybrid models; (4) began comparing these leading models' results with experimental data in aspects such as neutron yield, with good initial agreement; and (5) hired an additional postdoctoral researcher in this cutting-edge area of high-energy-density physics and published our results in *Physical Review Letters*.

Proposed Work for FY13

In FY13 we will (1) continue and complete the first-ever probe-beam experiments and compare results to predictions from our fully kinetic models to produce a new understanding of acceleration mechanisms in DPF devices; (2) determine the viability, through experiments and simulations, of using our DPF device as an injector for an induction linear accelerator; and (3) examine the feasibility of staging DPF devices for a next-generation high-gradient accelerator.

Publications

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Nuclear Plasma Physics

Dennis McNabb (11-ERD-069)

Abstract

To understand the evolution of the universe and fundamental underpinnings of high-energy-density plasmas, as well as fully 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 will help improve understanding of neutrino production in the Sun as well as enable a new diagnostic of plasma temperatures in National Ignition Facility

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 interaction 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 National Ignition Facility 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.

Mission Relevance

By improving our understanding of nuclear reactions in burning plasmas, we will enable new science capabilities important to stockpile stewardship, a key mission area at Lawrence Livermore, as well as further research in inertial fusion energy, in support of the Laboratory mission to enhance the nation's energy security. In addition, our research supports and enhances Livermore's core competency in high-energy-density science.

FY12 Accomplishments and Results

We (1) used an improved neutron time-of-flight detector to make the most detailed measurements to date of the neutron spectra emanating from the tritium–helium reaction $T(t,2n)^4He$—measurements were made at both the National Ignition Facility and the OMEGA laser at the University of Rochester at three different ion temperatures and data analysis from those measurements has begun; (2) made new alpha particle measurements with an improved spectrometer, although preliminary results indicated that backgrounds are still too high, and began using R-matrix methods to develop a set of spectral components to fit the data; (3) performed theoretical calculations of nuclear–plasma interactions, including nonlocal thermodynamic equilibrium effects; (4) used the calculations to design and carry out a thulium hohlraum experiment to search for plasma–nuclear coupling to the first excited state of thorium-169, with preliminary analysis indicating that metastable atomic states might have a similar signature to nuclear excitation; (5) began planning future experiments with osmium, which would allow us to disentangle these effects; and (6) made initial measurements of capture gamma-ray spectra on gold in collaboration with the University of Oslo.

Proposed Work for FY13

In FY13 we will (1) perform new, more-precise measurements and calculations of neutron- and alpha-particle spectra to better understand continuum states in $^4He,2n$ formed in the tritium–tritium reaction; (2) continue to develop an experimental approach to validate the theory of plasma–nuclear coupling; and (3) perform calculations of the evolving distribution of nuclear excited states expected to be present in an igniting capsule, and estimate the effect of these excited states on subsequent nuclear reactions.

Publications

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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 superconductors. These iron-based superconductors are mostly comprised of pnictides—materials that contain a pnictide element 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 infrastructure. We propose to couple high-pressure capabilities unique to LLNL with the highest-quality samples grown with state-of-the-art methods to experimentally determine structural properties conducive to superconductivity.

We expect to grow the highest-quality single crystals of ferropnictide compounds to date and map out the pressure conditions that maximize the onset of superconductivity in

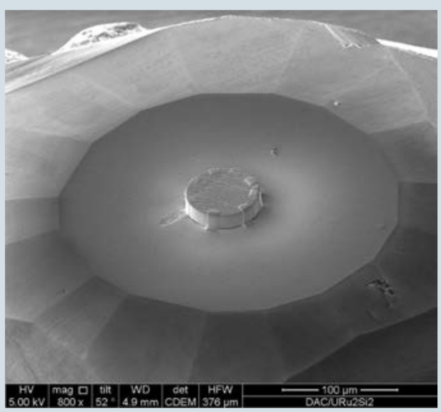
these materials. Using x-ray diffraction, we will reveal a never-before-seen correlation between structural properties and superconductivity. Furthermore, we will exploit the newly discovered structure–property relationships as feedback for maximizing superconductivity in ferropnictides at ambient pressure. 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.

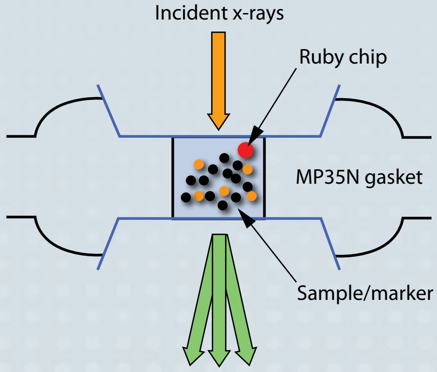
Mission Relevance

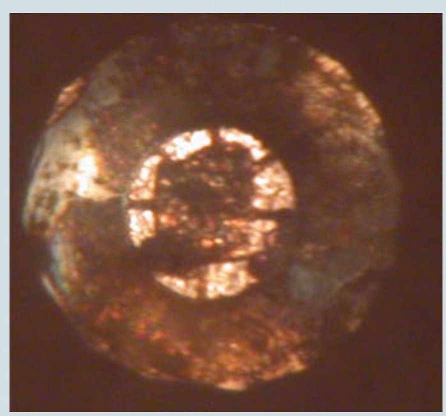
With this research, we aim to determine structure–property relationships of superconductors with the future goal of engineering materials suitable for use in the electrical power grid, in support of the Laboratory's core mission to enhance national energy security. In accomplishing this proposed work, we plan to support a graduate student, thus training a future scientist in mission-critical techniques that encompass many Laboratory thrust areas.

FY12 Accomplishments and Results

We completed and published our research on the electrical transport and structural information on (Ca,Sr)Fe_2As_2 under pressure. Specifically, we completed a series of studies, using x-ray emission spectroscopy, to examine the iron moment in (Ca,Sr)Fe_2As_2 through







We are using designer diamonds and synchrotron x-ray techniques to gain unprecedented insight into structure–property relationships over a wide range of phases. A scanning electron microscopy image of a sample attached to a designer diamond anvil using a focused ion beam is shown at 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.

the volume collapse transition, finding that the iron moment persists through the volume collapse, which suggests that observed superconductivity in the collapsed tetragonal phase may be mediated by spin fluctuations. We also completed electrical resistivity measurements of $PtAs_2$ under pressure, which appears to exhibit Mott insulator characteristics in its transport properties. In summary, this project has illuminated the relationships between the structural, magnetic, and electronic degrees of freedom that engender superconductivity in one class of ferropnictide superconductors. The knowledge gained from x-ray emission spectroscopy experiments on ferropnictide superconductors is now being applied to work at LLNL on permanent magnets based on critical materials, an emerging mission area for the Laboratory.

Publications

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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.

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Jeffries, J. R., et al., 2012. "The suppression of magnetism and the development of superconductivity within the collapsed tetragonal phase of $Ca_{0.67}Sr_{0.33}Fe_2As_2$ at high pressure." *Phys. Rev. B* **85**(18), 184501. LLNL-JRNL-536331.

Structure and Bonding in High-Atomic-Number Metals Under Extreme Conditions

Donald Correll (11-FS-003)

Abstract

We seek to develop a concept for National Ignition Facility experiments to study the structure and bonding of high-atomic-number metals at extreme pressures and temperatures using time-resolved x-ray diffraction methods. We expect to achieve

comprehensive insight into the pressure-induced electronic transitions in high-strength and high-melting-temperature transition metals in phases not previously considered. Areas we will explore include (1) the melting anomaly that explains an existing controversy, (2) the systematic phase transitions—from body-centered cubic to hexagonal closed packed and to face-centered cubic—that are predicted to arise from the pressure-induced valence-shell electron transitions, and (3) the low-symmetry crystal structures that may arise from core–shell electron hybridization.

In short, we will develop a concept for a National Ignition Facility experiment to probe the phase diagram of high-atomic-number metals under extreme conditions will have significant scientific and technological impacts on condensed matter theory.

Mission Relevance

This effort to develop a National Ignition Facility experiment to probe the phase diagram of high-atomic-number metals under extreme conditions will have significant scientific and technological impacts on condensed matter theory, and applications of these results are highly relevant to Laboratory efforts in stockpile stewardship science through exploring materials and chemistry at the extremes.

FY12 Accomplishments and Results

In collaboration with Choong-Shik Yoo and Minseob Kim of Washington State University and researchers from the University of California, San Diego, we carried out experiments to study the plastic response of shocked high-atomic-number materials using the Janus laser at LLNL. The laser was used to drive planar shocks into specially prepared samples of nanometer-scale indented high-atomic-number metals and the samples recovered for post-shock analysis. Our goal was to study how dislocations play a role in plastic deformation at the microscopic level. We also carried out experiments in tension by allowing thin samples to spall. Our results in FY12 have established the feasibility for further laser experiments to apply state-of-the-art optical spectroscopy to such shock-compressed material experiments.

Publications

Kim, M., S. Wall, and C. Yoo, 2012. *Structure and bonding in high-Z metals under extreme conditions*. LLNL-TR-608913.

Core-Collapse Supernova Explosions Mechanism Studies

Donald Correll (11-FS-004)

Abstract

We propose to study the evolution of a spherical, nearly stationary shock in a high-energy-density environment. The system parameters will scale to the conditions existing

shortly after the so-called "core bounce" stage (ultra-dense nuclear material rebounds) in a collapsing supernova progenitor and through the neutrino-driven supernova shock-revival phase, which lasts about one second. We will probe one of the crucially important elements of core-collapse supernova physics—namely, the standing accretion shock instability that is thought to trigger the actual explosion of a massive star. This proposal is unique in that it combines astronomical observations, fundamental theory, two-dimensional radiation-hydrodynamics simulations, and design of scaled National Ignition Facility validation experiments in a new paradigm of astrophysics research and methodology.

We will design a computational model of an experimental target system to examine photo-evaporation-front dynamics in radiatively driven molecular clouds that occur during the high-mass star formation process, and perform simulations and data analysis of this model's hydrodynamics. We intend to investigate shock stability, in particular with respect to bulk properties of the incoming flow, upstream flow perturbations, and design of the central outflow moderator (i.e., the size, degree of asphericity, and displacement of the dense, central target hemisphere). This effort will result in a new database of several dozen two-dimensional hydrodynamic models.

Mission Relevance

This research will help to resolve controversial issues in the theory of shock instabilities and develop the concepts for a new experimental platform in support of the Laboratory's commitment to pursuing high-energy-density science relevant to the Stockpile Stewardship Program.

FY12 Accomplishments and Results

Colleagues from Florida State University, under the direction of Tomasz Plewa, performed computational modeling and determined that spherical standing accretion shocks created in the high-energy-density laser experiments at the National Ignition Facility do not match characteristics of the exploding core-collapse supernova systems. In particular, relatively simple laboratory configurations of the type considered do not produce the standing accretion shock instability. This situation is different in the supernova setting because of the presence of greater gravitational forces. However, we generated plasma configurations that were stable to perturbations and remained stationary for tens of advective times (hundreds of nanoseconds). Such configurations may have applications in studies of basic plasma properties, plasma hydrodynamics, and nuclear plasma physics effects, including situations relevant to inertial-confinement fusion.

Publications

Plewa, T., and T. Handy, 2012. *Core-collapse supernova explosion mechanism studies on NIF*. LLNL-TR-608855.

Next-Generation Tunable Targets for Laser-Compression Experiments

Donald Correll (11-FS-005)

Abstract

We propose to develop new fusion-class laser targets that will allow tuning of sample properties over a wide range of initial densities and temperatures, prior to being dynamically compressed using laser-driven ramp, shock, and multishock waves. In combination with the unprecedented capabilities of the National Ignition Facility, such targets will provide access to unique states of matter at high densities and low temperatures, allowing high-quality measurements of broad interest in physics, chemistry, and planetary sciences. We will perform experiments at Livermore's Janus laser facility using cryogenically cooled and pre-compressed samples, along with a combination of ramp and multishock loading, to document the degree to which sample temperatures can be minimized—and hence density maximized—in laser-driven compression experiments. The experiments will be performed on methane, nitrogen, water, or hydrogen and helium mixtures—compositions of interest for planetary physics and astronomy. Such mixtures cannot, in general, be prepared as uniform samples through cryogenic cooling alone.

These experiments are intended as proof of principle in preparation for future applications at the National Ignition Facility. Each material to be investigated is of considerable scientific interest. However, some are easier to load than others—success in target preparation will be a key criterion for determining which samples are used for attempting maximum-density compressions. Nevertheless, we expect that each experiment will provide important data for planetary applications, documenting equations of state, molecular dissociation, and changes in chemical bonding over a broad range of pressures and temperatures.

Mission Relevance

Our efforts to generate accurate, ultrahigh-density states supports Laboratory core missions in both stockpile stewardship science and energy solutions by helping to develop the next generation of targets for laser compression experiments. Results from this study will also advance our fundamental understanding of the equations of state, over a wide range of initial densities and temperatures, for materials important to planetary physics and astronomy.

FY12 Accomplishments and Results

Colleagues from the University of California, Berkeley, under the direction of Raymond Jeanloz, made a significant discovery—that shock-compressed hydrogen–helium mixtures do not behave as would be expected by simply combining results for hydrogen and helium alone. Our result is the first experimental evidence for a longstanding theoretical prediction that hydrogen–helium fluid mixtures can undergo un-mixing at high pressures, with the implication that helium can separate from hydrogen inside giant planets. The resulting "differentiation"—compositional un-mixing—can release significant gravitational energy and is thought to have greatly influenced the evolution of Saturn relative to Jupiter. The next step would be OMEGA experiments on warm dense hydrogen isotopes as proof of principle in preparation for future applications at the National Ignition Facility.

