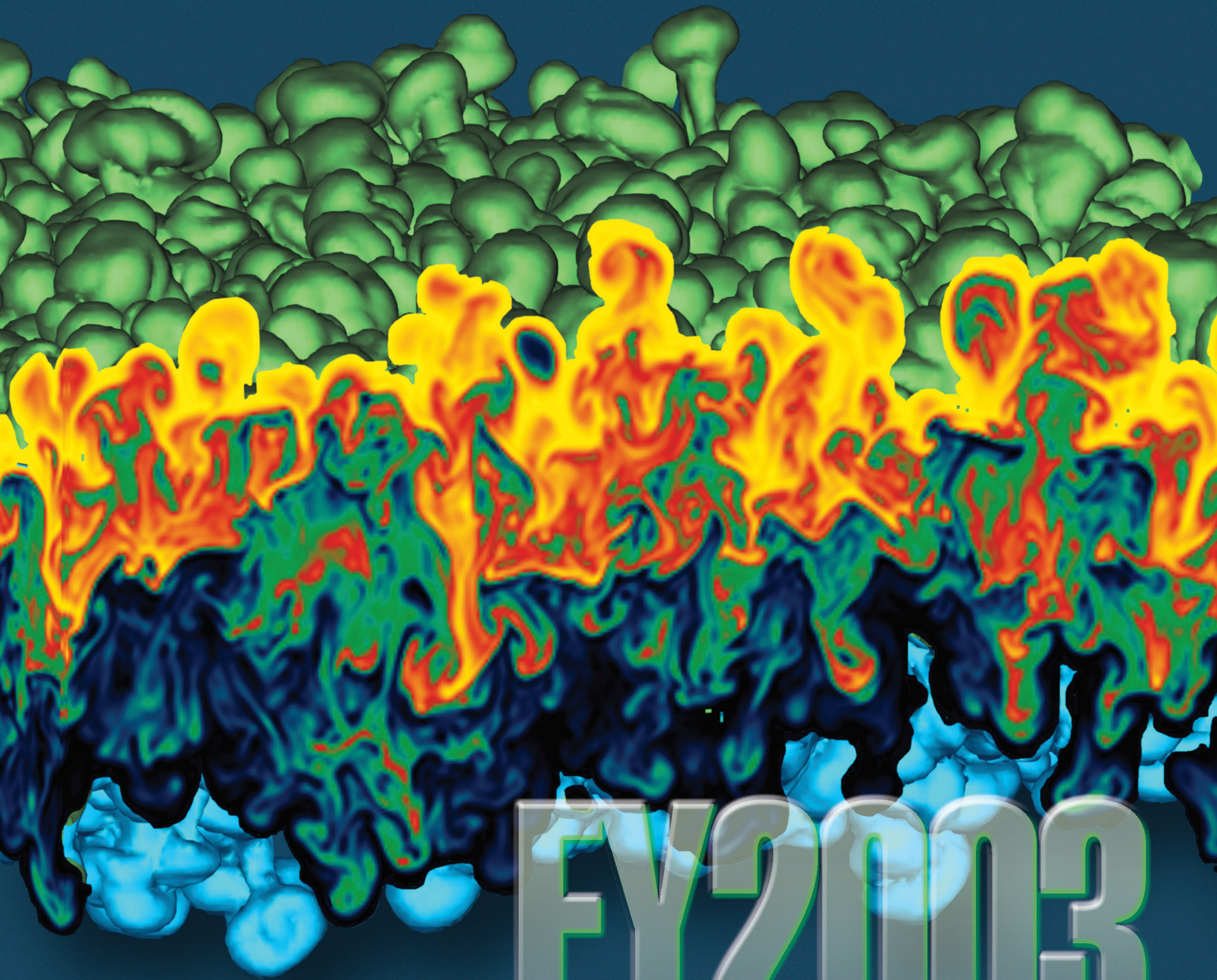
UCRL-TR-113717-03

LDRD

LABORATORY DIRECTED RESEARCH AND DEVELOPMENT



FY2003 ANNUAL REPORT

LAWRENCE LIVERMORE NATIONAL LABORATORY

Disclaimer

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Acknowledgments

This Annual Report provides an overview of the FY2003 Laboratory Directed Research and Development (LDRD) Program at Lawrence Livermore National Laboratory (LLNL) and presents a summary of the results achieved by each LDRD project. At LLNL, Laboratory Director Michael Anastasio and Deputy Director for Science and Technology Harold Graboske are responsible for the LDRD Program and delegate responsibility for the operation of the Program to the Associate Deputy Director for Science and Technology and the Director of the Laboratory Science and Technology Office (LSTO), Rokaya Al-Ayat. The LDRD Program at LLNL is in compliance with Department of Energy (DOE) Order 413.2 and other relevant DOE orders and guidelines.

The LDRD Program extends its sincere appreciation to the principal investigators of the FY2003 projects for providing the content of the Annual Report and to the publications team. A special thanks goes to Adam Schwartz for his generous assistance. The Program also thanks the following members of the LSTO team for their many contributions to this publication: Mary Callesen, administrator; Nancy Campos, database manager; Andrew Hurst, computer specialist; and Cathleen Sayre, resource manager.

Scientific Editors
Kenneth Jackson, Karl van Bibber

Publication Editors
Karen Kline, Paul Kotta

Publication Designer
Daniel Moore

Compositor
Louisa Cardoza

Michael R. Anastasio, Director

Director's Statement

The Laboratory Directed Research and Development (LDRD) Program, authorized by Congress in 1991 and administered by the Laboratory Science and Technology Office, is our primary means for pursuing innovative, long-term, high-risk, and potentially high-payoff research that supports the Laboratory's and the Department of Energy and National Nuclear Security Administration's missions in national security, energy security, environmental management, bioscience and technology to improve human health, and breakthroughs in fundamental science and technology. The accomplishments described in this Annual Report demonstrate the strong alignment of the LDRD portfolio with these missions and contribute to the Laboratory's success in meeting our goals.

The LDRD budget of $64.9 million for Fiscal Year 2003 sponsored over 200 projects. These projects were selected through an extensive peer-review process to ensure the highest scientific quality and mission relevance. Each year, the number of deserving proposals far exceeds the funding available, making the selection a tough one indeed.

Our ongoing investments in LDRD have reaped long-term rewards for the Laboratory and the Nation. Many Laboratory programs trace their roots to research thrusts that began several years ago under LDRD sponsorship. In addition, many LDRD projects contribute to more than one mission area, leveraging the Laboratory's multidisciplinary team approach to science and technology. Recent events underscore the importance of LDRD investments to national security. Safeguarding the Nation from terrorist activity and the proliferation of weapons of mass destruction will be an enduring mission of this Laboratory, for which LDRD will continue to play a vital role.

The LDRD Program is a success story. Our projects continue to win national recognition for excellence through prestigious awards, papers published in peer-reviewed journals, and patents granted. With its reputation for sponsoring innovative projects, the LDRD Program is also a major vehicle for attracting and retaining the best and the brightest technical staff and for establishing collaborations with universities, industry, and other scientific and research institutions.

By keeping the Laboratory at the forefront of science and technology, the LDRD Program enables us to meet our mission challenges, especially those of our ever-evolving national-security mission.

UCRL-LR-113717-03

LDRD

LABORATORY DIRECTED RESEARCH AND DEVELOPMENT

FY2003

ANNUAL REPORT

LAWRENCE LIVERMORE NATIONAL LABORATORY

LDRD FY2003 Table of Contents

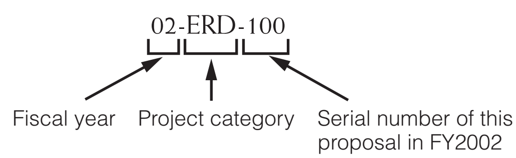
About the FY2003 Laboratory Directed Research and Development Annual Report

The *FY2003 Laboratory Directed Research and Development (LDRD) Annual Report* provides a summary of LDRD-funded projects for the fiscal year and consists of fourparts:

Overview: An introduction to the LDRD Program, the LDRD portfolio-management process, Program statistics for the year, and articles highlighting certain LDRD projects.

Project Summaries: An overview 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 FY2003, and a list of publications that resulted from the research in FY2003. [CD only]

Summaries are organized in sections by research category (in alphabetical order). Within each research category, the projects are listed in order of their LDRD project category: Strategic Initiative (SI), Exploratory Research (ER), Laboratory-Wide Competition (LW), and Feasibility Study (FS). Within each project category, the individual project summaries appear in order of their project tracking code, a unique identifier that consists of three elements. The first element is the fiscal year the project began, the second element represents the project category, and the third element identifies the serial number of the proposal for that fiscal year. For example:



Program Overview

Investing in our nation's future

About Lawrence Livermore National Laboratory

A premier applied-science laboratory, Lawrence Livermore National Laboratory (LLNL or the Laboratory) has at its core a primary national security mission—to ensure the safety, security, and reliability of the nation's nuclear weapons stockpile without nuclear testing, and to prevent and counter the spread and use of weapons of mass destruction: nuclear, chemical, and biological.

The Laboratory uses the scientific and engineering expertise and facilities developed for its primary mission to pursue advanced technologies to meet other important national security needs—homeland defense, military operations, and missile defense, for example—that evolve in response to emerging threats. For broader national needs, the Laboratory executes programs in energy security and long-term energy needs, environmental assessment and management, bioscience and technology to improve human health, and breakthroughs in fundamental science and technology. With this multidisciplinary expertise, the Laboratory serves as a science and technology resource to the U.S. government and as a partner with industry and academia.

One of three Department of Energy (DOE) National Nuclear Security Administration (NNSA) laboratories, LLNL has been managed since its inception in 1952 by the University of California (UC). This half-century association with UC has enabled the Laboratory to establish an atmosphere of intellectual freedom and innovation that attracts and maintains the world-class workforce needed to meet challenging national missions.

Laboratory Directed Research and Development Program

To fulfill its missions, LLNL must continually invest in areas that further develop and advance the skills and capabilities needed to fulfill those missions. The Laboratory Directed Research and Development (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 allow 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 LDRD Portfolio Management Process

The FY2003 LDRD portfolio-management process at LLNL had three major components that ensured the quality of the year's portfolio and its alignment with the DOE/NNSA and the Laboratory's missions: (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 FY2003, the top-level LDRD strategic planning process was guided by a newly issued DOE *Strategic Plan*[2] for the next 25 years, and by a long-range planning process at the Laboratory, begun this year, that will define the scientific and technical strategy for the Laboratory in the coming decade.

The 2003 DOE *Strategic Plan* articulates four strategic goals towards achieving the DOE mission of advancing the national, economic, and energy security of the U. S.; promoting scientific and technological innovation in support of that mission; and ensuring the environmental cleanup of the national nuclear weapons complex. In FY2003, the LLNL LDRD Program strongly supported all four DOE strategic goals:

1. **Defense Strategic Goal—**To protect our national security by applying advanced science and nuclear technology to the Nation's defense.
2. **Energy Strategic Goal—**To protect our national and economic security by promoting a diverse supply and delivery of reliable, affordable, and environmentally sound energy.
3. **Science Strategic Goal—**To protect our national and economic security by providing world-class scientific research capacity and advancing scientific knowledge.
4. **Environment Strategic Goal—**To protect the environment by providing a responsible resolution to the environmental legacy of the Cold War and by providing for the permanent disposal of the Nation's high-level radioactive waste.

[1] U. S. Department of Energy. Order O 413.2A. *Laboratory Directed Research and Development*. (January 8, 2001).

[2] U. S. Department of Energy. (2003). *Strategic plan*. http://strategicplan.doe.gov/ (retrieved March 5, 2004).

In FY2003, the Laboratory's first long-range strategic science and technology planning process began to inform the LDRD portfolio planning process in FY2003. Broadly inclusive, the process involved management and technical staff working in teams to elicit the most far-reaching and innovative ideas for the future shape of science and technology at LLNL. Six thematic areas emerged. These six areas are:

- Stockpile science and technology
- High-energy-density science and technology
- Nuclear, radiative, and astrophysical science and technology
- Science and technology at the intersection of chemistry, biology, and materials science and technology
- Information, simulations, and systems science and technology
- Energy and environmental science and technology (with fusion energy science and technology as a special subtopic)

The first published version of the plan is expected in FY2004.

The NNSA oversees LLNL's LDRD Program to ensure that it accomplishes its objectives. This oversight includes field and headquarters reviews of both technical content and management processes. As demonstrated in a memorandum (April 30, 2002) from the Secretary of Energy and the NNSA Administrator, the DOE/NNSA actively support the LDRD Program. In the memo, Secretary of Energy Spencer Abraham writes: "I believe that all sponsors, including other Federal agencies, benefit from the strong science and technology base provided by the Department's LDRD/PDRD programs. LDRD/PDRD is at the core of our ability to develop research capabilities and apply advanced technologies to effectively meet the Department's and the National's needs."[3]

Structure of the LDRD Program

Project Categories

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

Strategic Initiative

The SI category focuses on innovative R&D activities that are likely to set new directions for existing programs, help develop new programmatic areas within LLNL's mission responsibilities, or enhance the Laboratory's science and technology base. Projects in this category are usually larger and more technically challenging than projects funded in other categories. An SI project must be aligned with the strategic R&D priorities of at least one of the Laboratory's six thematic science and technology planning areas developed in FY2003.

Exploratory Research

The ER category is designed to help fulfill the strategic R&D needs of a Laboratory Directorate (ERD) or Institute (ERI). In this category, researchers submit their proposals to their Directorates

[3] Memorandum from Secretary of Energy Spencer Abraham. 2002-0077386. (Washington, DC, April 30, 2002).

and Institutes, which conduct their own screening process before forwarding their LDRD proposal portfolio to the ER selection committee for review. In FY03, the emerging science and technology plan also began to guide directorates in evaluating the ERD and ERI proposals.

Laboratory-Wide Competition

Projects in the LW category emphasize innovative research concepts and ideas and undergo limited management filtering. The LW competition is open to all LLNL staff in programmatic, scientific, engineering, and technical-support areas. Researchers submit their project proposals directly to the LW selection committee.

Feasibility Study/Project Definition

This special project category, FS, provides researchers with the flexibility to define and develop potential projects in the other three categories. To increase its responsiveness to Laboratory scientists and engineers, the LDRD Program funds FSs throughout the year.

Project Competency Areas

Although LDRD projects often address more than one scientific discipline, each project is classified into one of ten research categories that are 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

Window on Plutonium

Plutonium phonon dispersion curves bring LDRD researchers face to face with electrons on the edge

For many, the word "plutonium" evokes the danger, mystery, and power of nuclear energy and nuclear weapons. Plutonium (Pu), element 94 on the periodic table and a member of a series of 14 radioactive elements called the actinides, has earned this reputation due to the extraordinary nuclear properties and radiotoxicity of one of its isotopes—^{239}Pu. When it undergoes fission with thermal neutrons, the ^{239}Pu nucleus releases enormous energy. One kilogram is equivalent to about 22 million kilowatt hours of heat energy and, when detonated, produces an explosion equal to about 20,000 tons of chemical explosive. Plutonium also emits alpha particles, making it a radiologic poison.

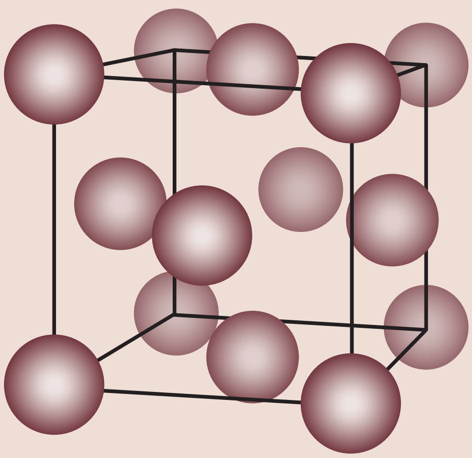


Figure 1. The crystallographic unit cell of face-centered-cubic delta-Pu. The structure consists of a cubic lattice, with Pu atoms at each corner and at the center of each face of the cubic lattice.

Yet, this powerful and dangerous element, so important for the Laboratory's national security mission, is also the most complex and perplexing in the periodic table, offering scientific challenges that have yet to be mastered. Though much has been discovered about it, Pu still guards many of its intriguing scientific secrets. Unlocking these secrets will provide valuable scientific insight to support the Laboratory missions; help develop methods for the safe handling, use, and long-term storage of Pu; and remediate environmental contamination.

Because of Pu's enigmatic behavior and the need for stringent safety and environmental procedures when handling the toxic radioactive material, much of the extensive characterization work done on other metals has not been performed on plutonium.

But in FY03, the LDRD project, *Determining Phonon-Dispersion Curves in Delta-Phase Plutonium–Gallium Alloys* (03-ERD-017), one of several LDRD-supported Laboratory research projects focusing on plutonium science, achieved a landmark experimental breakthrough in understanding this complex element. By mapping Pu phonon dispersion curves (PDCs) for the first time, principal investigator Joe Wong and his team have provided a window into Pu's elementary transitions. According to Wong, "Phonon dispersions are fundamental to understanding the properties of plutonium."

Phonons, the quanta of vibrational energy in the lattice structure of a solid,—are produced by the movement of the atoms that make up the lattice (Figure 1). The variations in these vibrations, as they disperse along different crystallographic directions of the solid, constitute PDCs, which provide fundamental information about the elastic and thermodynamic properties of the solid, including force constants, sound velocities, elastic constants, heat capacities, phase stability, electron–phonon coupling, and structural relaxation. Until Wong's pioneering experimental study, PDCs, well known for almost all other elements and many compounds, have remained unknown territory for Pu, defying both theoretical and experimental mapping attempts.

The Peculiarities of Plutonium

As Wong explains, "Although Pu's importance comes from the power of its nucleus, the element's complexity stems from the enigmatic properties of its electrons." These peculiar properties arise from Pu's position toward the middle of the actinide row in the periodic table (Figure 2).

Beginning with thorium (Th) and moving across the actinide row from left to right, the 5*f* electron shell is progressively filled. At the beginning of the row, the 5*f* electrons are chemically active, contributing to the bonding between atoms, much like the electrons in the transition metals. But starting with americium (Am), the 5*f* electrons are localized and chemically inert, more like the rare earth elements. Plutonium lies at the transition between the two types of 5*f* electron states, not belonging to either group (Figure 3). As a result of the unique behavior of its 5*f* electons, Pu possesses unusual properties and, until recently, resisted both experimental determination and theoretical computations using first-principles methods.

Pure Pu metal goes through six solid-state phase transformations—more than any other element—with large volume and density changes in its transition from the room-temperature

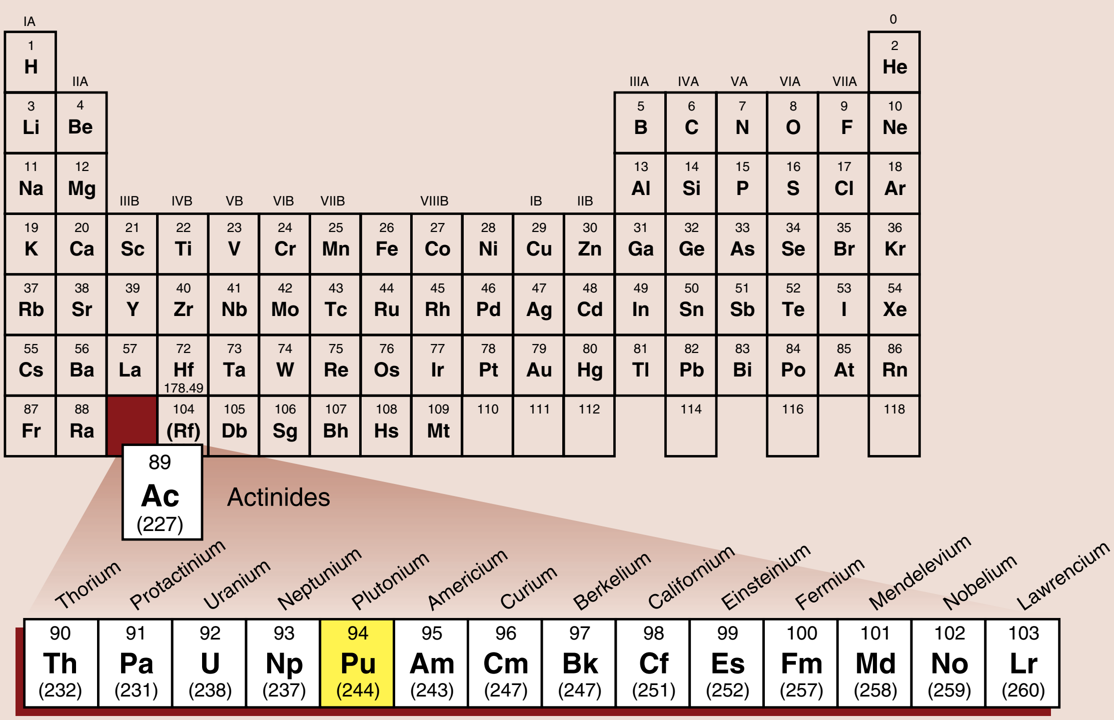


Figure 2. Plutonium (element 94) belongs to a group of elements—the actinides—in the periodic table. The group begins with actinium (Ac, element 89) and ends with lawrencium (La, element 103). All actinides are metals and all are radioactive, meaning they emit energy in the form of alpha particles, beta particles, or gamma rays. Plutonium, a silver-gray metal, has 16 isotopes. Most nuclear weapons are based on ^{239}Pu, which is fissionable, while ^{238}Pu is used as a power source in space probes.

alpha phase to the liquid state: alpha→beta→gamma→delta→delta′→epsilon→liquid. In addition, Pu melts at a relatively low temperature, about 640˚C, resulting in a higher-density liquid than the solid it came from (Figure 4).

Wong's team focused on the highly symmetric face-centered-cubic (fcc) delta phase of Pu, which has desirable mechanical properties like ductility and machinability, but is unstable at room temperature, i.e., it undergoes spontaneous fission. Although alpha-phase Pu is thermodynamically stable at room temperature, it is brittle and difficult to machine. But by alloying delta-Pu with small amounts of gallium (Ga), Pu can be stabilized at room temperature and below, an advantage for engineering.

Also, delta-Pu–Ga alloys transform directly to the alpha phase. Mapping the PDCs of the delta Pu–Ga alloy and alpha-Pu will reveal the fundamental nature of this delta-to-alpha transformation and its consequences. This information is key to understanding the structural and physico-chemical properties of Pu and its alloys, particularly phase stability and material dynamics, which are important for laboratory missions.

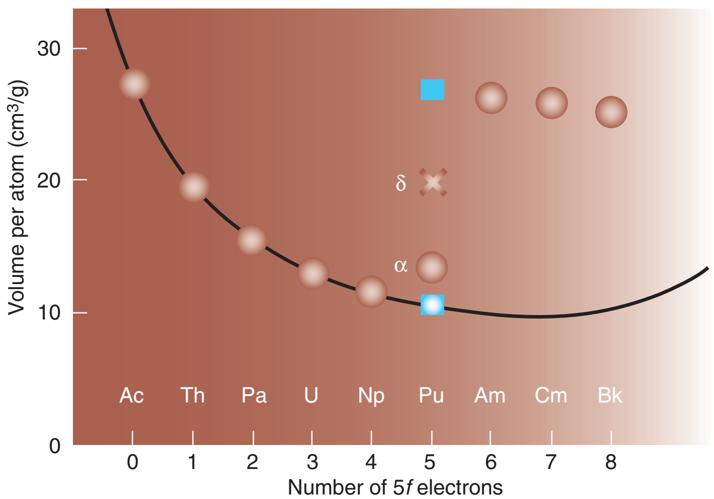


Figure 3. In this plot of atomic volume as a function of *5f* electrons, Pu is "on the edge," forming a transition in the behavior of *5f* electrons between the lighter actinides that precede it and the heavier actinides that follow. The plot shows the alpha (α) and delta (δ) phases of Pu. The squares indicate where Pu would be expected on the plot if it behaved like the other actinide elements.

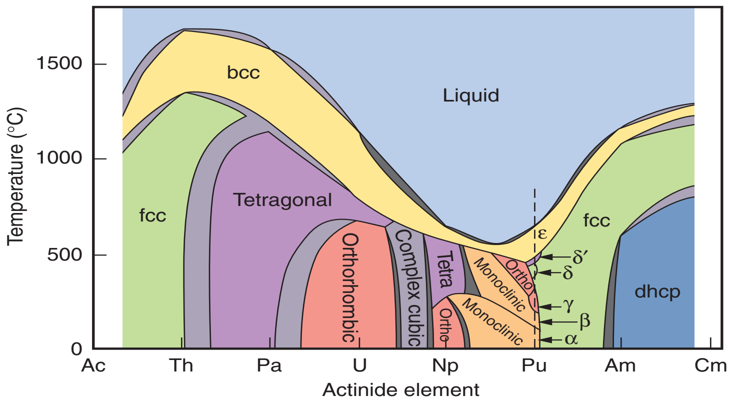


Figure 4. This temperature vs. composition diagram of the actinides demonstrates the transition in crystal structure from typical metallic structure at thorium (Th), to enormous complexity at Pu, and back to typical metallic structure at americium (Am). The dotted line indicates the six solid-state phase transitions of Pu: alpha (α) → beta (β) → gamma (γ) → delta (δ) → delta′ (δ′) → epsilon (ε).

Finding the Experimental Key

One of the major challenges of measuring Pu PDCs has been identifying the right experimental technique. Inelastic neutron scattering (INS) is the leading experimental technique for measuring PDCs in solids. In this technique, a neutron beam impinges on a single crystal of a specimen material. The energy of the neutrons scattered from the crystal is measured to determine the energy gained and lost by phonon creation and annihilation. But INS will not work for plutonium because of ^{239}Pu's high thermal-neutron absorption cross section and because INS requires relatively large single crystals—at least 100 mm^3—an impossibly large size for unalloyed Pu crystals.

With the advent of highly brilliant x-ray sources from third-generation synchrotron facilities and high-performance focusing optics that achieve milli-electron-volt resolution, another technique, high-resolution inelastic x-ray scattering (HRIXS), has recently become available. By using x rays rather than neutrons, HRIXS avoids the experimental problems of INS and makes it possible to study samples, like Pu, that have volumes as small as 10^{-4} mm^3.

Wong and his team fabricated large-grain polycrystalline samples of Pu–Ga alloy containing ~0.6 wt.% Ga for their experiments. In collaboration with scientists at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France, they designed scattering geometries for making PDC measurements with a HRIXS beamline at ESRF.

There, Wong and collaborators directed the 21-KeV beam onto single-crystal grains of fcc delta-Pu in the Pu–Ga alloy samples and measured the phonon energy along the three principal crystallographic directions: [001], [011], and [111]. Phonon energy is measured as a function of the scattering angle, which determines the wave vector of phonon propagation: either longitudinal (in the same direction as the motion of the atoms in the lattice) or transverse (at a right angle to the motion of the atoms).

These first-ever Pu PDCs confirmed the results of ultrasound experiments conducted almost 30 years ago, which indicated that the elastic properties of Pu are strongly direction dependent (anisotropic), more than any other fcc metal. Wong's results also showed good qualitative agreement with recent calculations of delta-Pu phonon spectra at arbitrary wavelengths. The calculations were based on the dynamical mean field theory (DMFT) of the correlation effects among electrons, a new first-principles theoretical approach that accounts for the behavior of the important 5f electrons. However, some quantitative differences between DMFT calculations and Wong's experiments point to the need for refining the theory and conducting further experiments that will, in turn, provide test data for the calculations (Figure 5).

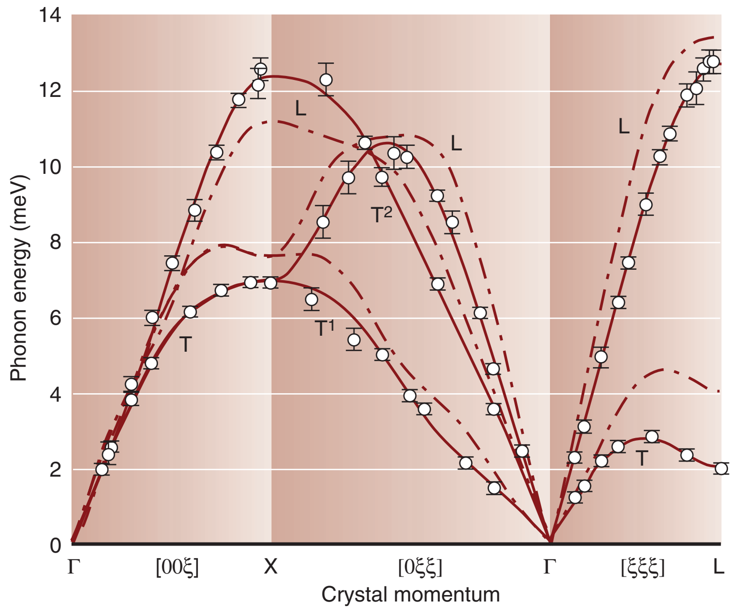


Figure 5. Phonon dispersions along-high-symmetry crystallographic directions [001, 011, 111] in a delta-Pu–0.6 wt% Ga alloy. Longitudinal and transverse wave vectors are denoted by L and T, respectively. Along the [011] direction, there are two transverse branches, T_1 and T_2. Circles indicate experimental data from this project; the solid lines are the data fit to the Born–von Karman model of lattice dynamics; and dotted lines are calculated dispersions for pure delta-Pu based on dynamical mean field theory (DMFT). The experimental data validate the main qualitative predictions of a recent DMFT calculation for pure delta-Pu, though some quantitative differences exist along T[001] near the X point, along T_1 [011], and T[111]. These differences provide the framework for refined theoretical treatments and further experiments in Pu and other actinides.

The experiments uncovered anomalies in delta-Pu–Ga PDCs that derive from electron interactions and are similar to anomalies found in other actinide systems. These results point to the instabilities in Pu's 5f electron structure and strong electron–phonon coupling as the underlying reason for both the phonon anomalies and the complexities of Pu phase transitions.

In FY04, Wong's team is concentrating on growing high-purity, large-grained delta-Pu–Ga crystals to explore these anomalies in a new series of scattering experiments at ESRF.

By providing important scientific insights into the basic elastic and thermodynamic properties of Pu through mapping its vibrations, Wong and team have blazed a new experimental path toward understanding the most enigmatic of all elements.

—Karen Kline

For more information, see the project summary (on CD only) for project covered in this article:

Determining Phonon-Dispersion Curves in Delta-Phase Plutonium–Gallium Alloys

Joe Wong, Principal Investigator, 03-ERD-017

Related projects on actinide science:

Metastability and Delta-Phase Retention in Plutonium Alloys

Adam J. Schwartz, Principal Investigator, 01-ERD-029

Plutonium and Quantum Criticality

Michael J. Fluss, Principal Investigator, 03-ERD-077

Properties of Actinide Nanostructures

Alex V. Hamza, Principal Investigator, 02-ERD-025

Thermodynamics and Structure of Plutonium Alloys

Patrick G. Allen, Principal Investigator, 01-ERD-030

Bioscience and Biotechnology in the Fight Against Bioterror and Disease

Four LDRD projects are advancing the state of the art in detecting and countering microorganisms and other causes of disease

This article looks at just some of the work that LDRD scientists are doing in the field of bioscience and technology, with a focus on countering harmful bioorganisms and biomolecules—through detection and neutralization—and on high-throughput experimentation and high-resolution imaging to find out what makes them tick in the first place. Part 1 looks at two projects that are producing knowledge and technology to counter disease; Part 2 focuses on two projects seeking to understand how organisms function at a molecular level—knowledge that will feed into more effective means of countering biothreats and treating disease.

Part 1: Countering

Detecting Biothreats at Ultralow Concentrations

For first responders and decision makers to deal effectively with a sudden outbreak of disease—whether spontaneous or due to a bioterrorist attack—they must first know what disease they are dealing with—and quickly. Whether it's identifying a suspicious substance, testing beef for mad cow disease, screening cancer survivors, or detecting botulism in milk, the nation requires detection technologies that are not only fast and accurate but also sensitive enough to detect concentrations too low for existing technologies. For instance, botulinum toxin is fatal at concentrations below the thresholds of current rapid-response detection technologies. In some cases, the target biomolecule can be amplified [e.g., by polymerase chain reaction (PCR)], but techniques such as PCR are time consuming and do not work on proteins, such as toxins and the prions that cause mad cow disease.

In this project, a team headed by principal investigator Chris Hollars is developing a system that is sensitive enough to detect target biomolecules at ultralow concentrations and fast enough to analyze samples quickly, for rapid detection. This approach begins with specially engineered biomarkers—fluorescent molecules attached to an antibody or oligonucleotide (a short fragment of single-strand DNA) that will bind only to the target biomolecule. The biomarkers are mixed into a solution that potentially contains the molecule of interest. Next, ion lasers are used to illuminate the solution as it is flowed through a microfluidics channel; a charge-coupled device (CCD) camera captures images of the solution at a high rate. The images are then analyzed to detect the presence of the molecule. Figure 1 shows the basic concept of the overall detection system.

The binding agents are two distinct antibodies or oligonucleotides chosen for their ability to bind to a specific target. Attached to the binding agents are organic chromophores that act as markers—spectrally distinguishable fluorescent molecules. Two different colors of emission are used, usually in the red and green spectral regions. The two biomarkers are attached to different binding molecules. Therefore, after the biomarkers are mixed with the solution, only the target molecules will have both a red and green marker attached. When numerous enough, these red and green pairs indicate the presence of the target molecule.

Imaging a Microflow

The next challenge is to detect such labeled individual molecules. Previous approaches to single-molecule detection in quantitative measurements involved imaging glass surfaces

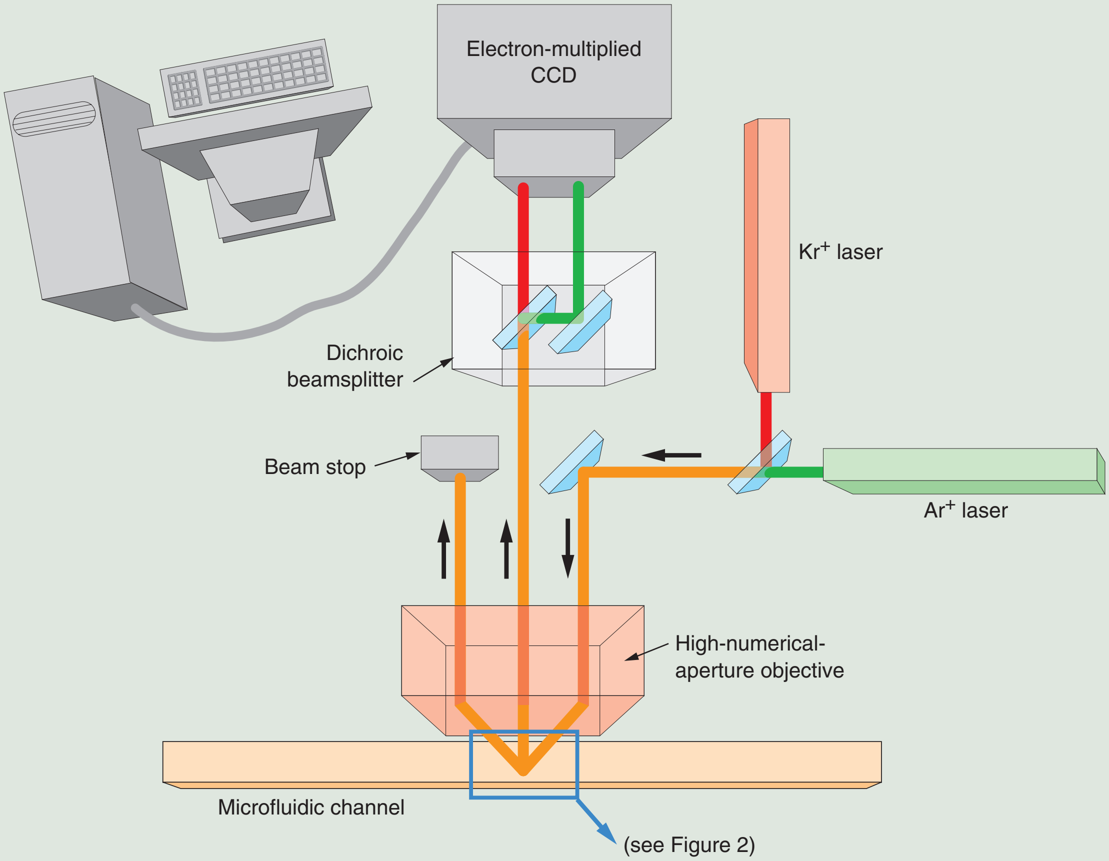


Figure 1. A diagram of a system designed for the rapid detection of biomolecules at ultralow concentrations. Beams from argon ion (Ar^+) and krypton ion (Kr^+) lasers illuminate a microchannel in which a solution containing the probe and target molecules is flowed. A beamsplitter separates the emission (amber) from the solution into two images in the red and green spectral regions, which are directed onto an electron-multiplied charge-coupled device (CCD) camera for detection.

to which probe molecules were attached. However, the performance of this approach was limited by excessive nonspecific binding—i.e., molecules other than the target binding to the surface. Hollars and team instead turned to microchannel devices. They fabricated a channel 100 μm wide and only 0.5 μm deep at its shallowest point—the detection region—where every marker-bound molecule can be imaged as it passes through.

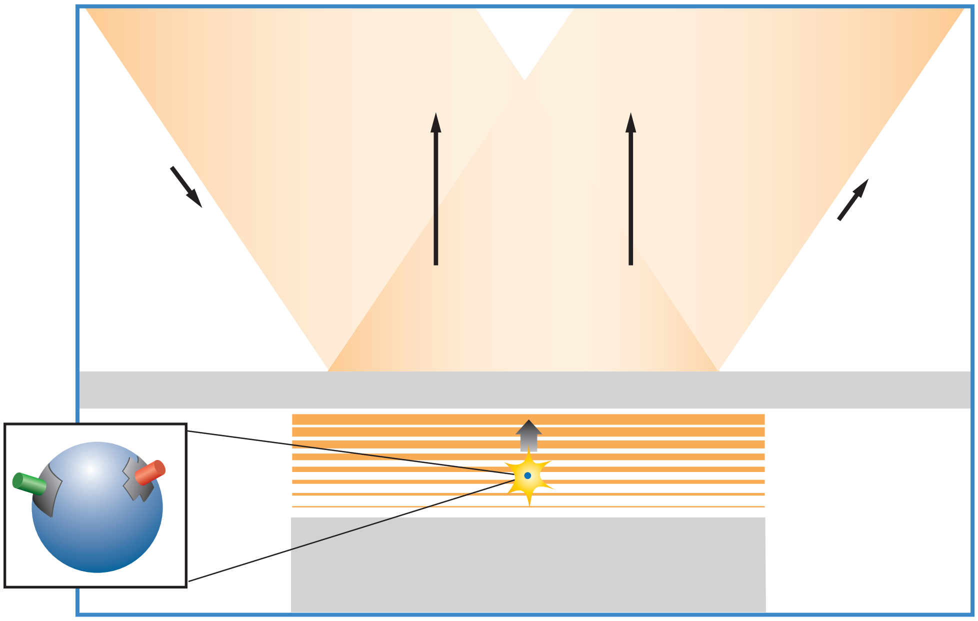


Figure 2. The single-molecule-detection system developed by Hollars and team illuminates a solution using an evanescent field. This occurs when a beam of light strikes a normally transparent surface at a large angle of incidence. Rather than passing through the material, the beam is reflected from the surface and generates a field that extends for a short distance away from the surface. The field causes the biomarkers attached to the molecule of interest (inset) to emit photons, which are imaged by the camera.

The CCD camera's frame rate is adjusted to the flow rate: the approximately 30-ms interval between frames is such that the majority of the molecules are imaged only once. In this manner, the entire volume of solution flowing through the detection region is imaged. In addition, this approach avoids problems with nonspecific binding because the probe and target molecules are not bound to the surface but rather continuously flow through the detection region.

This imaging system is the key to achieving the project's throughput goal—femtomolar-level analysis in less than 15 min. Normally, the large volume of solution—over 5000 times larger than in previous microstream devices—would greatly increase analysis time, but Hollars and team were determined not to sacrifice speed for sensitivity.

Detection is accomplished by first using a filter to split the fluorescence emission from the microchannel into separate red and green images—the same as the biomarker colors—before it enters the CCD camera, which produces a set of two pictures for each frame taken. These pairs of images are compared, and wherever a red biomarker precisely coincides with a green one, a target molecule is presumed to exist. Thus, detection occurs on the level of single molecules.

Utilizing Evanescent Fields

The microchannel is illuminated by means of total internal reflection (TIR), a technique that utilizes an evanescent field (Figure 2). An evanescent field occurs when a beam of light strikes a normally nonreflective surface at an angle greater than the critical angle of incidence. Rather than passing through the material, the light is reflected and generates a field that extends out from the surface for a short distance. In this detection system, the evanescent field illuminates the flow underneath the surface. The field excites the biomarker molecules, which emit photons in all directions; it is these photons that are captured by the CCD camera.

Together, these technologies enable detection on the order of femtomolar (10^{-15} moles/L) limits. This translates to as few as one target molecule in every 300 frames of images. This degree of sensitivity represents a 1000-fold improvement over current mainstream detection technologies, such as radioimmuno assays and enzyme-linked immunosorbent assay, the lower limit for which can be as low as picomolar concentrations.

Although single-molecule fluorescence technology is widely used, this project advanced the state of the art by overcoming various challenges, primarily the use of two colors of fluorescent markers, which requires coordinating the use of two different lasers, and the imaging of flowing single molecules. Comparable techniques involve repeatedly imaging a stationary solution, but this project had to develop the imaging capability to capture images of a flowing solution. This required a camera capable of capturing these images in less than 5 ms. Moreover, the camera must capture images at a frame rate rapid enough for a fast flow rate, which is required for the entire solution to be imaged and assayed in a relatively short amount of time.

Applications Already Under Way

Meanwhile, research that utilizes this technology is already beginning. Several research groups at the UC Davis Medical Center have expressed an interest in using this technology for the early detection of cancer recurrence. Such detection requires a highly sensitive method to analyze blood and urine samples for the low levels of certain messenger RNA fragments and proteins. Fluctuations in these expression levels can indicate early-stage recurrence, well before physical symptoms appear. In addition, the research community has also expressed a strong interest in this technology's potential in high-throughput drug screening.

Hollars and team are also exploring avenues for lowering the detection level even further, possibly into the attomolar (10^{-18} moles/L) range. One approach is pre-concentrating the sample using magnetic beads attached to the biomarkers. Before analysis, the beads would be recovered magnetically. The clumps of beads would then be released into a smaller volume of solution, increasing the chance of detection.

Once the team has demonstrated the feasibility of this detection for laboratory use, a device compact enough for use in the field could be possible with further engineering. Such a device could allow first responders to perform rapid analysis of suspected toxins or other bioterror agents, as well as uses in clinical settings.

Neutralizing Microbes at the Genetic Level

Until recently in humankind's fight against disease, many illnesses have presented only limited opportunities for prevention or treatment. Bacteria can be fought after infection with antibiotics, but preventative treatment is normally not possible. Viral infection can sometimes be prevented with immunizations, but once a person is infected, the only option often available is to treat the symptoms as the virus runs its course. Chemotherapy and other treatments intended to destroy cancer cells often have a devastating effect on the patient. As for other diseases that originate in our own damaged or defective genes, often only the symptoms can be treated. Today, however, state-of-the-art bioscience has pointed to the potential of preventing and treating disease by targeting the genes involved—whether those of a harmful organism or the patient's own. This approach is known as gene silencing and entails turning off specific genes to, say, kill microbes outright, make microbes or cancer cells vulnerable to other drugs, or turn off a defective gene in a patient's own cells.

One gene-silencing technique is ribonucleic acid (RNA) interference, which has been heralded as a breakthrough of such potential that recent surveys show that 75% of the scientific community either uses or plans to use RNA interference for research and clinical purposes. This technique uses a short segment of double-stranded RNA that corresponds to a specific gene; this segment is known as small, interfering RNA (siRNA). A gene is essentially a segment of deoxyribonucleic acid (DNA) consisting of a unique combination of the four nucleotides—adenine, guanine, cytosine, and thymine—that form the instructions to produce—or "express"—a specific protein.

Small, interfering RNA somehow targets messenger RNA—the exact mechanism is not known—to either completely stop or reduce expression. Because it affects only the specific messenger RNA for which it was designed, siRNA does not affect any other genes in the cell. In addition, because it works on RNA, siRNA that targets disease-causing viruses should also be possible.

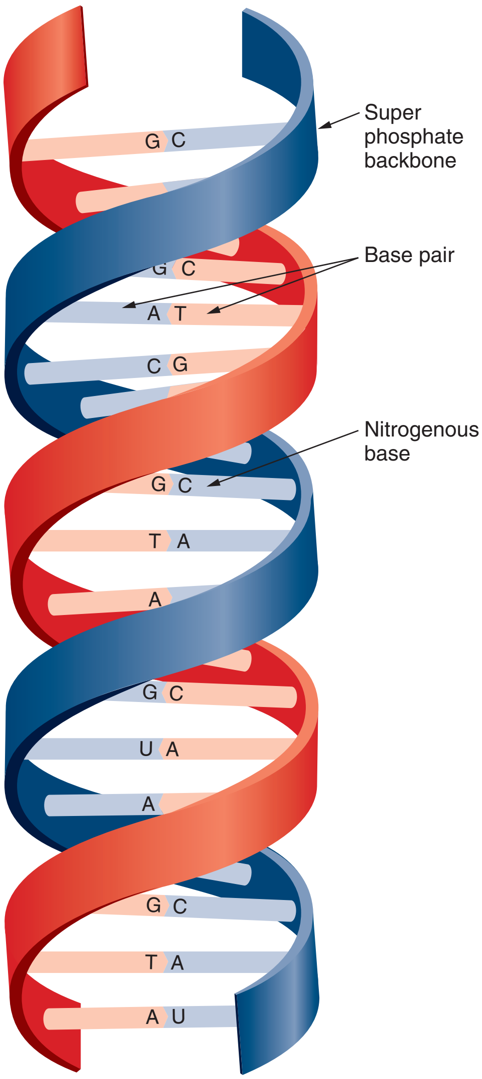


Figure 3. Illustration of an siHybrid—a hybrid of small, interfering RNA (siRNA) (blue) and DNA (red). Unlike siRNA, siHybrids are effective against bacteria and, in human cells, remain viable longer. The primary difference between the two is the way that the five nucleotide bases—adenine (A), thymine (T), guanine (G), cytosine (C), and uracil (U)—form base pairs. Depending on how they are engineered, siHybrids introduced into a human cell, bacteria, or virus can either silence a harmful or defective gene or kill the organism outright. SiHybrids are thus a single technology that can potentially combat all three major causes of disease—bacteria, viruses, and faulty genes.

The chief disadvantage of siRNA, however, is its limited lifespan: siRNA breaks down quickly in an organism and is effective for only 24 hours, which would require repeated doses to prevent or reverse infection. Another major drawback is that siRNA is effective only against eukaryotic organisms—cells with a nucleus—but not prokaryotic organisms—those without a nucleus, namely bacteria, which are responsible for some of the most frightening diseases out there, including anthrax, botulism, and the plague.

The Advantages of SiHybrids

In an LDRD project titled "An Agent That can Prohibit Microbial Development and Infection," principal investigator Allen Christian and his co-investigators have developed an RNA interference technique that is longer lasting than siRNA and is also effective against bacteria. Christian's efforts began with the realization that because DNA and RNA bind together strongly, conventional siRNA could be made more resistant to degradation by binding it with DNA.

The resultant RNA–DNA hybrid is called an siHybrid (for "small, interfering hybrid) (Figure 3). SiHybrids are inserted into a cell, where they remain quiescent for a considerable period of time, without affecting cellular processes, until the gene it targets is activated. At this time the siHybrids become effective, silencing the genetic response without affecting the host cell in any other way. This process can be used in both human and microbial cells. Moreover, siHybrids attack bacteria in a way virtually impossible for natural or engineered defenses to circumvent.

Testing has verified the effectiveness of siHybrids. In one test, siHybrids silenced the targeted gene of cultured cells by 80% (compared to 60% for siRNA) and 8 days later were still reducing the gene's activity by 60%. (Activity is measured as the amount of protein that the gene is still producing.)

In addition to attacking a microbe directly, siHybrids can also work indirectly. Christian's team has used siHybrids to remove antibiotic resistance from bacteria, rendering them sensitive to agents that were previously ineffective against them. Thus siHybrids could considerably extend the usefulness of existing antibiotics to which many disease-causing agents are becoming increasingly resistant.

How the Technique Works

Although the exact process is unknown, both SiHybrids and siRNA are thought to work by utilizing the cell's genomic defense mechanism: siHybrids entering a cell could trigger the same reaction triggered by viruses and harmful DNA that breach the cell wall. In responding to "invading" siHybrids, the cell destroys not only the siHybrids but also the messenger RNA that it resembles. In short, siHybrids "trick" the cell into mistaking its own messenger RNA as a hostile invader.

Because clinical trials are more easily conducted on a process of known mechanism, Christian and team are working to understand the mechanism by which siHybrids silence genes. Important unknowns about how siHybrids work include why they enter cells (including bacteria) more effectively than siRNA, and why the siHybrids split apart once inside the cell. (Understanding how siHybrids work entails research at the level of single cells and is one goal of the next LDRD project discussed in this article.)

Another major advantage of siHybrids over siRNA is that siHybrids do not require transfection for delivery into a cell. Transfection is a technique for getting the molecule through the cell membrane by latching it to fat molecules. The resultant glob binds to the cell in a manner similar to two soap bubbles combining upon contact. However, transfection can kill up to 50% of cells. SiRNA, although initially touted as a miracle technique that would enable wondrous new theraputics, was later found to work only when introduced by transfection, precluding its use as a therapeutic. SiHybrids, in contrast, can silence a gene up to 60% over 8 days without transfection. (With transfection, the rate is 100%.)

The Fight Against Disease

The significance of 60% silencing is illustrated by the example of certain drugs used to treat lung cancer. These drugs have been shown to be highly effective when a certain gene in the body has been silenced at least 50%. Administering these anticancer drugs with an siHybrid-based drug that silences the gene would therefore provide a more powerful means of fighting lung cancer.

Another example of using siHybrids against cancer is a certain form of prostate cancer that is caused by the mutated form of a gene. Collaborators at the UC Davis Cancer Center have found that an siHybrid matched to the mutated gene silences that gene 100% but does not inhibit the normal form of the gene. Figure 4 shows actual cancerous cells that have been silenced with siHybrids.

SiHybrids can also help basic research by allowing a fast, inexpensive alternative to gene knockouts. A gene knockout is a laborious, expensive technique of determining a gene's function by genetically engineering an organism (such as a mouse) that lacks the gene. SiHybrids could perform the same function by silencing a specific gene in genetically normal organisms. Multiplexed experiments are also possible—multiple-target siHybrids can shut off multiple genes simultaneously, to determine how those genes work together to produce a certain function.

Because they remain viable for some time in a cell, siHybrids can be used either as a post-infection treatment or as a preventative measure before infection. For instance, in a bioterror scenario involving the release of an infective organisms, responders in the affected area could administer an siHybrid-based drug to those already infected and everyone at risk; to those not infected, the drug would confer short-term immunity.

In addition to inhibiting infection, siHybrids may also be capable of killing the infectious agents outright. To this end, Christian is studying the various genetic pathways that, if interrupted, would induce apoptosis—programmed cell death—essentially inducing the bacteria to commit suicide. The team is examining this approach using a basic laboratory strain of *Escherichia coli*. Because the genetic pathways involved are fairly universal among the many species of bacteria, success in this work could produce a new class of broad-spectrum antibiotics and antiviral agents.

In FY03, the team demonstrated the ability to destroy the HIV virus in human T-cells. The team will continue to study various pathways that are common to all bacteria and which, if inhibited, cause the bacteria to die. They will also infect human cells with nonpathogenic viruses, such as baculovirus, to study the degree and duration of the anti-infection potential of siHybrids. Preliminary results are proving to be of seminal importance in the field of bioscience and pointing to the increasing potential of siHybrids as a single technology that can potentially combat all three major causes of disease—bacteria, viruses, and faulty genes.

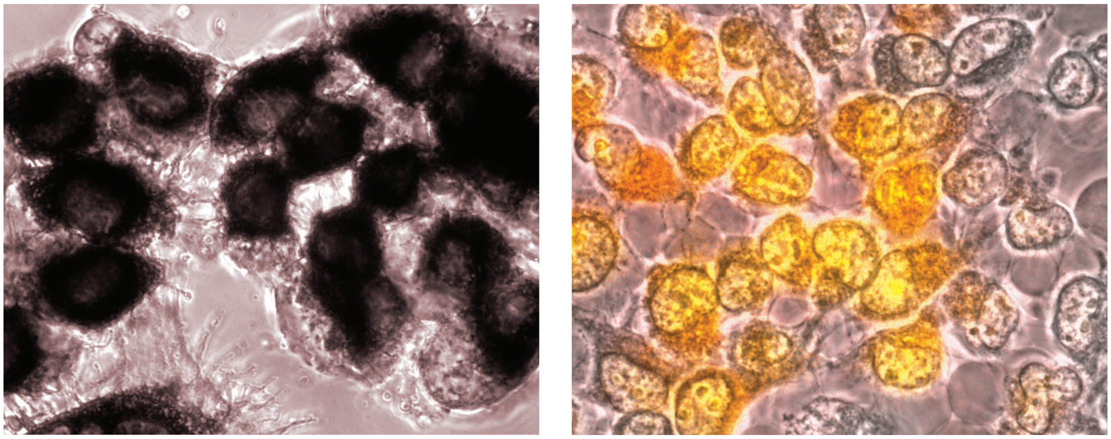


Figure 4. Cancerous human cervical epithelial cells before (left) and after (right) a single treatment with siHybrids. The black coloring indicates a protein produced by a gene in the cancerous cells. The absence of the protein in the treated cells indicates the complete silencing of the gene by siHybrids designed specifically to target that gene.

Part 2: Investigating Biological Processes

Understanding How Microbes and Other Cells Function at a Molecular Level

The cell is the basic unit of life, from disease-causing bacteria to humans and other higher organisms. To understand how drugs work in the human body, and how siHybrids silence genes, for instance, scientists must understand the chain reactions that those molecules trigger inside the cell. This, in turn, requires the ability to accurately simulate, measure, and manipulate the chemical state of individual compartments within a single cell. Such technology would revolutionize new-drug development and cell biology itself, providing the means to unravel the dynamic processes that underlie the cell functions and to precisely test hypotheses about biological functions.

An LDRD project with the potential to greatly advance the state of the art in this field is the Instrumented Cell initiative. The initiative seeks to develop a platform for capturing large numbers of living cells in organized arrays and carefully controlling the environment to keep the cells alive for experimentation. The project team, led by Allen Christian, is also developing the technology to introduce chemicals and other stimuli directly into the cells, as well as sensors for monitoring the cellular environment and software to control the overall system.

Modeling Cellular Activity at the Molecular Level

This project reflects a great movement underway in biology today—the transition from a qualitative, discovery-based science to what is termed "quantitative biology." In this field—still in its infancy—researchers strive to model the fundamental processes of life computationally and use those models to formulate and test hypotheses. The ultimate goal of this LDRD project is to produce data that would allow the computational modeling of the various chemical pathways of a cell. This would allow scientists to create predictive models of cellular responses to stimuli—such as new drugs—thus permitting the rapid, virtual screening of new-drug candidates, rather than the much slower and much more expensive method of preparing actual compounds and testing them in a laboratory. Other potential breakthroughs that these capabilities would enable include bioengineering cells with specific applications, such as breaking down oil slicks.

Although such capabilities would revolutionize biology, no method exists for acquiring quantitative data at the level of a single cell. Instead, current techniques dissolve cells into a homogeneous "soup" to measure average concentrations of target compounds in the cell, thus losing all information about how concentrations vary among different components of the cell. Differences in concentrations among individual cells are also involved in many fundamental biological processes, such as cell differentiation, carcinogenesis (the transformation of normal cells into cancer cells), spore creation by bacteria such as the anthrax bacteria, and cell-to-cell communication. Moreover, average concentrations cannot elucidate variation that exists among supposedly identical cells—such as the variation seen in cloned cells maintained under identical conditions. Average concentrations cannot determine what is behind this variation.

Living Cells on a Chip

Figure 5 is a photograph of a microchip that Christian and his team have fabricated to capture, keep alive, and experiment on individual cells. In each of the six wells, slightly more than 10 μm across, is a human cancer cell. Cell sizes vary considerably, but because each well must be just the right size to hold only one cell, the microchip will eventually be made with interchangeable wells.

To capture single cells in the wells, a liquid containing the cells is flowed into the chip. A syringe pump pulls the liquid through an opening in the bottom of each well that is small enough to trap the cells inside. Figure 6 is photograph of a chip with capture wells small enough to trap individual bacteria.

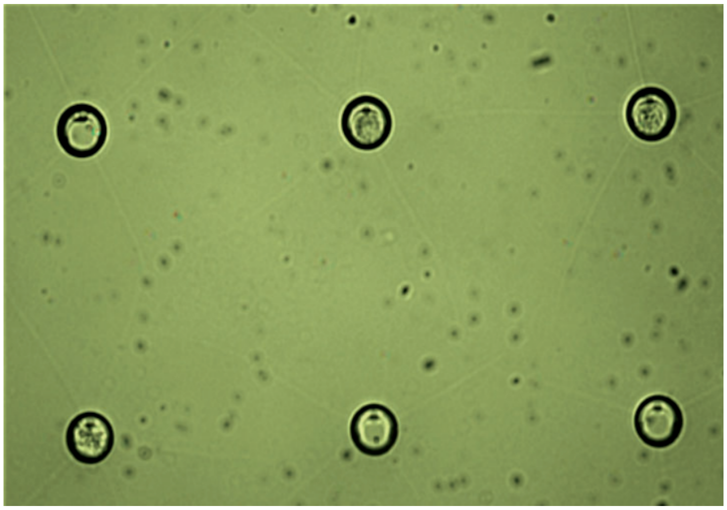


Figure 5. A photograph of a microchip designed to capture, keep alive, and experiment on single cells. Each capture well (circled) is approximately 10 μm across and contains a single human cancer cell. This and other technology being developed in this LDRD project will provide the ability to accurately simulate, measure, and manipulate the chemical state of individual compartments within a single cell, which would revolutionize the science of cell biology.

Keeping the cells alive after capture is one of the project's greatest challenges—and what makes this capability unique in the world. The liquid environment in which the cells are maintained amounts to an artificial blood stream, which must deliver nutrients and maintain the proper level of carbon dioxide (about 5%) and a temperature of 37°C, among other variables. The slightest deviation in any of these variables from what the cell is accustomed to can cause the cell to behave differently—which would defeat the purpose of studying the response of cells under normal conditions. For instance, a temperature rise of only 2°C would induce a "fever" effect on cellular metabolism as a fever.

The project team is also fabricating glass microneedles for injecting substances into the captured cells and sampling their interior. The needle shown in Figure 7 will be fabricated in arrays corresponding to the cell-capture wells, for the injection of multiple cells all at once. The ability to capture and manipulate large numbers of cells at once will assure the high throughput necessary to produce large amounts of data in a short amount of time. The team is also writing software to automate the entire process, further enhancing the accuracy of analysis while also reducing costs.

The device is built to fit onto the stage of a laser confocal microscope and has already been integrated with fluorescent and bright-field microscopy; surface-enhanced Raman spectroscopic nanoparticles and nanoneedle capability is being developed, as well as infrared spectroscopy.

The Instrumented Cell device can also be used in conjunction with other techniques being developed for quantitative biology, such as fluorescence tagging, to produce data on how concentrations of key molecules vary inside a single cell and how and where important molecular reactions occur in the cell. In addition, individual cells arrayed in the device could be subjected to multiple tests, thereby producing multiple sets of data from the same individual cells. This would enable the cross-checking of the data and produce a clearer picture of variation among individual cells.

The Near and Long Term

In FY2004, Christian and team will continue to test and refine the Instrumented Cell system. An immediate application will be studying the gene-silencing mechanism of siHybrids, beginning with quantitatively determining how siHybrids penetrate the cell wall and how long they remain viable inside. In the longer term, Christian sees researchers using the Integrated Cell to take on significant issues of biology by combining the technology with various sensitive methods of detection developed at LLNL. The resultant capabilities would revolutionize the science of cell biology, helping to unravel the dynamic processes that underlie cell functions and point the way to better defenses against disease and biological threats.

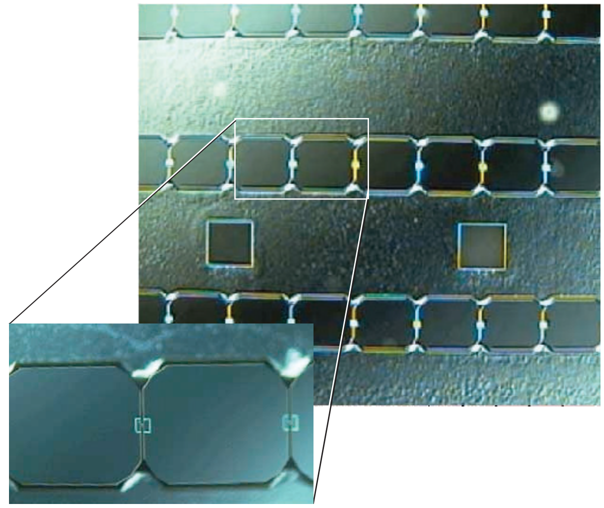


Figure 6. A side view of a microchip with arrays of capture wells small enough to trap individual bacteria. The inset is a magnified view of a single well, which is located in the vertical space between two adjacent rectangular bases. In the space is a narrow gap where a bacterium is trapped as the flow is drawn through.

Imaging Biomolecules at a Resolution Never Before Possible

No technique available today can determine the structure of uncrystallized proteins at atomic resolution. The low-intensity x rays generated by current third-generation synchrotrons can produce atomic-resolution images only when the target molecule has been crystallized—a process that is slow and expensive and cannot be used with many types of molecules. The Linac (Linear Accelerator) Coherent Light Source (LCLS), currently being constructed at Stanford University, will produce pulses of x rays so bright and focused that they will allow researchers to produce images of single molecules without the need for crystallization. It will be, in effect, the world's highest-magnification microscope, opening the door to a world previously far beyond the limits of imaging.

Under an LDRD project titled "A Revolution in Biological Imaging," Henry Chapman and his co-investigators are laying the groundwork for some of the first experiments to be performed at the LCLS. The project is performing fundamental research to image single biological molecules and particles such as viruses, as well as investigate the ultrahigh time resolution needed to image the sequence of molecular reactions, such as those between viruses and molecules in human cells.

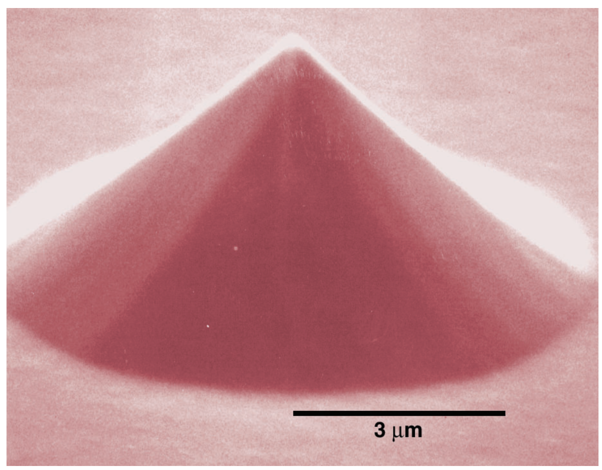


Figure 7. A glass microneedle fabricated for this project. Needles such as this one will be manufactured in large arrays and used to inject and sample large numbers of captured cells at once. This manipulation technology, combined with this project's technology for capturing and maintaining cells, will assure the high throughput necessary to produce large amounts of data quickly.

This project uses a combination of experimentation and computer modeling. In the first phase of the project—already completed—three-dimensional (3-D) numerical models of biomolecules were used to calculate x-ray scattering. Next, the team modeled the time evolution of a sample hit by an x-ray pulse to determine the maximum pulse duration that would enabling single-molecule imaging. In the coming phase, entire imaging experiments will be simulated, including sample damage and image generation. Algorithms for determining a molecule's structure from the patterns made by diffracted (scattered) x rays will then be developed and verified through experiments on a synchrotron and at other facilities.

X-Ray Free-Electron Lasers

When it becomes operational, the LCLS, a collaboration between LLNL and five other national laboratories and universities, will be the world's first full-scale x-ray free-electron laser (XFEL). (The Laboratory is responsible for all x-ray optics for the project.) A free-electron laser is a powerful combination of the physics of particle accelerators and lasers. The beam-generation process begins with an electron beam from a linear accelerator, which is passed through an undulator magnet. Inside, thousands of magnets arranged in alternating poles push and pull the electrons, causing them to emit a highly coherent beam of photons. Until recently, free-electron lasers have been operating in the infrared or near-ultraviolet wavelengths. But the technological advancements that are being incorporated into the LCLS will produce higher-power beams in the x-ray spectrum. These pulses will be so extraordinarily bright and fast that they will be capable of producing images of single molecules and measuring molecular processes such as melting, recrystallization, and light-induced structural change on time scales down to a femtosecond—one quadrillionth of a second.

Lensless Imaging

In these experiments, molecules will be imaged not with optics but with a lensless technique—x-ray diffraction. In x-ray diffraction imaging, an x-ray pulse is directed at a molecule. The x rays are diffracted (scattered) by electrons orbiting the atoms in the molecule. The resulting diffraction pattern is recorded, then analyzed by computer algorithms that translate the pattern into a 3-D structure. By using pulses of x rays that are intense enough and short enough in duration, a diffraction pattern can be recorded from a particle as small as a single molecule. Because the molecule explodes when struck by the x rays, the pulse duration must be shorter than the timescale at which the molecule begins to break apart. If it is, the diffraction pattern obtained will represent the undamaged molecule.

Ten billion times brighter than any existing x-ray light source, and with ultrashort pulses of 200 fs or less, the LCLS will overcome current radiation-damage limits to imaging. Initial molecular dynamics calculations indicate that pulse durations of 200 fs or less will increase the tolerable radiation dose of large single molecules by four orders of magnitude. This will ensure enough scattered photons to image single large molecules and viruses at a resolution of 2 Å or better. (At a resolution of 5 Å, amino acid sequences can be identified.)

A Revolution in Life Sciences

The atomic-resolution imaging of virtually any macromolecule, protein, or virus promises to revolutionize the life sciences, particularly structural biology and medicine. This capability could eventually lead to solving the entire human proteome—determining the complete structure of every protein expressed by the genome. The low-intensity x rays of today's third-generation synchrotrons can yield atomic resolution only when the target molecule has been crystallized. This "freezes" identical molecules in the same orientation, so that a single pulse produces numerous instances of the same diffraction pattern. However, many proteins cannot be crystallized, and once a protein is crystallized, scientists cannot study its interactions with other biological molecules. With the LCLS, researchers will be able to study such uncrystallizable proteins, including proteins linked to lipids (fats) and embedded in cell membranes. These cell-membrane proteins account for 30% of the more than 100,000 proteins in the human proteome and are the target of 80% of all pharmaceuticals. Removing the need to crystallize a protein will also shorten structure determination from years to weeks, leading to faster identification of biological agents in biodefense and other applications.

Setting the Stage

To enable single-particle diffraction experiments at the LCLS, Chapman and team are addressing a host of challenges. In particular, they are developing iterative, multiscale, multiresolution reconstruction algorithms and designing extended simulations—with experimental verification—to establish the requirements of the x-ray pulses and diagnostics. Another challenge is understanding how sample molecules respond to the x-rays, which will influence the achievable resolution for a given pulse duration and, consequently, the design of the x-ray sources themselves. To address this question, they are creating realistic photon–matter interaction models to understand the various interactions between the XFEL beams and samples and to verify that the beam pulse will end before the sample begins to break up. Figure 8 shows the results obtained with the team's hydrodynamic model in simulations of a protein molecule 20 Å in diameter hit by an XFEL beam.

The model, which includes more physics than previous molecular-dynamics models, has indicated that encasing the sample macromolecule in a water tamper will delay the molecule's Coulomb explosion enough to extend the time range of diffraction. The team then simulated the time-integrated diffraction pattern and compared this with the required signal to sort and average patterns of randomly oriented molecules. This yielded an estimate of the best achievable resolution for a given molecule size and pulse duration.

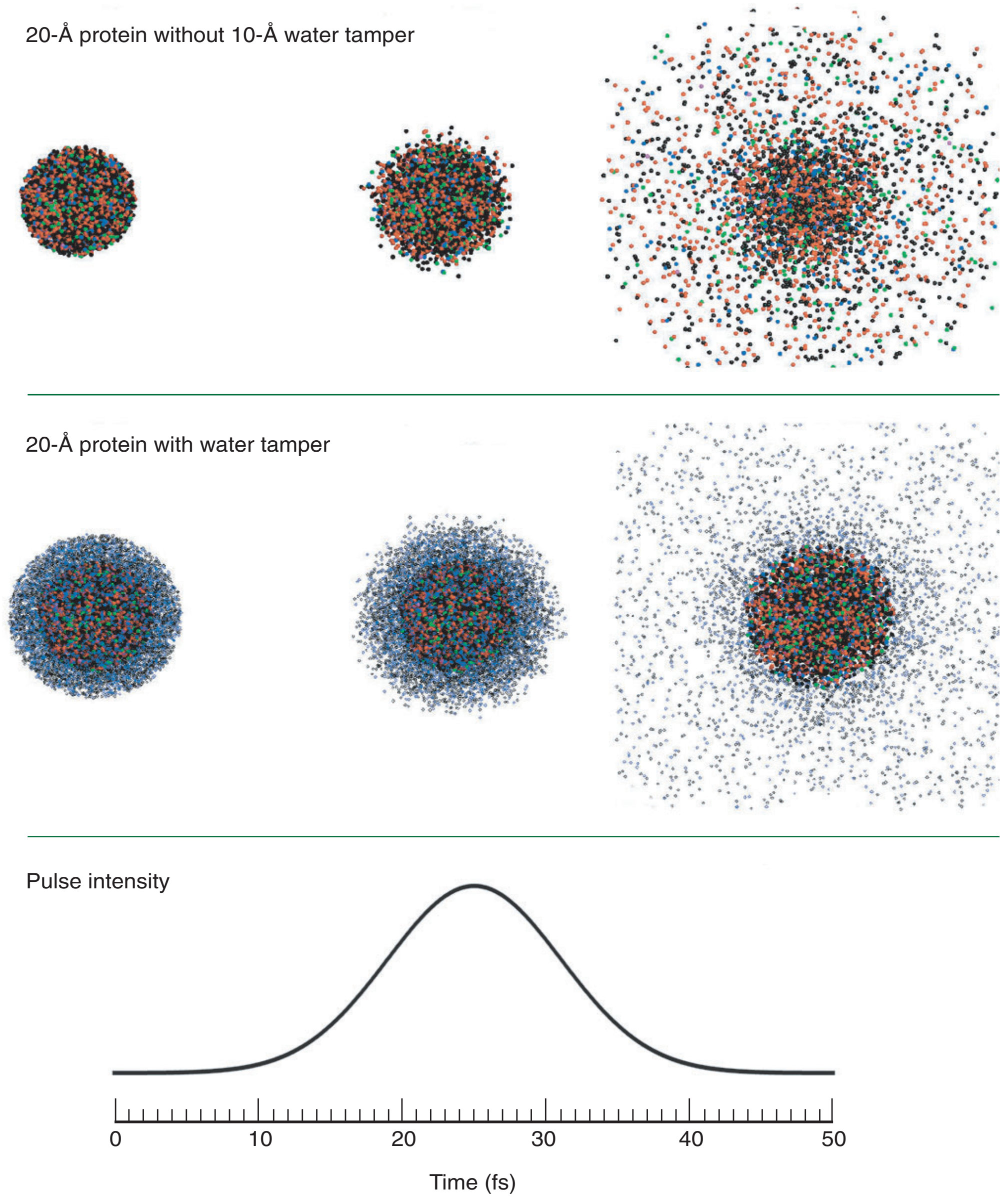


Figure 8. Simulation of a 20-Å protein molecule Coulomb-exploding due to a 20-fs, 12-keV x-ray free-electron laser (XFEL) pulse (top) with and (middle) without a 10-Å water tamper, which suppresses Coulomb explosion long enough to obtain an accurate diffraction pattern.

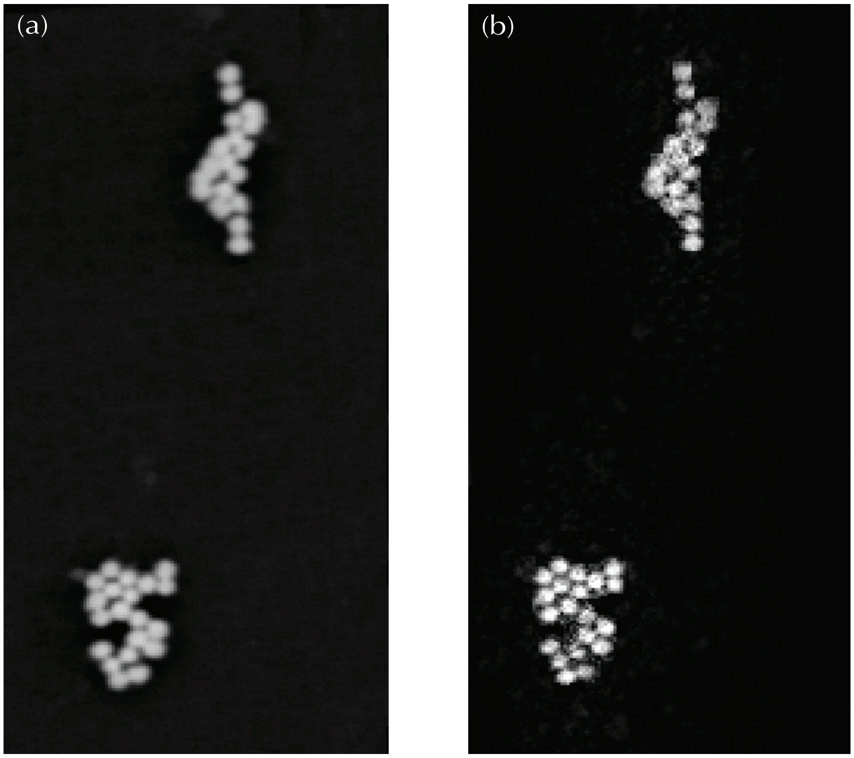


Figure 9. In preparation for single-molecule imaging at the Linac Coherent Light Source, Henry Chapman and team have developed computer algorithms able to determine the structure of a molecule based on its x-ray diffraction pattern. (a) Images of 50-nm-diameter colloidal gold spheres were reconstructed from x-ray diffraction patterns obtained at Lawrence Berkeley National Laboratory's Advanced Light Source. This is the first demonstration of true lensless imaging, and compares well with (b) an image of the same spheres obtained with a scanning electron microscope.

Other sample-preparation techniques that could allow longer and higher-intensity pulses include electrospray injection and using polarized laser light to orient the molecule or better determine its orientation.

A World First

A major achievement in this project is the first-ever demonstration of true lensless x-ray imaging. Although the reconstruction of images from experimental coherent x-ray diffraction patterns has been previously demonstrated, all work to date has required detailed and high-resolution images of the object boundary to assist the reconstruction algorithms. These images were acquired with x-ray microscopes at a resolution close to that required in the final reconstruction. Such a stringent requirement would render single-particle imaging in XFELs all but useless. Chapman's team has conceived a new algorithm that requires absolutely no prior knowledge about the spatial distribution of the sample. This algorithm was used to successfully reconstruct an image of clusters of 50-nm-diameter gold spheres from diffraction patterns recorded at the Advanced Light Source (ALS) at Lawrence Berkeley National Laboratory, in collaboration with colleagues there. As shown in Figure 9, a reconstructed image of the spheres compares extremely well with an image obtained using a scanning electron microscope, demonstrating the fidelity of the imaging technique.

Future Work

Although the LCLS will not be operational for some time, Chapman's team will continue with preparatory work that will enable researchers to make immediate use of the facility. At the ALS, the project team will create the first high-resolution lensless 3-D images. Samples will be prepared and characterized in advance, but images will be constructed from a series of diffraction patterns, without any prior images to assist the algorithms.

In preparation for testing at the Tesla Test Facility (TTF)—a soft-x-ray, prototype XFEL facility in Hamburg, Germany—the team will develop sample-preparation methods and test their hardware and concepts at a synchrotron source and at the LLNL Thompson source. In addition, they will extend dynamical models to the longer wavelengths and larger particle sizes of TTF to be able to fully interpret experimental results.

At the TTF, the team will perform the first x-ray diffraction imaging experiments ever performed on an XFEL, including 2-D images that break the radiation-damage resolution limit of x-ray microscopy, and 3-D images of unsupported, free-falling samples. In another series of experiments there, they will directly measure, through the x-ray scattering of aerosols, the Coulomb explosion of particles irradiated by the XFEL pulse.

Other work planned includes extending biomodeling to whole cells, to determine damage effects in cellular systems; extending damage models to large particle sizes (> 50 nm); improving accuracy by including the trapping of photoelectrons; verifying results against molecular dynamics calculations; and improving image reconstruction by developing methods for classifying diffraction data and extending a reconstruction code to handle objects such as a molecule with a heavy-atom marker—used to boost the diffraction signal and aid orientation.

Through these and other efforts, Chapman and team will continue to prepare the technologies, expertise, and experiments to make full use of the groundbreaking capabilities of the LCLS.

—Paul Kotta

For more information see the project summaries (on CD only) for projects covered in this article:

Development of Ultrasensitive, High-Speed Biological Assays Based on Two-Dimensional-Flow Cell Detection of Single Molecules
Christopher Hollars, Principal Investigator, 02-ERD-018

An Agent That can Prohibit Microbial Development and Infection
Allen Christian, Principal Investigator, 03-ERD-038

The Instrumented Cell
Allen Christian, Principal Investigator, 03-ERD-068

A Revolution in Biological Imaging
Henry Chapman, Principal Investigator, 02-ERD-047

Related projects on bioscience and biotechnology:

Development of Synthetic Antibodies
Julie Perkins, Principal Investigator, 01-ERD-111

DNA Detection through Designed Apertures
Sonia Letant, Principal Investigator, 03-ERD-013

Imaging of Isotopically Enhanced Molecular Targeting Agents
Judy Quong, Principal Investigator, 01-ERD-112

Microfluidic System for Solution Array-Based Bioassays
Peter Krulevitch, Principal Investigator, 03-ERD-024

Pathogen Pathway Project
Joseph Fitch, Principal Investigator, 01-SI-002

Single-Cell Proteomics with Ultrahigh-Sensitivity Mass Spectrometry
Eric Guard, Principal Investigator, 02-ERD-002

Visualizing ASC-Scale Simulations on a Laptop

ViSUS is a scalable, streaming visualization technology that is freeing scientists from large-scale systems.

Science-based stockpile stewardship, a core mission of the Laboratory, is based on computer simulations of the complex physics involved in nuclear weapons. Such simulations eliminate the need for actual weapons testing. The supercomputers on which such simulations are run process massive amounts of data, often many terabytes—trillions of bytes.

Once a simulation has been finished—which can take weeks or months even on the fastest supercomputers—scientists need to interpret the results. One way to do this is by visualization, which converts the raw data produced into a three-dimensional (3-D) visual format. Because the massive amounts of information involved would overwhelm the capabilities of conventional office computers, such simulations are usually visualized either on large display systems with massive processing power, or by using high-end systems to generate off line animations. However, the restrictions and limitations of the typical visualization system are many: they support only certain types of simulations or certain simulation systems, run only on high-end systems, and allow only limited modification of the parameters in the interactions they allow. In many cases, the output is a static movie that cannot be changed at all; to test other combinations of variables, or view a different cross section of the simulation, the researcher must have additional movies made by trained personnel.

When a large display system is used, the data set to visualize must be downloaded in its entirety to the visualization system. This can take hours or even days depending on the size of the data set, which is a function of the simulation's complexity. Multiply such wait time and heavy use of network bandwidth by thousands of scientists and thousands of simulations across the DOE complex, and the need becomes clear for the technology to use existing resources more efficiently.

Limited access to these conventional visualization systems creates a frustrating slowdown in the overall process of scientific discovery. Scientists must wait as animation files are generated. In the case of a high-performance display, they must "wait in line" for access and must travel to wherever the system is located to use it. This approach precludes testing a sudden flash of inspiration on an ordinary office computer or with a laptop from an offsite location.

Enter ViSUS

An LDRD project is putting sophisticated visualization capabilities in the hands of the researchers themselves. The ViSUS (Visualization Streams for Ultimate Scalability) project, led by computer scientist Valerio Pascucci, has developed a tool that lets researchers visualize massive simulation data sets remotely and adjust simulation variables on the fly to explore the 3-D simulations interactively. This powerful, flexible tool can run on an ordinary desktop or laptop computer.

To change a variable or the perspective during visualization, the user simply moves a lever on the program's graphic user interface, as shown in Figure 1. In this visualization of trace gases in the atmosphere made with the IMPACT global atmospheric chemistry model, users can visualize different concentrations of ozone (O_3), hydrogen peroxide (H_2O_2), and bromine monoxide (BrO). To test the effect of different combinations of variables, the user makes changes and sees those changes instantly reflected in the images. The testing of different scenarios—or the iterative process of finding the right set of variables to achieve a certain result—becomes much faster than it would by generating large numbers of static movies or having to run new simulations on a supercomputer.

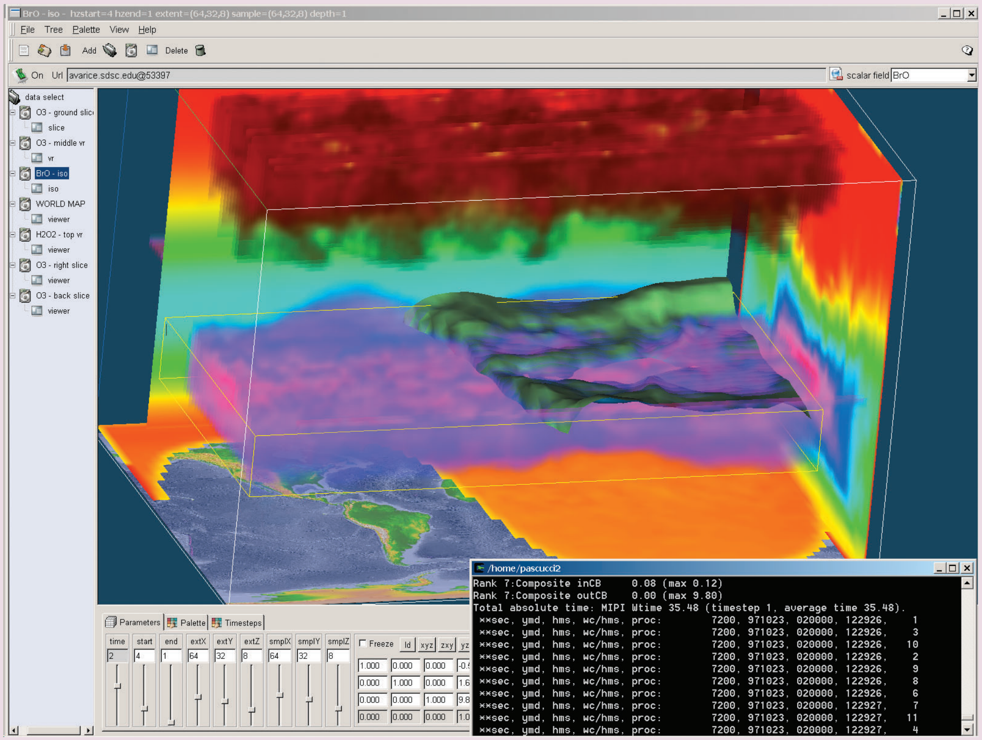


Figure 1. A screen capture of the Visualization Streams for Ultimate Scalability (ViSUS) system being used to visualize an atmospheric simulation done with the IMPACT atmospheric chemistry model. Unlike other visualization systems, ViSUS allows the user to change a variable on the fly by simply moving a lever on the program's graphic user interface. In this case, ViSUS can test the effects of different concentrations of ozone (O_3), hydrogen peroxide (H_2O_2), and bromine monoxide (BrO). With ViSUS, the testing of different scenarios becomes much faster than it would by generating large numbers of static movies.

Another important advantage of ViSUS is that it is a streaming solution that visualizes data in real time—as soon as retrieval of the data begins. This streaming concept is similar to the streaming audio and video now widely used on the Internet: instead of downloading a huge file, a data-streaming application plays the file as the data are received. Rather than requiring massive bandwidth between sender and receiver to download huge files quickly, all that is required is enough bandwidth for transmission to keep up with the rate at which the data are played.

Of course, ViSUS is much more complex than the Internet analogy suggests. The breakthrough technology at the heart of ViSUS is a set of algorithms that selectively choose the data that must be uploaded first in order for the images to be immediately useful. The algorithms first extract the data needed to produce coarse images, which can later be "fleshed out" into higher-resolution images. Depending on the memory and processor of the computer on which ViSUS is running, the higher-resolution images may be viewable almost immediately. But even if memory, processing power, or bandwidth are limited, ViSUS can enable the real-time imaging of massive, complex scientific datasets.

"Ultimate Scalability" Means Ultimate Flexibility

This is the "ultimate scalability" in the name ViSUS: the same version of the visualization engine can run on anything from a low-end laptop to a high-end workstation. In the latter case, a high-resolution version can be viewed without sacrificing time. But the flexibility to trade speed for resolution allows a scientist to use an ordinary desktop or laptop computer to visualize data remotely and in real time. He or she could run the visualization first at low resolution to get the big picture and then, if something intriguing appeared, view it again at high resolution. Because the local algorithms scale down to low-end computers, the ViSUS program is installed on the user's own computer, rather than on a server. This was a requirement for fast visualization.

For comparison, Figure 2 shows a simulation of Richtmyer–Meshkov fluid mixing—a phenomenon that arises when a shock passes through the boundary of a heavy fluid sitting atop a light fluid—being visualized with ViSUS both on a large, multi-projector, very-high-resolution display (a "PowerWall") and on a laptop connected to a standard presentation projector.

With its data-streaming and interactive capabilities, ViSUS could even be used to adjust the parameters of a supercomputer simulation in progresses, provided that that the data source is set up for such interaction. Unsuccessful simulations could therefore be terminated midway, a boon for conserving supercomputer resources. If a simulation would otherwise take hours or even days to finish—during which time the computational resources would be unavailable for other work—then in the long run, ViSUS could save both supercomputer time and the time of the people running the simulation and waiting for its results.

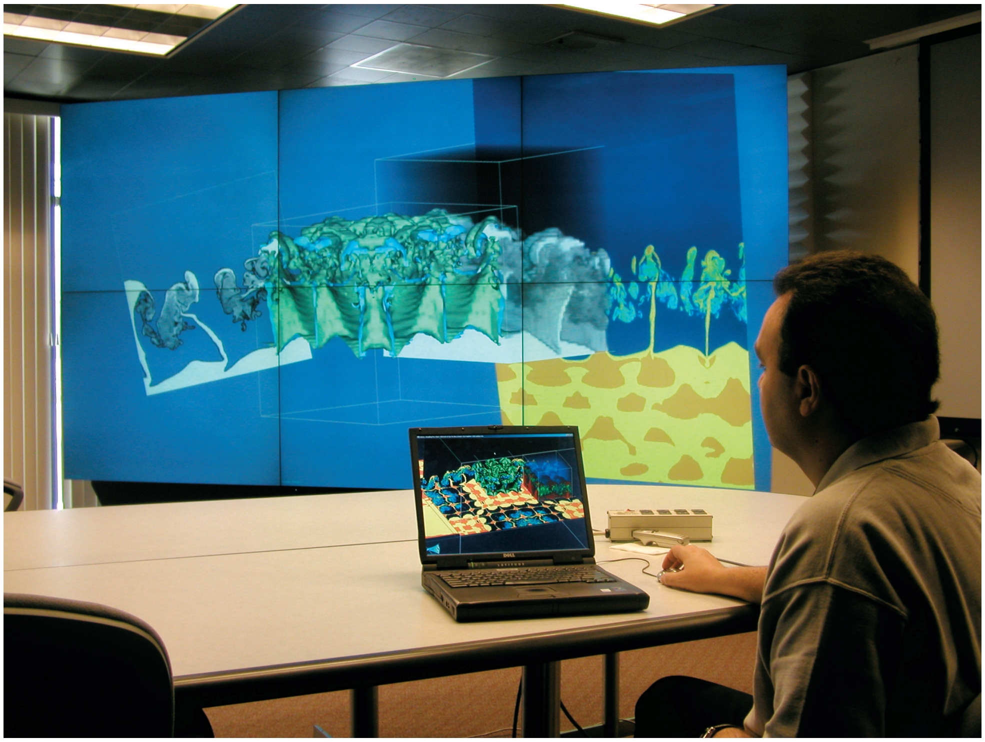


Figure 2. The same simulation—Richtmyer–Meshkov fluid mixing—being visualized on a large, multi-projector, very-high-resolution display and with ViSUS running on an ordinary laptop computer connected to a projector. Richtmyer–Meshkov instability is a phenomenon that arises when a shock passes through the boundary of a heavy fluid sitting atop a light fluid. This particular simulation won the 1999 Gordon Bell Award for best supercomputing performance.

The ability to terminate simulations before completion is particularly advantageous in climate simulation. Climate simulations are incredibly complex in that they involve numerous variables—such as wind speed, carbon dioxide content, temperature, and humidity—all interacting with each other; a change in any variable causes a chain reaction that affects all the others. Modeling climate events therefore entails some of the largest data sets in science. (For instance, the Earth Simulator, the Japanese supercomputer currently ranked as the world's fastest, was developed specifically for climate simulations.)

Figure 3 is a frame from a ViSUS visualization of Rayleigh–Taylor instability—another fluid-mixing phenomenon relevant to stockpile stewardship. The original data set, over

3 terabytes, took over 2 months to generate using the Miranda code on a nearly 2000-processor supercomputer at LLNL that ranks among the world's fastest. With conventional visualization technology, scientists did not know the results until after the simulation was completed. ViSUS, however, would allow scientists to visualize such simulations while still in progress; if the simulation was not progressing as desired, it could be stopped, or the variables could be adjusted.

This on-the-fly flexibility also permits researchers to explore large data sets to a groundbreaking degree. An ordinary visualization approach allows some changing of variables but is essentially a static movie. This approach may utilize only one tenth of a percent of the data generated in the simulation. ViSUS, on the other hand, allows researchers to change the variables of the experiment much more flexibly so that a much larger part of the data set can be utilized.

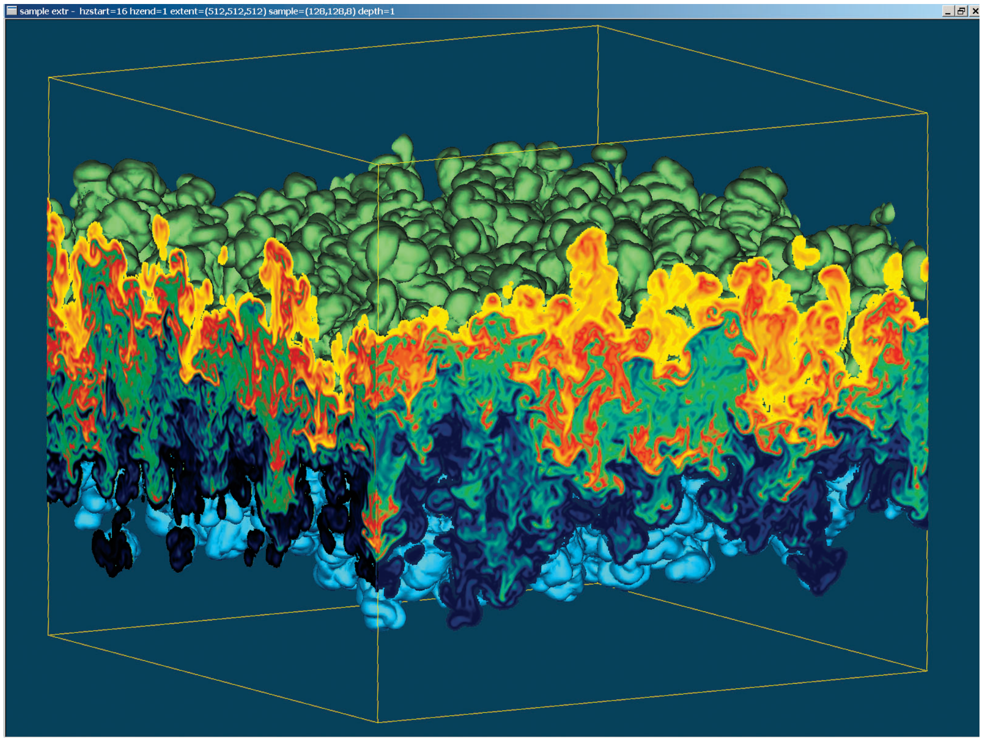


Figure 3. A frame from a ViSUS visualization of Rayleigh–Taylor instability. The original data set is over 3 terabytes in size and took over 2 months to generate using the Miranda code on a nearly 2000-processor supercomputer at LLNL. With ViSUS, scientists could visualize such simulations while still in progress and either stop the simulation or adjust the variables if it did not progress as desired.

The ViSUS Algorithms

The algorithms that selectively choose the data to transmit first—one of the major challenges in designing ViSUS—are known as progressive algorithms. Traditional visualization algorithms proceed fine to coarse—they first read the complete high-resolution information, from which users can generate lower-resolution approximations according to the capabilities of their computer. Progressive algorithms, in contrast, are algorithms that transition through multiple resolutions automatically. Input data are read from course to fine, and output is produced in the same order—coarse imagery is produced first, and from that high-resolution output can be produced. However, ordinary progressive algorithms have less accuracy at higher resolution because of loss due to compression. ViSUS, however, ensures that no accuracy is lost due to compression.

The visualization process begins with data retrieval, in which ViSUS analyzes the data and uploads the information needed to begin visualization right away. The next step is to build the mesh—the three-dimensional grid appropriate for the type of simulation and resolution. Next is the generation of filed data, which are the quantities computed by the simulation to describe the progress of a physical phenomenon in a given frame, such as the concentrations of two fluids at each location in the three-dimensional space in a fluid-mixing simulation. From these coordinates, isosurfaces are created. Isosurfaces are the 3-D surfaces that represent, for instance, the complex outer shape of twisting plumes of fluid. The final step is view-dependent rendering—selecting only the data that are "visible" to the user from the chosen perspective; everything else is excluded from the frame to conserve memory and processor use and thereby speed visualization.

In conventional visualization, data are accessed row by row. For example in a three-dimensional fluid-mixing simulation with a resolution of 2000 × 2000 × 2000 bytes, a single three-dimensional time step would contain 2000^3 bytes, or 8 gigabyes, which would be beyond the memory capacity of most desktop computers. A single two-dimensional cross-section would be 4 megabytes. If the cross-section is changed, the entire data set would have to be loaded again, 8 gigabytes at a time, to pick out the necessary points from each row. The ViSUS system, however, uses an advanced computational technique known as streaming Z curves to change the visualization without having to read the data all over again.

The algorithms that allow the ViSUS visualization engine to run on any computer are known as out-of-core algorithms. Because the data set is much larger than the computer's main ("core") memory, these algorithms move blocks of data in and out of the computer's memory as they are processed. Conventional out-of-core algorithms can only transfer blocks of data between the source disc and the computer's memory at a fixed ratio. ViSUS, however, uses a type of out-of-core algorithm called cache-oblivious algorithms, which are not limited to a predetermined ratio. This optimizes transfer directly from the storage disc to core on any computer platform.

Other Applications and Advantages

The potential uses of ViSUS go beyond stockpile stewardship and other advanced physics applications. In a medical-imaging application, a doctor could use ViSUS for real-time examination of magnetic resonance imaging (MRI) data, even from an office or other remote location. Although initial resolution would be low in such cases, the doctor could later zoom in on points of interest at higher resolution. And because ViSUS uses lossless compression, no accuracy is lost—potentially important details are preserved when moving from low to high resolution.

The ViSUS software package is also independent of any hardware configuration and has already been adapted for four operating systems—Sun, Irix, Linux, and Windows. This experience in adapting should make porting ViSUS to additional operating systems easy.

Using ViSUS can enhance data security, because the data set remains in one location, for centralized control. This also assures data integrity: ViSUS streams the data and does not actually download the data set to the user's computer. Therefore, a team of researchers can simultaneously use a single copy of the data set from different location, with no potential problems arising due to different versions of the data set.

Future Work

One of the new techniques that Pascucci and the rest of the ViSUS team are investigating as a means of accelerating the visualization process is occlusion culling, which means avoiding the unnecessary repeated constructing and drawing of portions of isosurfaces not visible in a given frame. Isosurface creation will also be sped up by adapting the algorithms to utilize the full capabilities of commercially available graphics cards in workstations.

As these and other upgrades enhance the robustness of ViSUS, the project team plans to allow LLNL scientists to use ViSUS in actual research, and to encourage use of ViSUS technology in other visualization projects. In addition to benefiting the research itself, this use will also provide valuable feedback for further enhancements.

This powerful, flexible visualization tool will enable researchers to study large, supercomputer-generated data sets quickly, flexibly, and thoroughly. ViSUS will free scientists from having to use application-specific software or compete for access to dedicated visualization systems. In short, ViSUS allows scientists to spend more time on the science and less time and money on the tools.

— Paul Kotta

For more information see the project summaries (on CD only) for projects covered in this article:

ViSUS: Visualization Steams for Ultimate Scalability
Valerio Pascucci, Principle Investigator, 02-ERI-003

Related projects on computational simulation:

Adaptive Mesh Refinement Algorithms for Parallel Unstructured Finite Element Codes
Dennis Parsons, Principle Investigator, 03-ERD-027

Long-Time-Scale Atomistic Simulations
Wei Cai, Principle Investigator, 03-LW-027

Strategic Initiative in Applied Biological Simulations
Michael E. Colvin, Principle Investigator, 01-SI-012

Water in California

LDRD research contributes to safe, secure, and sustainable water resources in California-from the source to the tap

Recent images from Mars showing robotic mobile laboratories roving the desolate, rock-strewn planet for ancient signs of water remind us that water is the essential ingredient of life. Unlike Mars, Earth has a seemingly abundant supply of this important resource, which covers more than 70% of the Earth's surface and supports a rich web of life. Yet, due to global warming and pollution—both results of human activity—along with natural climate variability, the supply, quality, and reliability of fresh water resources around the globe may drastically diminish within the coming decades.