Publications

Jeanloz, R., and M. Millot, 2012. *Next-generation tunable targets for laser-compression experiments*. LLNL-TR-608814.

Development of a New Materials Platform to Study Diamond and Iron at Ultrahigh Pressures

Donald Correll (11-FS-006)

Abstract

Diamond and iron are fundamental materials of profound interest in condensed matter physics, materials science, and planetary science. The behavior of these solids under ultrahigh compression is essential to understanding planetary interiors and for probing the limits to our understanding of solid-state behavior at extreme conditions. The National Ignition Facility (NIF) is now poised to provide an exciting new opportunity to uniquely extend the range at which solid materials can be compressed by more than an order of magnitude compared with existing static technology. We propose to develop the capability to subject solids to ultrahigh compression using ramp-loading techniques at NIF. We will build on our successful experiments at other laser facilities to develop new capabilities for the study of material properties including equation of state, strength, and crystal structure to unprecedented conditions that approach or exceed those found in the cores of giant or super-Earth planets.

With this project, we expect to achieve peak pressures of 30 Mbar in diamond and iron. Diamond anvil cell experiments—the technique of choice today for exploring the solid state at high pressures—are limited to approximately 3 Mbar, a capability that will not likely increase by more than twofold in coming years. Thus, our project has the potential to expand the pressure range by an order of magnitude and open up a wide new range of pressures and temperatures for materials study. We will exploit the newly demonstrated capability on NIF to quasi-isentropically compress samples, measure the resulting loading profile, and determine the equation of state of the sample. This will also enable us to identify phase transformations and examine their rate dependence.

Mission Relevance

Of particular importance to the Stockpile Stewardship Program is the development of advanced capabilities to predict physical properties of matter under an extremely broad range of dynamic conditions. Our project will develop new experimental capabilities as well as provide important data on materials in support of the Laboratory's mission in national security. This research will build on our past success at the Laboratory for Laser Energetics at the University of Rochester, where we have demonstrated the capability to obtain material stress-density compression paths with accuracy comparable to or better than pulse power or gas gun systems. Our work will also lead to fundamental scientific insight into the properties of materials under conditions never before experimentally investigated.

FY12 Accomplishments and Results

Colleagues from Princeton University, under the direction of Thomas Duffy, demonstrated a new experimental technique to create material conditions consistent with those found in the centers of giant planets. The achieved stress of 5 TPa lies between the expected pressure at the center of Saturn (4 TPa) and Jupiter (7 TPa), where a core of heavy elements is expected to reside. At this pressure, diamond is compressed 3.7 times to a density of about 12 g/cm^3, greater than that of lead at ambient conditions. In addition, we accumulated valuable information and experience related to the intersection of the laser pulse-shaping and target physics that will be highly useful in developing further high-pressure measurements on NIF. Future work will enable higher pressures to be reached with the use of higher laser power and provide more detailed measurements of material properties, as well as focus on understanding how sample microstructure affects wave propagation response.

Publications

Duffy, T., 2012. *Development of a new materials platform to study iron and diamond at ultrahigh pressures using NIF*. LLNL-TR-608856.

Dynamics of the Eagle Nebula

Donald Correll (11-FS-007)

Abstract

We propose to develop the first detailed design of a scaled dynamics laboratory experiment to study the Eagle Nebula interstellar cloud. The Eagle Nebula is an example of the common phenomenon of photo-evaporation front dynamics in radiatively driven molecular clouds that occur during the formation of high-mass stars. The directional radiation flux that drives the hydrodynamic flow also leads to new radiative hydrodynamic instabilities. We will design an experiment on the National Ignition Facility to quantitatively test models and two-dimensional simulations of the Eagle Nebula dynamics deep into the nonlinear regime. Photo-evaporation front dynamics and instabilities in astrophysics, such as those that radiatively drive the Eagle Nebula molecular cloud, are similar in many respects to radiatively driven (indirect-drive) ablation-front dynamics and instabilities in inertial-confinement fusion.

We expect to develop a detailed design and plan for a scaled Eagle Nebula dynamics experiment, including recommendations for specific high-energy-density facilities that

will be used to perform the work. We will determine the specific scale-transformation requirements for a laboratory experiment to accurately reproduce the astrophysical dynamics, including (1) detailed simulations and analysis of the conditions expected to be achieved to meet the physics goals, (2) the desired radiative dynamics to emulate proposed models of the Eagle Nebula, and (3) rigorous scaling criteria.

Mission Relevance

This research will benefit efforts in inertial-confinement fusion, in support of the Laboratory's mission in energy security. In addition, developing high-energy-density experiments and radiation-hydrodynamics simulations supports LLNL's science, technology, and engineering foundational work in materials and chemistry at the extremes. Finally, science related to spectacular astronomical objects such as the Eagle Nebula is expected to be an effective way to enhance the Laboratory's recruitment of new scientific and technological talent.

FY12 Accomplishments and Results

Colleagues from the University of Maryland, under the direction of Marc Pound, have used astronomical observations to investigate dense gas distributions in the Eagle and Pelican nebulae pillars. Observations were made with the CARMA (Combined Array for Research in Millimeter-Wave Astronomy) telescope at a high-altitude site in eastern California. We studied, in detail, the spatial and velocity morphology of the dense molecular material in the pillars to understand their dynamical evolution and to guide the design of astrophysics-relevant National Ignition Facility targets. We also explored the possibility of using 3-mm carbon–nitrogen transitions as a Zeeman probe, where a spectral line is split into several components in the presence of a static magnetic field, for future observations. In summary, this project helped define a design of credible, well-scaled, high-energy-density Eagle Nebula laboratory plasma experiments that can be fielded at the National Ignition Facility.

Publications

Pound, M., 2012. *Dynamics of the Eagle Nebula*. LLNL-TR-608916.

Relativistic Plasma Physics at the National Ignition Facility

Donald Correll (11-FS-008)

Abstract

There are several shortcomings of classical laser–plasma modeling in the regime under which the National Ignition Facility will operate that could affect accurate analysis of the experimental data that will be obtained, including reflectivity measurements. We propose a theoretical and computational investigation of plasma physics spanning the threshold between classical Newtonian physics and the relativistic concepts of Einstein in a controlled laboratory setting at energy densities never before realized. In particular, we will investigate relativistic modifications to plasma physics including laser–plasma interactions

and hot electron creation using theoretical and computational techniques. Practical assessment will be emphasized in the context of how these effects can be measured using traditional four-omega Thomson scattering and novel Thomson scattering techniques.

We expect to enable improved laser–plasma interaction tools for use at the National Ignition Facility and other large laser facilities by providing the relativistic growth rate and spectrum for forward- and backward-stimulated Raman scattering, as well as the growth rate for two-plasmon decay. We also intend to determine relativistic optical Thompson scattering. In addition, we will provide experimental requirements to explore relativistic plasma physics, as well as analysis of ongoing experiments.

Mission Relevance

By furthering the development of advanced experimental measurements and laser–plasma simulations, this project supports the Laboratory's core missions in national and energy security through stockpile stewardship science and fusion energy research.

FY12 Accomplishments and Results

Colleagues from the University of Maryland, under the direction of Thomas Antonsen and John Palastro, have derived a fully relativistic dispersion relation for nonlinear parametric processes in laser-driven plasmas. This dispersion relation can provide the relativistic growth rate for any manner of parametric instability and can be considered a generalization of the well-known Drake dispersion relation. Our newly derived relation differs in that no assumption about the relative timescales was made by performing a ponderomotive average, which is a nonlinear force that a charged particle experiences in an inhomogeneous oscillating electromagnetic field. Additionally, we included electrostatic parametric instabilities in our dispersion relation. Our results can be used to improve current laser–plasma interaction models and gain predictions for high-temperature experiments at the National Ignition Facility. Our research has therefore produced a fully relativistic dispersion relation for nonlinear parametric processes in laser-driven plasmas that does not rely on assumptions about relative timescales.

Publications

Palastro, J. P., and T. M. Antonsen, Jr., 2012. *Relativistic plasma physics at the National Ignition Facility*. LLNL-TR-608815.

Collisionless Shock Experiments at the National Ignition Facility

Donald Correll (11-FS-009)

Abstract

Collisions of supersonic flows are ubiquitous in astrophysics, and the resulting shock waves are responsible for high-energy radiation of many sources such as supernova remnants

and gamma-ray bursts. Because of the low density of astrophysical plasmas, the mean free path of Coulomb collisions is typically very large, so most shock waves in astrophysics are collisionless, in which the dissipation is mediated by plasma instabilities and other anomalous processes. In addition to applications in astrophysics, collisionless shock physics is important to many concepts of inertial fusion energy, especially fast ignition. Because of the complex plasma processes involved in shock formation and dissipation, the physics of collisionless shocks and mechanisms for efficient shock acceleration of particles remains unsolved. We propose to address several outstanding shock physics questions by documenting a series of proposed collisionless shock experiments at the National Ignition Facility and identifying future actions and resources required to carry out these experiments.

The results of our proposed collisionless shock project will have major impacts on both inertial fusion and astrophysics. We intend to provide a detailed report of laboratory experiments at the National Ignition Facility and computer simulations relevant to collisionless shock physics, including the structure of collisionless shocks as a function of upstream magnetization, composition, flow speed, and field geometry. We will also examine magnetic field generation and survival in shocks, mechanisms and efficiencies of non-thermal electron and ion acceleration processes, differences between relativistic and nonrelativistic shocks, and radiative processes.

Mission Relevance

This project supports LLNL's energy security mission by furthering knowledge of collisionless shock physics relevant to inertial fusion energy, and it 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.

FY12 Accomplishments and Results

Colleagues from Rice University, under the direction of Edison Liang, identified a new experimental platform for laboratory astrophysics involving multiple-beam, high-energy lasers focused in a circular, hollow ring pattern rather than a single spot. Our preliminary results on the FLASH free-electron laser at the DESY accelerator center in Germany show that because of the rocket nozzle effect, the hollow ring configuration produces a much more highly collimated supersonic outflow, with much higher axial density, temperature, and flow velocity than is achievable with single-spot focusing. We also found that by varying the radius of the hollow ring, a much larger dynamic range of jet parameters—including density, temperature, velocity, Mach number, opening angle, and collisional capability—can be achieved. In summary, this project demonstrated a new platform for laboratory astrophysics experiments utilizing supersonic outflows—a new hollow-ring platform with specific and important applications to the study of collisional and collisionless shocks, as well as many other laboratory astrophysics topics such as shear flows, jet propagation and stability, and magnetic field generation, to name a few.

Publications

Liang, E., 2012. *Collisionless shocks experiments using NIF*. LLNL-TR-608874.

High-Resolution K-Shell X-Ray Spectroscopy

Donald Correll (11-FS-012)

Abstract

We intend to investigate the type of x-ray spectrometer necessary to determine details of laser energy coupling to targets and the conversion ratio of laser energy to hard x-ray emission. Conceptual designs of experiments for recording high-resolution x-ray spectra will be evaluated by applying K-shell innermost orbital, spectroscopic diagnostic techniques for two types of laser targets. The first laser target considered, capsules with germanium in the fuel, will be used for observing implosion dynamics and possible mixing of germanium in the dense, hot core plasma. The second target capsule will incorporate silver back-lighters irradiated by Advanced Radiographic Capability beams on the National Ignition Facility that will produce a bright x-ray flux to be used for point-projection radiography of the dense core plasma.

We expect to determine the feasibility of using a three-crystal spectrometer with time-integrated K-shell spectra recorded by image plates positioned on one side of the spectrometer axis and covering the 6- to 511-keV x-ray range. We will accomplish this through conceptual experiment designs that will extend detector energy coverage down to 6 keV, which involves thinning the spectrometer crystal from the nominal 200 to 250 µm down to 100 µm and bending the thin crystal, and by extending the detector coverage up to 511 keV through use of a thick crystal or combination of crystals at least 1 mm in thickness. This work will contribute to measurement capabilities that may shed light onto how laser energy is coupled to the target through x-ray conversion in a hohlraum target-capsule by measuring the density, temperature, opacity, brightness, and time evolution of laser hohlraum plasmas.

Mission Relevance

This work contributes directly to a basic scientific understanding of how laser energy on fusion-class lasers is coupled to the target through x-ray conversion in a hohlraum. In doing so, the work supports NNSA missions in inertial-confinement fusion and in experiments that contribute to the Stockpile Stewardship Program.

FY12 Accomplishments and Results

In FY12 colleagues from Artep Inc., under the direction of John Seely, (1) provided a computer-aided design of an x-ray spectrometer comprised of three cylindrically bent transmission crystals that cover 3 channel ranges of 6 to 20 keV, 10 to 150 keV, and 50 to 511 keV; (2) measured the transmittance of quartz at 3 thicknesses, including 75 µm as tested for lower-energy coverage, 250 µm as used in the standard Cauchois spectrometers developed by Artep and fielded at major laser facilities, and 1 mm as considered for the high-energy channel; and (3) determined the design is fully consistent with diagnostic instrument manipulators on the National Ignition Facility target chamber. Design research took full advantage of the Naval Research Laboratory's dual-crystal spectrometer that has already been fielded at the LLNL Jupiter Laser Facility and the OMEGA facility at the Laboratory for Laser Energetics in Rochester, New York. Our success in providing a design fully consistent with the diagnostic manipulators on the National Ignition Facility target chamber has led to follow-up discussions with the diagnostic team.

Publications

Seely, J., U. Feldman, and L. Marlin, 201. *High-resolution K shell x-ray spectroscopy experiments*. LLNL-TR-608813.

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.