Developing sustainable and safe water supplies for the growing global population presents one of the major international challenges of the 21st century. Because water and energy are interdependent resources, a challenge of similar magnitude will be to secure reliable, affordable, and environmentally sound energy supplies—a strategic goal of the DOE and an LLNL mission area.

Global warming, caused primarily by burning fossil fuels for generating energy, is exacerbated by pressures of increasing population and economic growth, which in turn require more water. Energy is needed to pump, process, treat, and deliver water nationwide, while energy production follows agriculture as the largest consumer of fresh water in the U.S. Meeting the challenge of energy and water security for the nation—and the world—will require significant scientific advances in understanding complex global ecosystems and how human activity affects them.

As part of an integrated research program called the Water Initiative, three LDRD projects that began in FY2003 are tackling some of these scientific challenges. Their research focuses on water resources in California, a region where a semiarid climate, a highly variable hydrological cycle, increasing population, and a huge agricultural economy that consumes 75% of the State's water supply, make issues of water availability and quality especially critical. Although the Water Initiative concentrates on California water resources, the science that comes out of these three projects is certain to find application around the country and throughout the world.

The first project uses computational models of climate and surface hydrology to study the impact of global warming and natural climate variability on regional water resources. The second project characterizes the fate and transport of nitrate, the major contaminant in California and the nation's drinking water supply and simulates surface and groundwater systems to better understand the impact of land use on nitrate contamination. The third project investigates novel electrodialysis technologies for low-cost, energy-efficient purification of saline and contaminated water supplies.

Global warming turns up the heat

California, like much of the western U.S., has a Mediterranean climate—rainy winters and hot, dry summers. In California's historical hydrological cycle, the snowpack in the Sierra Nevada acts as a huge reservoir, storing winter precipitation and releasing it as the weather warms in the spring and early summer. The size of the snowpack and the rate of snowmelt determine water availability during the dry season, when the demand for water is the greatest.

To mitigate the natural winter flood and summer drought cycles and create a safe and reliable water supply for a growing population, California has built a complex water-storage and -distribution system over the past 150 years (Figure 1). Now, global warming is putting the region's hydrological equation at risk and raising questions about how to manage water resources for the future.

Computer models of Earth's climate suggest that temperatures in the California region will rise as much as 3˚C before the end of the 21st century. How much and how quickly temperatures increase depend on natural climate variability and, more importantly, on the rate of increase of greenhouse gases—mostly carbon dioxide (CO_2) from fossil fuels burned for transportation, industrial and commercial activities, electric utilities, and residential use.

Warmer temperatures will dramatically affect the hydrological cycle in California. During winter, more precipitation will fall as rain rather than snow, thus reducing the snowpack and changing the seasonal distribution of runoff. With greater runoff in the wet winter months when reservoirs are already full, and less snowmelt runoff during spring, the region will be at greater risk from winter flooding and summer shortages. As Figure 2 shows, a temperature increase of just 1.7˚C would have a significant effect on the timing and amount of winter runoff.

Most models show that doubling the level of atmospheric CO_2 could reduce the snowpack by as much as 50% by the year 2090. Recent climate changes are already having an effect on the region's hydrology. In response to a temperature increase of 0.8˚ C since the 1950s, snowpack levels have already decreased significantly, and spring runoff peaks earlier and at lower levels.

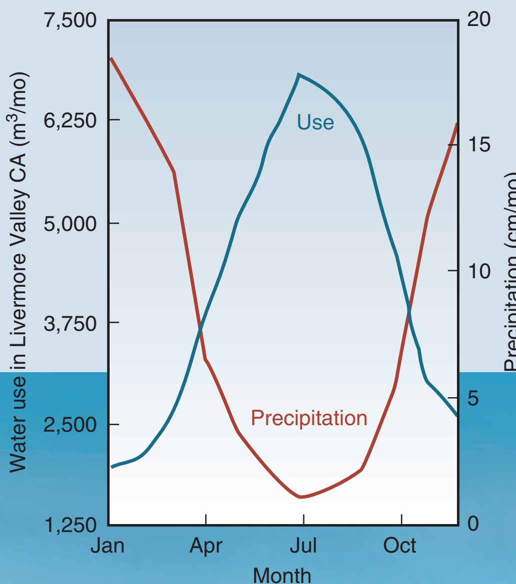
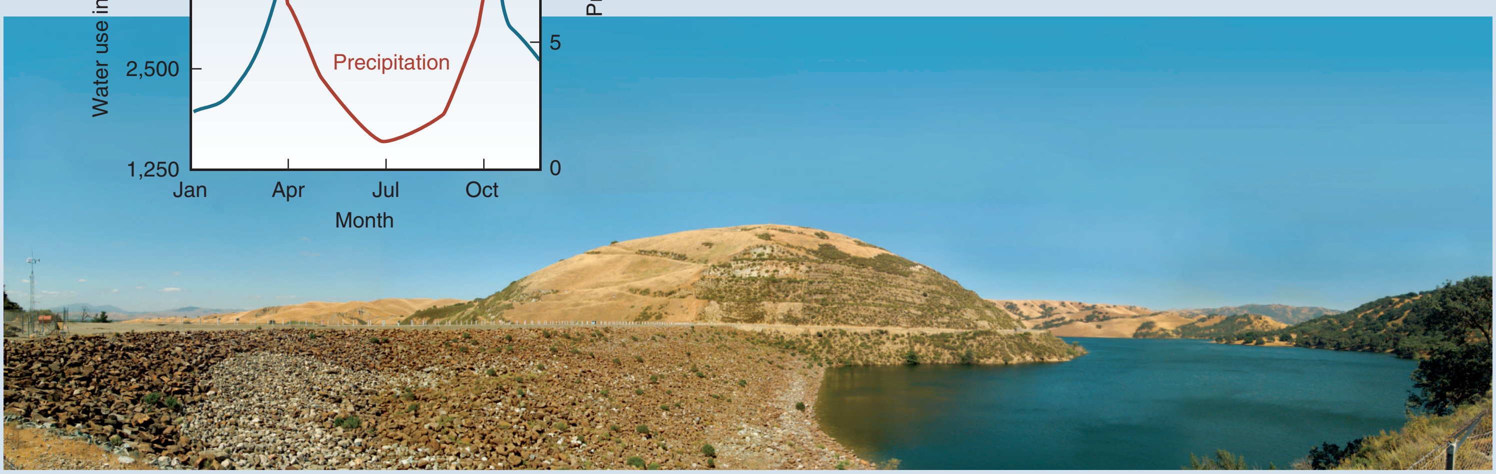


Figure 1. Two graphs, superimposed here, illustrate that water consumption and precipitation are out of phase in California. In this example, water use (red) in California's Livermore Valley peaks in the summer when precipitation (blue) is lowest, as measured in the Sierra snowpack (Donner Summit, CA, elevation 2150 m). The Sierra snowpack is the source of much of the Valley's water. Reservoirs, like the Del Valle reservoir in Livermore (below), store water from winter storms and the melting snowpack in the spring. Dams and reservoirs throughout California prevent flooding during the wet months and provide water during the summer, when it's needed most.



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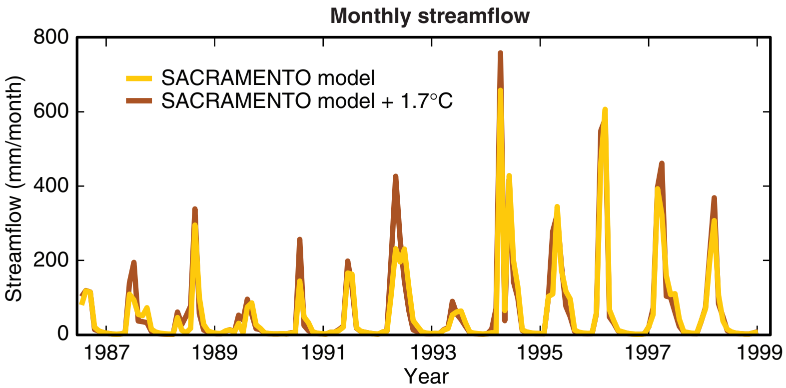

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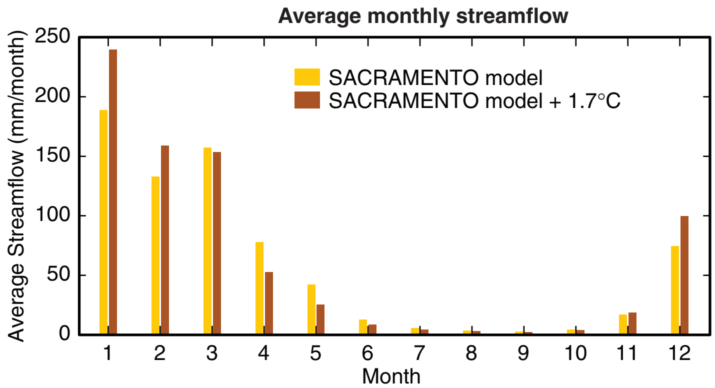


Figure 2. These graphs compare two SACRAMENTO regional climate model simulations of (a) monthly streamflow and (b) average monthly streamflow on the North Fork of the American River in California from 1987 to 1999. A temperature increase of 1.7°C (red line) is the only difference between the two simulations; precipitation is the same for both models. The increase in temperature, which could occur within several decades as a result of global warming, results in significantly higher peak runoff in (a), and changes the amount and distribution of the flow in (b), with increased flows in winter and reduced flows in the spring.

In one of three LDRD Water Initiative projects, "Predicting Effects of Climate Change/ Variability on Water Availability," Physicist Phil Duffy and his team of researchers are using a series of computational models, ranging from global and regional climate models to surface hydrology, to predict how climate change and year-to-year natural climate variability will affect surface temperatures, precipitation, and the amount and timing of water flow through rivers in California.

Their goal is twofold. First, they will use state-of-the-art models to predict likely effects of climate change on the hydrological cycle in California. Second, they will develop methodologies to quantify the uncertainties inherent in the predictions those models provide.

To make the best prediction of the effects of climate change on regional hydrology, the project team is running a sequence of high-resolution models, beginning at the global level, which provide parameters that drive regional models of finer resolution until they arrive at the surface hydrology of subregions within California. Global climate models (GCMs) divide the surface of the Earth into a grid, with each cell in the grid approximately 75 km on a side. Reducing the size of the grid acts like a microscope that provides higher resolution by including details of physical processes that are not "visible" at larger grid sizes. This "box-within-a-box" or "nested" method improves the accuracy of the model. By using a smaller

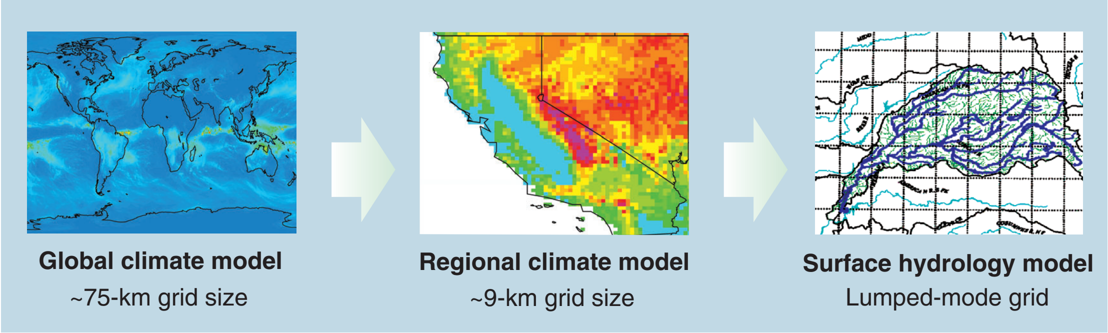


Figure 3. Simulations of California's hydrologic cycle are performed using a linked sequence of models. The global climate model, at 75-km grid size, simulates large-scale climate features and a nested regional climate model, at 9-km grid size, provides inputs to a surface hydrology model, which includes a more complete treatment of hydrological processes like river flow rates. The surface hydrology model uses a lumped-mode grid in which each watershed is represented as its own irregular grid cell.

(i.e., higher-resolution) grid in a regional model, the group will simulate the climate within the California region at a grid size of 9 km, and finally the surface hydrology of California watersheds (Figures 3 and 4).

The project has completed an initial set of simulations of the present and future climates in California using in-house models. To help assess the uncertainties in these simulations, the project also evaluated the results of 15 GCMs, produced at other research institutions, at 300-km resolution of a region that includes California, Nevada, and Oregon. Duffy and his colleagues also evaluated the results for this region in four nested regional climate simulations of present and future climate conditions at resolutions ranging from 36 km to 50 km.

Unlike their agreement on temperature increase, GCMs vary wildly in their forecasts of the impact of climate warming on precipitation, which is notoriously difficult to predict. Due to local topography and convective storms, precipitation can vary drastically over very short distances. Some GCMs show a 20% increase in precipitation, others show a 20% decrease (Figure 5). This level of uncertainty points to a need for improvements in climate models.

"There's no question that climate change and natural climate variability will affect the hydrological cycle in California. But how? Which model do you believe?" says Duffy. "The only way to make this information meaningful is to bound the range of outcomes and quantify the probabilities."

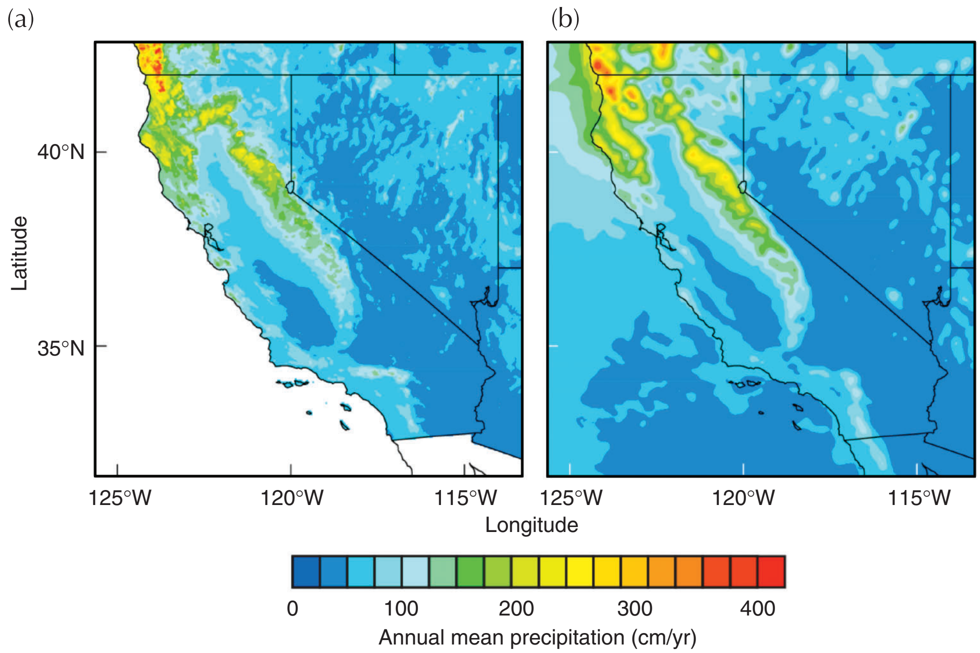


Figure 4. To validate the accuracy of their models, Duffy and his team compared (a) observation-based estimates of mean annual precipitation in the California region from 1971 to 2000 (4-km resolution) with (b) the results of their regional climate model (9-km resolution) of precipitation for the same period. The model closely matches the observed precipitation.

In an attempt to narrow the uncertainty in regional temperature and precipitation predictions, the project divided climate models into two groups: the "good" models—those that matched observed conditions—and the "bad" models—those that did not. However, a comparison of the predictions of the two groups showed no appreciable correlation between their past performance and their predictions for future climate change.

Next steps include collaborating with scientists at other research institutions to run simulations using a complete sequence of global, regional, and surface hydrology models, with multiple models of each type to offset the effect of variations between models.

In addition, work will focus on improving understanding of "extreme" hydrological events—droughts and floods—and how these events might be affected by global warming. Multiyear droughts, in particular, are poorly understood. California experienced four multiyear droughts in the 20th century, but geological data indicate that these events have been much more prolonged and severe in past centuries. Was the 20th century a "typical" century or an aberration?

"That's the advantage of computer simulations. We can use the model to investigate specific mechanisms that might cause or prolong multiyear droughts," says Duffy.

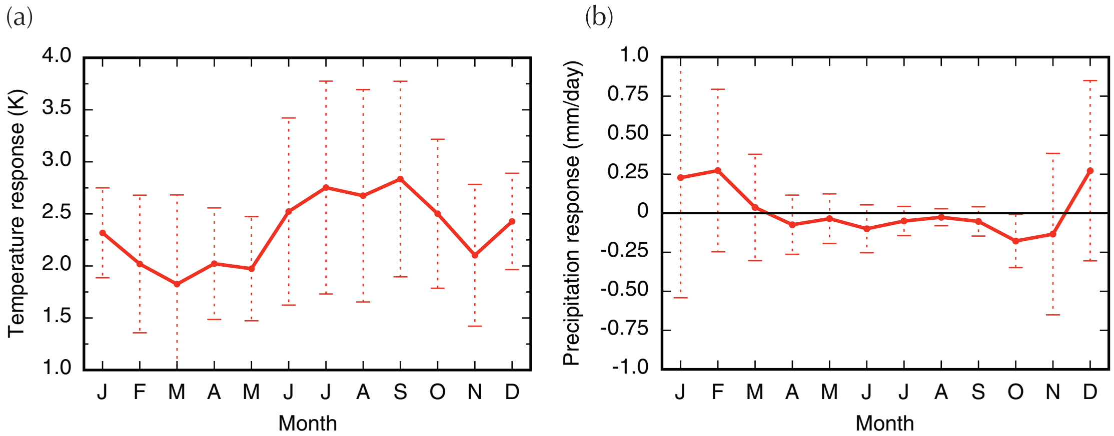


Figure 5. (a) Fifteen global climate models surveyed in this project all predict that doubling the atmospheric CO_2 will result in an increase in near-surface temperatures in the California region, though they disagree on how much temperatures will increase. (b) The same 15 global climate models disagree about the effect of doubled CO_2 on precipitation. In both graphs, the red curve is the mean of the 15 models and the error bars represent the standard intermodal deviation.

The final year of the project will also look at quantifying the uncertainties across simulations. In the future, this understanding could provide policymakers with better information about California's changing climate and help them determine if new infrastructure is needed to mitigate winter floods and summer water shortages.

Nitrate contamination puts drinking water at risk

Not all challenges to California's water supply will come in the future; some are here today. In recent decades, environmental and public health monitoring has revealed widespread and steadily increasing amounts of nitrate in California's vast surface and underground water resources. Nitrate contamination in water supplies, a pervasive and growing problem around the world, represents the leading drinking water contaminant in California and across the U.S. Nitrate is a nutrient, but too much nitrate can cause problems for human and animal health and the environment, with concomitant negative economic impacts. (See box on p. 48.)

Though nitrate—a water-soluble compound of nitrogen and oxygen—can come from natural sources like sediments, rocks, fixation of atmospheric nitrogen by plants, and lightening, the rapidly rising nitrate levels in groundwater have their origin in human activity. Major anthropogenic sources of nitrate include manure generated by concentrated animal production in feedlots and dairies, synthetic fertilizers applied to crops and landscaping, and discharge from septic systems and sewage treatment plants.

While other water contaminants, like trichloroethylene (known as TCE), are found in high-concentration plumes with easily identified point sources, nitrate contamination is often pervasive and low level, pointing to multiple nonpoint sources that are difficult to identify.

Nitrate contamination exceeds the federal regulatory drinking water standard of 45 ppm in about 10% of California public drinking water supply wells, and approaches that limit in a much larger fraction of wells (Figure 6). As many as 80% of the public drinking water supply wells in rural communities are affected by elevated nitrate levels.

Due to this contamination, California wells are being shut down at an alarming rate. About a third (8,600 of 25,000) have been closed since 1984. In the same period, the population of California has grown by over 40% and is projected to increase by another 50% in the next 20 years. As the population increases, the loss of drinking water supply to nitrate contamination will exacerbate California's water supply problems in the face of climate change and result in significant economic costs for the State.

To make informed decisions about managing the nitrate problem, water resource managers need both the ability to assess the impact of land-use practices on groundwater nitrate — to determine the source and timing of nitrate contamination — and the ability to simulate the evolution of groundwater — to understand the economic impacts of nitrate contamination and what steps would be most effective to mitigate the contamination.

The second project in the Water Initiative, "Nitrate Biogeochemistry and Reactive Transport in California Groundwater," led by chemist Brad Esser, is characterizing and simulating nitrate contamination with laboratory-, field-, and basin-scale experiments and computer simulations. Esser explains, "We're working to understand nitrate behavior in the subsurface from source to sink. Where did the nitrate come from? How and when did it get into the groundwater? Where is it going? And how is it transformed by biogeochemical processes along the way? Our ultimate goal is to gain enough knowledge to build scalable models of the reactive transport of nitrate in groundwater."

Esser's multidisciplinary team is taking an integrated approach, pooling their expertise in mass spectrometry, isotope hydrology, groundwater modeling, geostatistics, microbial kinetics, and molecular biology. About his team, Esser says, "Based on the capability the Lab's built up over the years for its national security and environmental missions—ranging from noble gas mass spectrometry, to parallel computing and geostatistics, to molecular biology — we're in a unique position to make scientific headway on the nitrate problem."

One of the major challenges facing Esser and his team is characterizing the heterogeneous nature of aquifers. Within a groundwater basin, the distribution of sediments and their associated hydraulic and geochemical properties can vary in three dimensions. Groundwater flowpaths, age distribution, and chemistry in the basin will reflect this heterogeneity.

To meet the heterogeneity challenge, Esser and his colleagues are bringing several techniques to bear. The team is using tritium/helium-3 age dating to determine the age of the water, geostatistical models of sediment distribution to address spatial heterogeneity in permiability, and massively

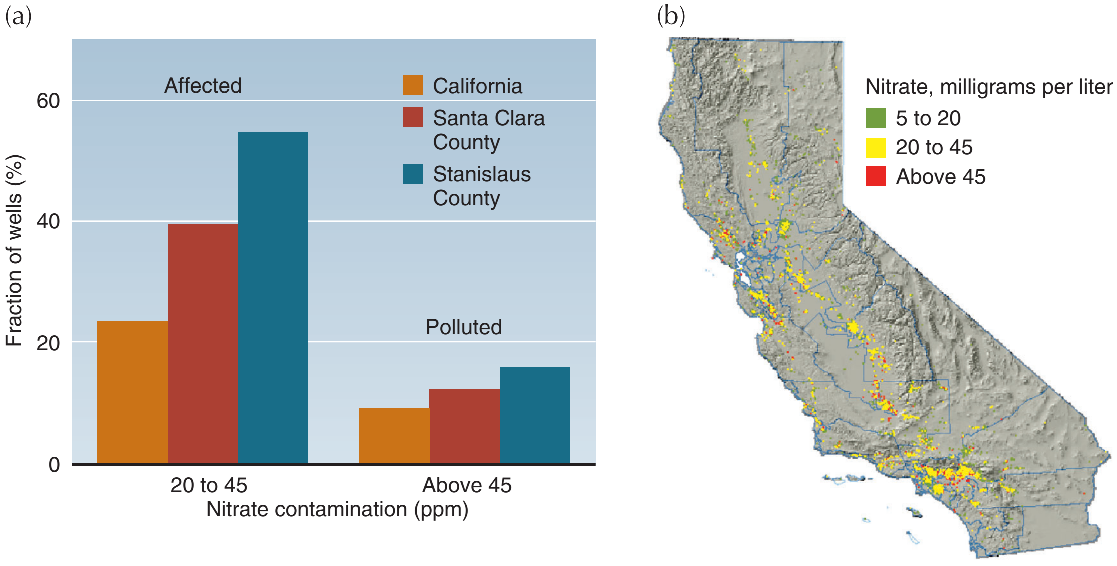


Figure 6. (a) In California, about 10% of public supply wells exceed the federal drinking-water standard for nitrate of 45 ppm. More than 50% of groundwater in Santa Clara County, formerly agricultural but now increasingly urban, is affected by nitrate. In agricultural counties, like Stanislaus, nitrate contamination is more acute, polluting up to 80% of groundwater. (b) Nitrate contamination in California groundwater is most prevalent in the States's major agricultural areas and around Los Angeles.

parallel models of groundwater flow, which are run on the Lab's supercomputers, to better understand groundwater flow paths and contaminant transport. With these models, water managers will be able to more accurately assess sources of nitrate contamination, the assimilative capacity of their basins for nitrate loading, the impact of different land-use patterns and management practices on groundwater nitrate, and the most cost-effective approach to remediation.

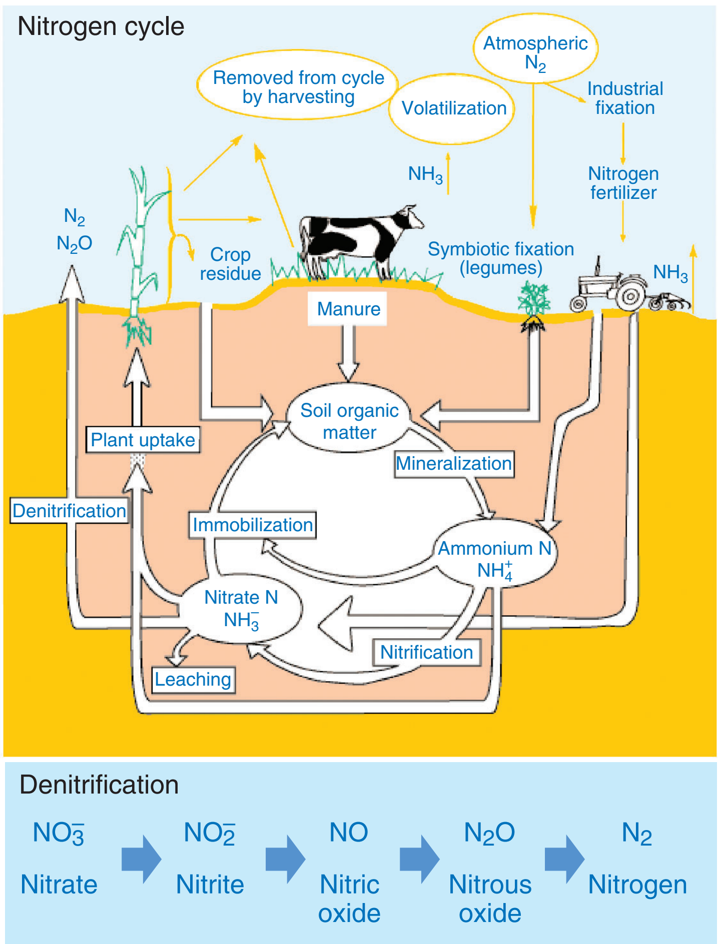


Figure 7. Agricultural fertilization, animal operations, and septic disposal systems are the three major contributors of nitrate to groundwater pollution. This diagram shows nitrogen cycling in the soil and vadose zone in an agricultural setting (NH_3 is ammonia). Septic system sources are not included in the diagram. The subsurface denitrification process converts nitrate to harmless atmospheric nitrogen.

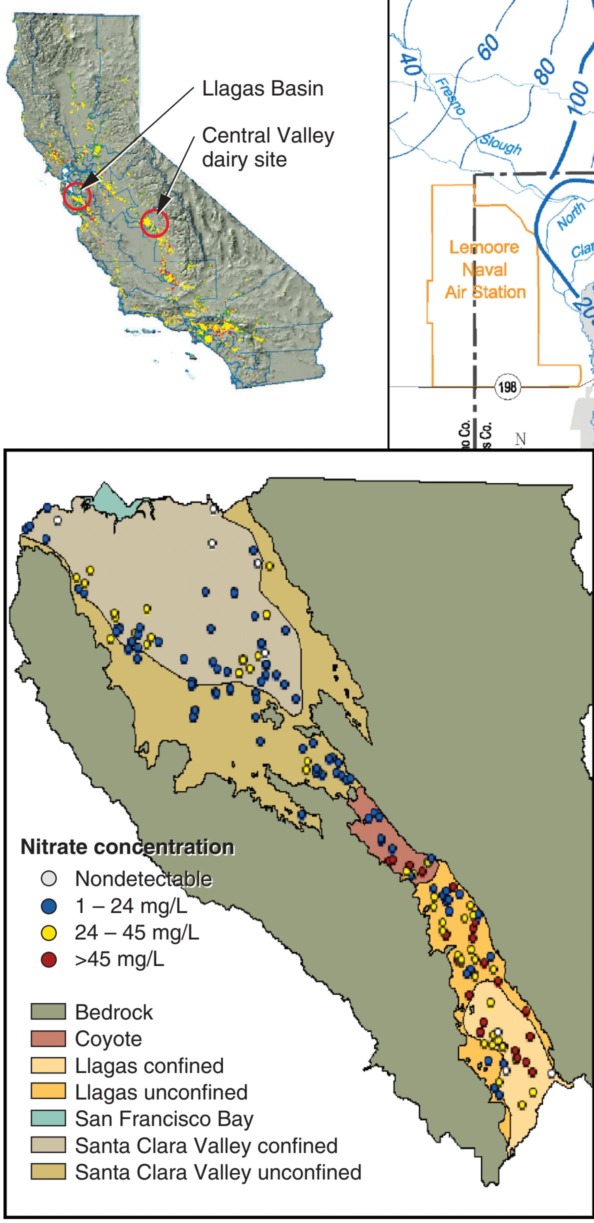
In addition to knowing how long the water has been in the subsurface and the path it followed to get there, understanding nitrate reactive transport and chemistry relies on knowledge of the biogeochemical cycling of nitrogen. Denitrification, the key process that influences the fate of nitrate in the subsurface, is a natural sink for nitrate. The rate of denitrification controls the capacity of a groundwater basin to assimilate nitrate (Figure 7).

The process of denitrification converts nitrate to nitrogen gas with the help of microbes, called denitrifiers, commonly found in soil. Although denitrifiers are ubiquitous, the geochemical conditions in an aquifer—low dissolved oxygen concentration and the presence of an electron donor—must be right for the denitrification to take place. While the denitrifying process is well understood, its role in controlling the assimilative capacity of California groundwater aquifers is not.

In FY2003, the team developed new tools to detect and characterize denitrification in groundwater. Researchers are using membrane-inlet mass spectrometry (MIMS) to measure the amount of excess nitrogen gas and dissolved oxygen in the groundwater, both indicators of denitrification. The MIMS technique, which only LLNL has applied to groundwaters, produces better estimates of excess nitrogen at lower cost than traditional gas chromatography methods. Determining excess nitrogen in groundwater has the added advantage of being able to estimate initial nitrate and the extent of denitrification over the entire groundwater flowpath. When combined with tritium/helium-3 age dating, the excess nitrogen method yields an integrated denitrification rate for the flow path, which is an important input for reactive transport modeling.

Real-time polymerase chain reaction (PCR) analysis provides another tool for detecting and characterizing denitrification. Though the Lab's PCR technology has most often been used to detect and quantify pathogenic bacteria for homeland-security missions, the technique can be used to quantify the population of denitrifying bacteria in a sample, based on a gene related to their metabolic activity rather than their phylogeny. Planned for FY2004, the next step—developing real-time PCR methods to analyze both denitrifier population and their denitrification activity—will lead to the ability to determine the rate of denitrification in aquifers as input to reactive transport models.

To demonstrate their integrated characterization and modeling approach at different scales, Esser's team is studying nitrate contamination at two field sites, one on a dairy farm set in the alluvial aquifers of the Central Valley and the other in a coastal aquifer, the Llagas Basin of the



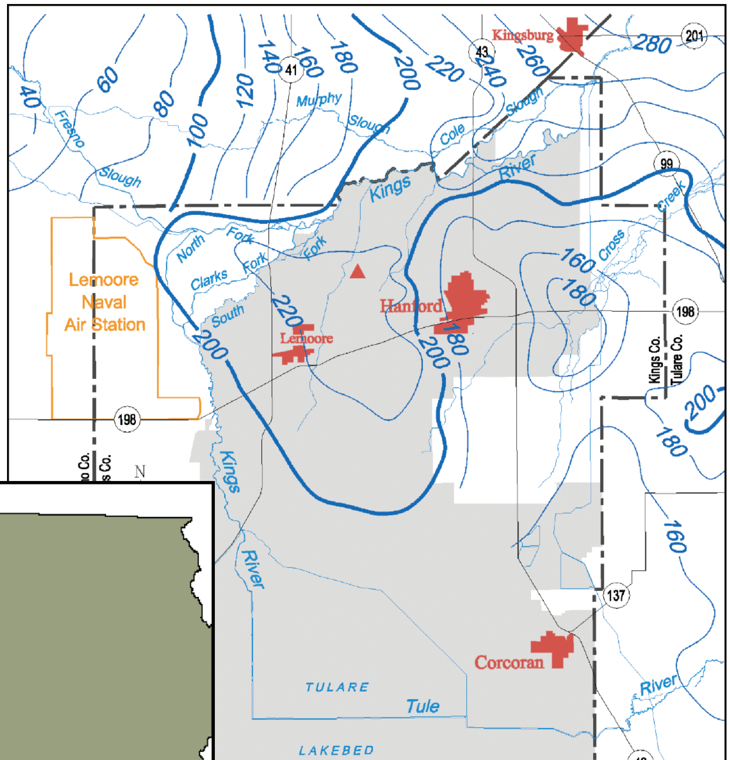


Figure 8. (left) The Llagas groundwater basin, which provides all the drinking water for Santa Clara County, is contaminated with nitrate. The LDRD team is developing a basin-scale reactive-transport model to determine if and when the pervasive nitrate contamination currently found in shallow aquifers will affect deep aquifers. (right) The red triangle on the map indicates the field-study site, located at a dairy farm in the Central Valley, where LDRD researchers will install monitoring wells to study groundwater flow paths, nitrate transport, and denitrification. Their goal is to understand how nitrate migrates from the source to the groundwater.

Santa Clara Valley, where land use is in transition from agriculture to urban. The Central Valley study focuses on shallow groundwater transport at the field scale (less than 10 km^2), while the Santa Clara Valley study focuses on basin-scale processes and the impacts of land-use change on groundwater nitrate patterns (Figure 8).

The research team chose the Central Valley for its field-scale study for several reasons. Groundwater is the single largest source of domestic and municipal drinking water in the region. Despite increasing urbanization, agricultural activities still dominate the economy and represent the most significant source of nitrate contamination in groundwater. Dairies, by far the largest type of confined animal operation in California and growing, are potentially significant contributors of nitrate to groundwater, producing 120 million cubic meters of liquid manure per year. Few detailed scientific studies have focused on the direct impacts of specific dairies on specific groundwater resources.

In FY2003, after surveying groundwater at 16 wells at different sites to characterize groundwater age and nitrate chemistry, the team identified a dairy farm in the Central Valley for its field-scale study and installed a multilevel monitoring well to collect shallow groundwater at discrete intervals.

The goal of this study is to improve understanding of nitrate movement between ground-level sources and shallow groundwater zones by characterizing spatial heterogeneity, groundwater recharge and flow, fixed nitrogen loading, and biogeochemistry. These data will be used to build a scalable stochastic model of the local flow system and to compare different methods for determining denitrification presence and rate. The work will be conducted in collaboration with the University of California, Davis; California State University, Fresno; and the California Water Institute.

What's So Bad about Nitrate?

Nitrate, a major component of fertilizer, is a nutrient that fuels plant growth and productivity. But when too much nitrate finds it way into the water supply, it can cause problems for human, animal, and environmental health. In babies, high nitrate levels can be fatal, interfering with the blood's oxygen-carrying capacity and causing blue-baby syndrome (methemoglobinemia). Elevated nitrate levels have also been associated with cancer and birth defects, though no firm link has been established.

Excess nitrate in the water can also cause environmental problems, many of which have economic impacts. Problems include oxygen-depleting algae growth in rivers and lakes, toxicity to aquatic life, increased animal abortion rates, and loss of quality in fruit and other crops.

Cleaning up nitrate contamination is costly. Wells must be deepened or shut down, clean water from another source must be added to lower contamination to acceptable levels, or water must be treated—all at considerable expense. Restricting the land-use practices that create nitrate contamination—intensive animal production and use of fertilizers—reduces agricultural revenues from those activities. Additional costs include loss of economic opportunity due to lack of an adequate water supply and a reduced tax base.

For their basin-scale study, Esser's team chose the Llagas Basin, which provides the sole water source for southern Santa Clara County. Since 1997, more than half the 600 wells tested have exceeded the federal drinking water standard for nitrate, with shallow nitrate and perchlorate contamination pervasive throughout the south County. The basin's water supply and storage capacity are key components of Santa Clara Valley Water District's (SCVWD's) plan for supplying clean water to its expanding population, and an active nitrate management plan has been in place for the past ten years.

Working with the SCVWD, the research team will develop a hydrogeologic reactive-transport modeling framework to analyze the source and timing of the contamination, what impacts land-use changes have and will make on the distribution of nitrate in Llagas Valley aquifers, and whether or not the contamination, which primarily affects shallow domestic wells, will migrate to deeper production wells in the future.

In FY2003, the project team sampled 60 wells in the Llagas and Livermore Basins to provide a comparison between groundwater in the two basins, and obtained extensive historical water-quality data and well logs from 330 Santa Clara wells. For the modeling effort, the team developed a three-dimensional permeability model of the Basin as part of a complete geostatistical lithology model and started testing geochemical inverse modeling techniques. In FY2004, work will begin on developing a basin-scale reactive-transport model.

Smart membranes to the rescue

Considering the pervasiveness of nitrate contamination in California groundwater, the most effective management practices implemented today will mitigate but not eliminate the nitrate problem. Treating the water to reduce nitrate contamination is the answer, but current water-treatment technologies, like reverse osmosis and electrodialysis, are energy intensive and nonselective—they remove all minerals from the water, not just the targeted contaminant, which creates an unnecessary waste stream. And ironically, the energy used to treat water can exacerbate the long-term water-supply shortage by increasing atmospheric CO_2 and adding to groundwater contamination. Using ion-exchange resins, another method for nitrate removal, means high labor costs associated with frequent regeneration and low selectivity for nitrate over other ions in the water.

Costly to operate and inefficient, all of these technologies are reserved for desalinating brackish water or treating only the most contaminated water where no other source of fresh water is available. An energy-efficient nitrate removal technology would have a major impact on water availability in California and around the world.

The third project in the Water Initiative, "Molecular Engineering of Electrodialysis Membranes," under the leadership of geochemist Bill Bourcier and engineer Kevin O'Brien, applies the latest advances in materials science, nanofabrication, and computer modeling to improve the energy efficiency and selectivity of an old water-treatment technology—electrodialysis—for treating nitrate contamination. Their goal is to improve energy efficiency by 50%.

Of the two commercial membrane-based water-treatment processes most frequently used to remove nitrate, electrodialysis and reverse osmosis, the project team targeted the electrodialysis process to improve on. "Electrodialysis has been around since the 1950s, but little work has been done to make it selective or more energy efficient since then," says Bourcier. "It's time to apply new materials science to water treatment."

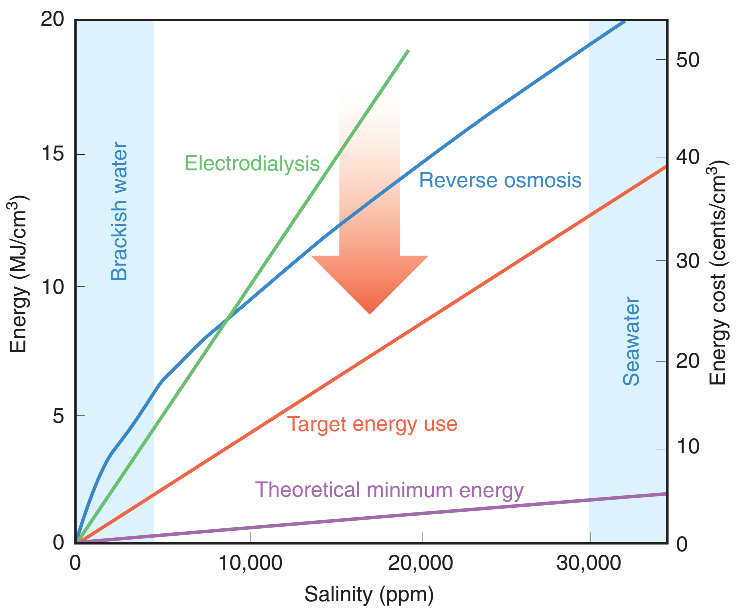


Figure 9. (S&TR w/ markup) Because smart-membrane electrodialysis uses much less energy than traditional electrodialysis and other water treatment methods, this technology promises to lower the cost of water treatment by as much as 50%.

With lower capital costs than reverse osmosis, electrodialysis requires less energy than reverse osmosis at low salt concentrations, but much more energy at high concentrations. Cutting the energy required for electrodialysis by 50% would make electrodialysis the more energy-efficient treatment technology at every concentration level (Figure 9).

Electrodialysis uses an electrical potential and electrically charged membranes to purify a liquid stream. Commonly used to desalinate brackish water, electrodialysis is one of the main processes for producing potable water in many areas of the world. Since the food, drug, and chemical industries also use electrodialyis extensively, a significant improvement in the process would benefit many sectors of the economy.

An electrodialysis cell consists of a series of alternating anion (negative) and cation (positive) polymer membranes separated by compartments about 1-mm wide. The electrodes are located at either end of the cell, the anode at one end and the cathode at the other. When an electrical potential is established between the two electrodes and an ionic solution is pumped through the cell, the ions in the liquid migrate toward the electrode with the opposite charge—

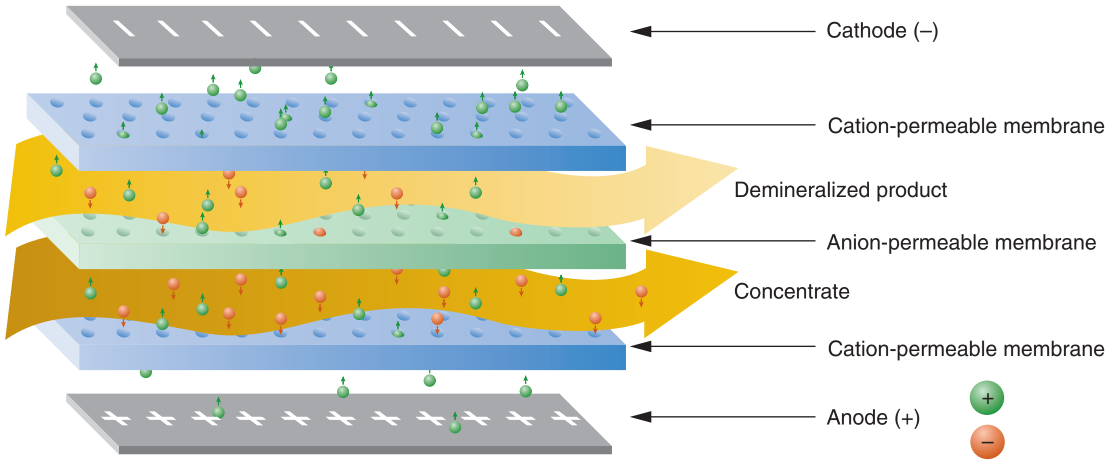


Figure 10. Schematic of a typical electrodialysis cell. Ions pass through membranes of opposite charge but are stopped by membranes with the same charge. Ions concentrate in alternating compartments while the solution in the other compartments becomes depleted of ions, or demineralized.

anions toward the cathode and cations toward the anode. The membranes that separate the compartments. If a membrane has the opposite charge of the ion, the ion moves through the membrane into the next compartment act as gates. But if the membrane has the same charge as the ion, the membrane blocks the ion from moving through. As a result, the ions become concentrated in the solution in alternating compartments, while the solution in the other compartments becomes depleted of ions (Figure 10).

The selectivity of the ion-exchange membranes determines how efficiently the cell removes the targeted ions, while the energy use depends on how much energy it takes to pump the solution through the unit and the amount of energy needed to force the ions through the membranes. The cost of the cell is a function of the membrane area needed to meet the target capacity.

To improve the selectivity, efficiency, and cost of the electrodialysis process, Bourcier and colleagues are developing an energy-efficient "smart" membrane to replace the polymer membranes used today. Smart membranes, so called because they target and remove specific contaminants, are fabricated from aligned pores in polycarbonate membranes produced using ion track etching. A sample 1 cm in diameter contains one billion nanopores with diameters of 10 nm. (Figure 11)

The membrane pores are either coated with a metal conducting layer that can be electrically charged, or functionalized with a chemical group that is tailored to attract a specific molecule. Although this project focuses on developing a smart membrane specifically to remove nitrate, the same methods can be used to design membranes to remove other species by varying membrane pore size, coatings, and electrical charge to match the targeted element. Work is in progress to determine the optimum membrane characteristics that are most effective for nitrate removal.

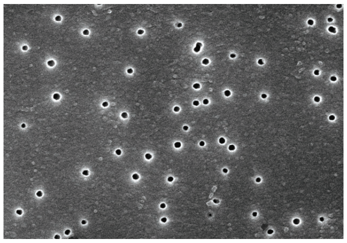


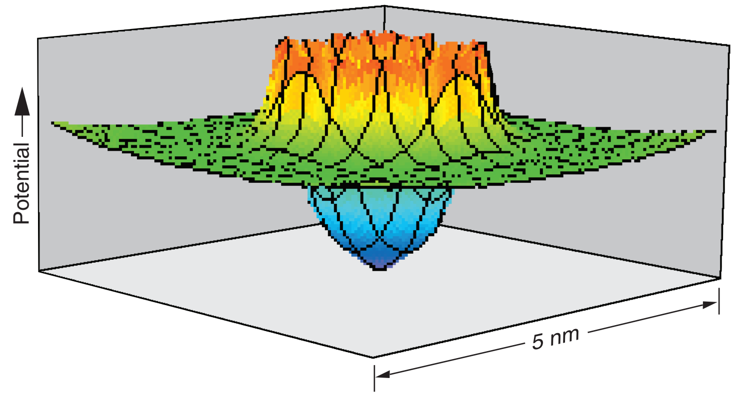
Figure 11. The 10-nm-diameter pores drilled in this "smart" membrane are the right size for nitrate ions to pass through.

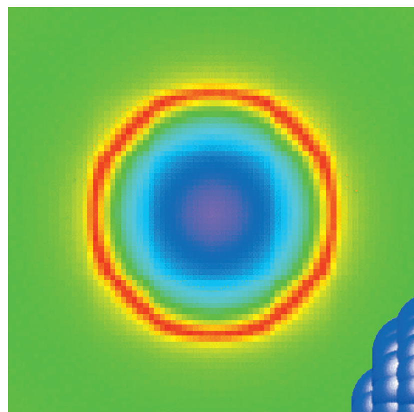
The team brings expertise in computer modeling, nanoscale fabrication techniques, and testing to the task of developing a smart membrane that targets nitrate. The ability to design and model the membrane on a computer before fabrication and testing avoids a lot of trial and error. The Laboratory has unique supercomputing capabilities and codes for modeling ion transport through membranes with charged or functionalized pore surfaces. For this project, physicist Bill Wilson is using Laboratory codes and a novel numerical method for calculating electrostatic fields in the vicinity of complex nanostructures. With quantum mechanical modeling, the team can simulate ion transport behavior under conditions of varying electrostatic charge, functionalized pores, membrane materials, and solvent medium to determine the optimum design for targeting nitrate or other contaminants (Figure 12).

Using a specialized nanofabrication technology developed at LLNL, in FY2003 the team synthesized porous polycarbonate membranes, with pores created by an ion-beam etching process. They developed modeling codes for calculating electric fields and ion transport through the membranes, and began testing to determine membrane selectivity and energy use.

In FY2004, they expect to produce a proof-of-principle smart membrane with pores functionalized to remove nitrate from the feed-water supply. If successful, the membrane will be incorporated into a benchtop electrodialysis unit for further testing.

Kevin O'Brien says, "Our next major challenge will be to bridge the gap between the laboratory and the field, where the electrodialysis unit will need to withstand the conditions found in the natural environment— corrosion, scaling and fouling, and microbes." Eventually, smart membranes will find many applications in water treatment, including resource extraction, pathogen detection, semiconductor manufacturing, and cleanup of boiler water for electrical power plants.





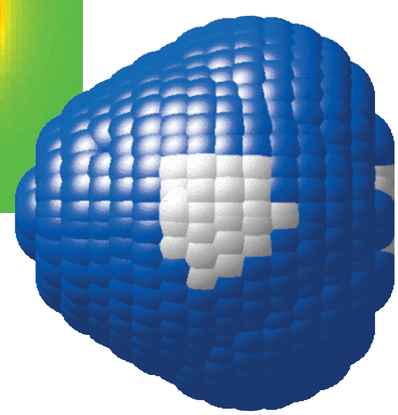


Figure 12. These quantum mechanical simulations of a smart membrane nanopore, viewed from (a) the side and (b) the top, show the electric field gradients around the pore surface, with blue being the lowest voltage and red the highest voltage. By taking into account the targeted ion's shape and charge distribution, quantum mechanical modeling allows engineers to design the pores with the exact size and electrostatic field to attract and isolate the targeted ion. (c) This model of a nitrate ion shows the charge distribution. White indicates an area of negative charge.

Water security begins at home

With a variable climate, a complex topography that contains both the lowest (Death Valley) and highest (Mount Whitney) elevations in the continental United States, the fifth largest economy in the world, and the biggest State population in the U.S., California offers a rich microcosm for studying water resources. The LDRD-funded Water Initiative brings the Laboratory's many scientific resources together to study water in California. Begun in FY2003, the three projects of the Water Initiative are investigating water resources from their source in the Sierra snowpack, through the fate and transport of nitrate contamination, to innovative water-treatment technologies.

As Bill Bourcier says, "The Water Initiative is a unique scientific look at end-to-end solutions for water availability and quality in California. What we learn here will be a modest step, but one that may benefit millions of people around the world who don't have access to clean water. It's exciting science that makes a difference."

— Karen Kline

For more information see the project summaries (on CD only) for projects covered in this article:

Predicting Effects of Climate Change and Variability on Water Availability
Philip B. Duffy, Principal Investigator, 03-ERD-042

Nitrate Biogeochemistry and Reactive Transport in California Groundwater
Bradley K. Esser, Principal Investigator, 03-ERD-067

Molecular Engineering of Electrodialysis Membranes
William Bourcier, Principal Investigator, 03-ERD-060

FY2003 Portfolio

Investing in our nation's future

The FY2003 LDRD Portfolio

The FY2003 LDRD portfolio was carefully structured to continue the Program's vigorous support for the strategic vision and long-term goals of DOE/NNSA and LLNL. The FY2003 projects described in this *Annual Report* underwent a stringent selection process and received ongoing management oversight.

In FY2003 the Program funded 210 projects with a total budget of $64.9M. Figure 1 shows the number of projects in each of the four categories. Figure 2 shows the distribution of funding among the four LDRD project categories.

Strategic Initiative

In FY2003, the LDRD Program funded 12 SI projects. Although the SI category represented only 6% of the total number of LDRD projects for FY2003, it accounted for 25% of the budget. SI projects ranged in funding from $283K to $2,161K.

Exploratory Research

In FY2003, 164 ER projects were funded. The largest project category, ERs account for 78% of LDRD projects for the fiscal year. Projects in this year's ER category ranged in budget from $21K to $1,133K.

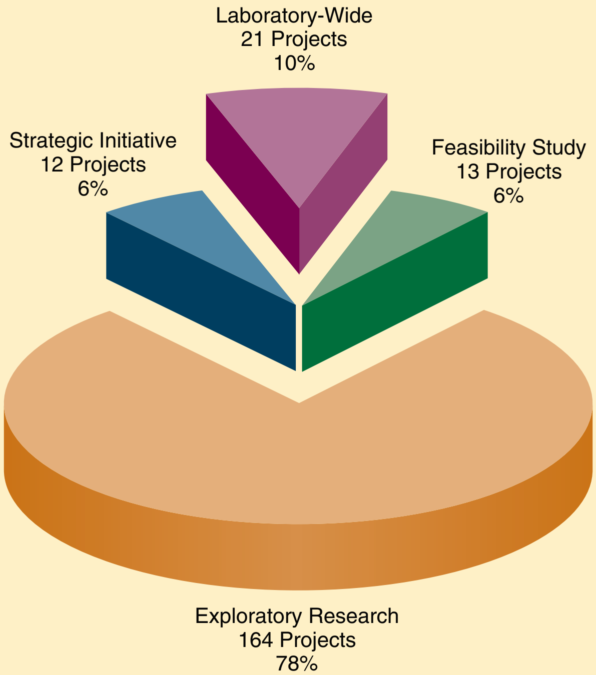


Figure 1. Number and percentage of the 210 LDRD projects in each project category in FY2003.

Laboratory-Wide Competition

Twenty-one LW projects were funded in FY2003, which represent 10% of LDRD projects for the year and 5% of the budget. Laboratory-Wide projects are limited to $190K/yr funding, with a few exceptions. In FY2003, LW projects ranged in funding level from $14K to $233K.

Feasibility Study

In FY2003, the Program funded 13 FS projects for 6% of the total. Feasibility Studies are limited to $75K and 12 months duration.

Figure 3 shows the funding distribution by dollar amount for the 210 FY2003 projects. Seventy percent of the projects were in the $101K to $500K range, with 21% falling below $100K. This lowest funding level included all the FS projects. Eight percent of the projects were in the $501K to $1M funding level and only 5% of the projects received more than $1M. The average funding level for the 210 projects was $309K.

Figure 4 shows the percentage of Program funding and number of projects in each competency for FY2003.

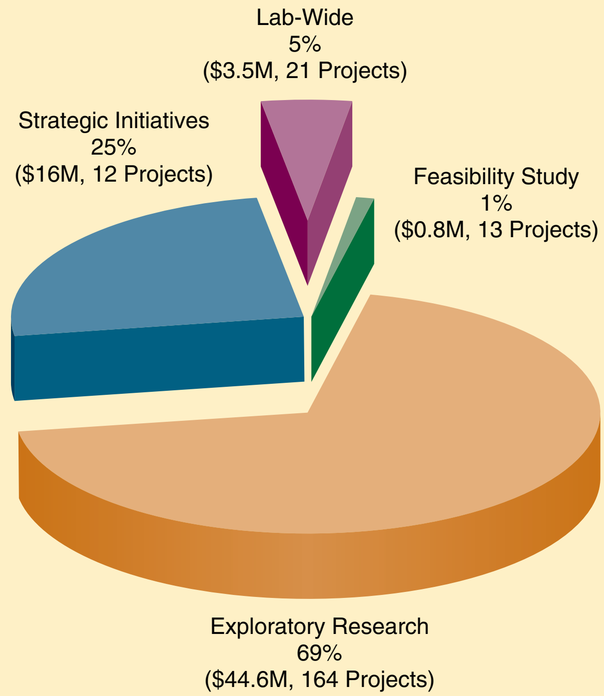


Figure 2. Distribution of funding (in $M) among the four LDRD project categories. Total funding for FY2003 was $64.9M.

FY2003 LDRD Program Accomplishments

Over the years, LDRD-sponsored projects have realized major scientific and technical breakthroughs that have been widely reported in the technical community. This section presents a few indicators of the achievements attained by FY2003 LDRD projects and principal investigators. The "Highlights" section of this year's *Annual Report* presents four in-depth articles that showcase the scientific highlights of the FY2003 portfolio.

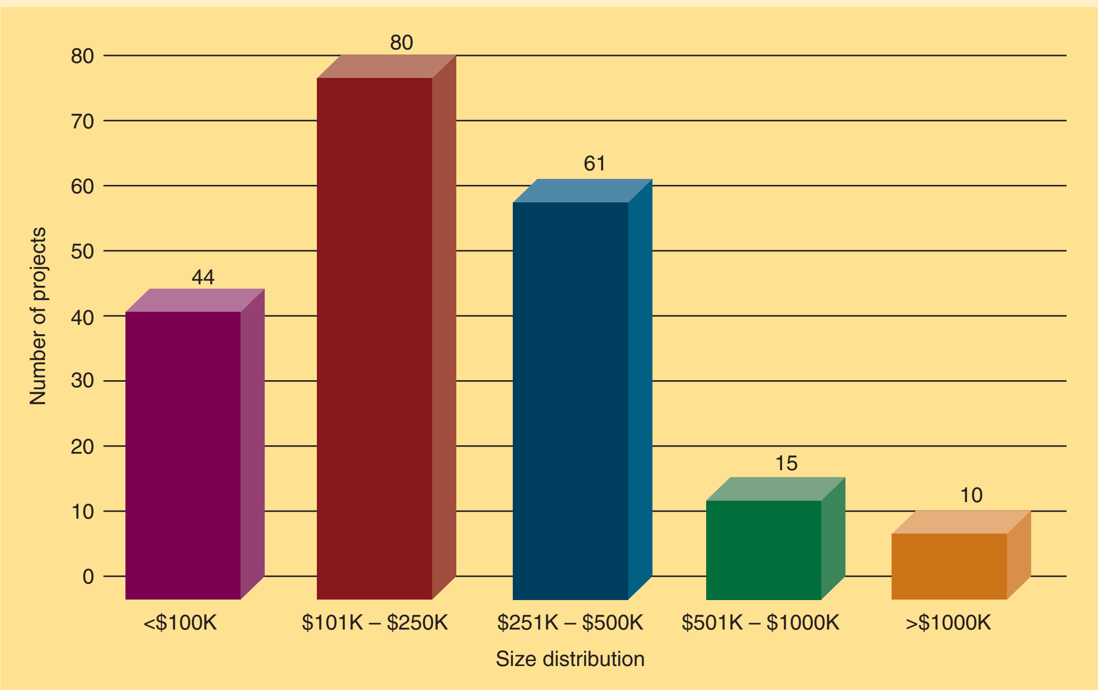


Figure 3. Number of projects and levels of funding (in $K). The average funding level for an LDRD project in FY2003 was $309K.

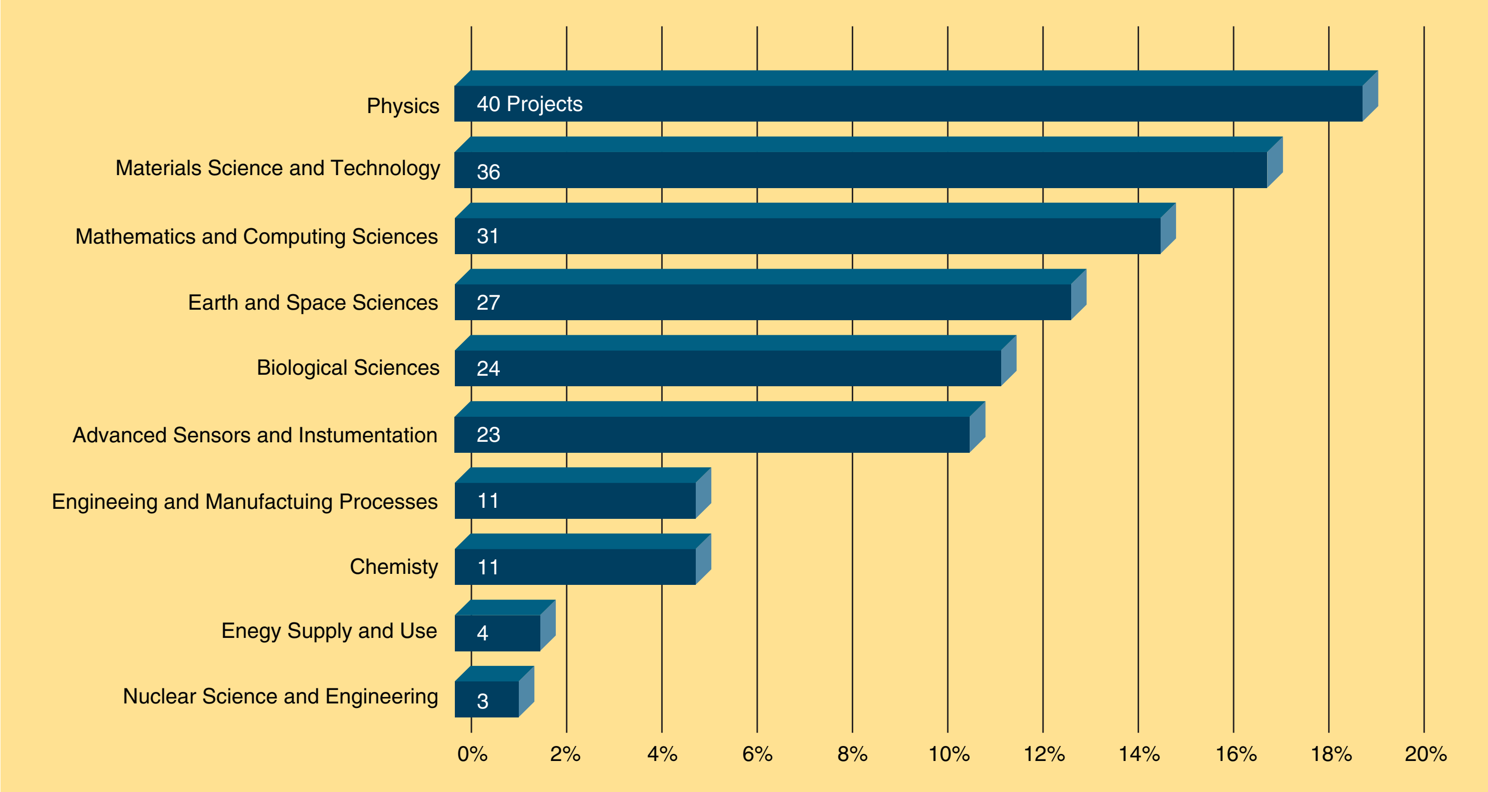


Figure 4. Percentage of LDRD funding and number of projects in each research category in FY2003.

Table 1. Patents resulting from LDRD-funded research as a percentage of all LLNL patents from 1996 to 2003.

	1996	1997	1998	1999	2000	2001	2002	2003
All LLNL patents	83	64	78	84	93	89	97	59
LDRD patents	35	29	39	45	35	42	27	25
LDRD patents as percentage of total	42	45	50	54	38	47	28	56

Patents

Projects sponsored by LDRD consistently account for a large percentage of the patents issued for LLNL research, especially considering that the LDRD Program funding represents 6% of the total LLNL budget, with the exception of FY2000, when the Program received only 4% of the budget. Table 1 shows the number of patents resulting from LDRD-funded research since 1996.

Awards

R&D 100 Awards

In 2003, LLNL technologies won six R&D 100 awards. Of these, past LDRD support directly contributed to three:

- High-Average-Power Electro-optic Q Switch (LDRD projects 93-ER-108 and 97-SI-014)
- MEMS-Based Adaptive Optics Phoropter (LDRD project 01-LW-036)
- The Biological Aerosol Sentry and Information System (BASIS) (with Los Alamos National Laboratory) (LDRD project 01-SI-002)

Professional Society Fellows

In 2003, the following recent LDRD principal investigators were recognized by their national and international peers for outstanding scientific contributions in their profession.

- William Durham, Fellow, American Geophysical Union
- Giulia Galli, Fellow, American Physical Society
- Siegfried Glenzer, Excellence in Plasma Physics Award, American Physical Society
- Richard Klein, Fellow, American Physical Society
- Erich Ormand, Fellow, American Physical Society

LLNL FY2003 Science and Technology Awards

One of two LLNL FY2003 Science and Technology Awards went to teams whose research was conducted with substantial LDRD support. While these awards are internal, they are based on national and international recognition of LLNL scientific achievements.

Complete Phonon Dispersion Curves in Gallium-Stabilized Delta-Plutonium
Team: Joe Wong (lead), Carl Boro, Dan Farber, Florent Occelli, Adam Schwartz, Mark Wall

Phonon dispersions are fundamental to understanding the properties and behavior of plutonium (Pu) and its alloys. Under FY2003 LDRD funding (project 03-ERD-029), a Livermore group prepared thin, polycrystalline Pu targets and measured both longitudinal and transverse phonon spectra by high-resolution inelastic x-ray scattering at the European Synchrotron Radiation Facility in Grenoble, France. The data show several unusual features related to phase transitions in Pu and to strong coupling between the lattice structure and the 5f valence instabilities. An article describing the results of this work has been published in *Science* magazine.[4] The new phonon data acquired in this project will greatly enhance scientists' basic knowledge of the transformations and phases Pu undergoes in different environments and over time. This understanding contributes greatly to the Laboratory's science-based stockpile stewardship mission to ensure the safety and reliability of the nation's aging nuclear weapons without testing.

[4] Wong, J. et al. (2003). "Phonon dispersions of fcc delta-plutonium-gallium by inelastic x-ray scattering." *Science* **301** (5636), 1078–1080.

Advanced Sensors and Instrumentation

SATRN: Secure Air–Optic Transport and Routing Network

Anthony J. Ruggiero

01-SI-010

Abstract

Information collection and communication technologies are crucial to national security. Because communication bottlenecks limit the timely synthesis and interpretation of data from sensor systems, maintaining information dominance requires secure, high-capacity communication networks. Laser communication (lasercom) links are a key part of this vision. The Secure Air–Optic Transport and Routing Network (SATRN) project develops and demonstrates enabling technologies for long-range, air–optic lasercom. Through a series of system-level horizontal-path link experiments we are establishing the viability of high-capacity data transport over long ranges. Link data from our horizontal lasercom demonstration test beds can be used to calibrate performance models and simulations of terrestrial links to airborne surveillance and reconnaissance platforms.

Mission Relevance

This work directly supports the NNSA's national-security mission by enhancing the ability to transport critical intelligence data from sensor platforms involved in proliferation detection. The technology resulting from this project will fulfill a national-security need for secure information collection and communication systems. Technology for secure, high-capacity, long-range communications will help overcome the current communication bottlenecks that limit timely synthesis and interpretation of intelligence, surveillance, and reconnaissance data.

FY03 Results

Work conducted in FY03 included (1) optimizing the transceiver through multichannel differential phase-shift keying and matched filtering; (2) implementing advanced technology in 29-km link experiments to improve link availability and reduce the error rate; (3) conducting forward error correction air-link and microelectromechanical systems adaptive optics (AO) receiver system experiments; (4) developing nonlinear AO systems; (5) performing experimental work on erbium–ytterbium-doped photonic crystal fiber amplifiers for high-power wavelength division multiplexing applications; (6) conducting preliminary laser communications uplink and downlink experiments with mobile platforms; and (7) completing an end-to-end modeling capability that includes atmospherics, boundary-layer effects, and pointing and tracking.

The major goals of the SATRN project were achieved over the project's duration: We successfully demonstrated hyperspectral data transfer over an air link; evaluated conventional and unconventional AO approaches; developed and validated laser-beam propagation and performance models, advanced signaling and custom air–optic photonic transceiver components, and a fade-tolerant forward error correction technique; and completed numerous air-link experiments under a variety of atmospheric conditions. As part of this effort, a terrestrial air–optic distance–bandwidth record was set by closing a duplex 100 Gbit/s link over a 28-km ground-to-mountain range. In summary, we created a national resource and a ground validation test bed for tactical lasercom.

Publications

Johnson, G. W. et al. (2003). "Characterization of fiber channel over highly turbulent optical wireless links." *Proc. SPIE* **5160**, 37. UCRL-JC-151555.

Ruggiero, A.J. (2003). "Overview of air–optic laser communications efforts at LLNL." *Proc. SPIE* **5001**, 06. UCRL number.

Ruggiero, A. J. et al. (2003). "Application of a curvature-sensor-based modal adaptive optics system to long-range fiber coupled FSO links." *Proc. SPIE* **5160**, 18. UCRL-PRES-151552.

Ruggiero, A. J. et al. (2003). "High-capacity 28-km ground-to-mountain link experiments and FSO testbed." *Proc. SPIE* **5160**, 59. UCRL-MI-151553.

Scharlemann, E.T. et al. "Modeling of long-range atmospheric lasercom links between static and mobile platforms." *Proc. SPIE* **5160**, 35. UCRL-JC-151554.

Disposable Polymerase Chain Reaction Device

Elizabeth K. Wheeler 01-ERD-009

Abstract

Planning and equipping for responding to a biological attack requires detection devices that are robust in the field, easy to use, and relatively inexpensive. We have fabricated a low-cost disposable polymerase chain reaction (PCR) thermal chamber that uses thermal convective forces created by fixed, nonfluctuating, hotter- and cooler-temperature regions to thermally cycle the sample fluid to achieve amplification. Buoyancy forces cause the fluid to flow through the different temperature zones. The advantage of this approach is lower power requirements, ensuring that small batteries will suffice as the system's power source. The thermal cycler is also fabricated from inexpensive material, lowing the cost of manufacturing the device.

The device has been optimized to be the minimum system needed for its application, ensuring battery operation. Target users for the device are (1) first responders at the scene of a potential biological pathogen release and (2) medical personnel who perform clinical diagnostics. Biological samples have been amplified using this novel thermal chamber. Time for amplification is less than 30 min. More importantly, energy consumption is significantly improved over current handheld technology.

Mission Relevance

This project leveraged LLNL's expertise in microtechnology and instrumentation to provide new capabilities in support of LLNL's national-security mission. Specifically, the PCR device developed in our research will enable first responders (e.g., civilian or military personnel) at the scene of a suspected biological pathogen release, as well as medical personnel who perform clinical diagnostics, to quickly identify biological pathogens.

FY03 Results

During FY03 we tested and optimized the convectively driven PCR thermal cycler on a biological surrogate, starting with genomic DNA. Both 58- and 160-base-pair segments of *Erwinia herbicola* (relevant to both medical and biowarfare arenas) were amplified.

The convectively driven PCR device was shown to minimize heat losses to the environment and maximize the energy transferred to the fluid. Compared to existing field-deployable technologies, our convectively driven PCR thermal chamber lowers energy consumption per unit volume by an order of magnitude. Based on the low energy requirements of the convectively driven thermal cycler, we fabricated a battery-operated thermal cycler. For this iteration, the volume was decreased to 25 μL, further reducing the power requirements of the thermal cycler.

Publications

Wheeler, E. K. et al. (in press). "Convectively driven polymerase chain reaction thermal cycler." *Analytical Chemistry*. UCRL-JC-152544.

Wheeler, E. K. et al. (2003). "Convectively driven polymerase chain reaction thermal cycler." *Proc. Micro Total Analysis Systems Symposium 2003* **2**, 1133. UCRL-JC-152544 Abs.

Ultrawideband Communications

Farid U. Dowla 01-ERD-047

Abstract

The national-security need exists for short-range covert wireless communications based on low-power, small-size sensor communication hardware with a low probability of detection (LPD) and a low probability of interception (LPI). Moreover, many intelligence applications have the need to rapidly collect and transmit data covertly and reliably and for communication systems to easily interface with other systems for analysis or long-distance links. Commercial communication systems are not viable because they operate in fixed-frequency bands and are easily detectable. We are preparing our ultrawideband (UWB) communication systems for national security applications.

We have identified key operational requirements for covert communication systems: (1) voice communications (20 kbps) over 1 km for secure voice links; (2) high-speed communication through walls with multipath capabilities; and (3) network communication for a large number of sensors within a local area, including multipath.

Mission Relevance

Communications systems with LPD and LPI capabilities will have multiple covert-security applications in support of national and homeland security missions.

FY03 Results

In FY03, we demonstrated UWB communication for small, low-power transmitters and receivers at 2 Mbps over short distances (20 m). We also demonstrated longer-range communications over 1 km at significantly lower data rates, multinode hopping capability with sensors, and simple networks of UWB radios. We also built a portable battery-operated UWB bit error rate tester. Initial results validate performance models for LPD and LPI, as well as excellent bit error rate through walls at a distance of up to 20 m.

Stroke Sensor Development using Microdot Array Sensors

Chance Carter 01-ERD-101

Abstract

In the U.S., over 700,000 people annually suffer a stroke, which is a major cause of mortality and disability. Currently, the only approved therapy for treatment of acute ischemic stroke is intravenous thrombolytic drugs, which must be administered within 3 h of the onset of symptoms; drugs administered after this may result in a fatal intracranial hemorrhage. Consequently, only 2 to 3% of people with acute stroke are treated with these drugs. We propose to develop and demonstrate a minimally invasive, optical-fiber-based imaging sensor for rapid, in vivo measurements of multiple stroke biomarkers (e.g., pH, blood gas, and enzymes). Such a tool could extend the use of thrombolytic drug treatment to acute stroke victims outside the current limitation of 3 h.

Successful completion of this project will result in a unique multi-analyte biosensor platform. Specifically, we expect to (1) demonstrate the first reproducible fiber-based sensors and the first fiber-based enzyme biosensor; (2) develop a science and technology base in multi-analyte fiber-based sensors; (3) develop new biosensors for disease diagnostics; (4) apply this technology to stockpile surveillance; (5) develop a portfolio of intellectual property; and (6) recruit new postdoctoral researchers and provide topics for graduate students.

Mission Relevance

In support of LLNL's bioscience mission, the microdot-array sensor platform will complement existing stroke-related efforts, in which mechanical devices are being designed for removing endovascular obstructions in the brain. In addition, this technology is potentially applicable to stockpile surveillance for multi-analyte fiber-based sensing.

FY03 Results

In FY03 we developed (1) a biocompatible, printable polymer hydrogel with properties optimized for printing and polymerizing pH and enzyme sensors, (2) a custom software package for our nonpiezo printing system for printing the hydrogel microdots on fiber optics and other waveguide surfaces, (3) a technique for treating fiber surfaces before hydrogel printing to provide reproducible hydrogel shapes and adherence to the fiber surface, (4) a second-generation pH sensor, (5) a second-generation fluorescence resonance energy transfer-based polypeptide for detecting a target matrix metalloprotease (MMP) enzyme, and (6) chemistry for the oxygen sensor based on dye-doped nanoparticles trapped in a printable polymer hydrogel.

Proposed Work for FY04

In FY04 we will demonstrate (1) a blood-gas sensor, which requires trapping the oxygen nanoparticle indicators in a printable polymer hydrogel and tailoring the printing and polymerization conditions; (2) a reproducible enzyme sensor, which requires testing our second-generation MMP enzyme sensor; and (3) a calibrated reproducible multi-analyte biosensor for pH, O_2, and MMP. In addition, we will perform biocompatibility studies on our existing sensor design and investigate ways to resolve common problems with fiber-based in vivo sensors, such as degradation and encapsulation of the indicator chemistries attached to the fiber.

Retrospective Plutonium Biodosimetry by Modeling Urinary Plutonium-239 from Archived Occupational Samples

Kenneth T. Bogen

01-ERD-108

Project Description

Fundamental uncertainties about long-term plutonium (Pu) excretion by humans now seriously limits new applications of ultrasensitive Pu-detection technology to national security, counterterrorism, and DOE occupational health goals. This project applies accelerator mass spectrometry (AMS) to recover previously inaccessible information concerning long-term urinary excretion patterns of ^{239}Pu (which is specific to nuclear weapons) for 11 LLNL Pu workers. These workers had been monitored periodically via urine sampling analyzed by alpha spectrometry (AS) over a period of decades. Using AMS methods far more sensitive than AS, we are studying ^{239}Pu excretion biokinetics using archived AS discs with unprecedented sensitivity. We have identified a long-term excretion pattern that should be useful as a weapons-program signature for nonproliferation and homeland-security applications.

Mission Relevance

By developing a new Pu-detection method and compiling LLNL Pu-worker data, our project supports the Laboratory's occupational safety and national security missions by providing a unique national resource with which current fundamental uncertainties about human Pu biokinetics can be resolved. Furthermore, this work is highly relevant to LLNL's mission in nonproliferation and homeland security.

FY03 Results

In FY03 we conducted a total of three AMS runs, in which a combined total of 384 AMS targets (including unknowns, standards, AMS system backgrounds, tuning samples, and tracer tests) were analyzed for three Pu isotopes: (1) ^{242}Pu, added as a tracer isotope spike to indicate processing yield and to normalize AMS measurement of other Pu isotopes; (2) ^{239}Pu, the key Pu isotope of research interest; and (3) ^{240}Pu, another isotope of research interest. Of the AMS targets analyzed, 195 were derived from samples (e.g., unknown plates, blank plates, reagent blanks, and urine samples) containing an unknown test-isotope concentration.

In our AMS measurements, we studied AS discs that had been used historically to monitor the 11 current or former LLNL workers, of whom only seven were suspected of having previous ^{239}Pu intake. Measurements confirmed ^{239}Pu intake by all seven workers. Data for one worker confirmed a previously unsuspected and nondetectable intake of ^{239}Pu through an injection wound. These and other results support the unexpected conclusion that extremely low-level chronic occupational ^{239}Pu exposures can result in a lifetime ^{239}Pu burden that can be readily detected by analyzing urine samples with AMS. Further analysis of our data is expected to confirm long-term Pu excretion by persons chronically exposed to Pu levels that are below current or proposed DOE guidelines for Pu workers. This project thus not only demonstrated a unique capability to measure low-level Pu intakes years after exposure, but also paves the way for an improved scientific basis for Pu-related occupational safety and risk management, exposure assessment, and dose reconstruction, with significant homeland-security relevance.

Publications

Bogen, K. T., A. Marchetti, and T. A. Brown. (in press). "Use of a correlated compound binomial model to assess absence of non-counting noise in Pu-isotope ratios measured by AMS at LLNL. *Nuclear Instr. Meth. Phys. Res. B*. UCRL-JC-148648.

Imaging of Isotopically Enhanced Molecular Targeting Agents

Judy N. Quong

01-ERD-112

Abstract

A key to understanding complex biochemical pathways and mechanisms is determining their distribution: chemical imaging at the cellular and subcellular scale. For example, events leading from ingestion of the mutagen PhIP to mutagenicity are not known. Time-of-flight secondary-ion mass spectroscopy (TOF SIMS) measures elemental and biomolecular ions across a wide mass range. Because biological processes are dependent on their intracellular spatial distribution, determining the cellular distribution of PhIP and its metabolic byproducts can provide information for understanding PhIP mutagenesis; characterizing their mass spectra can determine the cell state to identify disease populations. This project will develop techniques to use SIMS to image the chemical composition of biological samples, focusing on optimizing sample preparation protocols and developing multivariate data analysis methods to study PhIP mutagenesis.

The capabilities developed under this project provide the methods and infrastructure to prepare biological samples suitable for SIMS measurements and to analyze the resulting complex mass spectra. Analysis of the mass spectra directly using the multivariate analysis techniques developed under this project will be used not only to aid in the analysis of chemical images but can also be used to analyze clinical samples to determine disease populations. In conjunction with measured chemical distributions, heterogeneous cell populations such as tumors and tissues can be characterized by measuring mass spectra from individual cells.

Mission Relevance

This project builds on LLNL's capabilities in analytical chemistry and computation to advance LLNL's mission in bioscience to improve human health.

FY03 Results

We first developed the protocols to measure ions generated from biological samples and measured the distribution of various biomolecules. The PhIP distribution was localized to the plasma membrane. Because data analysis methods are required for image interpretation, we next focused on developing a method using singular-value decomposition to distinguish mass spectra from different samples, including proteins, cell homogenates, and images. We prepared a publication describing a combination of singular-value decomposition and canonical analysis that enables the prediction of the samples from which mass spectra are generated. Finally, we also created a neural network that can classify mass spectra from cell homogenates.

Publications

Quong, J. N. et al. (2003). *Analysis of time-of-flight mass spectrometry data from cellular images*. UC Davis Cancer Center 9th Ann. Cancer Res. Symp., Oct. 2003. UCRL-PRES-201277.

Quong, J. N. et al. (2003). *Molecule-specific imaging analysis of PhIP in breast cancer MCF7 cells using time-of-flight secondary ion mass spectrometry.* UC Davis Cancer Center 9th Ann. Cancer Res. Symp., Oct. 2003. UCRL-PRES-200052.

Quong, J. N. et al. (2003). *Molecule-specific imaging analysis of carcinogens in breast cancer cells using time-of-flight secondary ion mass spectrometry.* 47th Ann. Meet. of Biophys. Soc., Mar. 2003. UCRL-JC-151797 Abs.

Quong, J. N. et al. (2003). *Secondary ion mass spectrometry for the analysis of biological materials.* 12th Intl. Mat. Res. Soc., Aug. 2003. UCRL-PRES-154757.

Quong, J.N. et al. (2003). *Molecule-specific imaging analysis of carcinogens in breast cancer cells using time-of-flight secondary ion mass spectrometry.* 14th Intl. Meet. on Secondary Ion Mass Spectrometry. UCRL-PRES-200052.

Wu, K. J. et al. (2003). *Molecule specific imaging analysis of carcinogens in breast cancer cells using time-of-flight secondary ion mass spectrometry.* Am. Vacuum Soc. 50th Intl. Symp., Nov. 2003. UCRL-JC-151797 Abs. Rev.

Single-Cell Proteomics with Ultrahigh-Sensitivity Mass Spectrometry

Matthias Frank 02-ERD-002

Abstract

The goal of this project is to measure the expression of biomolecules in single cells by ultrahigh-sensitivity mass spectrometry and to probe individual cells on a molecular level for the first time. To achieve this goal, the project will drastically improve the sensitivity and selectivity of time-of-flight mass spectrometry (TOF-MS) using a combination of advanced laser desorption and ionization techniques. These techniques will be used to study molecular changes in individual cells, investigate subtle differences between normal and cancerous cells, and detect and identify single biological aerosol particles in real time with TOF-MS. The project will obtain characteristic mass signatures from biological-warfare simulants and optimize the sensitivity and specificity of this detection technique.

The capability developed in this project will enable studies that probe individual cells at the molecular level to (1) understand molecular changes that occur during cell development; (2) investigate subtle differences between individual normal and cancerous cells; and (3) detect and identify single biological aerosol particles in real time.

Mission Relevance

The ability to detect and identify biological aerosols in real time and to study biochemical processes at the cellular level will lead to improved bioanalytical tools to support LLNL's missions in national security and homeland security to counter the threat of bioterrorism, and in bioscience and technology to improve human health.

FY03 Results

In FY03, major project accomplishments included demonstrating that unique molecular signatures can be obtained from several species of aerosolized individual bacterial spores and vegetative cells, thereby distinguishing bacteria from background aerosols; continuing progress with a two-step infrared laser desorption and an ultraviolet/vacuum ultraviolet ionization setup to explore sensitivity improvements; studying mass signatures from bacterial spores as a function of laser parameters; implementating a new mass-spectrometer design; and demonstrating the ability to follow metabolic markers within single bacterial cells. Another major accomplishment was the award of a $2.9M grant by DARPA to develop a spinoff from this project—an ultralow-false-positive trigger biodetector system based on aerosol mass spectrometry.

Proposed Work for FY04

In FY04, we plan to study well-known cell lines to measure proteins and other molecules contained in the cells and compare the mass signatures of cells from different lines. Single-cell mass spectrometry could detect and help understand changes in protein expression or cell composition occurring in early stages of a disease. We also plan to study the feasibility of measuring exhaled aerosol particles and droplets by aerosol mass spectrometry to screen them for bacterial or viral contamination. In addition, we will continue optimizing the desorption and ionization laser parameters and improve the design of the mass spectrometer to obtain higher mass signatures from cells. Finally, we will explore sporulation and germination processes of bacillus cells with mass spectrometry.

Publications

Fergenson, D. et al. (in press). “Reagentless identification of bioaerosol particles in seconds.” *Anal. Chem*. UCRL-JC-151115.

Steele, P. et al. (2003). ”Laser power dependence of mass spectral signatures from individual bacterial spores in bioaerosol mass spectrometry.” *Anal. Chem*. **75**, 5480. UCRL-JC-153095.

Ultrasonic Nondestructive Evaluation of Multilayered Structures

Michael J. Quarry 02-ERD-010

Abstract

Multilayered structures such as weapons assemblies and components, cladded boiler tubing, and high-energy laser targets pose challenges to nondestructive inspection techniques, especially if defects occur two or three layers below the surface. This project will use arrays with phasing characteristics to demonstrate ultrasonics techniques for (1) evaluating multilayers with preferentially excited individual layers using phased arrays or (2) using the multilayered structure as a wave guide and propagating guided-wave modes along a particular layer to evaluate its integrity, which is useful if the layer has inaccessible areas. The project will develop the theory and experimental configurations to preferentially excite guided acoustic waves in desired layers for inspection purposes.

This project will result in a prototype of an array system with phasing characteristics for evaluating multilayered structures and systems for nondestructive evaluation (NDE), and will demonstrate feasibility for programmatic inspection applications, including detecting cracked ceramics or voids in layers of multilayered weapons systems.

Mission Relevance

By developing new state-of-the-art technology, this program will establish new NDE methods for inspecting weapons in support of LLNL’s stockpile stewardship mission.

FY03 Results

In FY03, an electronic systems design for phasing an array, using a matrix switch and waveform postprocessing, was completed, and a record of invention was submitted for the device. We demonstrated detection with a guided wave of a 30% through-wall notch in the bottom layer of a multilayered structure from a position 20 cm away. With our bulk-wave array, we successfully imaged a void in a multilayer specimen using a full-aperture tomographic reconstruction algorithm and a time-domain, beam-forming algorithm.

Proposed Work for FY04

In FY04, after final modifications are made to the hardware, programmed defects will be fabricated in surrogate multilayer structures to demonstrate feasibility and to evaluate the capabilities of the guided-wave and bulk-wave techniques. Tests will include minimum crack-size detectability in the bottom layer for the guided-wave technique, and resolution and minimum flaw-size detectability for the bulk-array technique. By the end of the year, we expect to detect a 20% through-wall crack in the bottom layer of a surrogate multilayer with the guided wave technique and a 1-mm void with 3-mm resolution in a surrogate multilayer with the bulk-array technique.

Publications

Quarry, M. J. (2003). *Guided wave inspection of multi-layered structures.* Presented at the Review of Progress in Quantitative Nondestructive Evaluation, Green Bay, WI, July 27–31, 2003. UCRL-JC-152109.

Development of Ultrasensitive, High-Speed Biological Assays Based on Two-Dimensional Flow-Cell Detection of Single Molecules

Christopher W. Hollars 02-ERD-018

Abstract

This project is developing a practical device for rapid assay of biological species, such as proteins and DNA, at ultralow levels. Such a device will have applications in detecting biological warfare (BW) agents and would provide novel assays for medical applications, drug discovery, biotechnology, and basic biology research. Our objective is to detect and identify single, fluorescently labeled molecules using total internal reflection and an intensified charge-coupled device (CCD) camera. The analyte solution will be analyzed in a microfluidics device. Our goal is throughput 1,000 to 10,000 times greater and detection sensitivity 100 to 1000 times better than that of typical microstream devices. The device will be capable of performing biological assays in approximately 10 min at ultralow concentrations. This project represents a significant breakthrough in the analysis of small biological systems, e.g., large molecules, proteins, DNA, toxins, viruses, and bacteria.

Mission Relevance

This device will have applications in the detection of potential BW agents in support of the Laboratory's missions of nonproliferation, counterterrorism, and homeland security, and will provide a novel assay platform for medicine, drug discovery, biotechnology, and basic biology research in support of LLNL's mission in bioscience to improve human health.

FY03 Results

In FY03, we continued to optimize the planar microchannel and two-color imaging system. These efforts resulted in an increase in the detection signal-to-background ratio of the detected fluorescent probes at flow rates comparable to those required for the proposed assay conditions. The image-analysis program we developed was used to postanalyze data to determine the number of target molecules observed in an experimental run and to demonstrate single-color assays down to picomolar concentrations. Additionally, we evaluated the use of functionalized magnetic particles possessing a photocleavable linker as a method of sample purification and concentration prior to analysis and demonstrated ultrasensitive assays of dual-labeled target molecules in buffer solutions.

Proposed Work for FY04

In FY04, we plan to demonstrate assays in real applications. Areas of focus include the analysis of proteins and RNA fragments in body fluids, which can serve as indicators of cancer reoccurrence. Another area of interest is the analysis of the presence of surrogate toxins (proteins) in a variety of sample situations (i.e., drinking water, milk, and the surfaces of fresh produce) to detect natural outbreaks or the presence of BW.

Publications

Hollars, C. W., S. M. Lane, and T. Huser. (2003). "Controlled non-classical phton emission from single conjugated polymer molecules." *Chem. Phys. Lett.* **370** (3–4), 393–398. UCRL-JC-145314.

Development of a Fast Microfluidic Mixer for Studies of Protein-Folding Kinetics

Olgica Bakajin

02-ERD-040

Abstract

The goal of this project is to develop a robust, microfluidic laminar-flow mixer with at least 100 times lower consumption rate, shorter dead time, and better time resolution than currently available mixers. The mixer will be used to study kinetics of fast protein-folding reactions using ultraviolet- (UV-) light-induced fluorescence as a spectroscopic observation method. The project involves using computational modeling to develop mixer design, developing fabrication processes and techniques for assembly and characterization of the microfluidic mixer, and using the mixer to measure protein-folding kinetics. The mixers will be created with photolithographic patterning and wet and reactive ion etching of fused silica wafers.

If successful, this project will result in a robust microfluidic mixer that will offer a 500-fold improvement in mixing times and sample consumption compared to currently available mixers and will be compatible with most spectroscopic methods. This new capability will be useful for studies of DNA interactions with proteins, for single-molecule detection in low concentrations of toxins, and for studies of aggregation of prions or of the A-beta peptide implicated in Alzheimer's disease.

Mission Relevance

By developing new microfluidic devices with a wide range of potential applications, from detecting biological warfare agents to health care, this project supports the LLNL missions of counterterrorism and bioscience to improve human health.

FY03 Results

In FY03, we implemented computational modeling for design optimization; built a mixer in which we performed the first-ever measurements of kinetics on single molecules; fabricated a mixer made of silicon and glass with less than 10-ms time resolution; and performed microparticle image velocimetry measurements in the devices at velocities greater than 1 m/s, three-dimensional confocal microscopy measurements of mixing using iodide quenching of fluorescein dye, and first experiments to demonstrate fast mixing using fuorescent resonant energy transfer. We also published the results (in *Science*) with the single-molecule mixer. In collaboration with National Institutes of Health scientists, we were first to measure protein-folding kinetics at the single-molecule level. Finally, we completed fabrication of fused-silica mixers.

Proposed Work for FY04

We will attempt the first-ever measurements of the kinetics of the process of collapse of unfolded molecules using fluorescence-resonance energy transfer in our ultrafast mixer with our collaborator at the University of California, Los Angeles; improve the time resolution of the single-molecule spectroscopy and protein-folding measurement; and perform measurements of folding kinetics on cytochrome-c and other proteins by looking at tryptophan fluorescence using direct UV or two-photon or three-photon excitation.

Publications

Hertzog, D. E., J. Santiago, and O. Bakajin. (2003). "Microsecond microfluidic mixing for investigation of protein folding kinetics." *Micro Total Analysis Systems 2003* **1**, 891. UCRL-JC-153777.

Lipman, E. et al. (2003). "Single molecule measurement of protein folding kinetics." *Science* **301**, 1233. UCRL-JC-153057.

Development of a Quantum-Limited Microwave Amplifier using a Direct-Current, Superconducting Quantum Interference Device

S. Darin Kinion

02-ERD-071

Abstract

The goals of this project are to fabricate and demonstrate the first quantum-limited microwave amplifier based on a direct-current superconducting quantum-interference device (dc-SQUID), and to combine this amplifier with a single-electron transistor (SET) to ultimately achieve a quantum-limited electrometer. Both the amplifier and the electrometer are a vital component in all viable implementations of quantum computation and are important for many experiments now limited by the noise of microwave amplifiers.

The result of the project should be the first-ever demonstration of the standard quantum limit for a microwave amplifier, an important contribution to the field of quantum electronics. In addition, this work will result in high-profile articles in premier scientific journals.

Mission Relevance

This research and development in quantum information processing furthers LLNL's national-security mission by offering the potential for revolutionary techniques with application in secure information-communication networks.

FY03 Results

In FY03, we refined the SQUID fabrication process and produced SQUIDs that meet the design specifications. These devices have the highest gains measured to date. Major modifications to the dilution refrigerator were completed; the only remaining tasks to complete before noise-temperature testing can begin are system calibration and thermometry installation. Our SQUIDs were cooled to 25 mK with no performance problems, a very important step in the development. Finally, the performance of these devices was improved through better understanding of the microwave properties of superconducting strip lines achieved in this project.

Proposed Work for FY04

The proposed work for FY04 will (1) design and fabricate the final SQUID amplifier, (2) measure the noise temperature of these amplifiers at 25 mK, and (3) continue work on the SET development. If successful, the project will deliver the first-ever demonstration of the standard quantum limit in both a microwave amplifier and a charge detector.

Investigation of an Ultrafast, Direct Radiation-Driven, High-Speed Interferometric Detector

Mark E. Lowry 03-ERD-007

Abstract

We are developing a novel ionizing-radiation detector that provides an optical output, is capable of subpicosecond temporal response, has good sensitivity (even for hard x rays), and is scalable to imaging arrays. The detector uses an optical probe beam to detect the change in optical index that occurs when x rays impinge on a semiconducting medium and produce electron-hole pairs. This detector will enable a variety of radiography and self-emission imaging with hard-x-ray energies, as well as gamma and neutron detection important for experiments at future large laser systems. The technology will also meet requirements for x-ray detection with 100-fs temporal resolution at 8 to 24 keV for the Linac Coherent Light Source (LCLS) at the Stanford Synchrotron Radiation Laboratory (SSRL).

In this work, we expect to demonstrate a longitudinal single-pixel version of this detector that will allow x rays to efficiently enter from one side and the optical probe beam to enter from the other side. This single-pixel detector will then achieve high quantum efficiency (QE) while also having a fast temporal response. We will also attempt to construct array prototypes to demonstrate basic imaging.

Mission Relevance

This project supports LLNL's national- and energy-security missions by developing new radiation-detector technologies that will enable a variety of inertial-confinement fusion and high-energy-density physics experiments that are planned at existing and future large laser systems. High-energy-density experiments are a central component of the Laboratory's stockpile stewardship program.

FY03 Results

In two sessions at SSRL, we successfully fielded four different detectors having a geometry in which the probe beam was perpendicular to the x-ray beam. Each detector recorded significant index-modulated signals with very low absorbed x-ray fluences for ~9-keV x-ray photons. We studied the sensitivity as a function of proximity of the probe beam wavelength to the semiconducting band edge of the materials used. We were also able to measure the linearity of the detector response over two decades of x-ray fluence. In addition, we measured the rise time of the signal but found it to be limited by the ~180-ps x-ray pulse duration at SSRL. These are encouraging results that indicate the project goals are achievable.

Proposed Work for FY04

Work in FY04 will focus on developing detector-pixel technology based on the vertical-cavity radiation sensor concept. This technology will require growing top and bottom mirrors and detector-active volumes, as well as etching pixel mesas. The challenges to address include growing thick layers by molecular-beam epitaxy (MBE) and developing a low-temperature growth process that will enable high-speed operation. The research plan consists of three phases: (1) demonstrating a basic pixel using standard MBE growth techniques; (2) growing thicker layers to achieve better QE for high-energy x rays; and (3) developing the low-temperature MBE growth technology for fast response.

Publications

Lowry, M. E. et al. (2003). *RadSensor: X-ray detection by direct modulation of an optical probe beam.* Presented at SPIE Remote Sensing and Space Technology Conference, Aug. 3–8, 2003, Seattle, WA. UCRL-JC-243189.

DNA Detection through Designed Apertures

Sonia E. Letant 03-ERD-013

Abstract

Our goal is the selective detection of DNA signatures of harmful organisms using a rigid silicon aperture that mimics an ion channel. Our research plan has three main steps: (1) fabricating single-molecule-sized apertures on a silicon platform, (2) chemically functionalizing the aperture entrance, and (3) measuring DNA transport through the apertures by the current-blockade technique. The challenges are to design and build nanometer-sized apertures and to position functional chemical units with nanometric precision. We will use two original approaches: The apertures will be prepared by electrochemistry, and the functionalization will be accomplished by macromolecular chemistry.

Although other efforts have focused on building a single nanopore on various rigid substrates, no one has addressed either of the two key issues: finding a fast, reliable, and inexpensive technique to prepare single nano-apertures, and functionalizing a single nano-aperture with a single probe molecule. Our new approach, at the cutting edge of nanotechnology and supramolecular chemistry, should accomplish both and transform synthetic nanopores into extremely selective sensors able to detect and identify single DNA molecules in real time. Such a result would open the door to a new class of biosensors.

Mission Relevance

This project, at the intersection of chemistry, materials science, and biology, supports national security by enabling a new class of biosensors able to detect and identify DNA signatures from harmful organisms with single-molecule sensitivity—polymerase chain reaction is not required. Such fast and sensitive sensors will facilitate early response to terrorist or accidental biological contamination of air or water.

FY03 Results

We pursued two approaches to building nanometer-sized apertures on silicon: electrochemical etching (ECE) and focused ion-beam (FIB) drilling. We designed and fabricated silicon chips using FIB drilling, and developed an optimized ECE technique that produced through apertures with a top diameter of 10 nm and a length of 8 μm—the best aspect ratio on silicon by ECE to date. A state-of-the-art experiment with an electronic noise of 96 fA was conducted to measure current blockades in the picoampere, millisecond regime in real time. We chose a macro-cycle geometry and composition, ran a three-dimensional model, synthesized the building blocks and characterized them by nuclear magnetic resonance, and began assembly to form a closed macro-cycle.

Proposed Work for FY04

In FY04, we will optimize our apertures to achieve a top aperture diameter of 3 to 10 nm by electrochemistry and by FIB drilling; characterize these apertures and test them with our new experimental setup; and perform current blockade measurements on unfunctionalized apertures with modified T7 bacteriophages and with DNA strands on which bulky chemical adducts will be anchored to demonstrate a Coulter counter for viruses. When doing so, we will functionalize the macrocycle with a single strand of DNA and add various functional groups pointing inside and outside the ring to optimize the solubility of the molecule and its binding to the silicon surface.

Microfluidic System for Solution Array-Based Bioassays

Peter Krulevitch

03-ERD-024

Abstract

We are developing an instrument for performing multiplex bioassays in the field to detect biological warfare (BW) agents. Our integrated, reconfigurable system will perform user-specified assays based on nanobarcode (NBC) solution arrays—rod-shaped metal particles approximately 50 nm in diameter and 5 μm long and encoded with metal stripes for identification. A polymer-based microsystem will mix the NBCs with the sample and prepare the particles for optical readout. Other solution array approaches are under development, but none employs them in a way that meets the requirements for countering BW agents—multiplexed, user reconfigurable, inexpensive, portable, and easy to use. To guide development, we are performing fundamental studies of NBC behavior in microflows.