FY12 Accomplishments and Results

In FY12 we performed, for the first time, high-precision beta-delayed neutron spectroscopy using trapped ions. We used a californium-252 source to produce the fission products, which were then extracted as an ion beam suitable for capture in an ion trap. The trap was instrumented with plastic scintillator detectors for beta measurements and with position-sensitive micro-channel plate detectors for recoil-ion measurements. This detector array increased the beta-recoil ion coincidence efficiency by a factor of 12 over the initial array and allowed a measurement of the neutron energy spectrum with an energy resolution of about 5% (full-width, half-maximum). Our research enabled a new method for improving the quality and reliability of beta-delayed neutron measurements using trapped radioactive ions. This technique will now be used address topics of interest to the applied (nuclear energy and stockpile stewardship) and fundamental (nuclear astrophysics and structure) nuclear science communities.

Publications

Li, G., et al., 2012. *Tensor interaction limit derived from the alpha–beta–neutrino correlation in trapped ^{8}Li ions*. LLNL-JRNL-603533.

Scielzo, N. D., et al., 2012. "The Paul trap: A radiofrequency-quadrupole ion trap for precision studies." *Nucl. Instrum. Meth. Phys. Res. A* **681**, 94. LLNL-JRNL-555815.

Yee, R. M., et al., in press. "Beta-delayed neutron spectroscopy using trapped radioactive ions." *Phys. Rev. Lett*. LLNL-JRNL-511460-DRAFT.

Transport Properties of Dense Plasmas and a New Hybrid Simulation Technique for Matter at Extreme Conditions

Frank Graziani (12-SI-005)

Abstract

We propose to apply a recently developed world-class, massively parallel molecular dynamics code for hot dense matter—the ddcMD (domain decomposition molecular dynamics) code—to plasma physics model uncertainties about thermal conductivity and stopping power from ion–electron collisions. These issues are critically important to stockpile stewardship and laser fusion energy. In addition, the project will significantly extend the ddcMD code's current capability to the warm dense-matter regime where strong coupling exists and the electrons have significant degeneracy. The code will simulate

strongly coupled, nonideal, and degenerate plasmas known as warm dense matter, and will address species diffusivity and equation-of-state issues. It will provide insight into existing theories of complex plasmas, including mixtures, and motivate developments of new theories and experiments.

The proposed project will enable us to determine regime diffusivity for warm dense matter and the equation of state for low- and high-atomic-number plasma mixtures. We will study the behavior of these quantities as a function of atomic number and high-atomic-number impurity concentration. These results will be compared to current model implementations in the burn codes. In the regime of hot dense matter, the proposed project will produce stopping power and thermal conductivities for low- and high-atomic-number mixtures. We will also implement a new treatment of dynamic electrons in the regime of hot dense matter and study the implicit limit of electron kinetic equations and the utility of this approach for thermonuclear burn.

Mission Relevance

Strategically, this project will improve predictive capability for Livermore's Advanced Simulation and Computing codes and will facilitate the design of ignition experiments at the National Ignition Facility in support of stockpile stewardship science and energy security for the nation. These goals will be accomplished by enhancing our fundamental understanding of complex, nonideal plasmas and by validating models used in stockpile stewardship and National Ignition Facility codes.

FY12 Accomplishments and Results

In FY12 we (1) conducted an exhaustive study of models of stopping power and compared the results to those obtained with molecular dynamics, (2) completed our work on the theory of electron–ion coupling, including comparing the results to those obtained with molecular dynamics; (3) used a new Yukawa capability for the ion-only simulations of diffusivity in plasma mixtures, showing that the molecular dynamics behavior of plasma diffusivity does obey the standard laws of diffusion, known as Fick's laws; (4) began work on thermal conductivity; and (5) published a long review paper on our molecular dynamics capability in *High Energy Density Physics*.

Proposed Work for FY13

In FY13 we will (1) complete the thermal conductivity calculations, (2) extend the generalized Lennard–Balescu equation to thermal conductivity and perform comparisons with molecular dynamics, (3) perform thermonuclear burn simulations with the advanced integrators, (4) compare the various stopping-power theories and molecular dynamics simulations, and (5) perform comparisons between quantum molecular dynamics, molecular dynamics with statistical potentials, and average atom models of hydrogen properties at 1 g/cm^3 and 10 eV.

Publications

Benedict, L. X., et al., 2012. "Molecular dynamics simulations and generalized Lenard–Balescu calculations of electron–ion temperature equilibration in plasmas." *Phys. Rev. E* **86**(4), 046406. LLNL-JRNL-573852.

Ellis, I. N., et al., 2012. "Convective Raman amplification of light pulses causing kinetic inflation in inertial fusion plasmas." *Phys. Plasmas* **19**, 112704. LLNL-JRNL-517591.

Ellis, I. N., et al., 2012. *Relativistic particle wakes and their impact on electron stopping*. 42nd Ann. Anomalous Absorption Conf., Key West, FL, June 25–29, 2012. LLNL-ABS-552658.

Graziani, F. R., 2012. "Large-scale molecular dynamics simulations of dense plasmas: The Cimarron Project." *High Energ. Density Phys.* **8**(1), 105. LLNL-JRNL-516395.

Extreme Compression Science

Jon Eggert (12-SI-007)

Abstract

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

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

extreme pressures, enable a laboratory test bed for the wealth of new astronomical observations of planets outside the Solar System, and provide enhanced routes to inertial fusion energy.

Mission Relevance

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

FY12 Accomplishments and Results

In FY12 we (1) performed two pulse-shaping laser shots on NIF for diamond at 100 Mbar and iron at 25 Mbar; (2) began designing an x-ray diagnostic for use at NIF; (3) performed several gigabar-scale experiments at the OMEGA laser, obtaining Hugoniot data on plastic (carbon–hydrogen) at 60 Mbar; (4) measured temperatures for ramped iron using extended x-ray absorption fine structure spectroscopy and observed tantalum's L-shell structure; (5) identified phase transitions for tantalum at about 3 Mbar, tin at about 2 Mbar, and magnesium oxide at about 5 Mbar using x-ray diffraction; (6) studied

This project is building a large, interdisciplinary community to explore matter at near-atomic pressures.

pre-compressed hydrogen–helium gas and pre-compressed nitrogen gas and methane; and (7) conducted diagnostic development experiments on LLNL's Janus laser using a number of techniques, including transverse displacement interferometry, coherent anti-Stokes Raman spectroscopy, and two-dimensional velocity interferometer for any reflector.

Proposed Work for FY13

In FY13 we will (1) perform several equation-of-state shots for iron, for which we are well prepared because of our pulse-shaping shots in FY12; (2) conduct x-ray-diffraction shots on diamond and iron at 30 Mbar, as well as several gigabar-scale and cryogenic deuterium equation-of-state shots at NIF; (3) continue our pre-compressed hydrogen–helium experiments at OMEGA; (4) improve our calibrations of quartz, extending them to other silica polymorphs—specifically, coesite and stishovite; (5) continue to develop our gigabar equation-of-state experiments; (6) field both diffraction and extended x-ray absorption fine structure diagnostics on OMEGA and continue our work on advanced diagnostics for x-ray diffraction; and (7) continue our diagnostic development work on Janus.

Publications

Barrios, M. A., et al., 2012. "Precision equation-of-state measurements on NIF ablator materials from 1 to 12 Mbar using laser-driven shock waves." *J. Appl. Phys*. **111**(9), 093515. LLNL-JRNL-513776.

Erskine, D. J., et al., 2012. "Two-dimensional imaging velocity interferometry: Data analysis techniques." *Rev. Sci. Instrum*. **83**(4), 043116. LLNL-JRNL-513761.

Fratanduono, D. E., et al., 2011. "Index of refraction of shock-released materials." *J. Appl. Phys*. **110**(8), 083509. LLNL-JRNL-491933.

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Asteroid Deflection

Paul Miller (12-ERD-005)

Abstract

We propose to respond to a call by the presidential science adviser for significantly more analysis of asteroid deflection methods that could be used to protect Earth in the event of an impending collision. The Nuclear Regulatory Commission reported to Congress that nuclear explosives are currently the only option to defend Earth against large asteroids, or when the time before a collision is short. We intend to assess approaches employing nuclear devices to deflect near-earth objects identified to be on course to collide with Earth. Effects of such collisions range from localized disasters to massive global devastation. We will develop scenarios with a range of threat compositions, sizes, dynamics, and times to impact, and optimize parameters such as height of burst and yield for a nuclear deflection response. We will collaborate with experts in academia and the National Aeronautics and Space Administration. Much of our work will be unclassified, using generic sources for the studies.

We expect to develop threat scenarios and evaluate and optimize a variety of nuclear approaches, develop an asteroid breakup modeling capability, and increase the expertise in, and understanding of, the near-earth objects threat. We expect to publish our science in high-visibility journals and develop collaborations with universities and government agencies. We will assess U.S. devices for their applicability against a range of potential threats. This project will substantially improve our understanding of options available to disrupt or divert near-earth objects on a collision course with Earth.

Mission Relevance

Our proposed research draws directly on LLNL nuclear-design capability for a mission of national interest in ensuring international and domestic security. In addition, the work expands upon our traditional role of stockpile stewardship because nuclear explosives represent one of the major options for asteroid deflection.

FY12 Accomplishments and Results

We advanced our asteroid-deflection research in collaboration with academia and other government agencies, including NASA, the University of Washington, Harvard, Iowa State, and University of California, Santa Cruz. Specifically, we (1) developed scenarios spanning the range of near-earth object threats from comets and asteroids; (2) implemented a preliminary capability to model asteroid breakup, leveraging the specialized strengths of five different computer codes for energy coupling, hydrodynamic response, strength, and porosity; (3) conducted a series of energy-coupling simulations to explore several parameters, including composition, porosity, and energy source; and (4) developed and conducted a series of computational test problems as part of verification and successfully simulated the Stickney crater on Phobos—a moon of Mars—as a validation problem.

Proposed Work for FY13

For FY13 we will (1) continue to refine and implement our range of scenarios and their simulation within our suite of five codes, (2) continue use of our test problems to verify code results, (3) complete documentation of an error analysis of a dispersed object, (4) further develop orbital-dispersal simulations, and (5) conduct simulations of the Phobos Stickney crater as a validation problem.

Publications

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Probing Atomic-Scale Transient Phenomena Using High-Intensity X-Rays

Stefan Hau-Riege (12-ERD-021)

Abstract

Can we advance functional biology to prevent and cure diseases? Can we engineer storage technologies to harness renewable energy sources and obtain energy independence? These diverse, important issues can be addressed only through symbiotic basic research in physics, biology, chemistry, engineering, and mathematics, which is possible by using the newly built Linac Coherent Light Source (LCLS) at Stanford University that enables atomic-resolution imaging with femtosecond time resolution. However, samples exposed to the LCLS are severely damaged by radiation in an unchartered regime of x-ray and matter interaction. Without understanding the damage mechanism, the diffraction data cannot be analyzed correctly. Therefore, we propose to undertake fundamental research in the emerging field of x-ray-induced transient nanometer-scale phenomena. We will provide a validated predictive capability for high-intensity x-ray and matter interactions at the atomic level. At the same time, we will validate and improve x-ray plasma diagnostics methods for taking femtosecond snapshots of the dynamics of equilibration processes taking place at the nanometer-length scale.

We expect to greatly enhance our understanding of atomic-scale intense-x-ray and matter interaction as well as establish the foundational knowledge needed to enable LCLS imaging. We will develop experimentally validated, realistic models for high-intensity x-ray and matter interaction, obtain femtosecond snapshots of the dynamics of equilibration processes in materials excited by x rays, and develop an understanding of x-ray and matter interaction processes in nanostructures. This interplay of modeling and experiments forms the backbone of our proposal.

Mission Relevance

Our proposed high-visibility research program will sustain and further develop LLNL's international leadership in x-ray plasma probing and solid-damage physics through impactful, timely research resulting in high-profile publications. Our project is directly relevant to many Livermore strategic mission thrusts, including stockpile stewardship science and securing energy independence for the nation. In addition, this high-profile project will attract top postdoctoral researchers and allow us to engage in international collaborations.

FY12 Accomplishments and Results

In FY12 we were able to secure experimental time at the LCLS facility—one year ahead of schedule—so we shifted our focus from modeling efforts to an experiment to examine the dynamics of x-ray-excited materials. Specifically, we (1) designed and performed an x-ray free-electron laser experiment at the LCLS facility, (2) began model development by merging a tight-binding molecular dynamics code with a continuum model for atomic kinetics, and (3) developed a model to describe the photoelectron dynamics in materials irradiated by x-ray free-electron lasers.

Proposed Work for FY13

In FY13 we will (1) evaluate the scattering data obtained in our LCLS experiment performed in FY12, (2) compare the experimental data to our model predictions, (3) continue development of our x-ray interaction model, and (4) design—if time permits—a follow-up LCLS experiment for the third year of this project.

Publications

Hau-Riege, S., 2012. "The role of photoelectron dynamics in x-ray-free-electron-laser diffractive imaging of biological samples." *Phys. Rev. Lett.* **108**, 238101. LLNL-JRNL-537631.

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Computational Gyro-Landau Fluid Model for Tokamak Edge Plasmas

Xueqiao Xu (12-ERD-022)

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

If successful, the edge-plasma model created and validated in this project will generate a theoretical and simulation capability far beyond the present state of the art. An accurate gyro-Landau fluid simulation model would offer orders-of-magnitude savings in computational resources compared to a gyro-kinetic simulation, and make gyro-Landau fluid code very attractive as a component in an integrated, whole-device simulation. Moreover, a gyro-Landau fluid code could potentially be extended to treat core physics

and enable a global model of nonlinear plasma dynamics for the entire tokamak. This work will contribute to the validation and application of gyro-Landau fluid models that will be needed to design experiments at the international ITER fusion reactor in southern France as well as future reactors. This project also offers an excellent opportunity to forge alliances with emerging Asian fusion programs.