In addition, methods for putting functional biomolecular coatings on NBC particles will be valuable in future nanoparticle-based work for various applications. Studies of the transport behavior and control of nanorods in liquid flows are also of broad importance to the growing field of nanotechnology.

Mission Relevance

This project supports LLNL's national-security mission by developing technology to counter weapons of mass destruction. The instrument we develop will be used for detecting biological warfare agents in the field by first responders and has multiple applications, including surveillance, detection, and treaty verification. Our system fits well into the Department of Homeland Security's strategic plan for bioinstrumentation development. In addition, the project has clear applications in new medical diagnostics, supporting LLNL's mission in bioscience to improve human health.

FY03 Results

In FY03, two optical systems were set up at Stanford University—one for flow experiments and one for analyzing sedimentation—and a system was assembled at LLNL for performing automated optical readout of NBC arrays. We studied the Brownian motion, sedimentation, and pressure- and electro-osmosis-driven flow of NBCs, allowing us to write a program that predicts the migration of NBCs due to fluid shear and Brownian motion. Our surface coating investigations yielded an optimal coating composition of 80% mercaptoundecanoic acid, as well as a new technique for the high-performance attachment of antibodies and other biomolecules to NBCs. We obtained excellent initial results in demonstrating a working multiplex bioassay in the nanobarcode format.

Proposed Work for FY04

In FY04 we will (1) continue experimental studies of NBCs in microflows; (2) develop microfluidic subsystems for manipulating and transporting NBCs for readout and analysis; (3) continue development of NBC-simulation code; (4) enhance our optical system for particle analysis; (5) complete development of a multiplex test panel; and (6) demonstrate the multiplex test panel using NBCs. From our initial experiments, we expect to successfully demonstrate self-assembly of NBCs into two-dimensional arrays using nanopatterned surfaces and Brownian motion, providing maximum assay readout performance. Given the success of our initial surface functionalization and bioassay studies, we expect a straightforward transition of a full immunoassay panel to the NBC platform.

Cargo Container Security Sensor System

Stephen G. Azevedo 03-ERD-025

Abstract

Intermodal, containerized shipping has revolutionized world trade by allowing protected, point-to-point transport for manufacturers, but this type of shipping is also a threat to our nation's security. Any container that enters U.S. ports could contain weapons of mass destruction (WMD), whose detonation would have catastrophic consequences. Our project addresses this national-security problem by studying a sensor-system concept that could detect WMD in shipping containers before arrival at U.S. docks, and without impeding the regular flow of normal cargo. The sensors must be inexpensive and have a long life (10 yr) for permanent mounting. We focus on two major elements of such a system—low-power integrating radiation sensors, and robust, scalable, ad hoc wireless networks for extreme multipath environments.

This project will produce two key results. The first is a new radiation sensor concept that can integrate the radiation over time (during 7 days of ocean travel), has limited discrimination of energy bands, and is low in power draw. The leading candidate is a set of dosimeters (either

thermoluminescent or optically stimulable) that have varying amounts of shielding to crudely measure spectral content, and are remotely read and reset. The second result will be a new communication technique for networking thousands of sensors using energy-efficient signaling and routing protocols. The sensors must be queried and the data extracted by multiple hops to a ship's infrastructure using very low (sub-milliwatt) power levels. Such scalable, ultralow-power networks have never before been accomplished.

Mission Relevance

Our project will strengthen homeland security by researching and developing a sensor system concept to detect WMD components entering the U.S. in shipping containers. Furthermore, both the sensor and networking aspects of this project have relevance to other national needs in the military and intelligence fields. For instance, DOE/NNSA projects for the long-term storage of radioactive substances with small, low-cost sensors will also benefit from this research.

FY03 Results

In FY03, we studied possible radiation-detection sensors, performed several measurement studies of thermoluminescent dosimeters (TLDs) and advanced neutron detectors, measured background radiation on two container ships, performed radio frequency (RF) propagation studies aboard two ships, and began developing low-power communications and networking protocols. Measurements of radiation levels from a known source in a shipping container compared well to simulated results for various scenarios. These measurements have led to an initial design for the coming year. The RF experiments pointed out the need for an ultrawideband (UWB) physical implementation, and networking research has produced a new energy-efficient protocol mechanism.

Proposed Work for FY04

In FY04, our project will focus on the nuclear detector subsystem and networking issues. Producing a working detector will require integration of the sensor element, heater, and readout. We will perform larger experiments with passive detectors, Monte Carlo simulations to determine the best TLD configurations, begin integration of the TLD sensor package, and complete the device characterization. A prototype will be developed for testing. In communications and networking, we will publish our RF propagation results, experiment with ultralow-power versions of the UWB communications system, and develop networking scenarios for low-power conditions.

Photochromic Radiation Dosimetry

Charles G. Stevens

03-ERD-040

Abstract

The objective of our project is to develop a highly sensitive approach to radiation dosimetry that will enable the remote detection of radiation. We are developing two systems in which radiation converts photochromic molecules into fluorescers, which are then laser interrogated. Successful implementation will provide the technology to address important problems in homeland security and nuclear counterproliferation. The first system is a detector based on gradient-index millispheres with a photochromic coating that is sensitive to alpha

and beta radiation, (e.g., from UO_2 and ^{85}Kr) and laser interrogated remotely. The second is a radiation sensor that is sensitive to neutrons and gamma radiation and that uses a plastic scintillator coupled to a reversible photochromic material, which may be located remotely and periodically read.

This project will explore a new concept in radiation detection that offers a truly remote sensing capability for the first time. The product of this effort will be a determination of performance potential and demonstration of applicability for intelligence and defense community needs.

Mission Relevance

This project supports LLNL's national-security mission by furthering the development of advanced technical surveillance capabilities to provide early warning and assessment of the location of nuclear materials. The radiation sensor systems we seek to develop have unique features to enable detection of proliferation activities and remote sites. It will contribute directly to national and international security, counterterrorism, nonproliferation, homeland security, and military intelligence.

FY03 Results

We identified and evaluated effective photochromic materials; demonstrated proof of principle for two radiation sensor systems, including showing conversion efficiencies (radiation to photochromic molecules) and analyzing signal and background levels based on various homeland-security and proliferation scenarios; fabricated an alpha detector consisting of a thin-film scintillator and a photochromic detection layer and evaluated detection materials that undergo permanent photochromic conversion; fabricated a gamma-sensitive plastic scintillator unit coupled to a photochromic detector; and synthesized a special thioindigo dye, characterized it spectroscopically, and used it to develop solid and liquid thioindigo-based reversible photochromic detectors.

Proposed Work for FY04

In FY04 we plan to continue materials development and evaluation and determine performance characteristics, such as sensitivity, linearity with exposure, and fading of a stored signal over time. We will explore more candidate materials and study the integration of photochromic transducers with gradient-index spheres. For the gamma-detection system, we will optimize light concentration based on solar concentrator technology and continue to evaluate and develop reversible photochromic materials, including organic dyes and inorganic storage phosphors.

Long-Range, Passive Detection of Fissile Material

Lorenzo Fabris 03-ERD-048

Abstract

Recent events highlight the increased risk of an attack on the United States with a nuclear or radiological weapon. A key to countering such threats is the long-range detection of nuclear material. Detecting gamma-ray emissions from these materials at distances of greater than 100 m

is theoretically possible but has long been thought to be impractical due to fluctuating levels of natural background radiation. Recent work has shown that this problem can be overcome through the use of imaging gamma-ray detectors.

We expect to build a complete, large-area (6400-cm^2) gamma-ray imager based on individual 10-cm-square sodium–iodine scintillation detectors. The system will be mounted in a truck to conduct test searches for nuclear materials. The instrument will allow LLNL to be the first institution to demonstrate the ability to detect a 1-mCi-activity radiation source from a distance of 100 m and while traveling at a speed of 25 mph. A successful demonstration of this approach will lead to proposal of a full-size, fieldable unit.

Mission Relevance

One of the major challenges faced by the Department of Homeland Security is the remote detection of radioactive materials that could be used in a nuclear or radiological weapon of mass destruction. The detection of these materials is recognized as a top scientific priority of the Department. Our work will be an important verification that such detection can be realized. The national-security mission will benefit by allowing for rapid incorporation of this technology into the arsenal of deployed equipment.

FY03 Results

We came very close to demonstrating detection of nuclear materials at distances of 100 m. A working detector system was assembled and tested in the laboratory. We completed (1) 75 analog signal-processing board modules and the motherboard that hosts them, (2) the high- and low-voltage power supply systems, (3) the computer interface boards and most part of the software, (4) the cabin computer, system computer, and local-area network, (5) design, layout, and parts procurement for the digital back end, (6) the mechanical system comprised of the detector assembly and a coded aperture mask system, and (7) preliminary measurements, including the first image of a faint cesium-137 source. A paper was presented at a national symposium.

Proposed Work for FY04

In FY04, to complete the deployment we will fabricate and test the digital back-end board needed to acquire the converted signals from the analog and analog-to-digital converter boards, redistribute them to the computer interface card, and provide time-stamp information and vehicle position information. With most of the software having been written, we will address the interaction between the low-level drivers provided with the data-acquisition board and our software. When all preliminary measurements are complete, the system will be moved in its truck, and field trials and measurements will be performed to demonstrate mobile search and detection at large distances.

Publications

Ziock, K. P. et al. (2003). *Large-area imaging detector for long-range, passive detection of fissile material.* Presented at 2003 IEEE Nuclear Sci. Symp., Oct. 19–25, 2003. Portland, OR. UCRL-PRES-200367.

Carbon-Nanotube Permeable Membranes

David J. Eaglesham

03-ERD-050

Abstract

The motion of molecules through pores of molecular dimensions is a crucial problem in nanoscience. Diffusion down a nanotube has been frequently simulated but never measured. This project will measure the motion of molecules down the axis of a carbon nanotube (CNT). This will be accomplished by embedding ordered nanotubes in a thin film to make a semipermeable membrane with ideal pores. We will observe diffusion both by permeation and direct observation of single fluorescent molecules.

To provide protection for a detector (i.e., exclusion of a large molecule that would otherwise poison the sensor), we will create a semipermeable membrane with molecular size selectivity. Such a membrane would be a potentially powerful addition to the biological and chemical warfare armamentarium. If successful, this work could benefit other areas (including homeland security) and provide an avenue to address various basic- and applied-science challenges. This could include applications such as interfacing a bioactive sensor (e.g., for salmonella) to a complex sample (e.g., mayonnaise), eliminating complex molecules (e.g., oils) from air samples, and producing effective membranes for desalination.

Mission Relevance

The Laboratory's national-security mission would benefit from technology that enables development of more compact, highly integrated sensors. The Laboratory faces the challenge of interfacing a sensitive molecular detection and identification system to a complex environment. Current sensor devices employ compact macroscopic separation as the primary technique, but compact sensors with a very high degree of integration would advance the capabilities to a new level.

FY03 Results

We began the process of CNT deposition in magnetic-targeted carriers (MTCs). We demonstrated a process for putting extremely conformal silicon nitride (Si_3N_4) coatings onto CNT forests, and successfully made self-supporting membranes out of these nitride-coated nanotubes. However, the films have a undesirably high void fraction. The solutions we pursued include using (1) hot-wire-assisted deposition to achieve parallel nanotubes and (2) a spin-on-glass (SOG) technique to fill the gaps between the existing CNT films. Early results obtained with transmission electron microscopy of the films show we should be able to establish the conformation of graphitic cylinders.

Proposed Work for FY04

Multiwalled CNT arrays will be fabricated using existing recipes for iron-catalyzed growth. We will improve both homogeneity and ordering of these arrays by applying recipes for metal nanocluster arrays to the starting catalysts. We will embed these nanotubes in membranes using one of several thin-film techniques—including SOG and chemical vapor deposition and chemical mechanical planarization—then create replica nanotubes in the membrane material by combustion of the carbonaceous material and measure diffusion of tracers through these membranes by fluorescence and trace-gas analysis. The goal is to build a framework for a fundamental understanding of the behavior of molecules in and around pores of molecular dimensions.

Probing the Properties of Cells and Cell Surfaces with the Atomic Force Microscope

Michael W. McElfresh

01-ERI-001

Abstract

The goal of this project is to develop methods for characterizing the mechanical properties of living cells at the nanoscale using novel scanning probe microscopy-based techniques coupled with a sophisticated modeling approach. This project will develop new chemical force microscopy techniques (i.e., recognition microscopy) capable of probing the mobility of protein, lipid, and carbohydrate components of the membrane that are needed to map changes in the fluidity of the membrane, identify the forces and factors that play a role in receptor segregation and mobilization, and characterize how changes in the local environment of the membrane's protein components affect their structure and function.

The new type of recognition microscopy that we expect to develop will form the basis for detecting biological agents and diagnosing disease. Although currently available methods can identify specific sites on cells, distinguishing bonding between two specific molecules from nonspecific binding of molecules remains elusive. Recognition microscopy offers the prospect of using force spectroscopy to distinguish specific from nonspecific and to quantify the bonds under various physiological conditions. We also hope to shed some light on the issue of molecules having multiple bonding sites.

Mission Relevance

This work has the potential to form the basis of new sensor systems to enable the detection of biological and chemical warfare agents and their effects on host cells, the diagnosis of disease, or cell dysfunction in support of the Laboratory's mission in bioscience and human health.

FY03 Results

In FY03, we used a Concanavalin-A- (Con-A-) derivatized atomic force microscope (AFM) cantilever to study the physical properties of cell membranes. The derivatized tips were used to grab Con-A receptors, and lateral-force microscopy was used to study both the mobility of the receptor in the membrane and the receptor's range of motion. Quantitative comparisons of these results with earlier photobleaching experiments were made. In a separate experiment, the AFM was derivatized with farnesyl, a molecule that is easily incorporated into the cell membrane. After welding or gluing the tip into the cell membrane, we used lateral-force microscopy to directly measure membrane fluidity in order to understand more about the boundaries.

Generation of Single-Cycle Light Pulses

Brent C. Stuart

02-LW-001

Abstract

Most optical pulses, even at the 10-fs timescale, consist of several oscillations of the electric field. Our research proposes to produce and amplify an ultrabroadband continuum, thereby

producing single-cycle (<3 fs) optical pulses. Production of these single-cycle pulses will lead to new generations of experiments in the areas of coherent control of chemical excitations and reactions, high-order harmonic generation for probing materials and fast processes, and selective three-dimensional (3-D) micrometer-scale material removal and modification. Our approach includes generation of a broadband continuum in a hollow fiber waveguide, amplification of this continuum in a novel optical parametric amplifier (OPA), and adaptive control of compression to the single-cycle regime.

Demonstrating single-cycle (<3 fs) pulses would result in the shortest pulses ever produced in the visible/near-infrared spectral region. These pulses could be used in pump/probe experiments with unprecedented time resolution. In addition, single-cycle optical pulses can be used to produce single, subfemtosecond pulses in the extreme ultraviolet (XUV) region, which could be used to probe 100-attosecond timescale processes. Another application includes temporal shaping of longer pulses with resolution of 3 fs. These pulses can be applied to coherent control of molecular reaction pathways.

Mission Relevance

Completion of this project will create a new DOE capability in materials probing and manipulation on extremely fast timescales. Probing materials in such fast timescales is important for many Laboratory applications, such as weapons materials, and can be applied to three areas of particular relevance to the NNSA/LLNL stockpile stewardship mission: controlled excitation and detection of chemical compounds, selective submicrometer 3-D machining, and subfemtosecond XUV generation for materials probing.

FY03 Results

We demonstrated generation of a broadband continuum in a hollow capillary waveguide. The spectral content was optimized with a genetic algorithm that controls the intensity and phase of the input pulses. We then amplified a portion of this continuum in an OPA to create a 250-nm-wide spectrum, which should be compressible to 4 fs when the pulse compressor is completed. We also designed and procured the optics for two additional OPA stages to amplify additional portions of the spectrum. Calculations indicate that with all three stages, it should be possible to produce 2.5-fs pulses. A collaboration began with the University of Illinois on selective excitation of excited states in rubidium.

Coherent Anti-Stokes Raman Microscopy: Specific Molecular In-Vivo Imaging at Super Resolution without Fluorescence Labels

Thomas R. Huser 03-LW-056

Abstract

Coherent anti-Stokes Raman scattering (CARS) microscopy is a novel technique for imaging cells with spatial resolution on the order of 300 nm, without the need for extrinsic markers such as fluorescent labels. Microscopy with CARS provides three-dimensional sectioning capability at vibrational marker modes with very low background. Subcellular processes can be studied with picosecond time resolution. In this project, we are developing an inexpensive CARS

microscope by adding a fiber Raman laser to an existing femtosecond laser system, extending the microscope spatial resolution well beyond the diffraction limit, increasing the sensitivity of the instrument at the single-molecule level, and applying the technique to study the interaction pathway of viruses within cells.

The new imaging technique developed in this project will enable high-resolution imaging at the subcellular level. This technique also has applications in materials science for the investigation of stress or corrosion, the analysis of composite systems (alloys, high explosives), or in forensics.

Mission Relevance

The CARS instrument will establish a new imaging capability that supports LLNL's missions in stockpile stewardship and biotechnology. In addition CARS is directly relevant to DOE's Genomes-to-Life Initiative, the newly founded National Institutes of Health Institute for Biomedical Imaging and Bioengineering, and the National Science Foundation Center for Biophotonics Science and Technology at the University of California (UC), Davis.

FY03 Results

During FY03, we built a fiber Raman laser system based on photonic crystal fibers and demonstrated its lasing properties. The resulting amplified stimulated emission is very broadband, and parts of this broad emission can be selected by using bandpass filters and then used as a probe beam for CARS microscopy. We acquired two freely tunable, synchronized Ti:Sapphire laser systems, which have been incorporated into an existing confocal Raman microscope. Microscopy in an imaging mode was successfully demonstrated on individual polystyrene spheres and bacterial spores. We have further characterized the Raman spectrocopic response of a wide variety of biological samples, including virus-infected cells, cancer cells, and liposomes, that will be used to define characteristic marker modes for CARS imaging.

Proposed Work for FY04

In FY04, using our proprietary microscopy scheme, we plan to demonstrate CARS microscopy below the diffraction limit. Demonstrations of biological imaging applications with the CARS microscope will include following single living cells through all stages of a viral infection; imaging the interaction of a virus particle with a cell after the viral DNA has been silenced by the introduction of small interfering RNA (siRNA) into the cell; studying the interaction of lipid particles with endothelial cells, in collaboration with Prof. Jack Rutledge at UC Davis; and characterizing human cancer cells, in collaboration with the UC Davis Cancer Center.

Publications

Chan, J. et al. (in press). "Rapid, reagentless identification of single bacterial spores in aqueous solution by confocal laser tweezers Raman spectroscopy." *Anal. Chem.* UCRL-JC-155491.

Huser, T. et al. (in press). "Single-cell Raman spectroscopy of DNA packaging in human sperm cells." *PNAS.* UCRL-JC-153523.

Single-Use, Real-Time Bioweapons Detector

Billy W. Colston

03-FS-008

Abstract

Recent terrorist events, combined with post-Operation Iraqi Freedom hostilities, have highlighted the need for small, simple-to-use pathogen detectors, especially highly rugged, mobile, single-use systems to monitor sites in remote and hostile locations. Several portable polymerase chain reaction (PCR) pathogen-detection devices have been developed at LLNL. Although successful, the devices' lack of a portable sample-preparation component has limited their deployment in the field. To simplify the fluidics and reduce size and weight, we propose a design based on solid-phase capture and flow-through PCR detection. We demonstrated the use of such a design and evaluated its performance, including collection and analysis of a simulant aerosol, an investigation of sensor aerodynamics, and washing studies.

A successful demonstration of a small, reliable, portable, real-time, sample-preparation component for a PCR pathogen-detection device could result in a fieldable biodetector within 6 months.

Mission Relevance

This project can lead to a field-deployable PCR detection device for handheld decontamination detectors and inexpensive smoke-detector-size pathogen detectors that will enhance and extend the Laboratory's national security mission in bioweapons detection. In addition, portable pathogen detectors have application as a basic platform for bedside diagnostics in support of the Laboratory's mission in biotechnology to improve human health.

FY03 Results

Our FY03 work consisted of three components: (1) aerosol-release experiments to characterize spore collection and PCR detection, (2) wind tunnel experiments to determine the flow rates in different collector geometries, and (3) PCR compatibility tests of washing agents. Using tubes packed with polypropylene fibers, we captured aerosolized *B. subtilis*, var. *niger* (Bg) at efficiencies greater than 90%. We successfully detected Bg using real-time PCR with just a few sample-preparation steps and characterized the collected concentration as a function of tube inner diameter. Analysis of the wind-tunnel experiments demonstrated collection rates up to 70 L/min. Based on our results we proposed a sensor design for a drone aircraft.

An Early Warning System to Detect Contamination of Municipal Treated Water

John F. Cooper

03-FS-009

Abstract

Municipal treated-water systems are severely vulnerable to terrorist attack. Contrary to popular opinion, chlorination does not protect distributed water against biological weapons, as the chlorination may be readily countered by chemical or biological matter introduced concurrently. This project explored the feasibility of an autonomous, wireless array of sensors and communicators to detect and communicate biological toxin contamination of municipal treated-water systems. Since a biological intrusion in treated water effects a drop in oxidation

potential (ORP) of the water as the disinfectant chlorine is consumed, a sensor array can detect, measure, and communicate the drop to a management system via acoustic signals in the water. Using a lab flow system to simulate a municipal water system, the project will determine the response of an ORP sensor to chemical and biological reducing agents that accompany or simulate biological agents or growth media. Acoustic communication through water pipes will be tested using commercial "pingers" in the LLNL water-distribution system.

The results of this research could provide an early warning system to counter the threat of bioterrorist attacks on municipal treated-water systems.

Mission Relevance

The technology studied in this project has the potential to provide early detection and warning of potent biological agents, raw or refined, that have contaminated municipal treated-water systems. This project supports the Laboratory's counterterrorism and homeland security missions.

FY03 Results

Feasibility of ORP detection of biotoxin injection was established with response times of <50 s. Acoustic transmission of signals through water filled pipes was effective, but further advances are needed to offset losses from wall absorption and wave interference. We identified modeling techniques for determining wave propagation and commercial hydraulic flow codes for subsequent use for simulation and array optimization. The cost of the unit ($300) was estimated from retail components, while installation costs would add $500 to $1000 each. Array life was estimated to be 10 yr based on power requirements and commercial lithium batteries. Four inventions were recorded covering the basic technique, enhanced sensors, low-cost emplacement, and improved signal transmission and processing.

Biological Sciences

Pathogen Pathway Project

Joseph P. Fitch

01-SI-002

Project Description

Understanding virulence mechanisms of bacterial pathogens is vital to anticipating biological threats and to improving detectors, vaccines, and treatments. This project will characterize factors responsible for virulence of *Yersinia pestis*, the causative agent of plague and a biothreat agent, which has an inducible Type III secretion virulence mechanism also found in other animal, plant, and human pathogens. Our approach relies on genomic and proteomic characterization of *Y. pestis* in addition to a bioinformatic infrastructure. Scientific and technical capabilities developed in this project can be applied to other microbes of interest. This work will establish a significant new direction for biodefense at LLNL and expand our national and international scientific collaborations.

Results from genomic, proteomic, and modeling approaches to characterize *Y. pestis* will provide a more thorough understanding of the Type III secretion virulence mechanism in general while laying the foundation for next-generation detection of biothreat agents. Several spin-off projects have resulted from this work that encompass both an applied platform for pathogen detection as well as more basic science approaches to characterize virulence mechanisms for other relevant pathogens and microbes.

Mission Relevance

This project represents a national security investment that ultimately will help detect the presence of biowarfare threat agents and genetically modified organisms. Our research complements existing LLNL programs in counterbioterrorism and builds on LLNL's expertise in genomics, proteomics, biocomputing, instrumentation, and national security. The project will create a functional genomics and proteomics platform that will be useful for future applications relevant to national security, environmental management, and biotechnology missions in NNSA and the DOE Office of Science.

FY03 Results

Work completed in FY03 includes (1) genomic and proteomic characterization of *Y. pestis*; (2) analysis of host response to *Y. pestis* exposure; (3) characterization of *Y. pestis* regulatory proteins by surface-enhanced laser desorption/ionization mass spectrometry; (4) development of a real-time expression system to detect cascades and feedback loops that govern virulence; (5) siRNA knockouts of virulence factors to characterize virulence and protein interactions; (6) development of a novel fuzzy-logic-based gene network modeling approach; (7) gain of expertise in conventional pathway modeling, resulting in novel biological insights for virulence; and (8) implementation of bioinformatics for data management, mining, and interpretation in genomic, proteomic, and regulatory studies.

Publications

Chromy, B. A. et al. (2003). *Proteomic characterization of the mechanism of virulence in* Yersinia pestis. 6th Intl. Symp. on Mass Spectrometry in the Health and Life Sciences, Aug. 2003. UCRL-JC-153609 Abs.

Fitch, J. P. et al. (2003). "Biosignatures of pathogen and host." *Proc. IEEE Workshop on Genomic Signal Processing and Stat.* UCRL-JC-149741.

Fitch, J. P. et al. (2003). "Rapid development of nucleic acid diagnostics." *Proc. IEEE* **90**, 1708. UCRL-JC-148118.

Forde, C. E. and S. L. McCutchen-Maloney. (2003). "Characterization of transcription factors by mass spectrometry and the role of SELDI-MS." *Mass Spectrometry Reviews* **21**, 419. UCRL-JC-150675.

Heidbrink, J. L. et al. *Discovery of biomarkers from dialysis patients.* ProteinChip Technology Users Meet., May 4–7, 2003, Galveston, TX. UCRL-JC-152676 Abs.

Motin, V. L. et al. (in press). "Temporal global changes in gene expression during temperature transition in *Yersinia pestis*." *J. of Bacteriology.* UCRL-JC-150715.

Murphy, G. A. et al. (2003). *Discovery of protein biomarkers in dialysis patients using SELDI-MS.* 6th Intl. Symp. on Mass Spectrometry in the Health and Life Sci., Aug. 2003. UCRL-JC-153608 Abs.

Sokhansanj, B. A. et al. (2002). *Fuzzy rule-based modeling of* Yersinia*–host interactions.* DIMACS Workshop on the Pathogenesis of Infectious Disease, Sept. 23–25, 2002, New Brunswick, NJ. UCRL-JC-150426 Abs.

Sokhansanj, B. A. et al. (2003). "Exhaustive search for fuzzy gene networks from microarray data." *Proc. IEEE Engineering in Medicine and Biology Conf.* UCRL-JC-154129.

Sokhansanj, B. A. et al. (2003). *Modeling* Yersnia pestis *systems biology.* 2003 ASM First Biodefense Research Meeting, Mar. 9–12, 2003, Baltimore, MD. UCRL-JC-152137 Abs.

Sokhansanj, B. A. et al. (2003). "A quantitative model of human DNA base excision repair." *Nucleic Acids Research* **30**, 1817. UCRL-JC-146306.

Sokhansanj, B. A. and D. M. Wilson. (2003). "Mathematical modeling of human DNA base excision repair." *Proc. 47th Ann. Meet. of the Biophys. Soc.*, Mar. 1–5, 2003, San Antonio, TX. UCRL-MI-151945.

Quong, A. A. et al. (in press). "An indexed modeling and experimental strategy for biosignatures of pathogen and host." *J. of the Franklin Inst.* UCRL-JC-149741 Rev.

Zeller, L. et al. (2003). *Analysis of complex biological mixtures for disease detection and prevention.* 6th Intl. Symp. on Mass Spectrometry in the Health and Life Sciences, Aug. 2003. UCRL-JC-153512 Abs.

Pathomics

Kenneth W. Turteltaub

03-SI-005

Abstract

Pathomics is a comprehensive strategy to develop an understanding of an individual's response to infectious disease agents, particularly those that represent a threat from bioterrorism. Its overall goal is to predict possible routes of pathogenicity and to have detection systems that will determine that an attack or disease outbreak is beginning prior to people becoming overtly sick. Using a vaccine strain (vaccinia) for smallpox and the infectious cowpox virus, we will demonstrate that detection of disease is possible presymptomatically and that molecular signatures exist that can distinguish between pathogen types. We will propose a preliminary type of diagnostic platform and demonstrate its use in public health surveillance.

By the end of the project, we plan to demonstrate that molecular changes can be detected in blood within the first few hours after exposure, and that blood-borne signatures develop within the first few hours of infection by the vaccine strain. Our goal is to prove the feasibility of this approach.

Mission Relevance

This research, needed to meet gaps in our ability to detect and mitigate the effects of infectious disease, whether natural or from acts of bioterrorism, supports the Laboratory's national security and homeland security missions as well as missions of the National Institutes of Health.

FY03 Results

In the first five months of the project in FY03, we accomplished the following goals: (1) established the research team, including experts in mass spectrometry, protein chemistry, and genomics both internal and external to LLNL; (2) created a database that will assimilate the huge amounts of data that are expected from all aspects of the project; (2) developed a three-dimensional (3-D) liquid chromatography and mass spectrometry analysis system for characterizing plasma protein profiles; (3) designed, conducted, and collected samples, and began analysis of the first cowpox animal exposure; and (4) collected 15,00 data points from a human clinical study of the response to vaccinia vaccination. Initial data analysis is underway.

Proposed Work for FY04

Our FY04 goals are the following: (1) using human and animal models, determine the spectrum of biochemical components in blood that can be used to detect vaccinia infection before clinical symptoms are present and determine whether these spectra vary with different disease types; (2) conduct rodent experiments to determine how animals can be used as model systems for human biochemical signatures and provide data for developing and testing pathway models; initial studies will involve vaccinia and cowpox; (3) use in vitro cell cultures to determine how cultured cells can be used to model and test the relationships between signatures and individual steps in virulence; and (4) begin to apply a systems biology approach to whole-organism responses to infection.

Development of Synthetic Antibodies

Julie Perkins 01-ERD-111

Abstract

This project will provide the initial results for a novel approach to radioimmunotherapy, in which a small molecule with high affinity for a protein on the surface of the tumors delivers radiation treatment directly to the site of a metastatic disease. Our aim is to develop a synthetic high-affinity ligand (SHAL) that binds with high affinity and specificity to HLA-DR10, a protein found in abundance on the surface of non-Hodgkin's lymphoma, a cancer that affected 53,000 Americans in 2001. The attachment of a radionuclide to a SHAL would enable the targeted delivery of radiation to lymphocytes and could provide an effective therapeutic agent for this killer disease. Although the route to treatments against cancers is a long one, a small step in the development of such novel cancer treatments will clearly have a large impact in the area of radioimmunotherapy and subsequently in the area of cancer treatments in general. This multidisciplinary project uses LLNL's expertise in computations, nuclear magnetic resonance (NMR) spectroscopy and synthetic chemistry.

Mission Relevance

The scientific techniques required to develop a SHAL for HLA-DR10 can be applied to target or sense other proteins (e.g., biowarfare agents) that are relevant to LLNL's national security mission.

FY03 Results

In FY03 a SHAL that binds to HLA-DR10 was synthesized. This SHAL was shown by NMR to bind in vitro to HLA-DR10. Following these positive results, three derivatives of the SHAL were synthesized and also tested for their affinity to the target protein. The most promising SHAL to date was resynthesized with a group capable of chelating a radionuclide instead of a biotin tag. The SHAL was labeled with a radionuclide and tested in vitro by our collaborators at UC Davis Cancer Center. These in vivo experiments are currently being improved to determine the absolute affinity of the SHAL for HLA-DR10 in a real system.

Accelerator Analyses for Protein Research

John S. Vogel   01-ERI-006

Abstract

Proteomics depends on collecting, defining, and identifying proteins responsible for specific biological functions and will only succeed with highly specific separatory technologies. Common quantitative methods (chemical analyses) are dependent on the molecular properties of the specific protein and the protein's environment instead of only on the amount of protein. However, physical analyses can quantitate proteins without structural dependencies at the required sensitivity. We developed methods based on isotopic, mass, and elemental analyses that can be applied to protein research at extremely low concentrations. These approaches have the sensitivity that is required to analyze accurately even rarely expressed proteins, while coupling with accelerator mass spectrometry, proton-induced x-ray emission, scanning transmission ion microscopy, molecular absorption spectrometry, and matrix-assisted laser desorption–ionization analyses. Our goal in FY03 is to (1) streamline analysis procedures, (2) apply our techniques to study pesticide binding, (3) determine the absolute detection limits, and (4) publish our findings and begin collaborative work with universities.

Mission Relevance

This work supports the Laboratory's mission in bioscience to improve human health by providing advanced macromolecular quantitation and binding information in the studies of cell proliferation, carcinogenesis, and environmental toxicities.

FY03 Results

In FY03, analysis procedures were streamlined and applied in two other research projects. Proteins from animal exposure studies were isolated with bound pesticides and analyzed using our method. Preparations were made to apply this method in other work (e.g., DoD). The Lab has also filed two patent applications from this research.

Publications

Grant, P. G. et al. (2003). "Alpha-particle energy loss measurement of microgram depositions of biomolecules." *Analytical Chemistry* **75**, 4519. UCRL-JC-152090.

Vogel, J. S. et al. (2003). "Accelerator mass spectrometry in protein analysis." *Methods in Proteome and Protein Analysis*. R. M. Kamp. et al. (ed.). UCRL-JC-152167.

A Three-Dimensional Model of Signaling and Transport Pathways in Epithelial Cells

Andrew A. Quong   02-ERD-016

Abstract

This project is developing a fully three-dimensional (3-D) model of epithelial cells that includes ion transport across cell membranes. Epithelial cells, which line all of the cavities and free surfaces of the body, act as a barrier to the environment and prevent the entry of pathogens into the body. This project will model the cell by using experimental data at the largest length scale to identify the dominant components, develop a mesoscale model as a system of partial

differential equations, and use experimental data to determine the parameters of the model. Results of the model will be used to test hypotheses regarding the relationship between defective ion transport and disease, and will be the starting point for studying the response of epithelial cells to different pathogens.

A 3-D model of the epithelial cell will be completed that includes passive diffusion of ions within the cell and across membranes, the active transport of ions across the cell membrane, and structural predictions and molecular simulations of the proteins responsible for metal–ion chelation and transport processes. A validated model will allow us to understand how a single change in a complex network can change the phenotypical behavior of the system.

Mission Relevance

By helping to determine the pathogenic pathways of genetic diseases, a 3-D cellular model implemented in the ALE3D code will contribute to both LLNL's biosecurity mission and its mission of bioscience to improve human health.

FY03 Results

During this year, the calcium-signaling model was completed and validated against published experimental data for calcium wave propagation. In addition, immunological experiments to measure cellular geometries and receptor distributions were conducted and results incorporated into the model. Implementation of extracellular compartments and membrane transporters for study of vectorial ionic transport began.

Proposed Work for FY04

Having validated our model within the context of calcium signaling, ionic transport through the epithelial cell tissue will be studied in FY04. Adding ion channels will help elucidate the mechanisms by which cystic fibrosis transmembrane conductance regulator (CFTR) leads to infection in the lungs. Our simulations will address whether a dysfunctional chloride transporter leads to a driving force for water to enter the tissue and deplete the airway surface liquid, or to a compositional change within this layer, which lessens the immune response.

Publications

Golumbfskie, A. J. et al. (2003). "A three-dimensional model of calcium signaling in epithelial cells." *Proc. 25th Ann. Conf. IEEE EMBS*, 2694. UCRL-JC-149970-ABS.

Golumbfskie, A. J. et al. (2003). *A three-dimensional model of calcium signaling in epithelial cells.* Presented at the 25th Ann. Conf. IEEE EMBS, Cancun, MX. UCRL-PRES-154356.

Rapid Assay Development for Biological Weapon Agent Detection and Surveillance

Kenneth W. Turteltaub 02-ERD-045

Abstract

We propose to develop an approach that will rapidly lead to assays to detect protein and metabolite signatures to identify a biowarfare (BW) threat agent presymptomatically in surveillance populations. Our goal is to provide the science and technology for a sustainable, continuous, and comprehensive nation-wide medical surveillance-based early warning system for covert biological attacks. The approach will use genomic and proteomic methods to identify

unique molecular indicators of infection in animals and humans. Computational methods will be used to rapidly sort and prioritize targets into signatures to detect and identify BW threats. Selection based on validated function will lead to molecular signatures that specifically identify threat agents and predict infectious risk.

If successful, this project will lead to the capability to identify and characterize gene, protein, and metabolite indicators of exposure to infectious disease agents, which could result in presymptomatic treatment.

Mission Relevance

By developing and testing the feasibility of assays for screening populations for BW agent exposure and applying this technology in a surveillance system, this project is directly relevant to the Laboratory, NNSA, and Department of Homeland Security missions of protecting the civilian population against potential biological terrorism.

FY03 Results

In FY03, we initiated a discovery-based experimental program to delineate specific presymptomatic and/or precontagious biomarkers for a select number of pathogens. We started a preliminary study using a dialysis patient population and began analyzing the samples for gene expression and protein changes. Initial results suggest we can distinguish dialysis patients from healthy nondialysis controls. This work laid the technical foundations, defined goals, and established a team that has begun a follow-on Strategic Initiative on the broader topic of presyndromic disease detection for pathogens relevant to bioterrorism and human health.

Advanced Filtration and Regeneration Techniques Based on Nanoporous and Aerogel Technologies

Anthony J. Makarewicz
02-ERD-048

Abstract

This project carried out the enabling research and development for advanced filtration and separation technologies. Filters with specific molecular weight cutoffs and regenerable filtration units are broadly applicable across critical national needs in biological warfare (BW) defense, biosciences, and human health. Components of this research will include advanced-filtration devices, dialysate-regeneration devices, and portable fluidics systems. The filtration and regeneration devices rely on special LLNL technologies, nanoporous silicon and carbon aerogels, as well as newly developed materials, for the separation and sequestration of biological solutes.

Mission Relevance

This work supports national security by laying the groundwork for BW defense and by advancing bioscience to improve human health. Advanced filtration and separation technologies for biological solutes meet critical needs of several areas of BW defense, including filtration capture of BW agents, pathogen concentration for immunoassay or polymerase-chain-reaction (PCR) detection, and protein separations. These technological improvements will also impact medical diagnostic assays.

FY03 Results

For FY03, we characterized the ability of carbon aerogels as a component of a bioreactor system to regenerate used dialysate. Although the carbon aerogel deionized the dialysate solution, the capacity was insufficient to remove enough ions to regenerate the solution in a practically sized device. We developed new material samples to achieve tunable porosity, particularly using poly-NIPAAM. These materials are currently being applied to aerosolized particle collection for biodefense.

Publications

Bearinger, J. P. et al. (2003). *Control of biocompatible polymers and biomolecules using EC-OWLS (electrochemical optical waveguide light spectroscopy) and patterning techniques.* Presented at Euromat 2003. UCRL-JC-150976.

Dynamic Simulation Tools for the Analysis and Optimization of Novel Filtration, Sample Collection and Preparation Systems

David S. Clague

02-ERD-066

Abstract

The heightened attention on chemical and biological early-detection systems brings an increased need for high-efficiency filtration, collection, and sample-preparation systems. This research project is producing new computational analysis tools designed to optimize these critical operations and to characterize system efficiencies based on the details of the microstructure with environmental effects. To accomplish this we are developing new lattice Boltzmann simulation tools that include detailed microstructure descriptions, relevant surface interactions, temperature effects, and the ability to handle both liquid and gas phase systems. The new capability is being validated with experimental data and will be applied to real-world issues of concern to Laboratory programs.

Mission Relevance

This effort will directly contribute to the Laboratory's missions in homeland defense and counterterrorism by providing simulation tools for sample collection, filtration, and preparation systems for mitigating or preventing warfare or terrorist activities involving chemical and biological agents.

FY03 Results

In FY03, we developed (1) a generalized porous media module and a representation of porous membranes with specified pore size distribution; (2) a theory for colloidal interactions between particles and surfaces, including colloidal interactions, that was incorporated in the main multiphysics simulation; and (3) the capability to include a coupled energy equation for studying environmental and temperature effects on filter and collection efficiencies. In addition, we studied Brownian motion using a novel approach, characterized fluid and species transport properties, and investigated the capture efficiencies of porous structures.

Proposed Work for FY04

In FY04, we plan to (1) develop a simulation capability for particles in a gas phase and develop a theory for particle and surface interactions in a gas phase, (2) incorporate particle-surface interactions in the multiphysics lattice Boltzmann capability, and (3) apply a Brownian motion capability to characterize particle partitioning and transport in nanoporous membranes with colloidal interactions. In the second half of the year, we will perform numerical experiments investigating gas-phase flow with and without particles to validate the theoretical treatment used in the multiphysics code.

Publications

Clague, D. S. and T. H. Weisgraber. (2003). *Dynamic simulation of particle transport and adhesion in microenvironments*. Presented at the AIChE National Conference. UCRL-PRES-152543.

An Agent that Prohibits Microbial Development and Infection

Allen T. Christian 03-ERD-038

Abstract

Interfering RNA (RNAi) completely inhibits specific, targeted genetic activity within a cell. This process can be used in both human and microbial cells. As effective as the process is, however, it suffers from rapid degradation. Within hours, the effect is lost. We have developed a similar process that functions like RNAi but is extremely long-lasting. The agent that produces this effect can be inserted into a cell and remain quiescent for a considerable period of time, without affecting cellular processes, until the gene against which it is targeted is induced. At this time the agent becomes effective, silencing the genetic response without affecting the host cell in any other way. When given as an anti-infective, this process may have significant use as an antibacterial, antiviral agent.

We propose to test the use of our process as both an antimicrobial and an anti-infective agent. Since the genetic pathways that can be used to disrupt infection and kill microbes are fairly ubiquitous, success in this project will produce the equivalent of a new class of broad-spectrum antibiotics and antiviral agents. Our goal is to determine the capabilities of this process using both bacteria and viruses. We plan to use nonpathogenic organisms, specifically *Escherichia coli* and baculovirus, as test microbes. Each of these is extremely well characterized and offers multiple pathways to test.

Mission Relevance

This capability relates directly to the LLNL national-security mission area of combating weapons of mass destruction by rendering biowarfare agents null before or after infecting subjects. It is also relevant to DOE's Genomes to Life (GtL) initiative, as a high-throughput method for perturbing bacterial pathways, which is a necessary step in the types of measurements contemplated by the GtL program.

FY03 Results

In FY03, we demonstrated the ability of our constructs to silence gene activity within human and bacterial cells. We have ongoing collaborations with clinical, industrial, and academic partners. With Dr. Gary Jarvis at UCSF, we assessed the value of these bacteria-destroying molecules in a clinical setting. Early results indicate that siHybrids—hybrids of small, interfering RNA (siRNA) and DNA—may be an effective antibiotic. With Dr. David Mills of UC Davis, we demonstrated the ability to destroy bacteria that cause oxidation in wine. With Dr. Haynes Sheppard, we demonstrated the ability to destroy the HIV virus in T-cells. These capabilities have potential applications as reagents for research use and as therapeutics and antibiotics, among others.

Proposed Work for FY04

We will continue to pursue the aforementioned collaborations. Our goals are to publish more manuscripts (one is currently under review on bacterial siHybrid function, two others in preparation on, respectively, mechanism of function in bacteria and clinical efficacy based on unaided delivery of the siHybrids), to complete reagent licensing, and begin patenting a range of therapeutic uses.

Identifying Gene-Regulation Mechanisms using Rule-Based Classifiers

Krzysztof A. Fidelis 03-ERD-049

Abstract

We aim to develop a new approach to identifying gene-regulation mechanisms based on a supervised machine-learning method. The components and mechanisms of gene regulatory circuitry, by which genes are switched on and off in different cell types, in different stages of development, and under different metabolic and environmental conditions, are only poorly understood but are central to a real understanding of human disease, genetic susceptibility, and genome function. We will bring a new level of mathematical sophistication to the interpretation of these data. By addressing this critical problem, our proposal sets the stage for LLNL teams to build tools and expertise for a future program in gene-circuitry modeling.

The methods we develop will enable analysis of gene-regulation networks in any species, from microbes to humans. Cataloguing the associations between structural features of regulatory elements and the expression patterns of the corresponding genes will provide the basis for models of the mechanisms by which gene activity is controlled in living cells. The "language of gene regulation" is very likely to be universal in many fundamental respects that are shared by all organisms. The methods we develop will have wide application in the study of the mechanisms of cellular homeostasis and environmental response in a wide variety of living systems.

Mission Relevance

Analysis of gene-regulation mechanisms will yield significant insight into several topics relevant to the Laboratory's missions in homeland security (e.g., countering the threat of bioterrorism), environmental management (e.g., bioremediation), and human health (e.g., controlling and treating disease).

FY03 Results

We implemented our basic approach and tested it on yeast data. We designed four different tests to verify whether the scores obtained for the rules are significant, three with randomized data generated under different protocols, and one in which scores are computed for genes associated with motif sets that are already known to coregulate genes. Obtained results confirmed the validity of our approach. We also addressed the issue of local similarities in gene-expression profiles.

Proposed Work for FY04

We will adapt our methodology to model the circuitry that regulates human genes. Having already begun developing tools for comparative analysis of gene expression in closely related systems, we will test those tools with studies aimed at identifying differences between gene expression in human and mouse. We will also continue developing tools to model temporal interdependencies between patterns of gene expression.

Microbial Pathways

Michael P. Thelen

03-ERD-062

Abstract

Microbial processes can help solve complicated problems of soil and groundwater contamination that threaten human life. We intend to characterize signature biochemical pathways in selected microbes by developing novel biological and computational technologies. By increasing our understanding of microbial processes that are relevant to metal and radionuclide contamination in the environment, this project will bring a depth of expertise and a technical base to various national-security activities. We will do this by coordinating existing capabilities in genomics, microbiology, biochemistry, geochemistry, and computational modeling into a significantly new direction for bioscience.

Our goal is to create a bacterial gene clone resource for general microbiology studies and achieve scientific results pertaining to the molecular mechanisms underlying microbial processes that cause oxidation of uranium and other metals, as well as the mechanisms underlying the overall resistance of toxic metals to oxidation. Specifically, we will sequence the entire genome of *Thiobacillus denitrificans*, clone many of its genes, examine the expression of certain genes in response to environmental changes, and demonstrate the extent of this soil bacterium's metal-oxidizing capabilities. Proteins that are important in this process will be produced, purified, and examined for protein complex interactions. The proposed project is anticipated to encompass collaborations within and between the Laboratory and the DOE Joint Genome Institute.

Mission Relevance

We will further the emerging discipline of biogeochemistry that supports LLNL's mission in environmental management. Microbial processes active in the near-surface environment are important considerations in national security, especially issues concerning radionuclide and toxic-metal contamination of groundwater.

FY03 Results

As an initial step toward creating a bacterial clone resource for general microbiology studies, we characterized the soil bacterium *Ralstonia metallidurans*, using its known genome sequence as primary information for selecting sequences. We cloned several hundred genes (up to 1000 in total will be cloned); developed a database and web tracking tools; and created the proteomics tools for a later stage of the project—establishing dynamic light scattering in line with chromatographic purification of proteins, immobilizing proteins on chip surfaces using oligonucleotide aptamers, and analyzing bound proteins by surface plasmon resonance.

Proposed Work for FY04

In FY04, we will comprehensively characterize *T. denitrificans* using (1) geochemical analysis of microbially mediated metal oxidation–reduction (redox) reactions; (2) genome sequencing, gene expression analyses, and computational comparisons to identify metal resistance and redox genes; (3) homology modeling to predict protein structures and complex formation; (4) gene clone microarrays of protein-coding sequences; (5) production of proteins and purification by liquid chromatography with in-line, dynamic light scattering to determine physical properties; and (6) surface plasmon resonance coupled with mass spectrometry to analyze interactions in multiprotein complexes.

Protein Model Database

Krzysztof A. Fidelis 03-ERD-063

Abstract

We are developing the Protein Model Database (PMD), a global database of theoretically derived protein structures. Two main thrusts are envisioned for the PMD: (1) high-throughput, genome-scale automated modeling and structure prediction and (2) high-quality supervised modeling involving concerted efforts by multiple research groups worldwide. The increased ability to generate models of protein structure will be demonstrated by applying the PMD to existing projects. Livermore's lead role in the large-scale prediction experiments of the Critical Assessment of Protein Structure Prediction (CASP) effort and in organizing the Ten Most Wanted collaboration will be leveraged to implement this project.

For over 99% of all sequenced proteins, no data on the corresponding three-dimensional structure exist, opening a significant gap to be filled by modeling and prediction methods. The PMD, with its capacity to help generate high-quality, well-annotated models, will be a world-class capability that will help close this gap and make theoretical models more accessible to biologists. We will also demonstrate the significance of this initiative by applying the increased modeling capability to other science projects, such as those involving microbial pathways and pathomics.

Mission Relevance

Establishing the PMD will promote the development of protein-structure models and facilitate the ways they can benefit other projects, ranging from studies of the human genome and research on human disease, to biodefense and environmental studies. The basic nature and general applicability of protein-structure data is relevant to several Laboratory missions, from homeland defense to bioscience to improve human health.

FY03 Results

In FY03, we began to develop a rudimentary database containing models of protein structures with annotations of model quality, including estimates based on the performance of past models and on the difficulty of the modeling task. We completed the initial version of the database schema, including development of mmCIF (macromolecular Crystallographic Information File) protein-model-specific dictionaries, and held discussions about the future PMD with prominent members of the modeling community and the National Institutes of Health.

Proposed Work for FY04

In FY04, we will complete the rudimentary protein-structure model database. Whenever possible, specialist-oriented curation of models will be developed to lower model error rates and promote strong modeler–biologist interactions. Custom search capabilities will include specialized tools allowing for model comparison and visualization.

The Instrumented Cell

Allen T. Christian 03-ERD-068

Abstract

The goal of this project is to develop capabilities to measure, manipulate, and model life at the level of individual cells. To do this, we will develop a prototype device that will enable individual cells to be captured and held in place. The microenvironment will be controlled for temperature, pH, and other growth-related conditions, and will allow cells to be viably maintained while being continuously sampled. The device will be scalable, enabling many individual cells to be treated and sampled simultaneously—varying types of single cells, both prokaryotic and eukaryotic, and arraying them in a way that permits perturbation and analysis of intercellular and intracellular pathways and systems. We will use the system to test certain aspects of siHybrids, which are small, double-stranded molecules consisting of one strand of DNA and one of RNA, and which we have demonstrated to be highly effective in silencing genes.

This project aims to develop models of chemical interactions within and among cells, following perturbations made within the cell and to the cell's environment. The concept is that all of the pathways of a cell would be computationally modeled, allowing predictive models of cellular responses to stimuli to be created. Accomplishing this at the statistically relevant level of the individual cell would revolutionize biology, from basic research to industrial pharmacology. No method of acquiring quantitative data at the level of a single cell currently exists, nor does the technology to control and vary the environment of a cell while simultaneously injecting and sampling fluids and subcellular particles.

Mission Relevance

This project will strongly leverage LLNL capabilities in microfabrication, analytical techniques, and cellular biology to enable single-cell characterization of important biological processes. The result will be a capability that will lead to advances in bioscience and technology, in support of LLNL's missions in biodefense and in bioscience and technology to improve human health. For instance, a single-cell analytical platform will benefit DOE's Genomes-to-Life initiative, as it will allow measurement techniques to be focused on a single cell, kept alive in precisely controlled conditions.

FY03 Results

In FY03, we designed and built an engineering platform that will array and maintain individual cells, and integrated various measurement capabilities into the design to control the microenvironment of the platform. Tests were begun, and we obtained results that suggest that many more siHybrids are entering a cell than were first thought. This provides data for a hypothesis on the longevity of siHybrid-mediated gene silencing.

Proposed Work for FY04

In FY04 we will construct and test the platform. To elucidate the mechanism of action of siHybrids, and to demonstrate the feasibility of the prototype device for mechanistic studies, we will conduct various tests, including testing the following kinetic aspects of siHybrids in mammalian cells: rate of buildup in the cells, residence time in the cells, and rate of loss from the cells over time.

Elucidating the Mechanism of Gene Silencing using Small Interfering RNA–DNA Hybrid Molecules

Lawrence C. Dugan

03-ERI-004

Abstract

The objective of this project is to answer a number of important questions concerning a new gene-silencing technology known as small interfering hybrids (siHybrids), which have shown improvements over siRNA, a powerful tool for silencing gene function that is under consideration for use in clinical settings, including cancer and HIV therapy. In this project, cells are being treated with siHybrid molecules and then collected and analyzed for the presence of the siHybrids using accelerator mass spectroscopy (AMS) and nanosecondary ion mass spectroscopy (nanoSIMS). We are also using chromatin immunoprecipitation technology to determine whether histone methylation at the gene locus is involved in long-term gene silencing by siHybrids.

Successful completion of this project will provide a population-based answer to the question of how many siHybrid molecules are required to cause an effect in a cell, where the change occurs, and how long the molecules are present within the cell. Furthermore, we expect to identify which sequence within the hybrid plays a functional role in the gene-silencing effect. These answers will provide important clues for research to silence mutant p53 in prostate tumor cells, for instance. Finally, this project will provide important groundwork for other research such as the Instrumented Cell Strategic Initiative.

Mission Relevance

This research supports LLNL mission areas of (1) national security, by furthering research that could reduce the effectiveness of biological warfare agents, and (2) bioscience and biotechnology to improve human health, by providing mechanistic information for a process being tested for use in treating cancer and infectious diseases. This information will also help determine gene functions in cellular processes, e.g., development, differentiation, cell signaling, and cell death.

FY03 Results

We determined the experimental conditions needed for using AMS for carbon-14 (^{14}C) analysis of siHybrid molecules and identified unique mRNA sequences for a polymerase chain reaction- (PCR-) based DNA synthesis system. This system is now being tested in human HeLa cells. The first nanoSIMS images of biological samples at LLNL were also obtained. Our results showed that siHybrid molecules are degraded by cellular RNase III but not by cellular RNase H, as expected, and that stimulated cycling cells take up siHybrids while noncycling cells do not. Fluorescence imaging of siHybrid molecules indicates that cells readily take up these molecules and localize them within the cytoplasm. This work continues to track the siHybrids over many days in the cells.

Proposed Work for FY04

In FY04, we will incorporate ^{14}C into PCR-based siHybrid molecules and run AMS analysis of cells at varying timepoints after transfection to obtain population-based averages for number of molecules per cell. Using this same system, we will use ^{14}C or ^{15}N labeling of the siHybrids for analysis and localization on the nanoSIMS instrument. SiHybrid molecules labeled with different fluorochromes on each strand will be analyzed for subcellular localization using confocal microscopy with similar time points as the AMS analysis, which should allow us to determine the strand involved in inducing gene silencing. Finally, we will study the occurrence of changes in histone methylation states at the G6PD gene as a possible long-term effect of siHybrid molecules.

Cellular Response to Heat Stress: System Stability and Epigenetic Mechanisms

Halima Amer 03-ERI-005

Abstract

This project studies the mechanism of epigenetic stress response by monitoring single cells for the persistent activation of heat-shock proteins over many generations. This unique single-cell approach will remove the ambiguities associated with monitoring expression in cellular populations. The nature of the distribution of the cellular-stress response, and the subsequent establishment of clonal populations exhibiting a range of stress responses, will provide useful insights into the factors underlying such response and phenotypic heterogeneity. Developing the techniques for quantitatively monitoring such phenotypic change will provide a tool for subsequent studies and form the groundwork for systems-level modeling of cellular adaptation.

The project will further the development of a unique experimental platform in which the variation of protein-expression levels can be studied in single cells as part of a wider cellular population. In addition to examining the variation in cellular response, a deeper understanding of the mechanism of heat-shock response at the cellular level will be gained. Both of these factors will ultimately contribute to understanding phenotypic variation across an apparently genotypically identical population of cells under different environmental conditions.

Mission Relevance

By providing insight into the epigenetic activation of protective systems in cells that are exposed to stresses such as heat or radiation, this project will support LLNL's mission in bioscience to improve human health. The project sets will set the groundwork for future collaborations on a wide range of biological applications, including those relevant to NIH and DoD.

FY03 Results

We are using a special "immortalized" cell line (BJ1) that is stable over many generations. In FY03 we established this cell line and showed that we can make clones of these cells with adequate efficiency (>50%) for the proposed experiments. Next, we modified the BJ1 cells so that they contain a fluorescent indicator for the expression of a heat shock protein (HSP70). This was accomplished by creating a "vector construct" containing the DNA sequence activated by heat shock (the HSP70 promotor) linked to the sequence for a fluorescent protein. We successfully completed the first step, which involved making many copies of the component DNA sequences using DNA polymerase chain reaction. The final assembly of the vector construct involved "cutting and pasting" the DNA components together. We initially attempted to use a simpler "blunt-ended" approach to assembling the vector and were unsuccessful, but we succeeded using a more complex, but more precisely controlled "sticky-ended" approach. We completed preliminary assembly of the DNA components and will next perform DNA sequencing to confirm the integrity of the final product.

Proposed Work for FY04

A mechanism for selecting single cells after imaging will be developed, and a protocol for quantifying the amount of green fluorescent protein expressed as a result of heat shock will be established and correlated with an immunometric assay. The distribution of the response of single cells to heat shock will be determined under differing conditions of heat stress. The experiment will be repeated on generations of cells selected and cloned on the basis of their heat-shock response. The effect of additional stresses, such as radiation and oxidation, will be used to examine cell vulnerability. The resistance of vulnerable cells to thermal killing will also be studied. Finally, variation in the expression of the human heat-shock protein Hsp70 through the cell cycle will be examined.

Force Spectroscopy to Study Multivalent Binding in Protein–Antibody Interactions

Todd A. Sulchek

03-ERI-009

Abstract

This project is using the atomic force microscope (AFM) as a tool to measure the binding force between mucin1 (Muc1) protein, an indicator of some cancers, and a natural antibody. This binding interaction is the targeting mechanism in radioimmunotherapy—a technique used to treat cancer tumors. The Muc1 protein will be attached to a surface and the antibody attached to an AFM tip. The goals of this project are to determine the unbinding force and the shape of the interaction potential between one and more Muc1–antibody bonds, determine the optimal number of antibodies and tether length used to connect them in radioimmunotherapy, and finally, study details of multivalent binding.

Developing a force-measurement strategy opens the door to studying other molecular interactions that occur at the interfaces of cells, often mediated by multivalent interactions. For example, measuring the forces generated by these interactions should provide important information about mechanisms of antibody function. Also, multivalent antibody binding is the

targeting mechanism in radioimmunotherapy that determines the optimal number of antibodies and tether length.

Mission Relevance

The capability to measure antibody–protein interactions, combined with expertise in suspended bilayers will enable biological applications in characterizing cell–pathogen interactions for laboratory missions in counterproliferation and homeland security, and applications in drug–cell and cell–cell interactions for LLNL missions in basic bioscience research to improve human health.

FY03 Results

This year we successfully functionalized the AFM tips and surface. Our nitride cantilevers were coated with sputtered gold to ready the surface. Scanning electron micrographs revealed that the gold is uniform and completely coats the tip, even after repeated use. We attached antibodies to the tip using a polymer tether, which helps keep the antibody functional. We also covalently linked the Muc1 protein to the surface via a polymer tether. Force spectroscopy data indicate that binding interactions take place at a distance of approximately two tether lengths. Public-domain data-analysis routines for efficient analysis of the force distance curves were modified. Finally, using surface plasmon resonance measurements, we determined the binding strength of the Muc1 and antibody (antiMuc1).

Proposed Work for FY04

In collaboration with the University of California, Berkeley, we will obtain new antibody clones and help for multiple antibody attachment to the tip. Work to characterize Muc1 tethered to the surface with surface plasmon resonance and force spectroscopy will continue in FY04. The Muc1 will be incorporated in suspended lipid bilayers to create a model cell surface. Using the AFM platform, we will determine the unbinding force for a range of pulling rates. The relationship between unbinding force and the pulling rate will suggest a value for the characteristic bond length. This process will be repeated for one and more tethered antibodies. By varying the dwell times, we can determine the on-times for one and more bonds to form.

Intracellular Chemical Measurements: A Generalized Approach with High Spatial Resolution using Functionalized Nanoparticles

Ted A. Laurence
03-ERI-010

Project Description

The objective of this project is to develop novel methods to measure concentrations in chemical microenvironments in cells and tissues using recently developed, functionalized metal nanoparticles (50–100 nm). Current methods are limited by low spatial resolution and high cell toxicity. Surface-enhanced Raman spectroscopy (SERS) allows sensitive detection of changes in the state of chemical groups attached to single nanoparticles. A nanoscale pH meter, which has already been tested in a cell-free medium, will be tested inside cells to determine optimal conditions and methods for these measurements. When tested and calibrated, the nanoscale pH meter will be used to image the local pH inside and outside cells in tumor tissues (which contain acidic extracellular matrices), and to study the characteristic pH drop associated with the early stages of apoptosis.

Success with this novel measurement technique will demonstrate the ability of SERS nanoparticles to provide rigorous quantitation of cellular concentrations, opening new windows to cell characteristics and behavior and encouraging further development of SERS technology to measure chemical concentrations inside microenvironments. In tumor cells, it will be possible to monitor the effects of the local pH gradient on uptake of various therapeutic agents. A decrease in pH will serve as an indicator of apoptosis; correlation with stresses or other signals will help identify events, causes, and phenomenology of apoptosis.

Mission Relevance

Measuring chemical gradients (variations in concentration) across membranes is important for understanding basic biological processes and is relevant to developing disease treatments. This research supports development of a novel method for measuring chemical concentrations. Such probes of the intracellular environment represent a key enabling capability for the DOE Genomes-to-Life initiative and biosecurity applications. In addition, the new measurement technique supports LLNL applications related to bioscience and technology to improve human health, as well as breakthroughs in fundamental bioscience.

FY03 Results

We performed single-particle studies of SERS nano-pH meters freely diffusing in solution. To detect any signal-intensity fluctuations and detect heterogeneity, experiments were performed with high time resolution, at the sacrifice of spectral resolution. These experiments revealed heterogeneity of signal intensity from the particles, but no large distribution in translational diffusion times.

Proposed Work for FY04

To validate and perfect the SERS nanoparticle approach and apply it to specific biological questions, we plan to perform the following experiments in FY04: (1) the signal intensity and spectra of the nanoparticles will be tested in buffer conditions similar to those found in cells, (2) nanoparticles will be incorporated into liposomes to test the nanoparticles in a more cellular-like environment, (3) methods of incorporating the SERS nanoparticles into cells will be tested [for example endocytosis, which would use the liposomes from experiment (2)], (4) cells containing SERS nanoparticles will be monitored over periods of hours to days to test viability of the cells, and (5) the pH of the intracellular and extracellular environments will be measured in tumor cells and cells undergoing apoptosis.

Single-Molecule Techniques for Studying Chromatin Assembly and Remodeling

Christopher Jeans

03-ERI-011

Abstract

In eukaryotic cells, DNA is tightly packaged as chromatin through interactions with histone proteins. For processes such as DNA replication, repair, and transcription to occur, the structure of chromatin must be remodeled such that the necessary enzymes can access the

DNA. Our understanding of the remodeling process is hindered by a lack of knowledge of the fine structure of chromatin and how this is modulated in the living cell. We are carrying out single-molecule experiments using atomic force microscopy (AFM) to determine the nature of packaging interactions in chromatin and to measure the forces involved in such interactions. The effects of histone modifications on these forces will be determined, as will the effects of damage within the DNA.

The images of chromatin and the measured forces will be used to gain insight into the nature of the interactions that hold the chromatin fiber together under different conditions. This will give us valuable new information about the structure of chromosomes and the way remodeling affects this structure. Improved knowledge of the remodeling process will aid in understanding a number of cellular processes, from DNA replication and repair, to the mechanisms of gene activation and silencing.

Mission Relevance

This work will address questions of general biological interest through the development of novel techniques for the manipulation of single molecules and will establish a new capability. Specifically, the technology of physical manipulation of individual DNA strands may provide critical data for biodefense applications—in support of homeland security—and for DOE's Genomes-to-Life program.

FY03 Results

Chromatin fibers were isolated from chicken erythrocytes and Chinese hamster ovary cells. Agarose gel electrophoresis show the samples contain a broad range of fiber sizes, from single nucleosomes to chains of 20 nucleosomes or more. Sodium dodecyl sulfate–polyacrylamide gel electrophoresis (SDS-PAGE), a standard biochemical technique used to separate all the protein in a sample on a gel according to their size, determined that the samples contain only the core and linker histones, with no detectable contamination from cytoplasmic or nonhistone nuclear proteins. Preliminary AFM imaging was carried out on chicken erythrocyte chromatin. In these images, single nucleosomes and short chains of nucleosomes were readily identifiable, and regions of linker DNA between nucleosomes were seen. This work provides a strong basis for future analysis of AFM images of chromatin, and for experiments designed to stretch individual fibers with the AFM tip.

Proposed Work for FY04

In FY04, chromatin fibers isolated from chicken erythrocytes and Chinese hamster ovary cells will be imaged using AFM. The procedure used to prepare these samples for the AFM will be optimized so that individual fibers of known length can be manipulated. Chromatin reconstituted from DNA and histone proteins formedaccording to published protocols will also be investigated. Following sample optimization for AFM work, the AFM tip will be used to stretch the chromatin fiber away from the substrate, and the forces needed to achieve this will be measured. With the fiber extended, chemical and compositional modifications will be performed on the extended fiber, and changes in its tension will be measured. Post-modification images will also be taken.

Using Femtosecond Laser Subcellular Surgery as a Tool to Study Cell Biology

Nan Shen

03-ERI-012

Abstract

This project is investigating a novel surgical technique—femtosecond laser pulses—to alter processes in living cells. By tightly focusing laser pulses with nanometer-scale resolution inside human cells, the technique targets and vaporizes cellular material through nonlinear optical processes. We are using this technique to study the role mitochondria play in cell proliferation and apoptosis by selectively perturbing mitochondria in living cells. The experimental apparatus will allow us to selectively target a number of different subcellular structures for disruption and also to study cell behaviors in real time.

This research addresses questions about the long-term behavior of cells after laser surgery and the different cell responses that result from disruptions of selected cellular structures. With this setup, it may be possible to induce apoptosis by disrupting mitochondria in a cell and study the process of cell death. We also hope to determine if the number of functional mitochondria is critical to the survival of a cell, and whether mitochondria are capable of repairing structural damages. Functions of a number of proteins believed to be regulators of apoptosis will also be investigated. Our technique will provide a unique tool for the study of cell biology.

Mission Relevance

This research supports DOE missions in understanding cellular mechanisms, e.g., the Genomes-to-Life Program. Furthermore, the understanding gained in this project will ultimately prove important in helping to determine complete mechanisms for pathogenicity, in support of LLNL's homeland security mission.

FY03 Results

The project started late in FY03. The experimental setup, consisting of a Ti:sapphire femtosecond laser, a Nikon TE 2000 inverted microscope, and a CCD imaging system, was completed and the cell-culture facility was updated, where BJ1 and InfinityTM, human fibroblast and bovine endothelial cell lines, are maintained for our experiment. The BJ1 cell line was successfully transfected with plasmid DNA that encodes a fusion of enhanced yellow fluorescent protein (EYFP) and the mitochondrial targeting sequence of cytochrome c oxidase. Cells expressing EYFP were selected and cloned to obtain stably transfected cell population. We also compared fluorescent labeling of mitochondria using EYFP and commercially available fluorophore JC1 and in different cell lines.

Proposed Work for FY04

The first stage of the research in FY04 will focus on investigating how cells respond to femtosecond laser surgery. Using imaging techniques such as scanning electron microscopy, we plan to study the resolution limit of femtosecond laser surgery and determine the ultimate laser parameters for surgery for future studies. The next goal is to better understand the function of mitochondria and their role in cell apoptosis. We plan to probe mitochondria functions by selectively inducing physical disruptions on one or more mitochondria with laser pulses and to use imaging techniques to study cell responses.

Automated Three-Dimensional Protein-Structure Predictions Based on Sensitive Identification of Sequence Homology

Adam T. Zemla

02-LW-003

Abstract

Genome sequencing projects require many new proteins to be characterized in terms of their structure and biochemical function. Because the cost and time required to characterize such proteins experimentally is prohibitive, computational methods hold great promise for uncovering the structure and function of new proteins. Our research develops a computational homology-based protein structure prediction system that goes beyond current sequence-homology search capabilities. Our strategy is to predict protein structure by identifying unique folds, distinct conformations that are well conserved throughout all living organisms. The novelty of our approach lies in combining sequence and structure analyses to identify structural homologs of proteins for which sequence similarity alone is insufficient.

The chief aims of this proposal are to (1) develop a method of characterizing newly sequenced proteins in terms of their 3-D structure and function and (2) package this method into a fully automated Web-based system. The developments in sensitive identification of homology proposed in our research significantly improve and facilitate the template-building phase of protein structure prediction, thereby enabling scientists who search genomic sequences to more quickly identify a larger percentage of unknown genes. Our working protein structure predicting system (currently accessible at http://protein.llnl.gov/AS2TS) will be useful for biologists both at LLNL and worldwide.

Mission Relevance

Models generated by this system are used to predict the regions in protein structures where DNA signatures designed at LLNL landed, to determine potential unique protein signature candidates, to suggest locations for vaccine targets, and to suggest probable functions of unknown proteins. For example, the capabilities of this system have been applied to model critical proteins of smallpox, anthrax, plague, foot and mouth disease, monkeypox, African swine fever, and others in support of the national security and homeland security missions.

FY03 Results

Work in FY03 focused on methods of identification and analysis of structural similarities of proteins that do not have significant amino acid sequence similarity, development of modules to perform modeling of “difficult regions” in three-dimensional (3-D) models, and improvement of our modeling methods through collaboration with other researchers and experimental groups. Our modeling work included generating 3-D models of *Mycobacterium tuberculosis* and *Yersinia pestis* proteins to facilitate a molecular replacement process; modeling proteins and signature regions for plague, anthrax, and foot-and-mouth; and protein-structure modeling for the collaborative project “Plague plasminogen activator as a vaccine component” with the University of Texas Medical Branch.

Publications

Fitch, J. P. et al. (2002). *Biosignatures of pathogen and host?* Presented at the IEEE Wkshp. Genomic Signal Processing and Statistics (GENSIPS), Raleigh, NC, Oct. 12–13, 2002. UCRL-JC-149741.

Miller, K. A. et al. (in press). "Domain mapping of the Rad51 paralog protein complexes." *Nucleic Acids Res.* UCRL-JC-155506.

Miller, K. A. et al. (2002). *Analysis of RAD51 paralog complexes.* Presented at Maintenance of Genomic Integrity, Houston, Texas, Oct. 15-18, 2002. UCRL-JC-150531-ABS.

Slezak, T. (2003). "Comparative genomics tools applied to bioterrorism defense." *Brief. Bioinformatics* **4** (2), 133–149. UCRL-JC-151394.

Zemla, A. (2003). "LGA—A method for finding 3D similarities in protein structures." *Nucleic Acids Res.* **31** (2003), 3370–3374. UCRL-JC-152021.

Mutations that Cause Human Disease: A Computational–Experimental Approach

Peter T. Beernink

03-LW-017

Abstract

This project uses computational and experimental approaches to assess the effects of mutations that cause human diseases. Genetic variation is hypothesized to lead to differences in individual susceptibility to cancer, which can be caused by environmental carcinogens, radionuclides, or spontaneous mutations. The number of known human genetic-variant proteins has grown exponentially in the past 2 yr, and the ability to integrate these data will depend on computational methods. Because most disease-causing variants impact protein stability, we will predict the structural properties of disease-causing proteins in cases where the structure of the normal counterpart is already known. Predictions will be validated with quantitative measurements of protein thermal stability. The resulting methods will allow a rigorous analysis of over 100 proteins that cause single-locus genetic diseases and that have known wild-type structures.

We anticipate developing new computational approaches for structure-based predictions of variant-protein stability. In addition, the experimental data on stability and structure of certain proteins involved in cancer obtained in this study will elucidate the role of those variants in disease. The experimental studies will serve to validate the reliability of our predictions of the stability of variant proteins and their roles in disease.

Mission Relevance

This work supports the missions of LLNL in bioscience to improve human health and contributes to NIH missions by enhancing our understanding of natural human genetic variation, radiation biology, and cancer susceptibility. Genetic variation is hypothesized to lead to differences in individual susceptibility to cancer, which can be caused by environmental carcinogens, radionuclides, or spontaneous mutations. This work will identify specific effects of disease-associated genetic variation on protein structure and stability.

FY03 Results

We made significant progress in FY03. First, we applied a high-throughput, computational method to examine the stability of >220 protein variants using steric clash penalties and solvent accessible surface areas. Second, in experimental studies, we evaluated the thermal stability of cancer-associated variants of a DNA repair protein, Ape1, which are significantly less stable than the wild-type protein. Third, we established a correlation between experimental thermal stability and the root-mean-square atomic fluctuations in molecular-dynamics simulations. Given an atomic structure of the normal protein, this method has the potential to be used to predict variant protein stability computationally.

Proposed Work for FY04

In FY04, we will expand our computational energy predictions to include point energy calculations using an empirically derived energy force field; extend our intensive computational and experimental studies of protein stability to other disease-causing proteins, including p53, XRCC1, and RNaseH; and continue developing a high-throughput, protein-stability screen using cell free-protein production techniques. Once established, this method will allow hundreds of variants to be examined for their effects on protein stability.

Publications

Beernink, P. T. et al. (2003). *Cancer associated variants of human Ape1 exhibit reduced enzyme activity and thermal stability.* Presented at UC Davis Cancer Center Symp., Sacramento, CA, October 9–10, 2003. UCRL-POST-200148.

Ribozymes with Sequence Elements for Regulation of Expression

Harvey W. Mohrenweiser 03-LW-033

Abstract

Understanding the influence of complex patterns of genetic variation in healthy people at risk of disease is a high priority now that the DNA sequence of genes is known. This project focused on developing tools to study the effects of varying the activity of genes in the cell via regulation of their level of expression. The test platform was four genes of the Base Excision Repair pathway, which is critical for the repair of DNA damaged by oxygen radicals from cell metabolism and exposure to ionizing radiation. To minimize the risk of cancer, this pathway must function normally. Relating deviations in the levels of protein expression to quantitative estimates of the DNA repair capacity of cells establishes reference points relating genetic variation to cancer risk.

The success of this type of research could ultimately lead to increased understanding of the relationship between risk of cancer and genetic variation in the genes that determine individual capacity to repair damaged DNA. Determining individual risk addresses the hypothesis that most cancer occurs in a subset of the population with modestly elevated genetic susceptibility to exposure. Confirming this hypothesis would have a critical impact on dose-response assumptions and estimates of risk from low-level exposures, including exposures to ionizing radiation.

Mission Relevance

This effort supports LLNL's mission in bioscience to improve human health and NIH missions by furthering (1) the Human Genome Project's human susceptibility effort to understand how genes and environment interact; (2) efforts to develop a better scientific basis for estimating risk from low-dose radiation; and (3) efforts to identify radiation-sensitive subpopulations, which also contributes to goals of the Nuclear Isotope and Radiation Program in support of the Department of Homeland Security.

FY03 Results

In FY03 we designed and synthesized small inhibitory RNAs (siRNAs) for the genes POLB and FEN1; identified cell lines for experiments, verified their compatibility with siRNA transfection agents, and evaluated repair of radiation-induced damage; began development of additional repair-capacity assays; validated antibodies for immunofluorescence detection of POLB and FEN1 protein; designed primers for POLB and FEN1 messenger RNA quantitation; and published a paper in *Mutation Research*.

Publications

Mohrenweiser, H. W., D. M. Wilson III, and I. M. Jones. (2003)."Challenges and complexities in estimating both the functional impact and the disease risk associated with the extensive genetic variation in human DNA repair genes." *Mutation Res.* **526**, 93–125. UCRL-JC-152062.

Quantitative Carbon-Isotope Ratio Measurements of Individual Cells

Ted Ognibene

03-FS-011

Abstract

The ability to perform quantitative isotopic analyses of individual cells and components of individual cells could transform our understanding of many biological processes such as cell division, protein expression and localization, intra- and intercellular interactions, pharmaceutical targeting, and chemotherapeutic interactions. In this project, we set out to develop and demonstrate protocols that enable quantitative carbon-isotope ratio measurements to be performed on individual cells and components of individual cells by accelerator mass spectrometry (AMS). Yeast cells were cultured in carbon-14- (^{14}C-) glucose. We used micromanipulation techniques to isolate individual cells and quantified the amount of ^{14}C in each cell using AMS.

This project established techniques to isolate and quantitate individual cells and subcellular components for carbon-isotope ratio measurements by AMS and secondary ion mass spectroscopy. Such measurements could make significant contributions to basic understanding of biological processes at the cellular and subcellular levels.

Mission Relevance

This work will develop tools for modeling the cell and quantitating pathways in individual cells in support of the DOE's Genomes-to-Life program. This research also extends and enhances advanced analytic capabilities valuable to the Laboratory's nonproliferation and homeland security missions.

FY03 Results

In FY03 we established techniques for labeling individual cells with ^{14}C. Yeast cells were cultured on isotopically enriched culture media until up to 1% of the carbon atoms were ^{14}C-labeled. Cells were rinsed to remove unincorparated media and resuspended in growth media containing contemporary levels of ^{14}C (i.e., essentially depleted in ^{14}C relative to the ^{14}C-labeled media. Individual yeast cells were isolated with a micromanipulator for quantification of ^{14}C by AMS. Individual yeast cells, cultured in unlabeled media, were also prepared as controls. We demonstrated quantification of ^{14}C from individual yeast cells and determined the amount of ^{14}C glucose that is required to give a measurable signal with AMS.

Estimating the Person-to-Person Stability of mRNA Signatures of Radiation Exposure in Humans

Christine L. Hartmann-Siantar 03-FS-029

Project Description

This project aims to provide a preliminary estimate of the person-to-person variation in gene expression in human cells that have been exposed to ionizing radiation. Gene expression is known to change with time and dose after exposure, suggesting that it can be used to assess exposure dose in individuals after an exposure event. Understanding gene-expression variability among individuals is important in assessing the feasibility of messenger RNA (mRNA) expression profiles as biological dosimeters of exposure to radiation. We investigated gene expression variation in mRNA signatures after exposure to ionizing radiation using a well-characterized national collection of human lymphoblastoid (HLB) cell lines, obtained from a diverse population of adults through the National Institutes of Health Human Genetic Cell Repository.

The anticipated results of this work are a set of genes that are consistently up- or down-regulated in response to radiation dose and consistent across cell lines and individuals. This finding will be an important step towards establishing the feasibility of using gene-expression profiles as biodosimeters of exposure to ionizing radiation. As results are analyzed, they will provide some of the first evidence of variation in human gene expression following radiation and a preliminary assessment of which genes are most robust for post-exposure radiation dose assessment.

Mission Relevance

The study of radiation interactions with biological systems has been a long-standing component of the Laboratory's national security mission, which is a mainstay in the Laboratory's role as a resource to the U.S. government and partner with industry and academia. This project contributes to the ability to manage the medical consequences of using nuclear and radiological weapons of mass destruction.

FY03 Results

In FY03, we completed the irradiation and cell-preparation experiments to characterize the effects of variables important for the application of radiation dosimetry: exposure dose, time after exposure, and, most importantly, individual variation. Cultures of HLB cells representing the diversity in the U.S. population were grown from frozen stocks and expanded, then irradiated at two radiation doses plus controls, and then sampled at several post-irradiation times. We prepared over 500 vials of cell samples that represent separate time, dose, or cell-line replicates. RNA was then isolated from over 80 aliquotes and analyzed with gene transcript microarrays representing approximately 22,000 human genes per array. Analysis of the data was underway at the end of the year.

Chemistry

Chemical Deactivation of Reactive Uranium

Dianne D. Gates-Anderson

01-ERD-064

Abstract

Currently, LLNL has an inventory of at least 11,700 kg (158 drums, 33 m^3) of waste containing pyrophoric uranium metal that requires treatment prior to disposal. Approximately 12 m^3 of this waste stream is classified as mixed waste and must be treated for LLNL to remain in compliance with the Federal Facilities Compliance Act. Across the DOE complex, an additional inventory in excess of 40,000 kg of pyrophoric uranium waste also requires treatment. No commercially available, cost-effective, nonthermal treatment processes exist for this waste stream.

The objective of this project is to develop an integrated process to convert pyrophoric uranium metal wastes to a nonpyrophoric waste. This project's scope includes bench and pilot-scale studies using both low-level and mixed-waste depleted uranium. The reaction mechanism of the process developed is being determined with detailed kinetics and thermodynamic investigations. In addition, the solids and solutions formed during treatment are being characterized.

Mission Relevance

This project will determine the most effective physicochemical treatment process to stabilize pyrophoric uranium. Developing this technology supports LLNL's environmental-management mission and will enable LLNL and other sites in the DOE complex to treat their pyrophoric uranium wastes.

FY03 Results

In FY03, the third stage of the proposed depleted-uranium treatment process—stabilization—was studied in detail. Several different commercially available solidification and stabilization products were evaluated. A clay product was found to be the most suitable stabilization media and produced a final waste form that would not leach contaminants in a simulated landfill environment.

We also evaluated project scale and engineering considerations. The most critical design factor was determined to be the heat generated during depleted uranium dissolution. After completing several in-depth thermodynamic studies, a design for a full-scale treatment unit was prepared to asses the feasibility of this approach.

A full-scale, three-stage process is being built using the results of this project. The full-scale unit will be used to begin treating the legacy depleted uranium waste at LLNL. Having this unit operational will allow the Laboratory to meet the DOE mandate that all legacy waste, including depleted uranium, be treated by the end of FY05. Without the process development work in this project, this would not be possible.

Publications

Laue, C. A., D. D. Gates-Anderson, and T. E. Fitch. (in press). "Dissolution of metallic uranium and its alloys—part I: Review of analytical and process-scale metallic uranium dissolution." *J. Radioanalyt. & Nucl. Chem.* UCRL-JC-154536 Pt. 1.

Laue, C. A., D. D. Gates-Anderson, and T. E. Fitch. (in press). "Dissolution of metallic uranium and its alloys—part II: Screening study results: Identification of an effective non-thermal uranium dissolution method." *J. Radioanalyt. & Nucl. Chem.* UCRL-JC-154536 Pt. 2.

Mesochem: Chemical Dynamics on a Mesoscopic Scale

Laurence E. Fried

01-ERD-103

Abstract

This project proposes to develop a new modeling capability in the area of soft-matter chemistry and dynamics. The term soft matter describes systems in which diffusion, hydrodynamics, and possibly chemical reactions are strongly coupled. The crystallinity of polymers in explosives, grain structure aging in crystals such as pentaerythritol tetranitrate and other volatile materials, and the behavior of foams, pads, and cushions all involve soft matter effects. Soft matter is also characterized by dynamics involving domains between 10 nm and 1 mm in size. Moreover, the complex structure characteristic of soft matter leads to material behavior on a variety of time scales that are much longer than the 10-ns limit of molecular dynamics.

To create a framework to address this wide range of problems, we propose to expand upon a recently proposed method called dissipative particle dynamics (DPD) with the goal of solving several difficulties associated with application of DPD to real materials. One problem is determining an accurate mesoscopic representation of a system based on underlying microscale calculations. We plan to solve this problem by combining electronic structure and molecular dynamics simulations to derive effective DPD interactions and present the first truly multiscale approach to DPD. The result will be the ability to model microstructural changes in a variety of soft materials.

Mission Relevance

Development of a flexible code that can model soft matter on the mesoscopic scale aligns with the Stockpile Stewardship Program, as it will have wide-range application to goals related to material aging and constitutive behavior. If this work is successful, it will generate a scientifically unique model of nanoscale to mesoscale matter.

FY03 Results

In FY03 we developed a mesoscopic model of polymer crystallization that is closely based on the atomistic model of vinylidene fluoride. Effective chain interactions were developed to capture the essential feature of the microscopic models: the reduction in energy resulting from linear chain conformations. Large-scale atomistic and mesoscopic simulations were conducted using over 1000 CPUs on the Multiprogrammatic and Institutional Computing Capability Resource. The simulations showed the interactions of multiple crystalline regions for the first time and revealed that crystalline regions display a mixture of "adjacent re-entry," in which polymer chains re-enter the same crystallite, and "random re-entry," in which polymer chains re-enter more than one crystallite.

Publications

Gee, R. H. and L. E. Fried. (2003). "Ultrafast crystallization of polar polymer melts." *J. Chem. Phys.* **118**, 3827. UCRL-JC-242276.

Roszak, S. et al. (2003). "Molecular interactions of TATB clusters." *Chem. Phys. Lett.* **374**, 286. UCRL-JC-241598.

Modern Chemistry Techniques Applied to Metal Chelation with Medical and Environmental Applications

Mark Sutton

02-ERD-021

Abstract

This project aims to modernize and optimize the scientific approach to developing metal chelators using analytical and synthetic chemistry combined with thermodynamic and structural modeling. A specific screening process has been developed and used to choose beryllium (Be) chelators with promise for environmental, animal, and clinical trials. In addition, similar protocols will be used to assess chelation therapy for heavy-metal poisoning and other uses. This work is directly relevant to removing Be lung burden and has potential application in treating Chronic Beryllium Disease and for cleaning up environmental Be contamination in the DOE complex and industry.

By the end of the project, we will have rigorously investigated the chemistry of chelation therapy for reducing or removing Be lung or body burden, including optimum chemistry, structure, environment, efficiency, and selectivity. A potential new environmental cleanup application using chelator-incorporated aerogel will also be developed. In addition, our techniques will be applied to the medical treatment and environmental cleanup of uranium (U) and heavy metals such as lead (Pb), mercury (Hg), and arsenic (As). The results will benefit the health of DOE and industrial workers and contribute to medical research and environmental cleanup.

Mission Relevance

This work is a unique combination of medical toxicology and chemical science expertise and supports two DOE missions: bioscience to improve human health, with improved chelation therapy; and environmental management, with chelation techniques for environmental cleanup.

FY03 Results

During FY03, progress was made in researching both medical and environmental applications of Be decontamination. Accomplishments include (1) studying particle dissolution using selected chelators in simulated human biological fluids, including intracellular and interstitial fluids, plasma, saliva, sweat, urine, bile, gastric juice, pancreatic fluid, and airway-surface fluids; (2) improving our understanding of Be chemistry, sorption, and subsequent desorption with an aerogel/granular-activated-charcoal (GAC) material in both column and batch studies with applications in workplace and environmental cleanup; (3) bringing together additional LLNL capabilities to address workplace/environmental cleanup issues specific to LLNL; and (4) understanding U and Pb in biological fluids.

Proposed Work for FY04

We will continue to use chemical thermodynamics, synthetic chemistry, nuclear magnetic resonance, toxicology review, secondary ion mass spectrometry imaging, and laboratory application of chelators in pilot field studies to further investigate the effectiveness of selected Be chelators in both medical and environmental scenarios. This includes (1) animal trials to understand transport of Be/chelate complexes in a biological system; (2) additional work with variations of chelate/aerogel/GAC materials to improve cleanup applications; (3) removal of Be contamination from tools, work surfaces, and Site 300 facilities; and (4) exploration of potential applications of new chelators to wastewater treatment, bioagent decontamination, and heavy metals.

Publications

Sutton, M. and S. R. Burastero. (2003). "Beryllium chemical speciation in elemental human biological fluids." *Chem. Res. Tox.* **16**, 1145–1154. UCRL-JC-150385.

Local-Scale Atmospheric Reactive-Flow Simulations

Charles K. Westbrook
02-ERD-027

Abstract

This project is developing an atmospheric air-flow model to include reactions of gases and aerosols released into the air. The model describes chemical evolution over distances of a few kilometers of variable terrain, including obstacles such as buildings. This work builds on an existing finite-element atmospheric-flow model, FEM3MP, already in use on massively parallel computers, by adding subroutines to simulate gas-phase chemical reactions and aerosol-surface reactions of species carried along by airflow. Effects of humidity, sunlight, temperature, and concentrations of other chemical species on these reaction rates will be included in the evolution submodels.

The goal of this project is to produce a computational tool that will provide insight into the basic physics and chemistry of aerosols, which will help efforts to mitigate the adverse consequences of aerosol releases. The submodels will carry out basic science research on chemistry of toxic chemicals and aerosols in the atmosphere. Results of this work will be reported in scientific publications.

Mission Relevance

This work provides computational analysis tools for nonproliferation, counterterrorist, and environmental applications in support of LLNL missions in national security and environmental management.

FY03 Results

Additions were made to the existing FEM3MP code to include time-dependent chemical kinetics and aerosol evolution. These features were tested by examining problems with simplified but nontrivial kinetic rates of organophosphate-compound decomposition, and limited runs were performed with a surrogate of sarin, which could be benchmarked against experimental data. Scientific papers were published on the chemistry of sarin and nontoxic surrogates for chemical-warfare (CW) agents, as well as for chemistry simulations of surface chemistry of aerosol droplets. Full-system computational simulations were carried out successfully in realistic physical settings to test the model capabilities.

Proposed Work for FY04

In FY04, the computer model will describe reactive flow in the atmosphere on a spatial scale up to a few kilometers, including reactions involving gas-phase chemicals and particulates and aerosols. Work will focus on fundamental chemistry issues needed to simulate the evolution of aerosols in the atmosphere, with emphasis on chemicals and aerosols related to CW and biological weapons. Users of the FEM3MP code will be introduced to the new simulation capability.

Publications

Jayaweera, T. M., W. J. Pitz, and C. K. Westbrook. (2003). "Suppression of premixed C_3H_8-air flames by halogenated and phosphorus-containing compounds." *Proc. 3rd Joint Mtg. of the U.S. Sections of The Combustion Institute.* UCRL-JC-150885.

Transport and Biogeochemical Cycling of Iodine-129 from Nuclear Fuel Reprocessing Facilities

Jean E. Moran 02-ERD-058

Abstract

The unique biogeochemical properties of iodine make iodine isotopes excellent tracers of nuclear-fuel reprocessing activities on a local or regional scale. This project is developing methods for measuring the long-lived radioactive isotope iodine-129 (^{129}I) in the forms released from reprocessing facilities and in downstream environmental reservoirs. We are producing a capability to make fast measurements of ^{129}I and stable iodine in natural aqueous samples such as rainwater, river water, and groundwater. The results of these analyses will provide the scientific basis for predicting the transport and biological uptake of ^{129}I from releases from fuel-reprocessing facilities and will aide in unambiguous identification of its source.

Mission Relevance

The study of ^{129}I movement in the environment supports the Laboratory's counterproliferation mission by providing a tool for detecting clandestine nuclear-fuel reprocessing and supports the Laboratory's environmental assessment and restoration mission by providing a capability to determine the factors that control the fate and transport of iodine at several DOE sites.

FY03 Results

Chemical separation and analytical methods for determining the ratio of ^{129}I to stable ^{127}I by multicollector inductively coupled plasma mass spectrometer (ICP-MS) were developed, and measurement of $^{129}I/^{127}I$ by ICP-MS on untreated groundwater from an affected area was carried out. Successful analysis of solid and aqueous materials by x-ray absorption spectroscopy produced publishable results on iodine speciation and chemical form, and on the nature of the carbon–iodine bond in organic compounds. Key parameters that affect iodine transport and cycling in soils, such as total iodine concentration, iodine chemical form, soil-anion exchange capacity, and soil composition, were investigated in stir-cell column experiments.

Proposed Work for FY04

The results from benchtop soil-column experiments will be used to predict the environmental distribution of iodine. Environmental samples from affected areas such as Hanford Reservation and the Nevada Test Site will be analyzed for $^{129}I/^{127}I$ using the newly developed multicollector ICP-MS method. A comparison between plasma-source mass spectrometry and accelerator mass spectrometry techniques will be carried out and specific recommendations, based on new information from this project, will be made regarding the collection sites and matrix types for nonproliferation applications.

A Comprehensive Study of Surface Chemistry for Application to Engine NOx Aftertreatment

Salvador Aceves

03-ERD-020

Abstract

Diesel engines have been identified as high emitters of nitrogen oxides (NOx). New Environmental Protection Agency (EPA) regulations mandate a reduction in engine NOx emissions by a factor of 10, down to 0.20 g/hp/hr by 2007. This value is well below 1 g/hp/hr, which we have identified as the minimum to which NOx emissions can be reduced in a diesel engine with currently available technology. In this project, we are using our advanced modeling capabilities for chemical kinetics, fluid mechanics, and heat transfer to comprehensively study NOx traps, which represent the best available technical option. Scientific issues include developing chemical kinetic mechanisms for surface chemistry, analyzing sulfur poisoning of the catalyst surfaces, and countering the thermal aging of catalysts.

We are focusing on the fundamentals of NO_2 adsorption on barium oxide (BaO) surfaces and are using molecular-dynamics calculations to determine energy levels under different operating assumptions. We are also developing a computer code to link gas-phase temperatures and concentrations to surface temperatures and availability of adsorption sites.

Mission Relevance

This project supports the DOE energy-security mission by reducing dependence on foreign oil, and also supports the DOE's environmental mission. In addition, surface-chemistry analysis capabilities developed here may contribute to myriad future applications, including missile reentry, fuel cells, and sensors for detecting chemical-warfare agents and explosives.

FY03 Results

In FY03, we developed a simplified surface chemical kinetic mechanism for NOx traps. We used molecular dynamics modeling codes to study the microscopic mechanisms for trapping and release of NOx on the surface of a BaO catalyst—a computationally challenging task because many layers of the BaO surface must be included to achieve an accurate result. Initial results of our calculations of bulk BaO surface at finite temperature show that as the temperature increases from 0 to 300 K, charge transfer goes from paired to neutral NO_2 moieties. These results indicate that the kinetics of trapping at 300 to 500 K are very different than at 0 K. We revised algorithms, reaction models, kinetic parameters, and code interfaces to improve the simulation capability. The result was a detailed computer model that considers the effects of surface chemistry, temperature, and concentration in the gaseous phase, along with temperature distribution in the solid catalyst surfaces.

Environmental Fate of Organophosphorus Compounds Related to Chemical Weapons

M. Lee Davisson

03-ERD-022

Abstract

The environmental fate of the chemical weapon agent VX is an important component in threat assessment, civilian response, and counterterrorism. However, experimental data characterizing the environmental fate of VX are sparse, and the chemical's persistence and degradation pathways have not been accurately predicted, particularly at low concentrations. This work will determine plausible mechanisms of chemical interaction when VX and its associated byproducts are introduced into various aquatic matrices—dissolved ions, dissolved organic matter, and suspended sediments—and onto different mineral surfaces. Using the actual agent, we will conduct controlled experiments in a dilute-solutions laboratory and perform analysis with liquid chromatography mass spectrometry.

We anticipate gaining basic data that characterize the environmental behavior of VX and its precursors and byproducts. These data will provide a quantitative basis for predicting VX behavior under variable environmental conditions. Such prediction may reveal (1) preferential sites of sorption and signature persistence, (2) environmental conditions that promote VX persistence at toxic levels, or (3) conditions that form persistent toxic VX byproducts. We anticipate establishing a unique capability for research on dilute, live chemical-weapons agents.

Mission Relevance

This work addresses a knowledge gap in the LLNL mission areas of counterterrorism and homeland security: addressing the increasingly serious problem of the proliferation of weapons of mass destruction. This work will advance our understanding of the signatures of chemical weapons manufacturing and provide basic understanding of how nerve agents persist in the environment after release.

FY03 Results

We determined VX synthesis pathways; designed, developed, and tested an experimental apparatus for conducting studies of VX fate under closed conditions; and developed quantitative analytical protocols. Aqueous experiments buffered at pH 7 using 3-(N-morpholino)propanes ulfonic acid show that compared to distilled water, VX persisted approximately twice as long at pH 7 when dissolved organic matter concentration was 100 mg/L. We also found that (1) VX degradation rates vary only moderately over a wide pH range in solutions buffered with carbonate, compared to sodium hydroxide titrations over the same pH range; (2) VX readily adsorbs to clay minerals in distilled water, but not iron oxyhydroxides; and (3) VX readily degrades when dried down onto iron oxyhydroxides. These various experimental conditions were tailored to mimic key characteristics common in aquatic and soil environments that have been known to control other contaminant fate.

Proposed Work for FY04

We will derive the aqueous-sorption coefficients, degradation pathways, and half-lives of VX and its associated compounds under variable pH, ionic strength, initial spike concentration, and mineral–organic matter ratios; test VX persistence in water dosed with chlorine at typical drinking water treatment levels; conduct non-aqueous fate studies of VX on mixed goethite–organic matter substrates (i.e., simulated soil); and perform extractions and mass spectrometry measurements. Fourier-transform infrared spectroscopy will be used to measure VX–functional group interaction on solids.

Publications

Love, A. H. et al. (2003). *Determining environmental parameters of VX and related compounds for secondary exposure evaluation and signature detection.* 226th Am. Chem. Soc. Natl. Meet., Sept. 7–11, 2003, New York, NY. UCRL-JC-153600.

Development of a Virtual Crystallizer

Teresa A. Land 03-ERD-051

Abstract

The capability to predict how crystals grow is a valuable tool for any field requiring crystal production. We are developing a computer model capable of predicting growth from a 1-cm-size seed to a large, 60-cm crystal according to actual crystallizer system conditions. The model will use equations governing the fundamental physics and chemistry of crystal growth, along with the mass transfer to crystal faces determined using a computational fluid dynamics (CFD) model and a large-scale crystal growth model that calculates growth rates for the individual faces and allows them to evolve in time to form a three-dimensional (3-D) crystal. We will validate the model with actual potassium dihydrogen phosphate (KPD) crystals grown from 1 to 60 cm.

Expected results include (1) an understanding of the sensitivity of growth to various parameters; (2) a method for optimizing growth conditions; and (3) the ability to grow reproducibly a crystal with desired dimensions and properties. The immediate impact of our investigation will be to increase the quality and yield of rapid-growth KDP and deuterated KDP crystals used for frequency conversion and large-aperture, fast optical switches in large inertial-confinement-fusion (ICF) laser systems.

Mission Relevance

Developing the scientific foundation for growing crystals of the desired size and quality will benefit the broad array of efforts to develop crystalline materials for laser systems, optical components, and photonics applications. This technology will enable advances in high-energy-density physics that will benefit LLNL missions such as stockpile stewardship and ICF.