Mission Relevance

By filling critical gaps in theoretical understanding and simulation capability for fusion energy and other scientific fields such as astrophysical and space plasmas, the project supports LLNL's energy and national security missions and the focus areas of fusion energy, high-energy-density science, and high-performance computing.

FY12 Accomplishments and Results

In FY12, we (1) generated gyro-Landau fluid model equations using a vorticity formulation for electromagnetic "1 + 0" and "2 + 0" models, which refer to the number of parameter moments such as density and parallel velocity retained in the model equation; (2) developed a fast non-Fourier method for the computation of Landau-fluid closure terms; (3) implemented this method through solution of matrix equations in which the matrices are tridiagonal and demonstrated the method's computational efficiency; (4) implemented nonlocal gyro-average operators using Pade approximation; (5) implemented 3 + 0 electrostatic model equations; (6) implemented a 2 + 0 electromagnetic model; (7) benchmarked linear gyro-Landau fluid simulations with eigenvalue calculations and other linear fluid codes; and (8) validated simulated electromagnetic filamentary structures against camera images from the Experimental Advanced Superconducting Tokamak.

Proposed Work for FY13

We will (1) investigate requirements for models to behave well at large perturbations; (2) assess needs and methods for second-order-accurate closures; (3) implement 3 + 1 electrostatic and electromagnetic models; (4) implement realistic magnetic x-point geometry; (5) execute linear and nonlinear benchmarks—eigenfunctions, frequencies, growth rates, turbulence, and axisymmetric equilibria; (6) implement a nonlocal parallel transport operator in the BOUT++ framework; (7) develop a model of injected radio-frequency waves under a gyro-Landau fluid framework; (8) compare our 3 + 0 electrostatic model to turbulence data from the Korean National Fusion Research Institute and DIII-D; and (9) compare the 2 + 0 electromagnetic model to magnetohydrodynamic instability data from DIII-D and the Experimental Advanced Superconducting Tokamak.

Publications

Dimits, A. M., I. Joseph, and M. V. Umansky, 2012. *Fast non-Fourier methods for Landau-fluid operators*. 2012 Intl. Sherwood Fusion Theory Conf., Atlanta, GA, Mar. 31–Apr. 3, 2012. LLNL-POST-543370.

Liu, Z. X., T. Y. Xia, and X. Q. Xu, 2012. "ELMy H-mode linear simulation with 3-field model on EAST tokamak using BOUT++." *Phys. Plasma*. **19**(10), 102502. LLNL-JRNL-569455.

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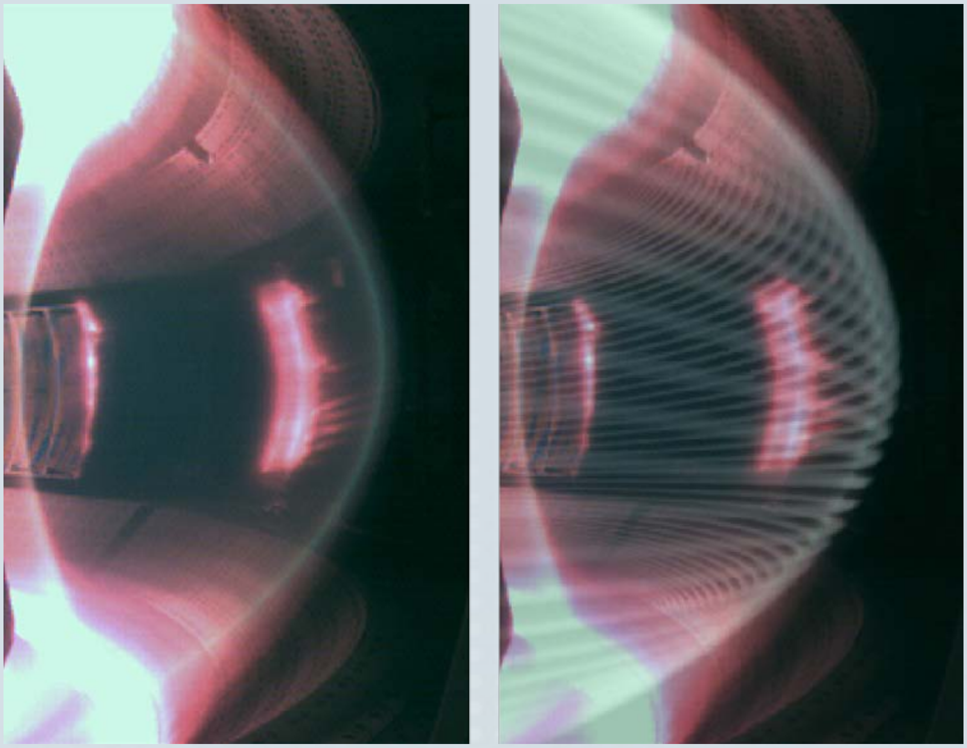

A high-speed camera captures ripples of cool plasma on the surface of the hot, but invisible, confined plasma in the Experimental Advanced Superconducting Tokamak in China (left). We developed a new supercomputer simulation (right) that produces the same surface ripples as the experiment, where both the filament orientation and spacing match.

Xu, X. Q., P. W. Xi, and T. Y. Xia, 2012. *Gyro-fluid simulations of large and small ELMs*. 22nd Pedestal and Edge Topical Group Mtg., Hefei, China, Apr. 2, 2012. LLNL-PRES-543362.

Hydrogen Ice Layers for Inertial-Confinement Fusion Targets

Evan Mapoles (12-ERD-032)

Abstract

Targets for inertial-confinement fusion comprise layers of condensed hydrogen fuel inside spherical capsules. The layers must be easily reproducible and very smooth. Numerous experiments have shown that these requirements can only be met by using a nearly perfect single crystal of solid hydrogen. The formation of these high-quality layers depends on creating and isolating a single crystal of the solid and then slowly cooling the melt to freeze the remaining liquid. The current success rate of this process is subject to the random nature of nucleation and the resulting seed crystal used to grow these layers. This method results in a range of layer qualities, many of which do not meet target specifications. This reduced yield currently constrains the shot rate for layered targets that can be achieved at the National Ignition Facility and other laser facilities. We propose to develop a deterministic seeding process leading to reproducible high-quality target ice layers.

If successful, this project will provide the scientific understanding needed to develop a layering process and modifications to the target that will allow high-quality layers to be grown reproducibly in a predictable time. In addition to the scientific insight into heterogeneous nucleation that will be gained, this work will also improve LLNL's ability to conduct inertial-confinement fusion research by reducing and enabling a more predictable time required to perform shots.

Mission Relevance

This project advances inertial-confinement fusion research and therefore supports the Laboratory's missions in stockpile stewardship, energy security, and high-energy-density science.

FY12 Accomplishments and Results

In FY12 we (1) completed the apparatus to test electro-freezing and conducted experiments that showed the current approach to electro-freezing is not a reliable method to induce controlled nucleation of solid hydrogen; (2) designed and constructed a cryostat to improve the process of creating target ice layers; (3) completed a large number of molecular dynamics calculations that encourage us to believe that a careful choice of substrate structure can lead to nucleation of hydrogen crystals of the desired phase and orientation—a substrate with prismatic hexagonal planes, close to the lattice constant of hydrogen, is predicted by simulation to produce only the hexagonal close-packed phase, whereas growth on the basal planes results in hydrogen with a mixture of face-centered cubic and hexagonal close-packed phases; and (4) began a new effort to optimize parameters used in our current layer formation process and develop metrics to predict the outcome of the layer process as early as possible—promising metrics have been developed that suggest bad layers may be identified as early as five hours into the process.

Proposed Work for FY13

We will (1) continue electro-freezing testing, demonstrating the improved techniques and beginning to customize the tips; (2) begin to study the nucleation of selected materials using the apparatus we built in FY12; (3) continue modeling deuterium–substrate interaction and deuterium crystal stress; (4) continue work on predicting layer growth in inertial-confinement fusion targets using metrics derived from analysis of x-ray images of the early evolution of these layers; and (5) begin analysis and experiments to integrate previously developed nucleation methods into the fusion targets.

Novel Multiple-Gigahertz Electron Beams for Advanced X-Ray and Gamma-Ray Light Sources

David Gibson (12-ERD-040)

Abstract

Our objective with this project is to design, model, assemble, and demonstrate a high-brightness electron-beam source that is capable of generating electron bunches in each wakefield period (bucket) of the drive radio frequency. Using X-band radio frequency (11.424 GHz) maximizes the number of bunches that can be generated, up to 1,000 bunches per pulse, at a 120-Hz repetition rate. Such an electron source would significantly increase the flux and/or brightness of electron-based light sources (such as Compton-scattering sources and free-electron lasers). The research objective will be accomplished by building a new multiple-gigahertz-compatible photo-injector and a gigahertz-compatible cathode illumination laser, which will be integrated with existing radio-frequency power and electron accelerator hardware to make beam measurements.

If the project is successful, we will have demonstrated a multiple-gigahertz electron beam suitable for use in a Compton-scattering-based gamma-ray (MEGa-Ray) source. This would allow the gamma-ray flux and overall source brightness to be significantly increased, while simultaneously simplifying the design of the associated photo-injector drive laser, accelerator, dark-current mitigation hardware, interaction laser, and interaction region. The time structure of the gamma-ray source opens the possibility of time-resolved or stroboscopic measurements with picosecond-scale resolution. In the process of conducting this research, we will also learn what changes, if any, are needed to the accelerator hardware to accelerate the high-repetition-rate bunch train to a few hundred megaelectronvolts.

Mission Relevance

This effort supports the Laboratory's thrust area in nuclear photo-science to address national nuclear missions. Generation of a multiple-gigahertz electron bunch train with small-per-bunch charge can allow a higher flux and higher brightness MEGa-Ray source than the current single-bunch system provides, improving the quality of desired fundamental nuclear physics measurements, as well as expanding the envelope of feasible nuclear photonics applications for national and international security.

FY12 Accomplishments and Results

In FY12 we (1) used a high radio-frequency modulator to carve a train of 500 laser pulses from a continuous-wave laser with a spacing of 87.53 ps (11.424 GHz); (2) amplified these pulses to 3.2 nJ each (75-mW average power) and used self-phase-modulation effects in 300 m of optical fiber to produce up to 4 nm of bandwidth—a pair of diffraction gratings then compressed the pulses to 850 fs; (3) determined, based on these results and further modeling of the self-phase modulation electron gun dynamics, that 40-GHz pulse shaping is not required to achieve the necessary electron beam performance; and (4) modeled electron beam dynamics in a 5.59-cell gun to understand the performance tradeoffs as a function of charge as well as the wakefield periods generated by individual bunches—this modeling indicates that the current radio-frequency design of the gun is likely to be adequate for a train of low-charge bunches and a redesign should be unnecessary. Our collaboration with Stanford's SLAC National Accelerator Laboratory has given us access to removable cathode gun designs that should meet our needs.

Proposed Work for FY13

Because our modeling indicates an existing gun cell design is sufficient (with the exception of the quantum efficiency limits) to support a multiple-bunch beam, in FY13 we will test that design with an actual beam. Specific proposed tasks include the assembly of existing radio-frequency power (the modulator and klystron), accelerator, photo-injector, magnet, power supply, vacuum, control, and other ancillary components already in hand into an X-band test station. We will couple this test station with existing laser hardware to enable beam generation, paving the way for a gigahertz electron-beam technology demonstration in FY14.

Publications

Gibson, D. J., et al., 2012. *A drive laser for multi-bunch photoinjector operation*. 2012 IEEE Intl. Particle Accelerator Conf. (IPAC), New Orleans, LA, May 20–25, 2012. LLNL-PROC-557582.

Marsh, R. A., 2012. *Ultracompact accelerator technology for a next-generation gamma-ray source*. LLNL-PRES-558291.

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Marsh, R. A., et al., 2012. "Modeling and design of an X-band RF photoinjector." *Phys. Rev. Spec. Topic. Accel. Beams* **15**(10), 102001. LLNL-JRNL-497831-DRAFT.

Marsh, R. A., et al., 2012. *Modeling multi-bunch X-band photoinjector challenges*. 2012 IEEE Intl. Particle Accelerator Conf. (IPAC), New Orleans, LA, May 20–25, 2012. LLNL-PROC-557580.

Marsh, R. A., et al., 2012. *Ultracompact accelerator technology for a next-generation gamma-ray source*. 2012 IEEE Intl. Particle Accelerator Conf. (IPAC), New Orleans, LA, May 20–25, 2012. LLNL-PROC-557854.

Strength in Metals at Ultrahigh Strain Rates

Jonathan Crowhurst (12-ERD-042)

Abstract

We intend to obtain accurate data on the dynamic strength of metals—such as aluminum, copper, vanadium, and tantalum—and the time-dependent evolution of shock waves at ultrahigh strain rates. Motivations for this work include the current lack of high-time-resolution data to allow direct comparison with molecular dynamics simulations and a lack of directly measured strengths and related properties necessary to refine and test dynamic strength and multiscale models. In addition, currently there is an inability to fully test the validity, and thus account for, fundamental scaling laws—for example, the fourth power law, which describes energy radiated per surface area per unit time for a black body. We will use a proven tabletop laser-shock apparatus to obtain strain rates consistent with time resolutions of 1 ps. Data acquisition will be rapid and inexpensive and results will be analyzed using state-of-the-art dynamic strength models.