FY03 Results

We developed a computer crystal-growth model based on our current understanding of the rate-controlling processes. The model includes the growth physics, mass transfer, impurity effects, maintains material balances, and crystal phases. Agreement was good between preliminary projections developed using the model with actual large-crystal run data. We developed a preliminary CFD model that includes physically accurate crystal, platform and tank geometries, fluid dynamics, transport parameters, system temperatures, and growth parameters. We thoroughly characterized thermal profile of the crystallizer and developed several rotation schedules to minimize solution turbulence and secondary nucleation formation.

Proposed Work for FY04

The CFD modeling will focus on four cases (30-cm and 60-cm sizes, each at two rotations) to explore extremes and to develop confidence in the results of the simulations. Half-scale experimental replications of these four cases will be performed to obtain a set of data, including instantaneous and time-averaged mass transport at the crystal surface using a heat-transfer analogy. The data will be used to verify the validity of the CFD model and will provide feedback for modifications to the model to produce results consistent with reality. The virtual crystallizer model will be updated with current kinetic and mass-transfer data and expanded to include 3-D growth projection capability.

Isotopic Tracing of Fuel Components in Particulate and Gaseous Emissions from Diesel Engines using Accelerator Mass Spectrometry

Bruce A. Buchholz

01-ERI-007

Abstract

Measurement of particulate matter (PM), nitrogen oxides, hydrocarbons, carbon monoxide, and sulfur dioxide emissions are routine, but determining which specific fuel components make these products is not. Subattomole sensitivity of accelerator mass spectrometry for carbon-14 (^{14}C AMS) allows tracing of specific fuel components in soot or gaseous emissions without the use of radioactive materials. The specificity and sensitivity of ^{14}C AMS make the technique a powerful tool in acquiring data for validating combustion models and investigating mechanisms for combustion regimes. In this project, we exploit the difference in isotope content to trace biofuel components (e.g., ethanol and biodiesel) and resulting combustion products in soot and gaseous emissions against a ^{14}C-free petroleum background. The successful completion of this work will result in the demonstration of a unique capability for assessing the fate of fuel components in emission products and will provide data for the validation of chemical kinetic combustion models.

Mission Relevance

This work contributes to the Laboratory's mission in energy security. Alternative renewable fuels expand fuel diversity and reduce the impact of CO_2 emissions on the climate. Renewable fuels are indirectly produced from CO_2 emissions and consequently promote the turnover of carbon in annual carbon reservoirs. Research into new combustion regimes such as homogeneous charge compression ignition (HCCI) expands the energy supply by enabling combustion of a broader quality of traditional fuels. This work has demonstrated ^{14}C AMS to the combustion community and laid the foundation for collaborative research.

FY03 Results

Gas and filter samples were collected at the University of California, Berkeley (UCB) combustion laboratory and transported to LLNL for AMS sample preparation and analysis. Simple experiments traced the contribution of lubrication oil to PM generated by diesel engines. After several months of unsuccessful attempts to get our catalyst-brick nitrogen oxide trap to function as designed, we abandoned catalyst kinetics and concentrated on tracing fuel components in the emission gases from an HCCI engine, which was modified to accept liquid fuel blends of ethanol and ether. Ignition timing, heat release, and combustion efficiency were manipulated and emissions traced by AMS.

Publications

Buchholz, B. A. et al. (2003). "Quantifying the contribution of lubrication oil carbon to particulate emissions from a diesel engine." *Soc. of Automotive Engineers Tech. Paper* 2003-01-1987. UCRL-JC-151294 Rev.

Buchholz, B. A. et al. (in press). "Tracing fuel component carbon in the emissions from diesel engines." *Nuclear Instruments and Methods B*. UCRL-JC-149036.

Buchholz, B. A. et al. (2003). *Measuring the effect of fuel chemical structure on particulate and gaseous emissions using isotope tracing*. American Flame Res. Committee Intl. Symp., Oct. 15–17, 2003, Livermore, CA. UCRL-JC-155257.

Cheng, A. S., B. A. Buchholz, and R. W. Dibble. (2003). I*sotopic tracing of fuel carbon in the emissions of a compression-ignition engine fueled with biodiesel blends*. Soc. of Automotive Engineers Tech. Paper 2003-01-2282. UCRL-JC-152168.

Cheng, A. S. et al. (2003). *Quantifying the contribution of lubrication oil carbon to particulate emissions from a diesel engine*. Third Joint Meet. of the U.S. Sections of the Combustion Inst., Mar. 16–19, 2003, Chigago, IL.
UCRL-JC-151294.

Laser-Initiated Nanoscale Molecularly Imprinted Polymers

Bradley R. Hart
03-LW-047

Abstract

This project is synthesizing nanoscale molecularly imprinted polymer (MIP) arrays that have complex patterns and structures and optimized selectivity for applications such as chemical and biological sensing and microscale separations and catalysis. Arrays of MIPs will be fabricated by using a focused laser source to pattern polymer features on a surface covered with a solution containing polymerizable monomers, a photoinitiator, and template molecules.

The work will establish an entirely new and versatile route to practical MIP systems and also overcome the limitations associated with traditional MIP systems. Advances in these areas would represent significant, publishable results in the area of molecular imprinting.

Mission Relevance

This work supports LLNL's national security mission in developing remote-sensing, monitoring, and assessment technologies for the detection of signatures of chemical, biological, and nuclear activities, as well as the detection of chemical and biological weapons themselves.

FY03 Results

In FY03, we successfully synthesized nanoscale polymers using common molecular imprinting formulations and demonstrated an understanding of the conditions required to prepare these materials in a controlled manner. We are now able to prepare large arrays of polymer features, as well as complex patterns and structures. Using a flow cell developed for this project, we also demonstrated that polymers with varying compositions can be prepared side by side on the same substrate. This will allow the preparation of MIP arrays made up of polymers that are selective for a variety of molecular species.

Proposed Work for FY04

Experiments aimed at optimizing and calibrating variables such as laser power, wavelength, and illumination time will continue in FY04. In addition, we will prepare MIPs using the fluorescent template molecules identified in FY03. Systems to evaluate binding by fluorescence will be developed and used to measure and optimize the selectivity of our MIPs. Alternative detection schemes such as Raman spectroscopy will also be examined. In addition, we will perform experiments aimed at forming multielement arrays of polymer features prepared using different template molecules.

Developing Multilayer Mirror Technology near 45 nm using Scandium–Silicon Interfaces

Joseph Nilsen

03-FS-003

Abstract

We plan to develop the process to stabilize the interfaces of nanolaminate structures using materials such as scandium (Sc) and silicon (Si). These materials will enable us to develop new multilayer mirror technology that can be used in the ultraviolet range, near 45 nm. To obtain this objective, the interfacial structure and reaction kinetics must first be well understood and then controlled for design applications. Hot-stage diffraction will be used to measure changes in the short-range order, thereby providing the bulk diffusivity and hence the activation energy of the Sc–Si system. Cross-section, high-resolution electron microscopy will reveal layer and interface microstructure. Dynamic behavior under extreme conditions will be assessed through tabletop x-ray laser (XRL) illumination at the COMET facility.

If multilayer mirrors can be fabricated at this wavelength, various imaging diagnostics can be developed to use with the COMET laser system. Our research will explore a regime of wavelength and target design that represent a new challenge to our understanding of interface stabilization.

Mission Relevance

Furthering our basic understanding in evaluating the interfacial structure of intrinsically metastable structures will broaden and deepen our understanding for applications that support Laboratory missions in stockpile stewardship and inertial-confinement fusion for energy security.

FY03 Results

In FY03, using the artificial concentration wave method, our analytic model was improved to assess the metastability of the multilayer by computing the interdiffusion rate for multilayers composed of dissimilar crystalline structures. Si-based nanolaminates were synthesized using a computer-controlled, planar-magnetron sputter-deposition system. Their reflectivity was measured at grazing incidence using 8-keV radiation, and at 45° using 1-keV radiation, and the structure of one nanolaminate sample was imaged in cross-section using atomic force microscopy. A set of prototype multilayer optics was prepared for a collaborative evaluation of normal-incidence reflectivity at 46.9 nm.

Publications

Jankowski, A. and C. Saw. (2003). *Interdiffusion in Ni/CrMo composition modulated films.* Presented at the 132nd Ann. Mtg. Metallurg. Soc., San Diego, CA, March 2–6, 2003. UCRL-JC-151896.

Earth and Space Sciences

INCCA: Integrated Climate and Carbon

Starley L. Thompson

01-SI-008

Abstract

The goals of the Integrated Climate and Carbon Initiative are to (1) develop an integrated climate and carbon-cycle model to predict the fate and climatic effects of fossil-fuel-derived carbon dioxide (CO_2) and (2) evaluate the climatic impact of proposed fossil-fuel burning policies. In current climate models, atmospheric CO_2 concentrations are prescribed, not predicted. In contrast, our completed model uses specified rates of fossil-fuel burning to predict future CO_2 concentrations and climates. This information can be used to address scientific and policy-related questions involving the climatic effects of burning fossil fuels. Our system—the most comprehenisve, and first American, fully-coupled climate-carbon simulation system—gives us the ability to model combined CO_2 concentration and climate scenarios from the 19th through the 21st century and beyond. This capability is being used to determine the strength of internal carbon cycle feedbacks on future climate change, including the effects of model science and socioeconomic uncertainties.

Mission Relevance

This research contributes to establishing the scientific basis needed to formulate national energy policies. This project builds on LLNL's core competency in atmospheric science and takes advantage of computing resources enabled by DOE's Advanced Simulation and Computing Program to extend LLNL's capabilities in climate simulations in support of the energy-security and environmental-management missions. In particular, the coupled climate and carbon-cycle modeling developed in this research initiative is needed to understand the fate and environmental impact of fossil-fuel burning as part of DOE's carbon management efforts.

FY03 Results

Milestones for FY03 included (1) completing the climate-carbon model; (2) performing century-scale climate production runs with prescribed CO_2 emissions scenarios; and (3) modifying our terrestrial biosphere model to interface to the new community climate system model (CCSM2). We also performed several 230-yr simulations with the full system model. For the first time we have started to explore parameter space with a comprehensive climate-carbon model. Initial results show that the strength of the positive feedback of carbon cycle change on climate is strongly affected by global vegetation response to CO_2 fertilization and the magntitude of global warming.

Publications

Delire, C., J. A. Foley, and S. L. Thompson. (2003). "Evaluating the carbon cycle of a coupled atmosphere–biosphere model." *Global Biogeochemical Cycles* **17**(1). UCRL-JC-149715.

Govindasamy, B. et al. (2003). "Impact of geoengineering schemes on the terrestrial biosphere." *Geophys. Res. Lett.* **29**(22). UCRL-JC-149732.

Thompson, S. L. (2003). *Climate-carbon feedbacks from a gcm-based model: Transient simulations to year 2100 using prescribed carbon emissions.* Intl. Conf. on Earth System Modeling MPI Meteorology, Sep. 15–19, 2003, Hamburg, Germany. UCRL-JC-152479 Abs.

Thompson, S. L. et al. (2002). *A comprehensive GCM-based climate and carbon cycle model: transient simulations to year 2100 using prescribed greenhouse gas emissions.* Am. Geophys. U. Fall Meet. 2002, Dec. 6–10, 2002, San Francisco, CA. UCRL-JC-149761 Abs.

Thompson, S. L. et al. (2003). *A coupled carbon cycle in PCM2: Transient simulations for 1870–2100.* UCRL-PRES-155265.

Adaptive Optics Imaging and Spectroscopy of the Solar System

Seran Gibbard 01-ERD-013

Abstract

This project uses the adaptive optics (AO) system and an infrared spectrograph at the Keck Observatory to obtain spectra of planets and satellites in our Solar System, including Uranus, Neptune, and Saturn's moon, Titan. Our project will investigate various aspects of their atmospheric features: altitude, composition, and connection to events occurring deeper in the planets' tropospheres.

Results from this project will include new insights on the structure of planetary atmospheres and planetary ring systems. Our observations of the atmospheres of Neptune and Uranus will enable us to determine the three-dimensional structure of their atmospheres by tracking cloud features in space and time. Using the AO system, we will be able to resolve the individual rings of Neptune and Uranus, which has never before been achieved from an Earth-based observatory. We will also provide new insights into the atmospheric and surface structure of the only satellite in the solar system with a substantial atmosphere, Titan. Titan has a solid surface located under its atmosphere, but the composition of this surface is still unknown. Our research will determine the surface reflectivity of Titan at near-infrared wavelengths. We will also study the vertical structure of Titan's atmosphere and will search its atmosphere for predicted methane clouds.

Mission Relevance

This project will result in the continued development of AO technology (e.g., new tools for high-precision optical characterization), which can be applied to other optical systems at LLNL, including AO systems and sensors for high-power lasers and all-optical, free-space communication networks and long-range surveillance capability in support of nonproliferation activities. In addition, success in this work will position LLNL as a world leader in observing and modeling giant planets and will support DOE's science mission by expanding basic scientific knowledge.

FY03 Results

Work accomplished in FY03 includes (1) analyzing the Neptune spectra and determining the altitudes of 17 infrared-bright features, (2) studying the dynamics of Neptune's clouds, (3) imaging the Uranian atmosphere and ring system with the highest resolution ever achieved from the Earth, (4) observing Neptune's very faint ring system, (5) obtaining high-resolution, spatially-resolved spectra of Titan, and (6) modifying the ARDRA code to use a point source irradiance source (to simulate incoming solar radiation) and to simulate a layered Titan atmosphere with optical depth varying with depth in the atmosphere. A NASA project to continue and expand on this work was pending at the end of FY03.

Publications

Gibbard, S. G. et al. (2003). "Determination of Neptune cloud heights from high-spatial-resolution near-infrared spectra." *Icarus* **166**, 359–374. UCRL-JC-155154.

Macintosh, B. et al. (2003). "Speckle imaging of volcanic hotspots on Io with the Keck telescope." *Icarus* **165**, 137. UCRL-JC-137182-REV-1.

Max, C. E. et al. (2003). "Cloud structures on Neptune obseved with Keck Telescope adaptive optics." *Astronomical Journal* **125**, 364. UCRL-JC-148432.

Constraining Nucleosynthesis Models: Mapping Titanium-44 in Cassiopeia A

William W. Craig 01-ERD-020

Abstract

Our project will obtain the first map of the titanium-44 distribution from a supernova remnant—the Cassiopeia A remnant—by imaging the emitted gamma rays to validate supernova nucleosynthesis and mixing models. A gamma-ray map of the titanium-44 distribution of Cassiopeia A will provide a direct constraint on supernova models and holds the promise of improving the codes used to model these events. Obtaining this first-ever map of nuclear line emission from a supernova remnant required an upgrade to the pointing system of the balloon-borne High-Energy Focusing Telescope (HEFT). Our work has included applying LLNL capabilities in precision engineering and metrology to reduce the pointing error of HEFT to 6 arcsec and extending the payload's operation to daytime target tracking by implementing a daytime star tracker. These improvements are complete, and a spring 2004 flight by NASA is planned for the HEFT payload. At a higher level, the technological demonstration of the ability to image objects at these energies by the HEFT program has already led to significant advances in target characterization and medical imaging.

Mission Relevance

The technology developed in our project has directly benefited LLNL's national-security mission by developing detector systems now being used for homeland security programs. In addition, the high-profile basic science will benefit LLNL's ability to recruit a high-quality scientific workforce.

FY03 Results

The payload was completed and was used to demonstrate the ability to track 40% more precisely than the original program goal. The daytime star trackers were completed and installed, and all support equipment was completed.

At NASA expense, the payload will be shipped to Ft. Sumner, New Mexico, in March 2004 for launching in April. After flight, the payload will be returned to LLNL and refurbished at NASA expense. The flight data will be analyzed by members of the HEFT collaboration, and the science results will be published. The HEFT concept was proposed to NASA as a Small Explorer satellite mission in June 2003, and was selected, in November 2003, for a NASA-funded implementation study that is now underway.

Novel Approaches for Monitoring Intrinsic Bioremediation

Harry R. Beller

01-ERD-063

Abstract

Intrinsic bioremediation (IB) is increasingly viewed as one of the most cost-effective options for restoring contaminated groundwater. A primary impediment to the acceptance of IB is the difficulty of demonstrating that decreases in the concentrations of contaminants in groundwater truly represent biodegradation rather than nondestructive processes, such as dilution. Using detailed laboratory microbial studies and focused mass spectrometric analyses of contaminated groundwater, we are identifying and testing the usefulness of unique metabolites that can be used in monitoring programs as signature indicators of the IB of specific contaminants. The contaminants, which are of concern at DOE and DoD facilities, include high explosives (HE), such as RDX (hexahydro-1,3,5-trinitro-1,3,5-triazine) and TNT (trinitrotoluene), as well as certain fuel hydrocarbons.

The results we anticipate include (1) field evidence addressing the potential for IB of HE in contaminated aquifers, (2) the bacterial physiology of RDX metabolism and the implications for IB, and (3) the usefulness of benzylsuccinates as specific indicators of the in situ anaerobic degradation of the hydrocarbons benzene, toluene, ethylbenzene, and xylene (BTEX) in fuel-contaminated aquifers. The research would contribute significantly to our understanding of the biological fate of HE compounds in subsurface environments. The research could also dramatically enhance the effectiveness of groundwater monitoring for in situ BTEX degradation by taking advantage of recent groundbreaking findings in anaerobic hydrocarbon biochemistry.

Mission Relevance

This project directly supports LLNL's environmental mission by contributing to HE-contaminated groundwater remediation at LLNL's Site 300, and is also applicable elsewhere in the DOE complex. The project also enhances LLNL's competency in the emerging field of IB and may be used in developing scientific knowledge that supports environmental remediation and regulation nationwide.

FY03 Results

In FY03, we demonstrated that bacteria enriched from Louisiana Army Ammunition Plant (AAP) soils were capable of metabolizing TNT and dinitrotoluene (DNT) under fermentative or methanogenic conditions in a simple growth medium with no added H_2 and no other organic compounds. The metabolites detected in these cultures reflected reduction of nitro groups to amino groups. These studies differed from typical TNT biodegradation studies that employ a complex medium and do not quantify the various electron donors and acceptors. We also demonstrated that in situ, anaerobic RDX metabolism in groundwater at the Iowa AAP is an ongoing, sustained process by detecting consistent concentrations of three nitroso-substituted metabolites over a period of more than 1 yr. The project resulted in four peer-reviewed articles.

Publications

Beller, H. R. (2002). *Sensitive and reliable monitoring of in situ RDX transformation.* Presented at DOE Technical Information Exchange Conference, Nov. 12–14, 2002, Oakland, CA. UCRL-JC-149821 Abs.

Beller, H. R. and S. R. Kane. (2003). *Advances in monitoring in situ BTEX biodegradation: Use of state-of-the-art mass spectrometric and molecular techniques.* Presented at Assoc. for Env. Health and Sciences, Mar. 2003, San Diego, CA. UCRL-JC-149343 Abs.

Beller, H. R. and S. R. Kane. (2002). "Monitoring in situ anaerobic alkylbenzene biodegradation based on mass spectrometric detection of unique metabolites or real-time PCR detection of a catabolic gene." *Proc. Am. Geophys.* U. Fall Meet., Dec. 6–10, 2002, San Francisco, CA. UCRL-JC-150696 Abs.

Reusser, D. E. et al. (2002). "In situ transformation of deuterated toluene and xylene to benzylsuccinic acid analogs in BTEX-contaminated aquifers." *Environmental Science and Technology* **36**, 4127. UCRL-JC-154324.

Evaluation and Optimization of Methyl-*Tert* Butyl Ether Biodegradation in Aquifers

Staci R. Kane

01-ERD-065

Abstract

Methyl-*tert* butyl ether (MTBE) is a widely used gasoline oxygenate that is present at over 250,000 leaking underground fuel tank (LUFT) sites nationwide. This project's focus is to evaluate and optimize intrinsic aerobic biodegradation of MTBE in aquifer sediments. The project objectives are to (1) determine how widespread aerobic MTBE degradation is at LUFT sites; (2) investigate microbial and chemical factors effecting MTBE degradation; (3) evaluate the effect of dissolved gasoline components on MTBE biodegradation; (4) isolate and characterize MTBE-degrading microorganisms and their genetic components coding for MTBE degradation activity; and (5) determine the environmental factors that favor increased aerobic MTBE degradation.

Results from this work will increase our knowledge of biological and chemical factors that may be controlling MTBE degradation and provide a foundation for DNA-based diagnostic methods to quantify MTBE-degrading microorganisms at LUFT sites. Results will also show whether other dissolved gasoline components, in particular benzene, toluene, ethylbenzene and xylenes (BTEX), which are often co-contaminants with MTBE, influence degradation of MTBE and its toxic metabolite, tert-butyl alcohol (TBA). MTBE-degrading isolates will be used to determine how MTBE degradation rates can be enhanced and thus provide insight into what conditions may favor optimal degradation rates in the field. Findings from this study will benefit the regulatory community, the academic community, and the petroleum industry.

Mission Relevance

This project supports the Laboratory's environmental-management mission by providing knowledge that could be used to develop novel, cost-effective technologies for bioremediation of MTBE-contaminated sites and to predict where engineered or intrinsic bioremediation may be successful. Furthermore, we will contribute information needed by state regulators and others in the public sector. Tools and methods developed for evaluating biodegradation of MTBE can be expanded to other drinking-water contaminants.

FY03 Results

In FY03, our MTBE degradation studies showed that under aerobic conditions, three of the eight sites tested had activity, and degradation was limited by microbial factors. From sediments with activity, we isolated MTBE-degrading pure cultures that were similar to a known MTBE degrading strain, PM1. These organisms were absent in sites without activity. MTBE degradation was a stable trait of the cultures even without consistent MTBE exposure. We determined MTBE degradation rates under different growth conditions and conducted transposition mutagenesis experiments targeting genes involved in MTBE degradation with the goal of designing specific gene targets for real-time polymerase chain reaction. Research findings were presented at national meetings and published in symposium proceedings.

Electromagnetic Imaging of Carbon Dioxide Sequestration at an Enhanced Oil Recovery Site

Barry A. Kirkendall

01-ERD-089

Abstract

This project will combine laboratory and field data to develop quantitative image interpretation techniques from a pilot carbon dioxide (CO_2) sequestration and enhanced oil recovery (EOR) injection site using borehole electromagnetic (EM) techniques. We will develop the inversion process in CO_2 studies by coupling results with petrophysical laboratory measurements and focus on new interpretation techniques for imaging sequestered CO_2. Our goal is to develop the ability to image injected CO_2 within the subsurface during EOR processes while simultaneously discriminating between pre-existing petroleum and water deposits. We characterize an EOR water flood through subsurface images of the CO_2 sequestration site at discrete intervals.

We are developing a field system and associated software to acquire in-field subsurface images in between two observation wells. These images show the EM signatures of CO_2, brine, and petroleum. We are developing a method to separate these individual components by incorporating laboratory measurements and numerical processing techniques that combine multiple-frequency images resulting in decreased error and higher resolution. This project is creating a unique capability that can be easily applied and adapted in future CO_2 sequestration and EOR studies.

Mission Relevance

This project directly addresses issues in the areas of carbon sequestration and oil and gas recovery enhancement in support of LLNL's energy-security and environmental-management missions. The results of this project can be applied to upcoming CO_2 EOR and sequestration studies.

FY03 Results

In FY03, we developed a time-lapse method to separate the CO_2, brine, and oil components in images; used multiple-frequency images to increase image resolution and decrease root mean square error in the inversion process; refurbished and modified the heated pressure vessel and flow-through system, including software, for conductivity measurements on clay-bearing rocks, including the Etchegoin Formation at the Lost Hills CO_2 site in California; and developed the ability to conduct permeability measurements in the laboratory. This laboratory capability was utilized by a visiting professor and resulted in a publication and an effort to examine the effects of finite-sized electrodes in the laboratory. These studies are expected to provide information on the applicability of comparing laboratory and field results.

Publications

Park, S. K. and J. J. Roberts. (2003). "Conductivity structure of the San Andreas fault, Parkfield, revisited." *Geophys. Res. Lett.* **30**, 1842. UCRL-JC-152099.

Accelerated Carbonate Dissolution as a Carbon Dioxide Separation and Sequestration Strategy: Continued Experimentation and Simulation

Kenneth G. Caldeira 01-ERD-091

Abstract

By conducting laboratory experiments and computer modeling, our project seeks to provide the science base required to evaluate the feasibility of the carbonate-dissolution method for sequestering carbon in the ocean. Theoretical studies suggested that this method of ocean carbon dioxide (CO_2) disposal is technically, geochemically, and environmentally advantageous for coastal power plants near limestone. This approach uses existing technologies, abundant carbonate minerals, and seawater to neutralize CO_2 acidity and largely converts fossil fuel CO_2 into a form not readily exchanged with the atmosphere. Laboratory experiments and three-dimensional (3-D) simulations will confirm our theories.

Mission Relevance

The success of our project will advance a novel concept for CO_2 separation and sequestration and provide additional knowledge in carbon-cycle modeling and geochemistry that could be expanded to prototype and larger-scale research in partnership with DOE and industry. By investigating the feasibility of the carbonate-dissolution technique for sequestering fossil-fuel carbon in the ocean, our project directly supports LLNL's energy and environmental-management missions.

FY03 Results

In FY03 we (1) demonstrated the carbonate-dissolution technique for sequestering CO_2 in the ocean at the bench-top scale using seawater, (2) validated and refined existing computer models, (3) equilibrated seawater with relatively concentrated CO_2 gas and used the resulting solution to dissolve crushed carbonate in a reactor, (4) monitored gas and liquid effluent chemistry (e.g., CO_2 partial pressure, pH, conductivity, alkalinity, and metal-ion concentration) to determine dissolution rates and reactor performance under a range of conditions, and (5) jointly developed a Lagrangian plume modeling code with the Massachusetts Institute of Technology.

Our results indicate that this method of ocean CO_2 disposal is technically, geochemically, and environmentally advantageous for coastal power plants near limestone. Our experimental work with both freshwater and seawater, along with computer modeling of both the fate of sequestered carbon on a global scale, helped to show that the carbonate-dissolution method could potentially be a major contributor to global carbon management and be applied economically on a large scale.

Publications

Caldeira, K., A. K. Jain, and M. I. Hoffert. (2003). "Climate sensitivity uncertainty and the need for non-CO_2 emitting energy sources." *Science* **299**, 2052. UCRL-JC-153428.

Caldeira, K. and M. I. Wickett. (2003). "Anthropogenic carbon and ocean pH." *Nature* **425**, 365. UCRL-JC-143232 Rev. 1.

Hoffert, M. I. and K. Caldeira. (in press). "Advanced technology paths to global climate stability: Energy for a greenhouse planet." *Encyclopedia of Energy*. UCRL-JC-144122 Rev. 1.

Ridgwell, A. J., M. Kennedy, and K. Caldeira. (2003). "Carbonate deposition, climate stability, and neoproterozoic ice ages." *Science* **302**, 861. UCRL-JC-153633.

Developing Smart Seismic Arrays: A Simulation Environment, Observational Database, and Advanced Signal Processing

Philip E. Harben 01-ERD-096

Abstract

We propose to develop a framework for solving near-real-time seismic imaging and tracking problems in heterogeneous, uncertain geologic media focusing on two applications: detecting and characterizing buried structures, and tracking vehicles in battlefield, border, and facility-monitoring environments. Our approach is to use the LLNL E3D elastic-propagation code to build a modeling capability for assessing the performance of imaging and tracking approaches. The algorithms extend existing matched-field processing approaches to handle medium uncertainty by matching data against a computed subspace of target fields. Field deployments validate this processing approach and demonstrate an integrated communication and computational infrastructure needed to support practical applications.

The goal of this project is to build capabilities in designing and using acquisition systems and in full three-dimensional, finite-difference modeling, as well as statistical characterization of geological heterogeneity. Such capabilities, coupled with a rapid field analysis methodology based on matched field processing, are applied to problems associated with surveillance, battlefield management, and deeply buried targets. Success will lead not only to direct applications for the above problems; it will lay the groundwork for how to deal with geological heterogeneity in forward modeling, a critical problem in other studies such as shock physics modeling for hard target defeat.

Mission Relevance

Developing seismic imaging and tracking capabilities will further the Laboratory's national-security mission area of supporting military and intelligence agencies (Defense Advanced Research Projects Agency, the Defense Threat Reduction Agency, and the U.S. Army). Such capabilities will have direct impact on emerging threats, vehicle-tracking applications, battlefield management, detection of hard and deeply buried targets, and portal monitoring.

FY03 Results

A simulation experiment was conducted to test the modeling-based approach to geological characterization using E2D, the K-L statistical methodology, and matched field processing applied to tunnel detection. The simulation validated the methodology. A mine-safety site in Lake Lynn, Pennsylvania, was modeled to produce a 32-million-node 3-D model grid for E3D. E3D-generated wavefield movies predicted reflection signal levels. Ability to model a 3-D geological structure and calculate synthetic seismograms that are in good agreement with actual data was demonstrated. We conclude that E3D is a powerful tool for assessing the conditions under which a tunnel could be detected. Lake Lynn in-tunnel explosion data showed that single detonations could be located, although simultaneous detonations would require a strategic placement of arrays.

Near-Real-Time Assessment of Health Risks from Simulated Containinant Wet Deposition using Real-Time Rainfall and Geographic Information System Databases

Richard Cederwall

02-ERD-036

Project Description

This project developed a prototype system for assessing potential health impacts of contaminants deposited on the ground by rain in near-real time, thus reducing a major source of uncertainty in assessing health risks from the contaminants. The project linked available real-time rainfall data with wet-deposition algorithms, performed case studies to validate the wet-deposition algorithms and determine levels of uncertainty, developed a health-risk assessment system based on proven methods and determined levels of uncertainty, and exercised the prototype system for case studies. A wet-deposition algorithm that makes use of model variables in the LLNL's National Atmospheric Release Advisory Center (NARAC) system was developed and tested.

Mission Relevance

The capabilities developed in this project will be used to support LLNL's national security and homeland security missions by providing predictions of the fate of airborne radiological or toxic material.

FY03 Results

In FY03, we incorporated improved treatment of in-cloud processes during convective storms in the rainout algorithm; quantified uncertainty estimates in the wet-deposition values from sensitivity studies and compared with data; added capabilities for assessing dose from short-lived radionuclides; extended the dose calculations to human health risk estimates; and completed development and testing of wet deposition and risk-assessment components of the prototype system, which has been made available to NARAC for implementation.

Publications

Arno, M. G. et al. (in press). "Extension of NCRP 129 to short-lived nuclides." *Health Physics*. UCRL-JC-152475.

Loosmore, G. A. and R. T. Cederwall. (2002). *In-cloud and below-cloud scavenging of aerosol particles in local to regional scale models used for emergency response*. Presented at the AGU Meeting, San Francisco, CA, Dec. 2002. UCRL-MI-149717.

Loosmore, G. A. and R. T. Cederwall. (in press). "Precipitation scavenging of atmospheric aerosols for emergency response applications: Testing an updated model with near real time data." *Atmosph. Environ*. UCRL-JC-155010.

Infrastructure Response to Natural and Man-Made Ground Motions: High Performance Computing and Distributed Sensing for Regional Scale Prediction and Response

David B. McCallen

02-ERD-044

Abstract

This project developed a rigorous, three-dimensional (3-D), computationally based framework for simulating regional seismic wave propagation and structural response. Traditional approaches to predicting seismic wave propagation and structural response subdivide and simplify the problem to make it tractable for limited computing engines. Such idealizations (e.g. assuming 1-D vertically propagating waves in site-response computations) do not permit accurate representation of the actual physics processes at work. This research developed a rigorous physics-based tool for addressing complex, 3-D wave propagation through the earth, and the coupled soil–structure interaction between a large structure and the ground. This work provides new understanding of the seismic hazard to structures located in sedimentary basins and provides an entirely new computationally based tool for simulating ground motion and infrastructure response.

Mission Relevance

The focus on Southern Nevada in this developmental effort will provide the knowledge base and tools with direct application to achieving NNSA mission objectives in enhanced nuclear test readiness at the Nevada Test Site and southern Nevada. The new simulation tools also have application to natural earthquake hazard assessment at any NNSA facility site and at sites and facilities of governmental agencies that support LLNL in Work-for-Others programs.

FY03 Results

Work completed in FY03 included: (1) implementation of a geologic velocity soil model for southern Nevada in our regional model; (2) computation of a suite of simulations to investigate the parametric effects of basin topography, basin velocity and shallow-soil site response on the severity of seismic ground motions in the basin; and (3) compared simulation results with observational data of basin response obtained from underground nuclear test and natural earthquake measurements. The model simulations provided insight into basin response and particularly to the distribution of surface motions expected in the Las Vegas basin. The simulations also indicated that inclusion of the shallow soil velocity structure (top 100–200 m) significantly increases the observed surface motions.

Publications

Rodgers, A. et al. (2003). *Las Vegas seismic response project.* Presented at the 2003 IRIS/UNAVCO Workshop. UCRL-JC-153010-ABS.

Rodgers, A. J. and D. McCallen. (2003). *Las Vegas Basin Seismic Response Project: Overview and site response.* Presented at the American Geophysical Union Fall Meeting 2003. UCRL-JC-149853-ABS.

Three-Dimensional Astronomy: Scientific Observations with the Livermore Imaging Fourier Transform Spectrograph

Ronald E. Wurtz 02-ERD-052

Abstract

This project plans to make a direct measurement of the mass density of the universe, one of the fundamental parameters of cosmology. The project will weigh the universe using a unique instrument, the Livermore Imaging Fourier Transform Spectrometer (IFTS), to determine the total mass and dark-matter fraction in clusters of galaxies. By conducting a census of galaxy dynamics, the project will resolve questions concerning the distribution of dark matter.

Conventional instrumentation imposes a limit on the faintest objects observed. The mass density obtained by conventional methods is skewed by the kinematics of the brightest objects in a cluster, which tend to move slower. Traditionally, attempts have been made to extrapolate the behavior of the faintest objects. In contrast to the conventional approach, the IFTS observes all objects in a field and tailors spectral resolution to the faintest objects. By measuring information about the faint objects directly, not extrapolated from models, the IFTS will provide new knowledge about galaxy clusters at high redshift. The success of this survey will influence the next generation of astronomical instruments, especially for multiple faint objects.

Mission Relevance

This project supports the Laboratory's mission in national security, specifically nonproliferation, by developing core competence in hyperspectral imaging, remote sensing, and mining large data sets.

FY03 Results

FY03 represented the first half-year of the project. During this period, we modified the instrument to operate as a regular visiting instrument at the Mt. Palomar 5-m telescope and observed successfully for four nights in January 2003. We acquired datacubes of multiple objects (two spatial coordinates plus wavelength) that enable users to estimate the scientific return from this unique instrument on the 5-m telescope. This work prepared us for a second observing run, scheduled for January 2004 under separate funding.

Adaptive Optics Views of the Hubble Deep Fields

Claire E. Max 03-ERD-002

Abstract

This project is using LLNL-developed laser guide star adaptive optics (AO) systems at the Lick and Keck Observatories to study galaxies in the early universe. The goals are to observe large galaxies in the process of initial assembly from subcomponents, to identify central active galactic nuclei caused by black holes, and to measure rates of star formation in galaxies throughout the past 10–12 billion yr. The study focuses on three small regions of sky—the Great

Observatories Origins Deep Survey (GOODS), Galaxy Evolution from Morphology and Spectral Energy Distributions (GEMS), and Coordinates, Sizes, Magnitudes, Orientations, and Shapes (COSMOS) fields—that will be intensively studied by the Hubble Space Telescope, Chandra X-Ray Space Telescope, X-Ray Multimirror Mission (XMM) Space Telescope, the Space Infrared Telescope Facility (SIRTF), and the Very Large Array Radio Interferometer.

This effort will vastly extend our knowledge of how galaxies formed and evolved and will detect significant numbers of active galactic nuclei and supernovae at high redshifts. The project will also develop new methods and procedures for efficient use of the LLNL laser guide star AO systems, which are new methods for data analysis and for modeling the early evolution of galaxies in the distant universe.

Mission Relevance

The AO technology and precision optics and controls developed in this project will have application to high-power lasers for inertial-confinement fusion and stockpile stewardship, in furtherance of the national and energy security missions. In addition, the project will contribute insights into galaxy formation and evolution, which support LLNL's mission in scientific breakthroughs. Being at the forefront of a highly visible and exciting area of scientific inquiry, this research will help attract new talent to LLNL.

FY03 Results

In FY03, we used the AO system at Lick Observatory to characterize luminous blue compact galaxies and nearby active galaxies to establish a baseline for our evolutionary studies the original Hubble Deep Fields were observed for more than a week with the Hubble Space Telescope. Our Keck Telescope AO work was extended to a more distant class of galaxies (redshift 0.8 and greater), and we developed computational methods for separating disk and bulge components of these galaxies. We tested new techniques for determining point-spread function and conducted extensive computer simulations of expected performance in the GOODS, GEMS, and COSMOS fields.

Proposed Work for FY04

In FY04 the next step will be conducting a sustained observational campaign using Keck AO to obtain the world's highest spatial resolution near-infrared data on distant galaxies, resolving key subcomponents of galaxies (i.e., bars, spiral arms, and small regions of star formation). We will continue to develop computer simulations to delineate error bars for our observed statistical data, and work will begin on a data-reduction pipeline that will make a homogeneous body of AO data available in a public archive for the first time.

Publications

Bogdanovic, T. et al. (2003). "Circumnuclear shock and starburst in NGC 6240: Near-IR imaging and spectroscopy with adaptive optics." *Astronom. J.* **126**, 2299. UCRL-JC-151686.

Brotherton, M. S. et al. (2003). "Hubble Space Telescope imaging of the poststarburst quasar UN J1025-0040: Evidence for recent star formation." *Proc. Astronom. Soc. of the Pacific* **114**, 593. UCRL-JC-151352.

Canalizo, G. et al. (2003). "Adaptitve optics imaging and spectroscopy of Cygnus A. I.—Evidence for a minor merger." *Astrophys. J.* **597**, 823. UCRL-JRNL-155674 Pt. 1.

Gavel, D. T. et al. (2003). "Recent science and engineering results with the laser guidestar adaptive optics system at Lick Observatory." *Proc. SPIE* **4839**, 354. UCRL-JC-147284.

Lacy, M. et al. (2002). "Observations of quasar hosts with adaptive optics at Lick Observatory." *Astronom. J.* **124**, 3023. UCRL-JC-151351.

Analyzing the Long-Range Transport of Asian Aerosols using an LLNL Atmospheric Model and CAMS/NOAA Measurements from Northern California

Philip J. Cameron-Smith 03-ERD-021

Abstract

Our objective is to improve our understanding of, and our ability to model, the intercontinental transport of dust and other species, both gas and aerosol. Dust and other aerosols have important roles in climate change and air pollution and can be important for homeland-security assessments of chemical, biological, and radiological releases. We plan to improve the atmospheric transport model IMPACT by adding an interactive dust-injection algorithm and by increasing resolution. We will then isolate and validate advection, diffusion, and gravitational settling processes in the model through comparison with time- and size-resolved aerosol samples taken from six sites in California, Oregon, and Korea. By validating the IMPACT physical parameterizations, we will improve LLNL capabilities for homeland-security concerns regarding atmospheric releases. This project will also elucidate the amount and sources of dust and other aerosols entering the U.S., which is vital knowledge for air-quality standards.

Mission Relevance

This project supports LLNL's mission in environmental management. Aerosols potentially represent a large term in the radiative balance of the atmosphere and may be masking effects from greenhouse gases. In addition, dust is known to fertilize the oceans and cause sequestration of CO_2 (which relates to carbon management), but the emission and distribution of dust remains uncertain.

FY03 Results

In FY03 we (1) measured time- and size-resolved elemental composition and mass from four of the samples from California and Oregon; (2) performed back-trajectory analysis to determine the source locations of selected 2002 dust events; (3) installed an interactive dust-injection parameterization into the IMPACT model; (4) adapted the model and input fields to run at higher resolution (1 × 1 deg); (5) labeled aerosols in the model according to source region; (6) simulated the spring 2001 and 2002 dust seasons using the interactive dust injection parameterization at the higher resolution.

Proposed Work for FY04

In FY04 we will (1) measure time- and size-resolved elemental composition and mass from two additional sites in California and Oregon from spring 2002; (2) perform back-trajectory analysis to indicate source locations for additional 2002 dust events (as necessary); (3) simulate the spring 2002 dust season with the IMPACT model using the labeled dust sources at high resolution (1 × 1 deg); (4) cross-validate the measured and modeled results to determine source strengths for the Asian dust-source regions and validate the advection, diffusion, and gravitational settling processes in the model; (5) run a full-year simulation to quantify the amount of aerosol entering the U.S. from outside sources.

Thermally Driven Processes and Atmospheric Transport and Dispersion of Stable Macroparticles

Roxana M. Greenman

03-ERD-039

Abstract

Weapons of mass destruction (WMD) pose a significant threat to our military forces and civilian populations. We will examine the fate and dispersion of chemical agents released by a properly functioning warhead and by successful low- and high-altitude missile intercepts, which are not well understood. We will computationally and experimentally investigate the coupled aerodynamic and thermal response of agents to produce the source term for atmospheric fate and transport studies. This research effort will provide (1) better understanding of the fate of chemical agents from an atmospheric release, (2) better quantification of the risk associated with different atmospheric release events, and (3) the capability for the rapid analysis of both types of release events within the atmosphere.

We will develop an integrated, multidisciplinary tool that can predict the fate, dispersion, and ground effects of low- and high-altitude releases of chemical agents. This project will also enhance our ability to address issues in homeland security.

Mission Relevance

This research will further LLNL's mission in national security by addressing the serious problem of proliferation of WMD, producing a new integrated capability to determine the fate, dispersion, and ground effects of chemical and biological agents released by either a properly functioning warhead or successfully intercepted missile.

FY03 Results

In FY03 we (1) conducted fluid-dynamics and thermal calculations using the codes ALE3D and MuSiC to investigate initial elastic response of particulate and coupled aerodynamic heating; (2) added mixed-element surface-tension capability to ALE3D; (3) extended existing three-dimensional (3-D) atmospheric particle-transport and -dispersion models to account for initial momentum; (5) began creating a set of weather classes for areas of interest to better understand the sensitivity of low- or high-altitude releases of agents; and (6) began extending existing 3-D atmospheric particle-transport and -dispersion models to simulate the equation of motions of macroparticles in a rarefied flow.

Proposed Work for FY04

In FY04, we will estimate mass loss due to surface vaporization; conduct fluid-dynamics and thermal calculations to investigate initial elastic response and coupled aerodynamic heating; examine the thermal behavior of stable droplets in supersonic flow; investigate the possibility of enhanced breakup; determine the parameter uncertainty for meteorological conditions for certain times of interest; investigate meteorological classes and conditions for general ensemble-averaged wind-field analyses; prepare the codes LODI and ADAPT for batch-mode calculations and run a series of Monte Carlo simulations for comprehensive uncertainty analysis and risk assessment; and finally incorporate physical processes for high-altitude release into LLNL's National Atmospheric Release Advisory Center system, whose functions include providing predictions of hazardous-material plumes to emergency responders.

Publications

Nourgaliev, R. R. et al. (2004). *Direct numerical simulation of disperse multiphase high-speed flows.* Presented at the 42nd AIAA Aerospace Sciences Meeting and Exhibit, AIAA 2004-1284, Reno, Nevada, Jan. 5–8, 2004. UCRL-CONF-202429.

Predicting the Effects of Climate Change and Variability on Water Availability

Philip B. Duffy 03-ERD-042

Abstract

Human-caused climate change and natural climate variability will affect the hydrological cycle in California. This will result in the need for new strategies to manage existing water storage and conveyance infrastructure, and may require the construction of new infrastructure. Predictions of how climate change might affect the regional hydrological cycle are needed to provide decision support for policymakers and water managers. Using a hierarchy of models (global climate, regional climate, and surface hydrology) we will predict how climate change and year-to-year natural climate variability will affect surface temperatures, precipitation amounts, snow amounts, and the amount and timing of flow through rivers.

This project has two technical thrusts. The first is to produce the best possible assessment to date of the effects of climate change and variability on the availability of water in California. We will emphasize the use of state-of-the-art models at the finest possible spatial resolutions. The second technical thrust recognizes the inherent uncertainty in projecting climate change and its impacts and will quantify the uncertainties in our projections of future climate and hydrological cycle. Our approach will be to perform and analyze simulations with multiple models, and to consider multiple climate-change scenarios. We expect our work to significantly advance the state of the art in the modeling of regional climates and surface hydrology.

Mission Relevance

This work supports LLNL's energy-security and environmental-management missions by developing prediction methodologies and evaluating and improving climate models in furtherance of DOE's long-standing commitment to understanding the environmental consequences of energy generation. This project focuses on one of those consequences, global warming, which is caused primarily by burning fossil fuels for energy generation.

FY03 Results

We analyzed simulations of present and future climates in the western U.S. using 15 global climate models. This is part of evaluating uncertainties in future-climate projections. Climate model simulations of North American and Indian monsoons were evaluated at varied spatial resolutions to evaluate and improve the subgrid-scale physics in global climate models. We completed 10-yr climate simulations with the code FVGCM at spatial resolutions of 2.0 × 2.5 deg., 1.0 × 1.25 deg., and 0.5 × 0.625 deg. as part of projecting the effects of climate change on the climate and hydrological cycle. In collaboration with the University of California, Los Angeles, we began to perform limited-domain climate model simulations of California using boundary conditions from our high-resolution global climate model.

Proposed Work for FY04

We will (1) complete an ultrahigh-resolution global climate simulation; (2) complete evaluation of aspects of subgrid-scale physics in high-resolution global climate models; (3) survey existing regional-climate model simulations to assess the range of predicted climate changes for California; (4) perform improved high-resolution global climate simulations; (5) perform limited-domain climate model simulations of California; (6) configure two surface hydrology models for California watersheds; (7) simulate river flow rates with these models and compare simulated to observed flow rates; and (8) evaluate a parameterization of subgrid-scale convection in a global climate model at high resolution.

Environmental Transport and Fate of Endocrine Disruptors from the Nonpotable Reuse of Municipal Wastewater

George B. Hudson 03-ERD-065

Abstract

Endocrine-disrupting compounds (EDCs) affect normal hormonal activity and are contaminants of concern in drinking-water systems. This project studies the impacts of EDCs introduced into groundwater by wastewater recycling. We will apply new technologies to accurately determine the transport and fate of EDCs as they move from the surface into groundwater and potentially impact drinking-water supplies. We will use advanced isotope-tracer techniques, such as dating based on tritium/helium-3 ratios, to track groundwater movement. We are developing methods for measuring EDCs using state-of-the-art tandem mass spectrometry and are adapting sensitive bioassay techniques to measure estrogenic activity from groundwater samples.

This work provides significant and necessary data for evaluating the use of recycled wastewater for irrigation. Water is a limited resource, and recycling of wastewater is an important option for enhancing water supplies. Issues related to contaminants governed by drinking water standards are well understood; however, impacts from EDCs are largely unknown. We will produce information and data needed to quantitatively account for the subsurface transport of EDCs. We expect two publications in a prominent peer-reviewed journal. The first will address the impacts of EDCs on groundwater from irrigation using nonpotable recycled water, and the second will address the use of EDCs as wastewater tracers.

Mission Relevance

This project supports the Laboratory's environmental-management mission by providing significant data for evaluating the use of recycled wastewater for irrigation. This interdisciplinary investigation matches the fundamental strengths of LLNL with significant and enduring national needs in management of water resources.

FY03 Results

We developed an ultrasensitive mass spectrometry method for nonylphenol (NP), tested a bioassay technique using a human cell line, and modeled exposure pathways. Samples were collected from a site irrigated with recycled wastewater. Isotopic analyses indicated similar amounts of dilution and evaporation for groundwater samples ranging in age from 5 to 25 yr. All samples had detectable concentrations of NP, ranging from 60 to 3500 ng/L. The bioassay results of groundwater samples and standards suggested that NP contributed a significant fraction of the estrogenic activity observed in the samples. Soil column studies showed that NP transport is markedly retarded by soil sorption. This observation suggests that soluble NP precursors were transported and metabolized to NP in situ.

Proposed Work for FY04

We will use our methods at other field sites to determine whether our findings are generally applicable. We will investigate bioassay quantification in groundwater matrices to assess whether the bioassay is sufficiently accurate and precise to determine the exact proportion of the bioassay response attributable to measured estrogenic compounds. We will apply our tandem mass spectrometric techniques for a range of EDCs (including NP) to determine whether we can use EDCs as source tracers to identify contamination originating from sewer and septic systems.

Publications

Beller, H. R. et al. (2003). *Environmental transport and fate of endocrine-disrupting compounds from non-potable reuse of municipal wastewater*. Presented at Groundwater Resources Assoc. Ann. Conf., Apr. 2003, Ontario, CA. UCRL-PRES-151537.

Nitrate Biogeochemistry and Reactive Transport in California Groundwater

Bradley K. Esser 03-ERD-067

Abstract

Nitrate contamination is a significant threat to California groundwater. Successful groundwater management requires modeling future impacts on affected aquifers and assessing practices for reducing nitrate levels. Our research will investigate the fate and transport of nitrate in groundwater at three different scales: microbial denitrification kinetics under controlled laboratory conditions, nitrate cycling in the vadose and shallow saturated zones at the field scale, and nitrate cycling in deeper groundwater at the basin scale. Research will focus on developing and applying rapid methods to quantify denitrification, assessing the role of denitrification in the saturated zone, understanding subsurface biogeochemical heterogeneity, and developing scalable reactive-transport models.