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

Mission Relevance

Shock waves, and the material data they provide, are of direct relevance to the Laboratory mission focus areas of stockpile stewardship science and advanced laser optical systems. Obtaining a more detailed understanding of the physics associated with high-strain-rate dynamic compression is also important for the foundational science of materials and chemistry at the extremes as well as for fundamental research in a range of disciplines.

FY12 Accomplishments and Results

In FY12 we performed studies concerning the behavior of iron, and in particular, the alpha-to-epsilon polymorphic transition for ultrafast timescales. Specifically, we (1) successfully measured surface velocity histories in iron films driven at strain rates in excess of 10^9 per second, (2) analyzed these data in the context of a comprehensive model designed to deal with nonsteady wave propagation and highly rate-dependent phenomena; (3) demonstrated that at these strain rates, and corresponding large deviatoric stresses, the alpha-to-epsilon polymorphic transition begins within tens of picoseconds after an initial very large and mostly elastic compression—by comparison with data obtained at much lower strain rates, we also show that the timescale of the transition clearly depends on strain rate; and (4) obtained ultrahigh strain-rate data from vanadium and beta-phase tantalum—in the case of the latter we observed very large elastic precursor strengths at strain rates of about 10^9 per second.

Proposed Work for FY13

In FY13 we will (1) continue our ultrahigh strain-rate measurements on vanadium, iron, and aluminum; (2) further experiment with drive profiles that have different rise times and durations to obtain faster rises in shocks, or at the opposite extreme, to drive ramps; (3) use our strength model and associated code to obtain dynamic strength of vanadium, iron, and aluminum at ultrahigh strain rates; and (4) compare the ultrahigh strain-rate behavior of beta-phase tantalum to the more familiar body-centered cubic phase.

Publications

Crowhurst, J. C., 2012. *Aspects of the behavior of aluminum and iron at ultrahigh strain rates*. Strain Measurement in Extreme Environments, Glascow, Scotland, Aug., 28, 2012. LLNL-PRES-563697.

A Model-Reduction Approach to Line-By-Line Calculations

Carlos Iglesias (12-ERD-047)

Abstract

The physical properties of stars depend upon the transport of energy from their nuclear cores to their surface. Although energy can be transferred out from the center by conduction and convection, radiation transport is the most important mechanism. In turn, the transport of photons depends on the transparency of the intervening matter, termed the radiative opacity. Consequently, opacity plays a key role in determining the evolution, luminosity, and instabilities of stars and even the eventual fate of the universe. We propose to improve opacity calculations by developing an accurate, efficient model-reduction approach to calculating bound–bound radiative transitions in many-electron ions. Opacity calculations for many-electron systems presently contain uncertainties because of the Gaussian approximation used to describe transition-array spectra. This research will bridge the extremes between the fast but potentially inaccurate Gaussian approximation presently

in use, and the exact but computationally expensive line-by-line calculation, with an expression that preserves higher moments of the transition array. We will develop an alternative method for computing the moments of the transition array and incorporate the algorithms into LLNL opacity codes.

Our main goal with this project is to develop a model-reduction approach to many-electron ions that is accurate and effective. If successful, this project will develop a new algorithm that will allow an accurate and efficient optimization of the number of effective lines required to generate a more realistic representation of bound–bound spectra. This result will significantly reduce computational time and allow opacity codes to handle larger problems without increasing the computational effort.

Mission Relevance

Stellar models and Laboratory applications, such as stockpile stewardship experiments on LLNL's laser facilities, rely on accurate descriptions of energy transport. By reducing the computational time required to calculate bound–bound spectra, this research furthers understanding of energy transport for the Laboratory's stockpile stewardship thrust area and supports the national security mission.

FY12 Accomplishments and Results

Instead of our original goal of developing a model-reduction approach to radiative transitions in many-electron ions, which we deemed impractical, in FY12 we (1) created a novel, simple concept to extend the existing unresolved transition array model with an approach that performs detailed line accounting for the most important subshells and treats the remaining subshells statistically, thereby conserving the array oscillator strengths of mean and variance and including initial population effects systematically; (2) extended our model to randomly generate lines and simulate detailed line-accounting calculations, achieving good agreement between these calculations and our new random, partially resolved transition array model, which is several orders of magnitude faster than the previous approach; and (3) published our work in peer-reviewed journals in the field of high-energy-density physics.

Proposed Work for FY13

In FY13 we propose to provide better physical justification for our random, partially resolved transition array model. Specifically, we will (1) achieve an improved understanding that may lead to even better agreement to detailed line-accounting calculations, (2) further improve and implement the model algorithm, and (3) fully test the new model in the OPAL stellar opacity and TOPAZ thermal diffusion codes.

Publications

Iglesias, C., 2012. "Statistical line-by-line model for atomic spectra in intermediate coupling." *High Energ. Density Phys*. **8**(3), 253. LLNL-JRNL-530533.

Iglesias, C., and V. Sonnad, 2012. "Partially resolved transition array model for atomic spectra." *High Energ. Density Phys*. **8**(2), 154. LLNL-JRNL-520413.

Forward Path to Discovery at the Large Hadron Collider

Douglas Wright (12-ERD-051)

Abstract

After nearly 20 years of construction, the Large Hadron Collider (LHC), situated near Geneva, Switzerland, is poised to explore the high-energy frontier of particle physics with the world's largest and highest-energy particle accelerator. The possibility exists, however, that the lightest new particles in the framework of today's proposed physics models will be too heavy to be discovered at the LHC. Lawrence Livermore is currently part of a detector upgrade project that would substantially enhance the physics reach of the Compact Muon Solenoid experiment at the LHC to eliminate such physics "blind spots." The upgrade will detect the anomalous production of W boson pairs (subatomic particles with a positive and negative electric charge that are each other's antiparticle) through their decay to muons (unstable subatomic particles with negative charge). This provides a model-independent means of observing the presence of new subatomic particles. In this project, we propose to adapt a feedback-stabilized, ultraprecise timing system, originally developed for the Advanced Light Source at the Stanford Linear Accelerator Center (SLAC), to synchronize the LHC forward-proton detectors required for the diagnostic upgrade.

We expect to enable the search for new fundamental subatomic particles by developing an advanced timing system that will be integral to a diagnostic upgrade to LHC. The key goal of the upgrade is to detect collisions in which beam protons remain intact and new physics signals are produced by the collision of photons produced from the proton beams. Detecting the intact outgoing protons requires small tracking detectors placed inside LHC beam pipes located a few hundred meters from the interaction point. Our staged plan starts with a basic feasibility demonstration and proceeds in two steps—including proving our approach with actual timing detectors in a test beam at the Fermi National Accelerator Laboratory—toward a fully capable LHC system.

Mission Relevance

This project applies LLNL science and engineering capabilities to address the highest priority mission of the particle physics program of the DOE Office of Science. A highly visible and unique role in both hardware and physics analysis at LHC will allow Lawrence Livermore to continue to attract and retain outstanding scientists, who ultimately make substantial contributions to national security programs. This frontier research project also supports Livermore's core competency in measurement science and technology by developing diagnostics able to view complex energetic dynamic processes.

FY12 Accomplishments and Results

In FY12 we conducted stand-alone timing tests at SLAC. Specifically, we (1) prepared a long length of low-loss coaxial cable and tested it with the spare system at SLAC designed for the Linac Coherent Light Source to validate the longest cable length required for the upgrade, (2) fed a reference signal and stabilized signal at the end of the cable into an existing phase comparator detector and sampled every few seconds over an extended period to demonstrate stability of the timing for the LHC proton-tagging detectors, and (3) developed initial analysis of exclusive muon pairs with missing-energy topology using existing LHC detectors. In addition, the principal investigator for this LDRD project was appointed U.S. principal investigator for the entire proton detector upgrade for the Compact Muon Solenoid experiment at LHC. This position entails significant responsibilities in leading the project and demonstrates how the U.S. has, by far, the largest technical role in the proposed upgrade.

Proposed Work for FY13

In FY13 we propose to (1) test the prototype reference timing system with a time-to-digital converter data-acquisition system to demonstrate timing suitable for use with the actual Cherenkov proton-timing detectors, (2) design an LHC-capable timing system based on the LHC clock signal and lessons learned from prototype measurements, and (3) obtain and publish results demonstrating the central, exclusive production of muon pairs using an existing detector from the Compact Muon Solenoid experiment and rejecting events that have evidence of proton breakup at the edge of the forward hadron calorimeters.

Compton-Scattering Optimization for Ultra-Narrow-Band Nuclear Photonics Applications

Frederic Hartemann (12-ERD-057)

Abstract

The main goal of this project is to optimize Livermore's Compton-scattering gamma-ray source technology for a potential 100,000-fold bandwidth reduction by combining gigahertz gamma-ray pulse trains with ultrahigh-resolution stabilized-crystal spectrometers. Gamma-ray lens technology also will be utilized for fractional bandwidths of 100 parts per million. This bandwidth reduction will enable the direct, isotope-specific detection (with nuclear resonance fluorescence), assaying, and imaging of special nuclear materials and other materials of interest. Key deliverables include detailed computer models of gamma-ray production, tracking, and interaction with gamma-ray optics, along with optimization algorithms. In addition, we will perform the experimental characterization of gamma-ray optics.

If successful, this project will bring the emerging new field of nuclear photonics closer to maturity by providing tunable, mega-electronvolt photons within sub-electronvolt bandwidths at high average flux. Our proposed approach is two-pronged. First, an intensive theory and modeling component will combine advanced three-dimensional electron beam dynamics codes with LLNL's Compton-scattering models, laser design tools, gamma-ray particle tracing and interaction codes, and sophisticated gamma-ray crystal diffraction models. Second, we will characterize a precision gamma-ray crystal spectrometer on loan from the Institut Laue-Langevin in France, and we may integrate

the device with LLNL's Compton gamma-ray source, along with performing characterization of gamma-ray optics.

Mission Relevance

Our project is expected to have maximum impact for the Laboratory's strategic mission in advanced laser optical systems and applications by developing a Compton source with higher flux and brightness than the current single-bunch system to provide improved quality of desired nuclear physics measurements and expanded research possibilities. By enabling imaging of special nuclear materials, this research also directly supports LLNL's national security mission, with applications to nuclear threat elimination.

FY12 Accomplishments and Results

In FY12 we (1) developed a Compton scattering code capable of calculating scattered photon conditions integrated over a given solid angle, quantifying nonlinear spectral broadening effects, and handling very long laser pulses without loss of performance; (2) began work on the nonlinear, three-dimensional optimization of laser parameters for single and multiple electron bunches; (3) defined quantitative, covariant parameters for the weakly nonlinear ponderomotive dephasing—discovered by our group—which limits the brightness of Compton scattering light sources; (4) acquired output data sets from three-dimensional, parallel, finite-element accelerator codes at the Stanford Linear Accelerator Center and used them to model disturbances known as wakefield oscillations in an x-band radio-frequency gun; and (5) established a collaborative effort on gamma-ray interactions with matter with the Ludwig Maximilian University of Munich and the Institut Laue-Langevin, and exchanged gamma-ray simulations.

Proposed Work for FY13

In FY13 we plan to (1) finalize the Compton scattering code we began developing in FY12 and use it to optimize gamma-ray production, with the goal of generating minimal on-axis spectral bandwidth—in conjunction with optimizing the electron beam multi-bunch pulse format and matching it with the appropriate laser pulse; (2) complete the gamma-ray code, along with interfaces to input electron phase and output gamma rays; (3) finalize a theoretical single-electron maximum-brightness study; and (4) participate in ongoing refractive index measurements at the European Synchrotron Radiation Facility and conduct characterization experiments with a precision spectrometer at the Institut Laue-Langevin.

Publications

Albert, F., et al., 2012. *Development of Compton scattering gamma ray sources at LLNL*. 15th Advanced Accelerator Concepts Workshop (AAC 2012), Austin, TX, June 10–15, 2012. LLNL-ABS-557757.

Albert, F., et al., 2012. *NRF-based isotope material detection experiments with the LLNL T-REX MEGa-ray machine*. 53rd Ann. Mtg. Institute of Nuclear Materials Management, Orlando, FL, July 15–19, 2012. LLNL-ABS-468934.

Anderson , S. G., et al., 2012. *Compact linear accelerator sources for mono-energetic gamma ray (MEGa-ray) generation and NRF-based nuclear materials management*. 53rd Ann. Mtg. Institute of Nuclear Materials Management, Orlando, FL, July 15–19, 2012. LLNL-ABS-468571.

Marsh, R. A., et al., 2012. *Ultracompact accelerator technology for a next-generation gamma-ray source*. Intl. Particle Accelerator Conf. 2012 (IPAC 2012), New Orleans, LA, May 20–25, 2012. LLNL-PROC-557854.

Wu, S. S. Q., and F. V. Hartemann, 2012. *Transient evolution of a photon gas in the nonlinear QED vacuum*. LLNL-TR-503123.

Early-Phase Hydrodynamic Instability Development in National Ignition Facility Capsules

Daniel Clark (12-ERD-058)

Abstract

Recently, the importance of the effect of early-phase Richtmyer–Meshkov instability growth on later Rayleigh–Taylor instability has been appreciated in determining the pulse shape needed to achieve fusion ignition in National Ignition Facility (NIF) experiments. We will develop a quantitative understanding of the role of this early-phase growth in determining the phase and amplitude of later Rayleigh–Taylor growth in NIF fusion implosions. To this end, we will combine simulations using the radiation-hydrodynamics code HYDRA with published and original analytic work. Although the immediate scope of the project is limited to indirectly driven NIF implosions and to theoretical and simulation-based investigations, our results will be relevant to other applications in stockpile stewardship science, NIF uses of ignition, and target design for laser ignition fusion energy.

Our three main deliverables will be (1) a detailed and quantitative understanding of the connection of Richtmyer–Meshkov phase growth to subsequent Rayleigh–Taylor growth in NIF implosions; (2) an understanding of the influence of equation of state, opacity, and other physics uncertainties in how Richtmyer–Meshkov instability growth affects Rayleigh–Taylor growth; and (3) a comparison of our results with mixed-model results previously obtained at LLNL. An additional result possibly will be experiment designs to test our theoretical results on NIF. Our results will be significant to the NIF ignition campaign, post-ignition NIF target designs, target designs for laser ignition fusion energy, stockpile stewardship, and the inertial-confinement fusion community at large.

Mission Relevance

By helping achieve a more complete understanding of hydrodynamic instabilities and mix, this project will reduce uncertainties in stockpile stewardship science applications, may uncover new physics relevant to stockpile stewardship, and also further LLNL's missions in nuclear and energy security, including NIF ignition, in which hydrodynamic instabilities play a central role. In addition, by understanding the details of instability development more thoroughly and by precisely determining acceptable stability boundaries, this project will also help improve ignition-capsule hydrodynamic stability and therefore enable high-gain target designs required for laser ignition fusion energy.

FY12 Accomplishments and Results

In FY12 we performed and analyzed simulations focusing on evolution of the perturbation spectrum in time at both the ablation front and the fuel–ablator interface, as well as at the propagating shock front for instability growth of both early and late phases. Our initial analysis resulted in the novel discovery that Rayleigh–Taylor growth appears not to be determined by the phase set in the initial, shock Richtmyer–Meshkov growth, but by the returning rarefaction fan from the capsule inner surface. This result is preliminary, but if verified will substantially modify our understanding of how Rayleigh–Taylor instability in inertial-confinement fusion is controlled. In collaboration with a colleague from the CEA megajoule laser facility in France, we have also made a close comparison of our simulations results against existing analytic theories for Rayleigh–Taylor and ablative Richtmyer–Meshkov growth. We find that the salient features extracted from the simulations, particularly evolution of the phase or sign of the perturbation, can be captured remarkably well by a simple analytical description informed by simulation results.

Proposed Work for FY13

For FY13 we plan to continue the phenomenological, simulation-based study of Richtmyer–Meshkov and Rayleigh–Taylor instability growth phase effects and their dependence on the strength of the foot of the radiation drive pulse. Specifically, we will focus primarily on verifying or refuting our newly developed appreciation of the role of the return rarefaction in setting the perturbation phase for subsequent Rayleigh–Taylor growth with the use of analytic theory and development of a quantitative, semi-analytic description of multiple-shock Richtmyer–Meshkov and Rayleigh–Taylor growth.

Publications

Peterson, J. L., et al., 2012. *Altering the hydrodynamic mixing of NIF capsules with shocks and rarefactions*. 54th Ann. Mtg. APS Division of Plasma Physics, Providence, RI, Oct. 29–Nov. 2, 2012. LLNL-ABS-563711.

Peterson, J. L., et al., 2012. *The phase of hydrodynamic growth in inertial confinement fusion capsules*. 42nd Ann. Anomalous Absorption Conf., Key West, FL, June 25–29, 2012. LLNL-ABS-552711.

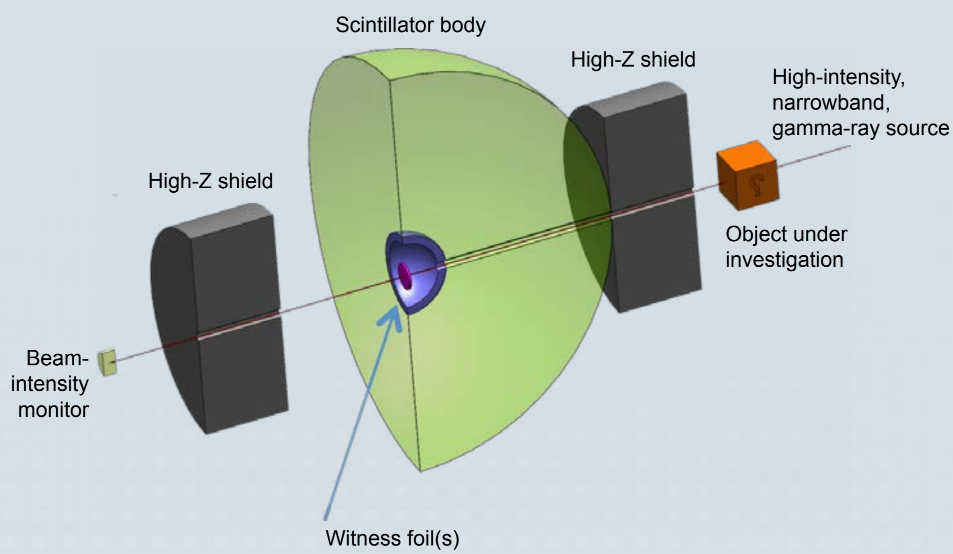
The Next Generation of Gamma-Ray Sources: Dual-Isotope Notch Observation

Christopher Ebbers (12-ERD-060)

Abstract

Our objective is to model, develop, design, assemble, and demonstrate a detection system capable of efficiently using the next generation of laser-based gamma-ray, or MEGa-ray, sources being developed at LLNL and elsewhere. Created by Compton scattering of short-duration laser pulses interacting with relativistic electrons, MEGa-rays are a new class of light source with extraordinary qualities. We propose the dual-isotope notch observation (DINO) system, a revolutionary detector arrangement that, when coupled with a MEGa-ray source, enables the unambiguous detection of special nuclear materials, isotope-specific imaging, and nuclear assay. We aim to create the computational tools necessary to model such detectors including enabling LLNL's radiation transport code Mercury, or equivalent, to handle nuclear photonics. Using those tools, we will model DINO detector activities specific to the MEGa-ray source under development, and build and test a DINO detector for source characterization.

Upon successful completion, we will have developed the physics code base, modeled, built, and demonstrated a DINO detector configuration for characterizing MEGa-ray sources, with an unprecedented level of isotopic specificity detection that is two to three orders of magnitude faster than current technology. The suite of computational tools for modeling and optimizing DINO-type detectors will enhance the physics modeling capability of the Mercury code for nuclear resonance fluorescence.



The advent of high-intensity, narrow-band gamma-ray sources (MEGa-rays) will enable the rapid identification, assay, and differentiation of nuclear isotopes. However, these new gamma-ray sources under development also require a new detection paradigm. Our dual-isotope notch observation detector concept, schematically shown above, uses a fast calorimetric-type detection scheme with high-atomic-number (high-Z) shields as opposed to the existing high-resolution spectroscopic detection method.

Mission Relevance

By developing technology critical to the success of MEGa-ray sources, this effort supports the Laboratory's strategic goal for advanced laser optical systems of establishing nuclear photo-science as a new scientific discipline to address nuclear security missions. In addition to their potential as basic research tools in nuclear photonics, narrow-bandwidth photon sources such as MEGa-ray are expected to have a number of applications in key LLNL mission areas including detecting highly enriched uranium for counterterrorism, precision assay of nuclear fuels and nuclear waste for counter-proliferation, and surveillance for stockpile stewardship.

FY12 Accomplishments and Results

In FY12 we successfully (1) modeled the effectiveness of scintillator and detector architectures in preparation to experimentally verify the angular scattering properties of nuclear resonance fluorescence at the High Intensity Gamma Source at Duke University in North Carolina; (2) began modification of the atomic and nuclear data frameworks for optimized codes that include nuclear resonance fluorescence in optimized, modern (data-driven) codes; (3) initiated implementation of coherent elastic scattering and beam polarization physics into the Mercury Monte-Carlo simulation code; and (4) benchmarked simulated nuclear resonance fluorescence results between the standard available codes (MCNP and COG) and the newly modified code.

Proposed Work for FY13

In FY13 we propose to continue to develop a novel nuclear resonance fluorescence detector and modeling code achieved through (1) modification of the Mercury modeling code to include hot-electron transport, coherent, and incoherent scattering; (2) add polarized gamma-ray interactions to the Mercury modeling code; (3) use the optimized numerical simulation codes to design and model the construction and layout of a DINO detection system (shielding, Compton liner, foil, and scintillator); and (4) construct a numerically modeled, segmented-architecture detector and test using an existing or surrogate source of nuclear resonance fluorescence photons.

Publications

Johnson, M. S., et al., 2012. *Detector array performance estimates for nuclear resonance fluorescence applications*. LLNL-ABS-563014.

Johnson, M. S., et al., 2012. *Preliminary study of the efficacy of using nuclear resonance fluorescence with quasi-monoenergetic gamma-ray sources for nuclear safeguards assay*. LLNL-TR-483851.

Semenov, V., C. Barty, and M. Johnson, 2012. *Differential cross-section of nuclear resonance fluorescence: Proposed measurements and applications*. LLNL-PRES-567354.

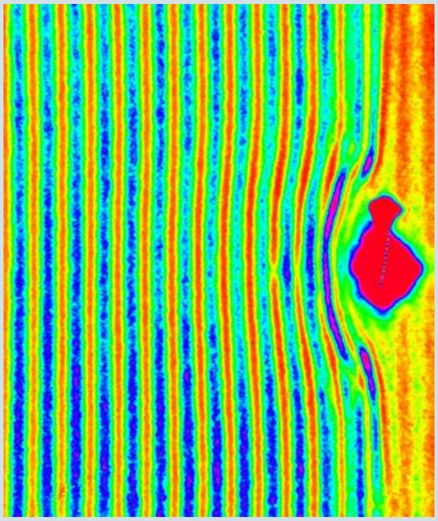
Pair-Plasma Creation Using the National Ignition Facility

Hui Chen (12-ERD-062)

Abstract

Relativistic electron–positron (antimatter) pair plasmas and jets are believed to exist in many astrophysical objects. Their presence would explain energetic phenomena related to gamma-ray bursts and black holes. Yet the ability to study these dense relativistic electron–positron plasmas in the laboratory has been elusive until now because no experimental platform has been capable of producing the high-temperature pair plasma and high-flux pair jets required to simulate astrophysical positron conditions. Experimental scaling based on the data from smaller, short-pulse laser experiments shows that, when completed, Livermore's Advanced Radiography Capability at the National Ignition Facility will be the only facility where a pure pair plasma can be created. We propose to initially use short-pulse laser experiments and ultimately use the Advanced Radiography Capability to create and study electron–positron pair jets and plasmas and the consequent gamma-ray bursts that result from pair annihilation.

If successful, this project will build a new experimental platform that will create, for the first time ever in a laboratory, the high-temperature pair plasma and high-flux pair jets required for understanding exotic, ubiquitous, yet little-understood astrophysical phenomena. The results will have a significant impact in the high-energy-density field and may establish the plasma physics of laser and antimatter interactions as a new subfield in high-energy-density science. Multiple, high-profile publications are expected.



Interferogram measured when the Livermore Titan laser interacts with a target in pair-plasma experiments. The laser irradiates the target from the left to the right. The bright red area is the self-emission of the target, and plasma density is inferred from this measurement.

Mission Relevance

These novel, high-energy-density laboratory experiments will aid in understanding astrophysical phenomena in support of basic and stockpile stewardship science. In addition, high-density, laser-produced positrons produced in this project will also enable new applications for the Laboratory's nuclear security mission. Such applications include diagnosing high-energy-density plasmas and providing a source of pulsed, mono-energetic gamma rays for radiography of dynamic processes in materials.

FY12 Accomplishments and Results

In FY12 we (1) performed experiments on pair-plasma scaling versus energies up to 1.4 kJ using the LLNL Titan laser and the OMEGA EP laser at the Laboratory for Laser Energetics in Rochester, New York; (2) designed and performed the initial pair-jet experiments on the LFEX (laser for first ignition experiment) short-pulse laser system at Osaka University in Japan; (3) began design of the electron–positron spectrometer for the National Ignition Facility; (4) completed our first computer simulations for design of a laboratory astrophysics experiment on the interaction between a relativistic pair-plasma jet and a dense plasma produced by nanosecond-pulse lasers; (5) and submitted papers on positron emittance, LFEX experiments, and annihilation measurements to relevant peer-reviewed journals.

Proposed Work for FY13

In FY13 we will (1) optimize the pair-plasma density and temperature using the LLNL Titan laser and the OMEGA EP laser; (2) carry out a pair-plasma experiment at Osaka University on pair scaling as a function of laser intensities; (3) construct a prototype electron and positron spectrometer for the National Ignition Facility, and prepare for a pair-plasma experiment there; and (4) design astrophysics-relevant experiments using relativistic pair-plasma jets for Titan, OMEGA EP, and Advanced Radiography Capability laser systems.

Publications

Chen, H., R. Tommasini, and J. Seely, 2012. "Measuring electron–positron annihilation radiations from laser plasma interactions." *Rev. Sci. Instrum*. **83**(10), 10E113. LLNL-JRNL-529821.

Chen, H. et al., 2013. "Emittance of positron beams produced in intense laser plasma interaction." *Phys. Plasma*. **20**(1), 01311. LLNL-JRNL-538314.

Chen, H., et al., 2012. *First electron–positron pair experiments using the Osaka LFEX laser*. LLNL-JRNL-564459.

Volume Collapse: Finger on the Pulse of the f Electrons

Magnus Lipp (12-LW-014)

Abstract

We propose to examine a very important correlated-electron phenomenon: electron-driven volume collapse. Volume collapse is related to one of the most fascinating and important properties of f-type electrons, which is their ability to transition between localized-like and delocalized-like behavior. This transition is a key element in determining important material properties, including the temperature phase diagram of plutonium, the great power of rare-earth magnets, and exotic, condensed-matter effects like heavy fermions, valence-mediated superconductivity, and quantum criticality. We will employ new experimental probes—pressure-dependent advanced x-ray spectroscopy techniques—to distinguish between competing theoretical descriptions of the f-electron delocalization and volume collapse. Direct examination of the microscopic physics of f-electrons promises to create a fertile feedback loop with theory, in direct contrast with the standard paradigm, where theory is often expected to derive f-electron physics from crystallographic structure alone.

If successful, we will provide a systematic description of the delocalization process of f-electrons, in order of increasing pressure, for classes of rare-earth materials. Such a description would be an immediate benchmark for the theory of correlated f-electron

systems, which is still under development. The project will also offer the x-ray community a new and powerful methodology for addressing questions of electronic structure under pressure. Furthermore, LLNL will have developed the infrastructure and expertise to examine and explain the electronic structure of more complex rare-earth materials, which have important applications that depend on the subtle details of their microscopic electronic states.

Mission Relevance

Because of the strategic importance of fundamental research on rare-earth metal systems, this project has relevance to LLNL's nuclear and energy security missions. In the future, this work can be extended to the actinides, where chemical and equation-of- state modeling both depend strongly on understanding intimate details of the microscopic physics of f-electrons.

FY12 Accomplishments and Results

In FY12 we (1) designed, built, and commissioned a resonant x-ray emission spectrometer for measurement of the light rare-earth element neodymium; (2) performed high-pressure resonant x-ray emission spectrometry and praseodymium spectrometry, as well as high-pressure x-ray Raman measurements, on cerium metal; and (3) built and commissioned a spectrometer for improved non-resonant emission spectroscopy, and made initial measurements on neodymium—the results were coordinated with theory and published in three peer-reviewed journals.

Proposed Work for FY13

We propose to perform experiments and measurements of rare-earth metals and other materials with f-type electrons to elucidate the role of electrons in the process of pressure-induced volume collapses using three methods: (1) resonant x-ray emission spectrometry, (2) non-resonant x-ray emission spectrometry, and (3) x-ray Raman spectroscopy.

Publications

Bradley, J. A., et al., 2012. "4f electron delocalization and volume collapse in praseodymium metal." *Phys. Rev. B* **85**(10), 100102R. LLNL-JRNL-600273.

Lipp, M. J., et al., 2012. "X-ray emission spectroscopy of cerium across the γ-α volume collapse transition." *Phys. Rev. Lett.* **109**(19), 195705. LLNL-JRNL-600255.

Pacold, J. I., et al., 2012. "A miniature X-ray emission spectrometer (miniXES) for high-pressure studies in a diamond anvil cell." *J. Synchrotron Rad*. **19**(2), 245. LLNL-JRNL-600752.

Physics Beyond Feynman

Peter Beiersdorfer (12-LW-026)

Abstract

We intend to determine whether the theory of quantum electrodynamics (QED) is complete, its incompleteness having been suggested for the first time in an experiment measuring differences in energy levels in hydrogen published in Nature in the latter half of 2010. Of profound importance to all of physics and to our very understanding of the universe, QED is the relativistic quantum-field theory of electrodynamics. The recent reported experiment, which measured the charge radius of the proton, raises the question of whether QED theory is incomplete at the level of two-loop Feynman diagrams, which are representations of the mathematical expressions governing the behavior of subatomic particles. To answer this important question, we will determine the QED two-loop term with an accuracy that is ten times greater than what has been achieved previously. These measurements will be carried out at the Livermore SuperEBIT electron-beam ion trap facility, which will produce the necessary lead ions. Very-high-resolution grating spectrometers will be installed at this facility for observational and calibration purposes.

With the success of this project, we will demonstrate the first-ever test of two-loop QED in a bound atomic system not encumbered by the finite size of the nucleus—in the case of atomic hydrogen, this will be the proton. If we determine that QED predictions differ from measurement on the two-loop level of Feynman diagrams, our results will mean that QED theory will need to be revised. Our results could also imply that the interpretation of the recent measurements of the proton radius is flawed, likely requiring changes in the Standard Model of particle physics concerning nuclear interactions.

Mission Relevance

This project may profoundly alter our understanding of the fundamental forces of physics. It may thus affect the underpinnings of the physics of many LLNL missions and scientific thrusts, including atomic physics in stockpile stewardship, high-energy chemical compounds with national security relevance, energy manipulation, and materials on demand.

FY12 Accomplishments and Results

In FY12 we (1) installed and tested two high-resolution grating spectrometers and calibrated their energy scales; (2) determined that the count rate on one of the spectrometers was reduced, redesigned the instrument to increase collection efficiency, and reinstalled the device; (3) completed conversion of SuperEBIT to high-energy operation and determined initial operation at high voltage was successful—in addition, injection of lead into the trap was successful; (4) completed analysis of data on lithium-like praseodymium, which is less charged than lead but is in the same isoelectronic sequence; (5) performed associated QED calculations; and (6) began measurements of lithium-like lead.

Proposed Work for FY13

In FY13 we will (1) optimize production and trapping of large numbers of lead ions in SuperEBIT, with the goal of collecting enough photon counts for deriving the QED contribution for determining the two-loop QED term for behavior of subatomic particles at a tenfold increase in accuracy over what has been previously achieved; (2) process the spectral data by optimizing the signal-to-noise discriminator in the analysis software; and (3) complete QED calculations for comparison with data and prepare the results for publication.

Publications

Trabert, E., et al., 2012. *Hyperfine splitting of the* $^2S_{1/2}$ *and* $^2P_{1/2}$ *levels in Li- and Be-like ions of the open-nuclear shell isotope* ^{141}Pr. LLNL-JRNL-594313.

Nonlinear Evolution of the Weibel Instability of Relativistic Electron Beams

Donald Correll (12-FS-002)

Abstract

We propose the first-ever systematic study in the laboratory of nonlinear evolution of Weibel instability present in electromagnetic plasmas. This instability produces counter-streaming beams in plasma flow that could impact ignition for inertial-confinement fusion. We will use computer simulations to develop the concept for a National Ignition Facility (NIF) experiment in which electromagnetic Weibel instability and the associated magnetic-field structures are studied using long-pulse NIF lasers to create large background plasmas with tailored density and temperature scales, and using high-intensity NIF Advanced Radiographic Capability beams to produce fast electrons. The creation of large hot plasmas with various density profiles using a long-pulse laser will be modeled with radiation-hydrodynamic codes. With the available energy and various configurations of the NIF nanosecond-pulse beams, millimeter-scale large plasma samples with a density range spanning five orders of magnitude can be assembled. This will provide the needed density platform to study the evolution of Weibel instability.

We intend to address several key questions on Weibel instability: (1) the dependence of instability on background plasma density; (2) the long-term evolution and equilibrium state of instability, including how strong the magnetic field is, how much magnetic energy can be generated, whether filaments merge, and what spatial structures exist and how long they last; and (3) the effects of Weibel instability on relativistic electron beam propagation, including whether instability could result in significant anomalous energy stopping.

Mission Relevance

By building underlying science and technology regarding an important phenomenon related to electromagnetic plasmas, this project supports the Laboratory's missions in national security, energy security, and scientific discovery. In particular, the Weibel instability of relativistic beams is of central importance to plasmas in the laboratory and in the cosmos. Success in achieving fast ignition in inertial-confinement fusion may depend on understanding this type of instability and preventing its occurrence.

FY12 Accomplishments and Results

Colleagues from General Atomics and the University of California, San Diego, under the direction of Mingsheng Wei, performed hybrid particle-in-cell simulations to characterize the nonlinear growth of Weibel-like instabilities and magnetic structures in a laser–plasma interaction for NIF Advanced Radiographic Capability laser parameters. We determined that (1) low-background plasma density (0.2 g/cm^3) leads to rapid growth of the Weibel fields near the beam creation plane that significantly inhibits fast electron transport, (2) the effect of increasing plasma temperature (20 to 100 eV) was found to be negligible, and (3) decreasing fast electron-beam divergence (30 to 15°) produces stronger Weibel-like and global magnetic fields that can collimate fast electron transport.

Publications

Wei, M., 2012. *Study of nonlinear evolution of the Weibel-like instability of relativistic electron beams relevant to fast ignition and astrophysics*. LLNL-TR-608854.

Recovering Large Volumes of Homogeneously Shocked Samples

Donald Correll (12-FS-003)

Abstract

The formation and modification of water-bearing minerals is central to understanding the history of water in the solar system. Shock processes played a major role in the transport of water—for example, in the impact delivery of water to growing planets, the collisional formation of the first small celestial bodies, and the removal and redistribution of crustal water during the early period of intense planetary bombardment. Despite previous experimental attempts aimed at understanding shock-induced structural and spectroscopic changes of water-bearing minerals, the data currently in hand are sparse and difficult to interpret because of heterogeneous, non-planar shock loading, improper recovery capsules, and unknown and complex loading history in the sample. We propose to develop large-volume recovery experiments for friable materials, taking advantage of simultaneous in situ diagnostics at the National Ignition Facility (NIF), where large volumes of recovered sample are necessary for precise post-shock analysis of hydrated minerals. We will combine the computational simulation of NIF experiments with concept-validation experiments on the Janus platform at Livermore's Jupiter Laser Facility.

The loading history of a rock during a natural impact event is straightforward: the material is shocked up to a peak pressure and then immediately begins to release to ambient pressure. The NIF laser platform is the perfect drive mechanism for such work, capable of accurately

recovering samples subjected to "natural" loading histories. We expect to determine the planarity and roughness of shock waves over a large area using a spatially resolved VISAR (velocity interferometer system for any reflector) device. We will also develop a design for large-volume recovery with in situ diagnostics and provide a summary of design considerations. In addition, we will summarize design considerations and tips for future NIF users wishing to propose large-volume recovery experiments.

Mission Relevance

By developing a large-volume recovery system, we can address the poorly understood response of hydrated minerals to shock compression and release, in support of the Laboratory's core competency in high-energy-density science.

FY12 Accomplishments and Results

In FY12 colleagues at Harvard University, led by Sarah Stewart, (1) designed and tested a recovery system for laser targets subject to Richtmyer–Meshkov or Rayleigh–Taylor instability growth; (2) subjected samples of tantalum and iron to shock states ranging from 20 to 150 GPa on Livermore's Janus laser; and (3) successfully recovered rippled samples of tantalum and iron, observing Richtmyer–Meshkov instability growth in both tantalum and iron, and analyzing the samples to better understand the physics and scaling of instability growth in materials with strength and to investigate microstructural evolution during the stages of instability growth. In summary, we used the Janus laser to shock rippled samples of tantalum and iron and successfully recovered and analyzed the samples to improve our understanding of the physics and scaling of instability growth in such materials, as well as determine microstructural evolution during instability growth. Results indicate that future shock-recovery experiments on higher-energy laser platforms such as NIF and the OMEGA laser at the University of Rochester are likely to be successful.

Publications

Stewart, S. T., 2012. *Recovering large volumes of homogeneously shocked samples*. LLNL-TR-608816.

Imaging of Scattered X-Ray Radiation for Density Measurements in Hydrodynamics Experiments at the National Ignition Facility

Donald Correll (12-FS-004)

Abstract

The National Ignition Facility (NIF) has the capability to create complicated hydrodynamics experiments relevant to astrophysical objects and turbulent systems such as plasmas. However, the limitations of x-ray radiography, which is traditionally used for hydrodynamics experiments, underscore the need for a more sophisticated diagnostic technique. We propose to develop a new diagnostic technique—imaging x-ray scattering (IXRS)—in which intense x rays are used to probe a dense plasma by imaging the x rays that scatter from

that plasma. The goal is to obtain images in which intensity is nearly proportional to total electron density, thereby visualizing variations in material density. This technique can be used to study experiments on complex hydrodynamics such as radiation hydrodynamics, astrophysical turbulence, and complex, spherically divergent hydrodynamics.

We will develop a novel imaging x-ray technique to diagnose hydrodynamics experiments at NIF. This includes hydrodynamic instability experiments, such as those investigating multiple coupled interfaces in divergent geometries or planar radiation hydrodynamics. In such experiments, the proposed IXRS technique could detect variations in densities and thereby reveal information about the structure resulting from instability growth. For instance, in an experiment with instability growth at multiple interfaces in a divergent geometry, IXRS will enable researchers to diagnose the structure at different interfaces by probing with various x-ray energies.

Mission Relevance

By developing a technique that will enhance the science obtained in high-energy-density experiments at NIF, our research into IXRS supports multiple mission areas at Lawrence Livermore, including stockpile stewardship, laser inertial fusion energy, and advanced laser optical systems.

FY12 Accomplishments and Results

Colleagues from University of Michigan under the direction of Carolyn Kuranz explored x-ray scattering as a superior alternative to transmission radiography for supernova hydrodynamics experiments conducted on NIF. In particular, fluorescence imaging was considered, and we studied an approach based on a uniformly doped single-atomic layer on a NIF target. A sheet of x rays would pump this layer, doped with a fluorescent tracer. The fluorescent intensity could then be measured to create a density map of the doped material as it mixed with other layers. This research has confirmed that developing x-ray scattering with fluorescence imaging diagnostic capabilities would create a powerful tool to characterize hydrodynamic experiments with complex geometries.

Publications

Juranz, C. C., 2012. *Imaging scattered x-ray radiation for density measurements in hydrodynamics experiments on NIF*. LLNL-TR-619598.

Concept Development for Astrophysically Relevant Turbulence at the National Ignition Facility

Donald Correll (12-FS-005)

Abstract

The Rayleigh–Taylor instability is ubiquitous in fluidlike systems, including the highly compressible systems found in astrophysics. In such environments, and elsewhere in

nature, the phenomena of interest very often involve the evolution of Rayleigh–Taylor from complex, multimode initial conditions into a deep nonlinear state. Examples include supernovae, the evolution of supernova remnants, and the interaction of astrophysical shocks with extended molecular clouds. Our goal in this project is to produce, diagnose, and simulate Rayleigh–Taylor instability structures in which merger occurs in a highly compressible system, by observing structures over a sufficiently long time and with spatial resolution that spans more than two orders of magnitude. This will enable us to use simulations to develop, test, and apply the behavior of these laboratory-created structures to astrophysics sub-grid-scale models for highly compressible fluid turbulence.

The proposed project addresses the development of structures produced by Rayleigh–Taylor instability during the phase when these structures interact, merge, or otherwise produce turbulence across a wide and changing range of spatial scales. Beyond its intrinsic scientific interest, this work is relevant to numerous astrophysical systems. We will use the National Ignition Facility (NIF) to produce a system that will have far more Rayleigh–Taylor growth—sufficient to produce merger events—than previously was possible, and to diagnose this system by making local measurements that span more than two orders of magnitude in spatial scale, which is not possible at any other existing facility. Moreover, these experiments will be in the highly compressible regime most relevant to astrophysics.

Mission Relevance

This project is fully aligned with the Laboratory's core competency in high-energy-density science. High-energy-density experiments at NIF promise to revolutionize our understanding of astrophysics and space physics, hydrodynamics, nuclear astrophysics, material properties, plasma physics, nonlinear optical physics, radiation sources and radiative properties, and other mission-relevant areas of science.

FY12 Accomplishments and Results

Colleagues at the University of Michigan, led by R. Paul Drake, developed the preliminary design of a NIF experiment to investigate the evolution of multimode Rayleigh–Taylor instability by directly measuring two-dimensional bubble front evolution in the hydrodynamic regime. Rayleigh–Taylor instability growth with the proposed design was then analyzed with one-dimensional direct numerical simulations in the HYADES plasma hydrodynamics code and with a comparable behavior model, verifying that the proposed design assures generation of three to four bubble mergers, bringing the bubble front to a self-similar stage, and that the design takes advantage of NIF capabilities to provide a sufficiently large laser spot area along with a sufficiently low drive to keep preheat effects small. In summary, this project developed the preliminary design of an experiment to investigate the evolution of multimode Rayleigh–Taylor instability on NIF, taking advantage of the platform's capabilities to provide a sufficiently large laser spot area along with a low drive.

Publications

Drake, R. P., 2012. *Concept development for astrophysically relevant turbulence on NIF*. LLNL-TR-608953.

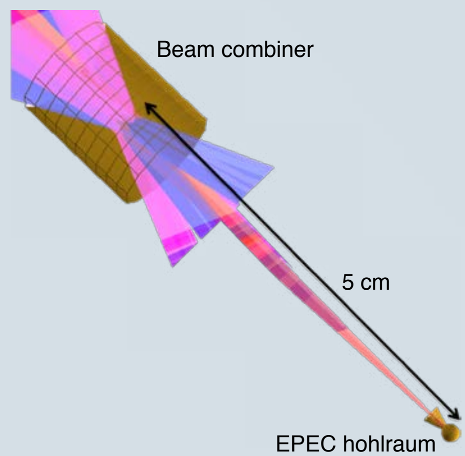
High-Energy Beams for Simulating High-Yield Nuclear Events

Robert Kirkwood (12-FS-012)

Abstract

We propose to design and test a laser beam combiner to assess the feasibility of increasing the available energy range for planned nuclear event simulations at the National Ignition Facility (NIF) by a factor of three to four and allow events as large as 10 kT to be simulated. Ongoing efforts of the Energy-Partitioning Energy-Coupling (EPEC) program at NIF are limited by current capability and our new technical approach could dramatically improve on this. The technique we intend to investigate will combine the laser beams prior to best focus via scattering from stimulated ion waves in a plasma, which is an adaptation of the ion-wave beam process that is presently being used for NIF ignition target experiments and has been demonstrated to have high reproducibility and controllability.

The EPEC program is presently carrying out NIF experiments that use a single quad of beams with 10 kJ of energy to simulate a number of important issues representing a nuclear device 2.5 kT in size. We intend to design a target that will combine multiple quads of NIF beams into a single beam, which will allow simulation of events up to 10 kT, making the capability applicable to a much broader set of problems. A larger energy range will enable an expanded data suite for more accurate weapon physics codes. The increased energy simulation is intended to provide valuable information relevant to burst height, cavity de-coupling, optical signals, electromagnetic pulse, and chemical fractionation for high-yield nuclear events—all important issues in the study of weapons physics and relevant to shelter-in-place or evacuation measures in the aftermath of a nuclear explosion.



A plasma-wave beam combiner is being developed to combine the power and energy of six National Ignition Facility beam quads into a single beam, using the stimulated ion-wave scattering process developed for the laser-ignition hohlraum target capsules. The technology is expected to enable a range of high-energy-density experiments benefiting from increased beam energy and power, including those in the Energy-Partitioning Energy-Coupling program (EPEC) at the National Ignition Facility.

Mission Relevance

The proposed research would allow NIF weapon simulations to address a wider range of potential situations than possible with the EPEC program as currently envisioned, which directly supports the Laboratory's core mission thrust in national security and stockpile stewardship science.

FY12 Accomplishments and Results

In FY12 we (1) optimized the plan for a target that can combine six quads of beams into a single beam and performed initial target design calculations; (2) illuminated, with the six beams and an additional ten heater beams, a gas-filled tube with windows in a Hydra radiation-hydrodynamics code simulation; and (3) analyzed the efficiency of beam transmission through the entire plasma profile and the gain for the energy transfer process. The target design produced shows that absorption can be kept to 30% with only 10% in the plasma column where the beam amplification occurs, and 20% in the wings of the profile. The peak stimulated Brillouin scattering gain is also sufficient for power transfer under the simulated conditions.

Proposed Work for FY13

In the previous year we demonstrated the feasibility of producing high-transmission plasmas that can combine five or more quads at NIF, and produced a target design adequate for testing. In FY13 we propose to optimize heater pointing to produce a uniform profile and further minimize absorption in the wings. When complete, our target design will be testable on NIF as part of the present EPEC program, with the results used for proposing additional high-energy EPEC experiments to the Office of Nonproliferation Research and Development of the NNSA, as well as to other interested agencies.

Publications

Kirkwood, R. K., et al., 2012. "Increasing beam power and energy with the SBS forward energy transfer instability." *Bull. Am. Phys. Soc.* **57**(12), CP800089. LLNL-PRES-595873.

Deep Penetration in Aerospace Composite Materials Using Near-Infrared Laser Radiation

Sheldon Wu (12-FS-014)

Abstract

We propose to investigate the feasibility of laser-weapon lethality against composite materials through deep penetration of near-infrared-wavelength radiation. Laser–composite material interactions to date have focused on 3.8- and 10.6-µm-wavelength radiation, which is strongly absorbed at the surface, forming a carbonaceous char layer that thermally insulates underlying material and limits further damage. We suggest that in composite materials, near-infrared laser radiation may be absorbed more deeply, and thus have enhanced lethality over longer-wavelength lasers. We propose to determine the feasibility of using near-infrared-wavelength laser radiation to deeply penetrate composite materials to enable more lethal effect against missile radar housings and other composite aerospace components. We will also test the use of laser radiation produced by systems like the diode-pumped alkali laser currently being developed by LLNL for missile defense.

If successful, we will characterize laser–composite material interactions with 0.8-µm-wavelength laser light and demonstrate the feasibility of laser lethality against composite materials through deep penetration of near-infrared-wavelength radiation. We also expect to develop a technique for accurate calibration of infrared cameras for remote temperature measurement, and a numerical model for propagation of laser light in composite materials that relates linear absorption of fibers and epoxy resin to composite absorption. Such a model is needed to accurately characterize absorption in weakly absorbing composites where the scattering length can significantly exceed the absorption length.

Mission Relevance

Successful demonstration of deep penetration of near-infrared-wavelength radiation in composite materials will contribute to maintaining LLNL's technical vitality in the field of advanced laser optical systems and multiphysics simulation. This study in the field of directed energy weapons continues the Laboratory's long history of collaboration with the Department of Defense and other government agencies to provide research and development support to meet emerging national security needs.

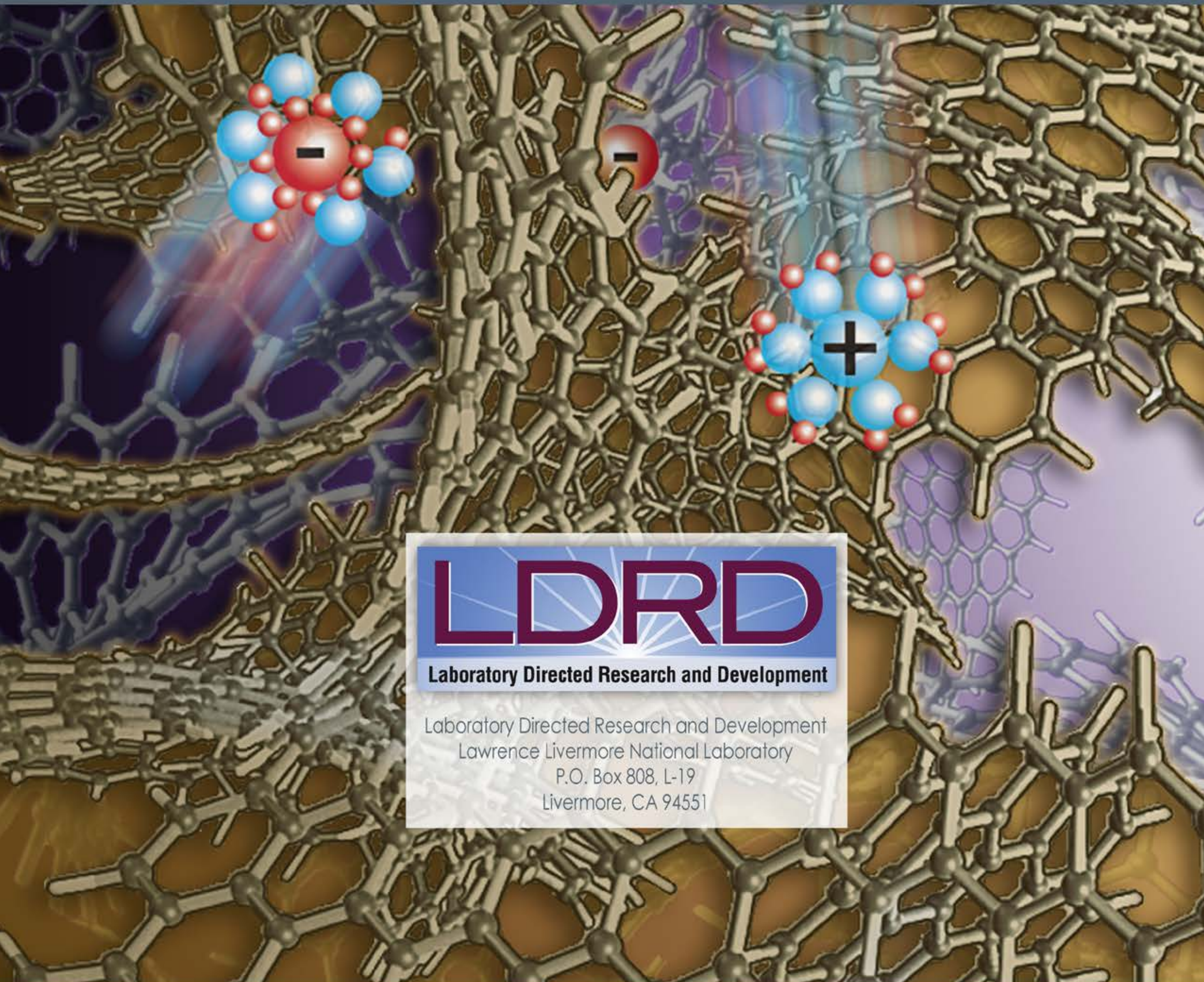
FY12 Accomplishments and Results

In FY12 we (1) performed measurements to characterize fiber-reinforced composite materials under laser illumination at 0.8-μm wavelength; (2) measured temperature at varying depths in the samples and developed and demonstrated a method to provide accurate infrared camera calibration for remote sensing; (3) developed a detailed ray-trace model to calculate absorptivity, reflectivity, and localized field enhancement from internal scattering; and (4) determined that volume heating can induce failure of aerospace structural composite materials under compressive load within several seconds of irradiation by a near-infrared laser at intensities much lower than that required to produce lethal effect by target penetration or material removal. The successful conclusion of this study showed that volume heating either by direct in-depth absorption of laser energy (e.g., near-infrared laser irradiation of translucent fiberglass composite) or by rapid conduction of surface-deposited energy (e.g., as in opaque carbon composite) can induce structural failure under load in seconds by softening the resin matrix at irradiance less than 100 W/cm^2. Composite material failure by any mechanism at such low flux has not been previously reported and has significant laser lethality and vulnerability implications. Our findings have generated interest from external entities and will lead to continued efforts in the field of laser–material interaction of fiber composites under load.

Publications

Wu, S. S., et al., 2012. *Near-IR laser interaction with composite materials*. 15th Ann. Directed Energy Symp., Albuquerque, NM, Nov. 26–30, 2012. LLNL-PRES-606013.

Wu, S. S. Q., et al., 2012. *Interaction of composite materials with 0.8-micron-wavelength light*. 15th Ann. Directed Energy Symp., Albuquerque, NM, Nov. 26–30, 2012. LLNL-ABS-570113.



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