The motivation for this work is the need to develop science-based approaches to characterizing and managing nitrate contamination in groundwater. If successful, we will develop a much more robust approach to characterizing nitrate biogeochemistry in the subsurface. When coupled with LLNL's ability to characterize groundwater flow and to model reactive transport, this approach will allow significantly more accurate assessment of the future distribution of nitrate in California aquifers, and of the impact of different management practices on nitrate inputs to groundwater. Such assessments are vital to making cost-effective management and policy decisions regarding land-use regulations and the water-treatment infrastructure.

Mission Relevance

This project supports the Laboratory's mission in environmental management by developing models and technologies to assess the environmental consequences of toxic materials and to manage risk. This type of multidisciplinary investigation matches the fundamental strengths of LLNL with significant and enduring national needs.

FY03 Results

In FY03 we (1) initiated a state-wide survey of the extent of saturated-zone denitrification in representative groundwater basins of California; (2) installed a grid of nested, multilevel samplers at a dairy farm site in the Central Valley in collaboration with California State University, Fresno, the California Water Institute, and the University of California, Davis; (3) performed initial sampling and characterization of groundwater flow and nitrate biogeochemistry in the Llagas Basin of Santa Clara county; (4) and developed a conceptual model for groundwater flow in the Llagas Basin.

Proposed Work for FY04

In FY04 we will develop real-time molecular probes based on polymerase chain reaction tailored for the in situ detection of microbial denitrification; determine the kinetics and physiology of autotrophic denitrification with pyrite; and continue our survey of the extent of saturated-zone denitrification in representative groundwater basins in California. At the Central Valley site, we will characterize spatial heterogeneity of denitrification in cores collected from the site; characterize groundwater flow and nitrate biogeochemistry in shallow groundwater from the site; begin development of a field-scale reactive-transport model; and investigate scaling using stochastic methods. In the Llagas Basin, we will complete characterization and begin developing a basin-scale reactive-transport model.

Starburst Galaxies

Willem Van Breugel

01-ERI-003

Abstract

Starbursts—large bursts of star formation—signal key episodes in the formation and evolution of galaxies. This project proposes to study massive forming galaxies to measure the physical conditions in collimated outflows (winds) triggered by these starbursts. The objective is to learn how these superwinds may affect the formation and evolution of subsequent generations of stars and galaxies. The project will use narrow-band filters on ground- and space-based observatories, numerical simulations, and adaptive optics (AO) to (1) study giant emission-line halos around massive forming galaxies and use narrow-band filters to estimate how common starburst superwinds are; (2) determine how superwinds are ionized and whether they contain nuclear-fusion material produced by starbursts; (3) determine the space densities of starburst galaxies at high redshift and the duty cycles of starburst and radio-source activity; (4) study nearby starburst galaxies with AO to determine the connection between galaxy collisions and starbursts; (5) perform numerical simulations and explore designs for possible laser experiments to help understand the effects of jets and radiation on star formation and the chemical enrichment in galaxies and their environment.

Mission Relevance

This project will advance special LLNL capabilities in AO, instrumentation, and tunable filters and will develop data-analysis and observational techniques that are applicable to the

Laboratory's nonproliferation mission. In addition, this project will contribute to stockpile stewardship by helping to validate hydrodynamic codes for galaxy formation and starburst astrophysics and laser experiments of starburst outflows and jets. The scientific appeal of this research will also attract new talent to the Laboratory.

FY03 Results

We completed our studies of giant emission-line halos around massive forming radio galaxies using narrow-band filters. From a comparison of deep optical and x-ray images, we found that the gas in the halos may be ionized by inverse Compton x-ray photons, which are due to scattering of infrared photons from dust by relativistic electrons from the radio source. We completed our study of the effects of jets on dense ambient material and found that jet-induced star formation is possible under certain conditions.

Publications

Arav, N. et al. "Hubble Space Telescope STIS observations of PG 0946+301." *Astrophys. J.* **561**, 118. UCRL-JC-155413.

Brotherton, M. et al. (2002). "Hubble Space Telescope imaging of the poststarburst quasar UN J1025-0040." *Astron. J. of the Pacific* **114**, 593. UCRL-JC-151352.

Brotherton, M. et al. (2002). "Multiband VLA observations of the faint radio core of 3C 68.1." *Astronom. J.* **124**, 1943. UCRL-JC-155405.

de Vries, W., W. van Breugel, and A. Quirrenbach. (2002). "AO observations of three powerful radio galaxies." *Proc. SPIE* **4834**, 310. UCRL-JC-149438.

Reuland, N. et al. (2003). "Giant Ly-α nebulae associated with high redshift radio galaxies." *Astrophys. J.* **592**, 755. UCRL-JC-153465.

Reuland, M., W. van Breugel, and H. Rottgering. (2002). "SCUBA observations of high redshift radio galaxies." *Proc. Conf. "Radio Galaxies: Past, Present, and Future,"* Nov. 2002, Leiden, Netherlands. UCRL-JC-152104.

Reuland, M. et al. (2002). "An obscured radio galaxy at high redshift." *Astrophys. J.* **582**, 71 UCRL-JC-151307.

van Breugel, W., et al. (2003). "Bright Lights, Big City: Massive galaxies, giant Ly-α nebulae and proto-clusters." SPIE Conf. Proc. **4834**, 24. UCRL-JC-149640.

Lithic Astronomy: Absolute Chronometers and Correlated Isotopic Anomalies in Meteorites

Ian D. Hutcheon 01-ERI-004

Abstract

A major discovery of the past 30 yr is the observation of isotopic anomalies in calcium–aluminum- (Ca–Al) rich inclusions (CAIs) in chondritic meteorites, the oldest known solid objects in our solar system. These nuclides have been attributed to seeding of the solar nebula by supernovae ejecta or irradiation by protosolar cosmic rays. The origin of these anomalies has important implications for the evolution of our solar system and, by comparison, to all systems with Sun-like stars. The evaluation of competing models is hindered by incomplete inventories of the various anomalies and the absence of an absolute chronometer. We investigate

correlated isotopic anomalies associated with short-lived radionuclides and apply an absolute chronometer based on uranium–lead decay to constrain the ages of CAIs and chondrules. The results of this research will allow us to constrain the time scales for the formation of the first solid bodies in the Solar System and provide more stringent constraints on astrophysical and cosmochemical models for the evolution of other planetary systems surrounding Sun-like stars.

Mission Relevance

This work supports both the stockpile-stewardship and basic-science missions of the Laboratory by providing an important testing ground for the new microanalytical instrumentation required by many DOE programs and is closely related to ongoing investigations of optical components and fissile materials for stockpile stewardship.

FY03 Results

We conducted experiments to examine the correlation between enrichments in oxygen-16 (^{16}O), a diagnostic signature of primitive Solar System material, and ^{26}Al. For the first time, we demonstrated that igneous (i.e., crystallized from a melt) inclusions in an especially primitive class of carbonaceous chondrites show a heretofore unrecognized correlation between ^{16}O and ^{26}Al. The ^{16}O-rich inclusions are also ^{26}Al rich, whereas ^{16}O-poor inclusions contain at least 10 times less ^{26}Al. This difference points to early formation of ^{16}O-rich inclusions and either delayed formation or remelting of ^{16}O-poor inclusions. The ^{16}O-poor inclusions appear closely related to chondrules both in time of formation and in oxygen isotope composition. Finally, the ability to analyze individual CAIs using high-spatial-resolution ion microprobe and high-precision multicollector mass spectrometry is an important milestone for this project.

Natural Variability and Anthropogenic Influence on Climate: Surface Water Processes in the Indonesian Seas over the Last 120 Yr

Thomas P. Guilderson 01-ERI-009

Abstract

To ascertain the influence of human activity on climate in the late 20th century, anthropogenic influences must be separated from natural variability in the climate system. This project focuses on the Indonesian throughflow, although our general approach and results will have broader applicability. Carbon-14 (^{14}C) time series, in conjunction with model experiments, are used to study the processes governing water-mass evolution in the Indonesian seas over time scales long enough to study the linkage between processes there and decadal modulation of the El Niño Southern Oscillation (ENSO), Asian monsoons, and global climate.

Implicated in the genesis of the ENSO are sea-surface temperature anomalies in the West Pacific that appear to be modulated by wind-forced vertical mixing. A related mechanism that may affect ENSO variability is long-period modulation of the vertical mixing within the Indonesian seas due to the dissipation of tidal energy. If vertical mixing in the Indonesian seas is a mechanism to modulate ENSO variability, we expect to find a decadally modulated ^{14}C seasonal cycle. Modelling actual ^{14}C values will be difficult, but we expect that seasonal and interannual variability will be robustly captured. Differences between the observed and

modeled tracer fields, in amplitude or in the time domain, will reveal flaws in the model physics and parameterization schemes. The combined data–model approach will lead to insights of the systematic biases in coupled ocean general circulation models used in climate prediction simulations.

Mission Relevance

The research combines three critical research themes: carbon cycle studies, natural vs. anthropogenic climate variability, and improved prognostication of future climate. All three of these broadly defined themes are relevant to existing programs within the DOE and other national agencies. This work supports the Laboratory's mission of environmental management and promotes collaborative research with several universities.

FY03 Results

We have completed a ~115-yr bimonthly coral-^{14}C time series from the Makasaar Straits and ~55-yr records from the Lombok Straits and the Sulawesi Sea. Exploratory analyses were made on novel sclerosponges. The ^{14}C records exhibited a significant pre- to post-atomic bomb testing transition, with large (up to 60 per mil) seasonal variability. The radiocarbon time series occasionally displayed a small seasonal signal (10–15 per mil) between 1890 and 1954. We found little evidence for decadal modulation of vertical mixing. Modeling experiments focused on two themes: (1) sensitivity of the model (tracer field and CO_2 uptake) to vertical mixing schemes and (2) wind forcing. The mixed-layer parameterization had the most influence on the seasonal cycle but was of secondary importance to the mean state and CO_2 uptake.

Publications

Fallon, S. J., T. P. Guilderson, and K. Caldeira. (2003). *Surface water processes in the Indonesian Seas as documented by a high-resolution coral delta-^{14}C record*. 18th Int'l Radiocarbon Conf., Sept. 2003, Wellington, New Zealand. UCRL-JC-153975.

Fallon, S. J., T. P. Guilderson, and K. Caldeira. (2003). "Surface water processes in the Indonesian Throughflow as documented by a 115-year high-resolution coral delta ^{14}C record." *Proc. 2003 Fall Amer. Geophys. Union Meet.*, Dec. 8–12, 2003, San Francisco, CA. UCRL-JC-155227 Abs.

Fallon, S. J., T. P. Guilderson, and K. Caldeira. (2002). "Radiocarbon constraints on oceanic mixing, Banks Island, Vanuatu: a coralline sponge perspective." *Proc. 2002 Am. Geophys. U. Fall Meet.*, Dec. 6–10, 2002, San Francisco, CA. UCRL-JC-149778.

Grumet, N., T. P. Guilderson, and R. B. Dunbar. (2003). "Meridional transport in the Indian Ocean traced by coral radiocarbon." *J. Marine Research* **60**, 725. UCRL-JC-150987.

Grumet, N., T. P. Guilderson, and R. B. Dunbar. (2003). "Pre-bomb radiocarbon variability inferred from a Kenyan coral record." *Radiocarbon* ***44***, 581. UCRL-JC-150919.

Guilderson, T. P. et al. (2002). "Post-bomb surface water radiocarbon history in Indo-Pacific corals." *Proc. 9th Intl. Conf. on Accelerator Mass Spectrom.*, Sept. 9–13, 2002, Nagoya, Japan. UCRL-JC-151787.

Direct Imaging of Warm Extrasolar Planets

Bruce A. Macintosh

02-ERI-005

Abstract

More than 100 giant planets orbiting stars outside our Solar System have been detected via spectroscopy. However, this technology can detect only those planets located in the inner part of their systems. Although the knowledge gained from these observations has overturned models of planet formation, it does not reveal habitable, Earth-like worlds. By contrast, direct imaging of the planets would be sensitive to planets in wide orbits. However, mature planets are a billion times fainter than the stars they orbit, making direct imaging a challenge. In this project, we are using adaptive optics (AO) to look for infrared (IR) emission from young planets orbiting young stars. With AO, we search for young planetary companions to 200 stars and develop image-processing techniques that will make it possible to detect sources in the scattered starlight.

Our large-scale survey of young stars has the potential to produce some of the first direct detections of IR emission from planets orbiting other stars. We have the ability to detect planets with masses two to five times that of Jupiter in orbits of 10 to 20 astronomical units—planetary systems slightly bigger than our own. Even if such systems do not exist, their absence will strongly constrain models of planet formation, showing that giant planets form only in the central parts of solar systems and then migrate inwards. This project will also develop new techniques for high-contrast processing of AO images. These techniques will exploit correlations in noise patterns and will be applicable to a wide variety of imaging applications.

Mission Relevance

This project maintains and extends AO as a key technology for a variety of LLNL and DOE applications that require high-contrast imaging and image processing, especially for secure communications and remote sensing in support of the national security missions of nonproliferation, arms control, and international security.

FY03 Results

Using the Keck AO system, we observed approximately 120 young stars from several distinct populations, began analysis of these data, and identified several candidate companions for follow-up observations of their spectra or motion. We also observed, at very high sensitivity, stars surrounded by dust rings that could be signposts of a planet. Follow-up observations of these stars were completed, and we derived and published upper limits on planets in these systems. Our work on image processing exploited the correlated nature of the noise in the high-contrast AO images of stars by calculating local correlation statistics and searching for deviations in images caused by the presence of a faint planet. The results demonstrated increases in imaging sensitivity of two to five times.

Proposed Work for FY04

During FY04, we will use Keck AO to observe the remainder of our young-star sample. Follow-up observations of candidate planetary companions will continue, either by follow-up spectroscopy or by studying the motion of the planet itself. We plan to apply a new observing strategy to our most interesting stars by observing them at thermal IR wavelengths where

AO performance is best. Our correlation-based image-processing software will be refined and applied to our observational data set. Finally, we will study promising image-processing techniques that could be applied to hyperspectral data cubes.

Publications

Macintosh, B. et al. (2003). "Deep Keck adaptive optics searches for extrasolar planets in the dust of epsilon Eridani and Vega." *Astrophys. J.* **594** (1), 538–544 Part 1. UCRL-JC-150951.

Sivaramakrishnan, A. et al. (2003). "Speckle decorrelation and dynamic range in speckle noise limited imaging." *Astrophys. J.* **581** (1), L59–L62 Part 2. UCRL-JC-150950.

Tectonic Morphochronology of the Southern San Andreas Fault System

Frederick J. Ryerson 03-ERI-001

Abstract

The San Andreas Fault (SAF) accommodates a major fraction of the motion between the North American and Pacific tectonic plates. How slip is distributed among the many segments of the fault is not well understood and has implications for seismic risk. The SAF divides into a number of strike-slip segments and thrust zones south of the Transverse Range. Geodetic observations performed there focus on deformation, but only two millennial (long-term) slip-rate measurements exist for the SAF. We will begin developing a millennial slip-rate map of the western U.S., initially focusing on sites within the SAF system. Slip rates will be determined by using satellite imagery and field data to measure landscape offsets caused by active faults and then using cosmic-ray surface-exposure dating to determine their age.

This project will add to our understanding of the interactions of fault motion in continental settings, with important implications for the assessment of earthquake risk. We will use cosmogenic dating, which has not been widely used in the western U.S., to develop a method for determining millennial slip rates. Along with paleoseismic data and geodetic slip rates, these millennial slip rates will help to identify stress accumulation on the various segments of the fault. Disparities in long- and short-term slip rates influence earthquake frequency. If successful, this method will provide a new tool in evaluating crustal deformation that can be widely used in seismically active regions throughout the world.

Mission Relevance

The analysis of satellite imagery to identify tectonic features furthers radar interferometry and contributes to understanding the mechanics of the Earth's crust. The dating of landforms also provides information on their stability. These topics are related to explosion monitoring and analysis for the Laboratory's mission in nonproliferation. In addition, the structure and tectonics of the western U.S. are important in enhanced test readiness, geothermal energy, and the disposal of radioactive waste, in support of missions in national and energy security.

FY03 Results

During FY03, we completed the dating of an offset alluvial fan at Biskra Palms on the southern SAF. This is the first time that slip rate on the fault was determined by cosmogenic dating, and extends the interval of millennial slip-rate observation for the SAF by a factor of three. The ages determined are tightly clustered and, with an offset, yielded a rate of 17 mm/yr.

In addition, we began dating surfaces that constrain the rate on the San Jacinto Fault at Anza. The dates from surface samples show more dispersion than we had hoped, and additional sampling of subsurface samples is required. We also collected samples from the Mojave segment of the SAF and from Cajon Pass for dating.

Proposed Work for FY04

During FY04, work will focus on the San Gorgonio Pass and the Mojave segments of the SAF, the Anza region on the San Jacinto Fault, and uplift along the Sierra Madre-Cucamonga thrust system. We expect to make the first long-term rate determinations in each of these regions. The San Gorgonio Pass and Anza data will constrain the relative deformation accommodated by these features and can be directly compared to geodetic data to assess stress accumulation. The Mojave and Sierra Madre data will provide a new assessment of the relative roles of thrusting and strike-slip faulting in the Los Angeles Basin. We will also date samples from Cajon Pass to compare radiocarbon and cosmogenic dates.

A Next-Generation Microlensing Survey

Kem H. Cook

03-ERI-002

Abstract

This project is conducting a microlensing survey, SuperMACHO (Massive Compact Halo Objects), that will show a tenfold improvement over the MACHO project in the detection rate of microlensing events. The main goal of the SuperMACHO project is detecting microlensing events toward the Large Magellanic Cloud (LMC). The research addresses one of the outstanding problems in the physical sciences: the nature and distribution of dark matter. In MACHO, the detected rate of gravitational lensing events indicated that MACHOs comprise, at most, 20% of the dark matter halo. Our goal is to determine the nature of this lensing population, which may be more massive than all other known components of the galaxy. We are using the 4-m telescope at Chile's Cerro Tololo International Observatory's to conduct this survey.

We expect to locate a significant fraction of the lenses in microlensing events toward the LMC. This will determine the baryonic fraction of the Milky Way's halo dark matter. If the lenses are in the halo, we will have detected the first significant component of dark matter. If not, we will make the best determination yet that dark matter is composed of exotic subatomic particles and will achieve significant experience in handling large quantities of large-format images and analyzing them in real time.

Mission Relevance

The project supports the development of software and techniques for LLNL's national-security mission by advancing wide-field imaging capabilities with real-time analysis of large data streams. The project also supports DOE's and NNSA's goal of doing great science by characterizing the nature of dark matter and provides the technical foundation that will support the development of the National Academy of Science's highly recommended project for astronomy, the Large-Aperture Synoptic Survey Telescope. This research is a collaboration involving several universities and national facilities.

FY03 Results

We have produced a list of more than 300 microlensing-event candidates. About ten are very strong cases for true microlensing. To reach this milestone, we developed a custom database and search tools. Positions for all identified MACHO variables on the UCAC1 system were loaded into the database to be cross-correlated with SuperMACHO variable stars and microlensing candidates. Light curves are easily extracted for any variable object given only right ascension and declination. We developed two photometry pipelines capable of reducing many gigabytes of image data in near real time and integrated these pipelines with the Cerro Tololo 4-m data-acquisition system. We also developed remote-observing capabilities to optimize operations in Chile. The team reported on the project through three conference presentations.

Proposed Work for FY04

In FY04, we will participate in the collection and analysis of data using both image-difference techniques and object-based photometry. We anticipate producing real-time detection of microlensing events and real-time identification of exotic events, and we plan to implement real-time detection efficiency determination and optimize the survey procedure to maximize detections. When events are detected, we will make rapid announcements to the astronomy community to allow smaller telescopes to better characterize exotic events.

Publications

Drake, A. J. et al. (2003). "A next generation microlensing search: SuperMacho." *Dark Matter in Astro- and Particle Physics: Proc. Intl. Conf. DARK* **20**, 71. UCRL-PROC-200594.

Hiriart, R. et al. (2003). *The SuperMacho+SuperNova Survey database design: Supporting time domain analysis of GB to TB astronomical databases.* Presented at the Am. Astronom. Soc. 201st Mtg. UCRL-ABS-200538.

Stubbs, C. W. et al. (2003). *The SuperMacho Project: An LMC microlensing search using the Blanco 4 m telescope and the MOSAIC imager.* Presented at the Am. Astronom. Soc. 201st Mtg. UCRL-ABS-200632.

Resolving the Earthquake Source Scaling Problem

William R. Walter 03-LW-004

Abstract

How does the energy released by earthquakes scale with size? Is a magnitude-8 earthquake just a scaled-up magnitude 2, or does the fault energy density change with earthquake size? Disagreement exists within the geophysical community on this subject, with recent papers presenting conflicting evidence for and against constant energy density. The resolution of this issue is complicated by the difficulty of accurately accounting for attenuation, radiation inhomogeneities, and bandwidth, and of determining the seismic energy radiated by earthquakes over a wide range of event sizes in a consistent manner. In this project, we propose to resolve the issue by consistently applying two different LLNL-developed state-of-the-art techniques over common paths for a wide range of earthquake sizes.

Our goal is to produce the best regional study of earthquake energy estimates to date. By examining events covering many orders of magnitude in size with consistent techniques, paths, and stations, we expect to resolve the question of constant earthquake energy density. Earthquake scaling is a key property both for better understanding the physics of the earthquake source and as a tool to predict ground motion for future large events. Nonconstant energy-density scaling of earthquakes would lead to significantly larger predicted groundmotion for future big earthquakes, when such ground motion is estimated by scaling up existing smaller events.

Mission Relevance

This work supports the nonproliferation and environmental risk-reduction missions of LLNL. The project will provide a more accurate energy-scaling model of the earthquake source, which can then be coupled with LLNL-developed propagation techniques. This model has important applications for existing earthquake seismic-hazard prediction and event identification for nuclear monitoring. For hazard mitigation, the model provides more accurate prediction of future large-magnitude earthquake amplitudes from the more common, existing small earthquakes.

FY03 Results

The project had two major accomplishments in FY03. First we organized and ran a workshop on earthquake energy scaling on July 24, 2003, which brought together many experts in the field to better define the problem and examine the conflicting data available to date. Second, we completed an earthquake energy study using direct and coda (i.e., continuous spectral wave trains in the tail portion of seismographs) techniques on several hundred earthquakes in the Western U.S. and in Turkey. We found that earthquake energy scaling breaks down, either through increasing apparent stress or through a change in spectral shape scaling as earthquake size increases. These results were presented at the 2003 Fall American Geophysical Union meeting and in a planned peer-reviewed paper.

Publications

Mayeda, K. and W. R. Walter. (2003). *Earthquake apparent stress scaling: A comparative study*. Presented at the EGS/AGU/EUG Mtg., Nice, France, April 6–11, 2003. UCRL-JC-150198-ABS.

Mayeda, K. and W. R. Walter. (2003). *Energy scaling for the Hector Mine and Landers sequences*. Presented at the Fall AGU Mtg., San Francisco, CA, Dec. 8–12, 2003. UCRL-JC-155206-ABS.

Walter, W. R. and K. Mayeda. (2003). *Earthquake apparent stress scaling for the 1999 Hector Mine sequence*. Presented at the Fall AGU Mtg., San Francisco, CA, Dec. 8–12, 2003. UCRL-JC-155081-ABS.

Walter, W. R. and K. Mayeda. (2003). *Earthquake Energy Scaling Workshop*. http://earthscience.llnl.gov/scaling-workshop/. (retrieved Jan. 9, 2004). UCRL-WEB-155236.

Walter, W. R., K. Mayeda, and S. Ruppert. (2002). *Earthquake apparent stress scaling*. Presented at the Fall AGU Mtg., San Francisco, CA, Dec. 8–12, 2003. UCRL-JC-149777-ABS.

Walter, W. R. et al. (2003). *Earthquake apparent stress scaling*. Presented at the IRIS Ann. Workshop, Fish Camp, CA, June 19–21, 2003. UCRL-JC-150196-ABS-REV.

Melting Studies of Simple Planetary Ices

Jae-Hyun Park 03-LW-020

Abstract

Methane, CH_4, the simplest organic molecule in which carbon is tetrahedrally bonded, is known to be a major component of some of the Jovian planets. Recent discovery shows that methane–water clathrates are also abundant near the floor of deep oceans, where they are potentially useful as a source of gas. Given the importance of methane to terrestrial investigations and astronomic models, this study delivered fundamental experimental data on the melt and phase diagrams of methane in an extended region of high pressures and temperatures. In this study, we observed phase transitions in situ at high pressures and temperatures by using both conventional and coherent anti-Stokes Raman spectroscopy applied to laser- or ohmic-heated diamond-anvil cells (DACs). Structures of methane clathrates were characterized using intense, third-generation synchrotron x-ray diffraction.

The melt and phase information from this study will have an immediate impact on three major scientific areas: the constraint of planetary models for the outer-giant planets, Uranus and Neptune; discovery of exotic phases; and insights for many-body intermolecular interactions of nonhard sphere and nonspherical molecules for which no reliable theory exists as yet.

Mission Relevance

Because simple methane molecules are created behind a detonation front, their melt and phase data are fundamental to understanding detonation chemistry, which is important for stockpile stewardship. Improved understanding of methane, which has potential as an energy source, contributes to DOE's mission in developing new sources of energy. In addition, methane is a potent greenhouse gas; its exchange with the atmosphere has implications for global climate change, another DOE mission area. Also, addressing the stability and properties of fundamental units of hydrogen-bonded biosystems in a wide pressure and temperature range is critically important for understanding the origins of life on Earth and for countering bioterrorism.

FY03 Results

Methane samples loaded into DACs were studied up to 55 GPa in the temperature range of 25 to 450°C by using Raman spectroscopy and synchrotron x-ray diffraction. Contrasting the results from a condensation-loading method and one using a bellows shows strong evidence that the presence of a nitrous oxide impurity can shift the pressure–temperature phase boundaries. Sample heating was also observed to shift the phase boundaries metastably. These experiments resolve the conflicting results of previous literature reports. Pure methane at room temperature does not show any clear spectroscopic or cystallographic evidence of phase transition at least to 50 GPa. Laser-heating experiments demonstrated that methane decomposes into carbon and hydrogen rather than producing higher-order carbon chains.

Publications

Iota, V., J.-H. Park, and C. S. Yoo. (in press). "Phase diagram of nitrous oxide: analogy with carbon dioxide," *PRB*. UCRL-JC-153092.

Park, J.-H. et al. (2003). "Crystal structure of bent carbon dioxide: Precursor to polymerization." *PRB* **68**, 014107. UCRL-JC-151312.

Park, J.-H. et al. *Crystal structures of bent CO_2 and N_2O at high pressures*. Presented at the APS 2003 March Mtg., Austin, TX, March 3–7, 2003. UCRL-JC-150171-ABS.

A Novel Antimatter Detector with Application to Dark Matter Searches

William W. Craig

03-LW-059

Abstract

We propose to demonstrate an entirely new technique, the gaseous antiparticle spectrometer (GAPS), for the detection of antimatter in cosmic rays. The concept will allow an orders-of-magnitude increase in searches for these elusive particles. The sensitivity of our technique has an immediate and significant impact on the detection of the mysterious dark matter component of the Universe and could answer one of the most pressing questions of modern cosmology: What is the composition of the vast majority of the gravitating mass in our Universe? In the second year of our project we will complete construction of a prototype detector that validates the physics and efficiency of the GAPS concept. The prototype will be tested in an antiproton beam line. Once beam-line testing confirms that the approach is valid, this novel and scientifically important research will result in several publications.

Mission Relevance

The technologies in the GAPS prototype instrument will be directly applicable to the national-security mission. In particular, the data-acquisition system being developed for the prototype will be used by a large, coded-aperture instrument being developed for nuclear material search programs. The GAPS background rejection scheme is now under study for potential use in detecting heavily shielded high-*Z* material. If scientifically successful, GAPS will be a high-profile achievement that will help recruit quality scientists interested in detector concept development.

FY03 Results

In FY03, the physics optimization of the prototype instrument was completed, and the initial configurations of the GAPS chamber and detector were fixed. A change in the geometry of the detectors allowed more uniform coverage of the interactions occurring within the gas cell. All mechanical and electronic systems, including detectors, were specified by the end of the sixth month. Beam time for FY04 was approved in collaboration with scientists at the Japanese High-Energy Accelerator Research Organization (KEK). Construction began on the gas containment vessel, the detector support structure, the detectors, and the data-acquisition system. Detailed Monte Carlo work was begun, taking into account the beamline geometry. Publications describing the instrument were drafted and will be submitted in FY04.

Proposed Work for FY04

We plan to spend the first 6 mo of FY04 completing the construction, laboratory testing, and calibration of the GAPS prototype. A primary task will be to generate the software for real-time data processing; the first release will be completed by end of the third month. Testing will use laboratory sources and simulated triggers and vetoes, and will validate instrument performance before installing the prototype at KEK, where 14 shifts of beam time in March of 2004 will be used to perform system validation. Data analysis and publications, including instrument description and beam test results, will continue through the end of the year, with a possible second beam test scheduled for year's end.

Permeability Enhancement in Fine-Grained Sediments by Chemically Induced Clay Fabric Shrinkage

Ananda M. Wijesinghe 03-FS-004

Abstract

Contaminant entrapment in fine-grained, low-permeabilty, high-sorptivity sediments is a major impediment to timely, cost-effective remediation of groundwater to regulatory standards. We propose to inject ethanol, an EPA-approved organic solvent, to induce shrinkage cracks to form in clay strata, thus reducing by orders of magnitude the time required to complete remediation by diffusive transport alone. We are testing clays at different confining stresses and ethanol concentrations to measure by x-ray and ultrasonic methods (1) the shrinkage coefficient and compression modulus and (2) the crack propagation speed, effective permeability increase, and the geometry of the created cracks.

The proof-of-principle experiments undertaken will determine whether ethanol-induced shrinkage can overcome the confining stresses and increase the effective permeability by orders of magnitude much faster than by diffusive transport in unaltered clay sediments. Previous studies have not measured or explained the mechanisms responsible for rapid cracking of clays by organic solvents. If successful, the results from this project will yield significant savings for site remediation at LLNL and elsewhere. We have patented this clay-cracking technology (U.S. Pats. 5,593,248 and 5,906,748), but the feasibility of key processes is not yet established.

Mission Relevance

Environmental remediation is a major mission for DOE and LLNL. Existing technologies cannot cost-effectively overcome the barriers to mass transfer posed by fine-grained sediments and currently require decades, even centuries, of treatment. As pollution of large bodies of water by small amounts of toxic contaminants is the greatest threat to the nation's groundwater supply, the impact of successful deployment of this technology would be immense.

FY03 Results

Shrinkage and crack-propagation experiments were designed, and the test cells were fabricated. Four instrument clusters for continuous measurement of ethanol–water fractions, sodium bromide and sodium perchlorate tracer concentrations, electrical conductivity, pH, and temperature were designed and fabricated. A data-acquisition system was developed to automatically acquire data into a spreadsheet. X-ray imaging was found to be more effective; ultrasonic imaging was deferred. We devised methods for preparing bentonite clay test samples, calibrated all transducers, and evaluated the impact of leached chemicals on the fluid chemistry. The clay shrinkage and cracking experiments were set up for the most favorable case of 100% ethanol perfusion, and measurements are currently being acquired from the experiments.

Proposed Work for FY04

The cracking experiment in FY04 will determine the minimum ethanol concentration needed to initiate cracking, the increase in permeability, and the crack velocity, as functions of ethanol concentration and confining stress. If the cracking time is much smaller than the time for diffusive mass transfer, the feasibility of the concept will have been demonstrated, thus justifying further development of the process. If the results are promising, we plan to prepare and submit papers on the results of this project for publication in peer-reviewed journals.

Energy Supply and Use

Innovative Carbon-Dioxide-Separation Technologies

James R. Smith

03-ERD-058

Abstract

A challenge facing the U.S. energy-research community is to develop technology to reduce the U.S. industrial carbon intensity by 18% by 2012 without adverse economic effects. Fossil-fuel electric plants—especially coal-fired processes—are the single largest component (41%) of U.S. carbon emissions. This project is investigating novel approaches to carbon dioxide (CO_2) separation from the flue gas streams of fossil-fuel power plants. Our aim is to identify an innovative technology that can significantly reduce CO_2 separation costs and that can be implemented within a decade.

Mission Relevance

This project supports LLNL's energy-security and environmental-management missions by establishing effective technologies for significantly reducing CO_2 emissions and finding a permanent means for sequestering the captured carbon.

FY03 Results

In FY03 we investigated four approaches to CO_2 separation. The first was to reduce coastal fossil-fuel power plant CO_2 emissions using plant cooling-water flows, enhanced carbonate dissolution, and bicarbonate carbon sequestration. We found that the technique would not significantly reduce separation and sequestration costs. The second approach was the use of polymeric membranes to separate CO_2 from flue gases. Our findings indicate that this method could significantly reduce separation costs. Advanced membrane development was identified as a future task. The third method—combining unit processes for oxygen enrichment and CO_2 extraction using an electrochemical cell with low-temperature hydrated molten carbonate salt—was found to reduce separation costs somewhat. The fourth approach was an engineered natural analog process of converting gas-phase CO_2 to graphite; we identified the likely process conditions.

Molecular Engineering of Electrodialysis Membranes

William Bourcier

03-ERD-060

Abstract

We plan to develop an energy-efficient membrane for use in the electrodialysis treatment of brackish waters. Synthesis of a nitrate-selective membrane will be guided by testing and modeling membranes with nanotubes as well as membranes with functionalized pores. An

energy-efficient nitrate-removal technology will have a major impact on water availability in California and elsewhere.

Completion of this project will advance the state of the art in membrane functionalization specific to water technology applications, provide innovative ways to improve membrane process-energy efficiency, and establish new membrane-fabrication techniques, all of which can then be applied to a wide variety of additional applications in water treatment and resource extraction from saline brines.

Mission Relevance

This effort will support the Laboratory's homeland-security mission by developing a means to protect water, a resource that is vital to national security. To meet the goals of a safe, economic supply of water, it is important that clean water be generated in a way that consumes the least possible amount of energy. Technologies developed under this research project would provide clean water in a cost-effective manner for the U.S.

FY03 Results

In FY03, we synthesized ion-track-etched porous polycarbonate membranes, developed modeling codes for calculating electric fields and ion transport through membranes, designed and fabricated a test cell for measuring ion movement through permselective membranes driven by electrical potential gradient, and evaluated porous alumina membranes.

Proposed Work for FY04

In FY04, we will produce a proof-of-principle polycarbonate membrane (~1–3 cm in diam) that has 10-nm holes made by ion track etching. The membrane will have a conducting layer deposited onto its surface, such as gold. The pores will be functionalized to facilitate the transport (removal) of nitrate from the feed water supply. This membrane will then be incorporated into a benchtop electrodialysis unit. In addition, we will develop a molecular-level model of ion transport through nanoporous membranes with explicit provision for an electrostatic field, a flow field, diffusion, and quantum mechanical descriptions of molecular species.

Persistent Monitoring Platforms

Charles L. Bennett 03-ERD-076

Abstract

In this project, we plan to build and test a model of a stratospheric aircraft powered by thermal energy from the Sun. Such an aircraft could maintain station over a particular ground location almost indefinitely, since it would not need fuel. We are developing a thermally coupled system with an efficiency nearly an order of magnitude better than the state of the art (Helios) by creating the technology for a sun-tracking solar-heat collector, thermal storage reservoir, and a direct-drive Stirling heat engine. Physics models will be developed for thermal transport, materials interactions, loss mechanisms, and engine performance in the stratosphere.

The physics models developed and validated in this project will prove the principles involved in a solar thermal-powered aircraft in preparation for constructing a prototype scale-model aircraft to demonstrate station-keeping capability at sea level. Successful demonstration of

this technology would set the stage for building a stratospheric-altitude prototype capable of circumnavigating the globe.

Mission Relevance

Persistent surveillance—having unlimited dwell time over a region of interest—would enable the acquisition of a qualitatively new type of intelligence information for the purposes of a variety of national-security applications, such as countering the proliferation of weapons of mass destruction, and for border monitoring for homeland-security missions.

FY03 Results

The project began with only a few weeks left in FY03. During this time, we produced a preliminary design of a prototype unit.

Proposed Work for FY04

In FY04, we propose to construct physics-based models for solar thermal-powered aircraft having a hot reservoir of lithium hydride (LiH) at ~1000 K in the 200-K, 0.1-bar environment of the stratosphere. By studying the materials science associated with LiH, the tribology of a direct-drive Stirling engine using ambient air as its working fluid, and the physics of hydrodynamic foil air bearings, we expect to produce a system physics model with sufficient fidelity to establish proof of principle. This work will guide follow-on system engineering for actual construction of the thermal-powered aircraft. Scaled models of at least some of the system components may also be constructed to validate the theoretical models.

The Kinetic Stabilizer: A Route to a Simpler Magnetic Fusion System

Richard F. Post

02-LW-043

Abstract

This research studies a new plasma-stabilization concept, the Kinetic Stabilizer (KS), as applied to magnetic mirror fusion systems. The KS stems from theory by Ryutov, who showed that magnetohydrodynamically (MHD) unstable, axisymmetric mirror systems in magnetic fusion systems are stabilized by the presence of low-density plasma in the field outside the mirror. The KS allows this concept to be used to make tandem-mirror (TM) fusion systems that would be much simpler than present systems. The KS injects ion beams into the magnetic field outside the mirror. Compression and reflection of these ions form the stabilizer plasma. The basic feasibility of the KS having been shown, we are now using analyses and computer codes to verify and extend these results in preparation for proof-of-principle experiments.

Success in these studies could result in a major change in the prospects for fusion power. Magnetic fusion research has been dominated by a search for ways to handle plasma turbulence, endemic to closed approaches such as the tokamak. Turbulence determines the scale of such systems and impedes theoretical prediction of performance, thus requiring a largely empirical approach involving increasingly large and expensive apparatus. The KS TM offers the possibility of fusion power systems that are not dominated by turbulence, making

the systems simpler and amenable to prediction through the use of theory and computer-simulation codes.

Mission Relevance

This work supports LLNL's energy-security mission by responding to a pressing need, expressed by the DOE Magnetic Fusion Program, for simpler, smaller, and less expensive approaches to fusion energy. The KS would allow MHD-stable operation of TM fusion power systems, which should be able to exploit fully the potential of magnetic confinement, something not possible with current magnetic fusion systems such as the tokamak.

FY03 Results

In FY03, progress included optimizing the magnetic field configurations used in the MHD stability studies, updating and benchmarking the FLORA MHD code, using FLORA to confirm and extend the stability analyses, and acquiring a Fokker-Planck code and adapting it for use on our computer systems. Progress was also made toward developing transport codes to assess conditions required for ignition of a fusion plasma in a KS TM fusion system. Results from these codes predict that ignition of deuterium–tritium fusion fuel could be achieved with magnetic fields and plasma radii that are small compared to those of "closed" magnetic fusion devices, such as tokamaks. A study of means to suppress non-MHD instability modes in a KS TM system was also performed resulting in specific stability criteria.

Proposed Work for FY04

Work planned for FY04 includes using distribution functions from Fokker-Planck and/or Monte Carlo code calculations as input for calculating the MHD stability of fusion-relevant TM plasmas, using analysis and transport codes developed in FY03 to assess radial and axial transport using fusion-relevant boundary conditions, and at the same time systematically considering the means to avoid non-MHD instability modes (such as temperature–gradient modes) through control of the potential gradients and boundary conditions. Finally, combining our theoretical and computational results, we hope to provide a credible prediction of the performance of a KS TM fusion power system, including preliminary economic analyses of the cost of fusion power in the KS TM as compared to tokamak-type systems.

Publications

Post, R. F. et al. (2003). *The Kinetic Stabilizer tandem mirror: Analyses and new options*. Presented at Innovative Confinement Concepts 2003, Seattle, WA, May 28–30, 2003. UCRL-JC-153408-ABS.

Post, R. F. et al. (2003). *Progress toward the analysis of the kinetic stabilizer concept*. Presented at the 5th Symp. on Current Trends in Intl. Fusion Research, Washington, D.C., June 24–28, 2003. UCRL-JC-151779-ABS.

Engineering and Manufacturing Processes

Advancing the Science and Engineering of Mesoscale Nondestructive Characterization

Harry E. Martz

03-SI-004

Abstract

This project will provide tabletop technologies for nondestructive characterization (NDC) data on the structure and geometry of mesoscale (millimeter-size) objects. The goal is to obtain 1-μm resolution over a 1-mm field of view. The plan is to advance amplitude- and phase-contrast imaging to achieve the spatial resolution, penetration, contrast, field of view, and acquisition times needed. We expect to achieve this by researching and developing physics and engineering models and a Wölter-optic x-ray microscope. As-built, two-dimensional (2-D) and 3-D models will incorporate the characterization data to bridge the gap between simulation and experimental results.

The results of this project will form the basis for a new mesoscale NDC capability with micrometer resolution, high contrast (0.999), useful fields of view (1 to 3 mm), and rapid acquisition times (minutes) that will make future NDC instruments practical in fabrication environments and enable 2-D and 3-D as-built models for simulations.

Mission Relevance

Increased understanding of radiation–matter interactions for mesoscale-object NDC may ultimately be applied to high-energy-density targets in support of stockpile stewardship experiments on large lasers. In addition, mesoscale-object NDC capabilities are applicable to explosives characterization, sensor technologies for counterterrorism, advanced fuel cells, and biological research to advance human health.

FY03 Results

The mechanical design of the 8-keV prototype instrument was nearly completed, and several replicated optics fabricated for testing purposes. One developmental 8-keV Wölter multilayed optic was fabricated. Preliminary designs for a 59-keV prototype instrument were carried out and preliminary object-recovery techniques were developed and tested. Phase-contrast imaging techniques in both point-projection and in the Wölter optical microscope have been investigated. Results show that phase contrast is observed for both methods. Modeling of x-ray imaging to evaluate and optimize instrument designs was advanced to incorporate phase contrast.

Proposed Work for FY04

In FY04, we will research tabletop-imaging technologies in the 8 to 60-keV x-ray regime that will provide information on the structure, geometry, and alignment of mesoscale objects. This will include demonstrating the utility of a high-collection-efficiency 8-keV Wölter multilayer optics microscope to achieve the highest possible amplitude-contrast data in the shortest amount of time and exploring the use of this microscope for phase-contrast imaging. We plan to develop a high-efficiency x-ray detector and complete the optical design of the 59-keV Wölter microscope. Object-recovery algorithms will be developed for the Wölter microscope. Our x-ray simulation codes will incorporate Wölter optics ray-tracing and scalar-diffraction methods. The as-built modeling requirements will be defined.

Publications

Chapman, H. et al. (2003). *Is high brightness required for phase contrast X-ray imaging?* Presented at the 2nd Intl. Conf. on Noncrystalline Phase Retrieval, Palm Cove, Australia, July 2003. UCRL-PRES-154215.

Reconfigurable, Optical Code-Division Multiple Access for Fiber-Optic Networks

Steven W. Bond
01-ERD-002

Abstract

This project aims to develop code-division multiple-access (CDMA) technology using phase modulation to encode wideband, ultrashort-pulse data, thereby enabling multiple users to transmit data on wavelength-division-multiplexing networks. The objective of this project is to demonstrate the reconfigurable, physical-layer optical CDMA encoding and decoding of signals to provide multiuser information transmission over fiber-optic networks.

Our deliverable is an integrated electro-optical chip, composed of a gigahertz modulator monolithically integrated with an arrayed waveguide grating (AWG) and capable of supporting these data streams. We have already developed a concept that allows transmission of secure signals with optical CDMA formats that utilize complex, time-varying codes.

Mission Relevance

This project entails the development of advanced technologies, such as secure, high-speed data transmission, with potential applications that support LLNL's national-security mission. This project will result in a new implementation of CDMA as a physical-layer encoding technique for secure communication and data transmission, which in addition to national-security applications have the potential for a number of commercial spinoffs, e.g., enabling network operators to rapidly reconfigure networks based on user demand.

FY03 Results

In FY03 our effort focused on fabricating an integrated chip capable of supporting data streams within a CDMA platform. To this end, we (1) developed a state-of-the-art process for the chlorine and hydrogen dry etching of indium phosphide and indium-gallium-arsenic-phosphide; (2) fabricated low-loss waveguides (loss = 1.2 dB/cm); (3) fabricated AWGs with eight channels, a signal-to-noise ratio of 20 dB, and a channel spacing of 3.4 nm; and (4) integrated designs for AWGs with high-speed modulators.

Modeling Tools Development for the Analysis and Design of Photonic Integrated Circuits

Tiziana C. Bond
01-ERD-010

Abstract

Photonic integrated circuits (PICs) are poised to become the next generation of high-speed secure information systems and high-performance diagnostic instrumentation. Recent advances in material processing and device engineering are leading to the integration of several functions on single chips. Design tools are of paramount importance to reduce the cost and time of developing such optical chips. Commercial photonic design tools are currently narrowly

focused on the telecommunications industry. We are creating sophisticated, user-friendly PIC simulation tools by enabling the design of ultrafast switching devices for high-power laser diagnostics, high-density optical interconnects for advanced computing, and all-optical devices for secure communication networks.

Mission Relevance

This project is developing computational tools to support core areas of LLNL's national-security mission, including stockpile stewardship, laser science, surveillance, and high-bandwidth diagnostics that rely heavily on photonics for secure communication and remote sensing.

FY03 Results

We focused on novel lasers and nonlinear devices—such as logic gates—for all-optical digital circuits and signal processing, as well as highly sensitive sensors for analog and digital operations. To design such devices, we produced a suite of codes (with manuals) that range from zero-dimensional to one- and two-dimensional transient and steady-state simulations. We cross-checked them for consistency and validated them against literature. We investigated the feasibility of building PICs based on gain-quenched laser logic, in which light injected into the side of a laser should quench the gain enough to stop the device from lasing. Although it is possible to design devices that could switch states, the gain-quenching effect was not found to be large enough to make the devices practical.

Further Development of Wet-Etching Tools for Precision Optical Figuring

Michael C. Rushford

01-ERD-072

Abstract

This project developed a process to figure <1-mm-thick glass sheets for multiple applications. These include ultralight-weight space optics, disposable protection for expensive optical systems, phase plates that avoid self-focusing in high-power laser beams, and eyeglasses for removing or adding aberrations. Traditional processes rub glass with pressure and can break thin sheets. In addition, small tools based on dead reckoning need repeated measurement and figuring on multiple machines.

To figure these glass sheets, we are developing a two-dimensional (2-D) etcher that figures more quickly than a small tool by differentially heating an etchant using resistors on a circuit board. Figuring and measuring are conducted on one machine and in one location. Our tools figure the optic component without mechanical stress in parallel regions and with in-place metrology, for closed-loop processing, which saves time and therefore the cost of fabricating such thin optics.

Mission Relevance

Our project will develop new low-cost, wet-etch techniques for finishing high-quality, large-aperture optics. These techniques will support LLNL's stockpile-stewardship mission by enabling the fabrication of continuous-phase plates and disposable debris shields for high-power lasers. The technology developed in our project will enable the production of low-cost, high-quality optics for a wide range of other national-security applications.

FY03 Results

We developed an interferometer, a circuit board, and a wet-etch heater to flatten thin glass by the optical path difference (OPD) technique. After a 5- to 10-min warmup, the etchant was hand-poured onto the heated glass. After 30 min, the 1-cm thickness of etchant solution lost thermal contrast and needed to be removed by vacuum pump. Buoyancy-driven flows indicated a need to remove the etchant and reestablish the heating pattern more often than originally planed. However, in one run, 10 to 30 µm of glass was removed over a 25-cm-square area in 1 to 2 h; faster rates are also possible. Finishing took less than 8 h; alignment of the resistor with the interferometer matrix gave the best results. We also calculated the encircled energy for this glass and developed a high-energy-laser phase plate that made a 1-mm-diam blue focal spot that was within 200% of the energy loss allowed. This project demonstrated the dewedging and OPD flattening of borosilicate glass for large-scale lasers. Further issues to address include reengineering the glass and acid handling and measuring the glass OPD more quickly, which would help speed up the process.

Integrated Microfluidic Fuel Processor for Miniature Power Sources

Ravindra S. Upadhye 01-ERD-080

Abstract

The lifetime of today's unattended and remote sensors is limited by the life of the associated power sources. Our goal is to develop, as a longer-lasting power source, a microfluidic fuel processor producing sufficient hydrogen to feed a 500-mW fuel cell. Current batteries and fuel cells have limited energy density, about 500 W·hr/kg, whereas the energy density of liquid hydrocarbon fuels is about 10,000 W·hr/kg. The technical motivation of the project is to approach the energy densities of liquid hydrocarbons. To do so, we plan to develop catalytic microreactors, reform methanol at temperatures of about 300°C, and demonstrate integrated operation of the reformer with a proton-exchange membrane (PEM) fuel cell.

If the project is successful, we expect to develop an integrated microfabrication and microelectromechanical systems- (MEMS-) based microfluidic fuel processor, converting a solution of methanol and water to produce sufficient hydrogen to feed a fuel cell producing 500 mW of electrical power at about 50% efficiency. We also expect to develop a thermal management system to reject all the excess heat, while keeping the external surface at reasonably low temperatures. The significance of this is that such a device will be able to power unattended sensors for longer durations than is currently possible.

Mission Relevance

This project leverages LLNL's microfluidic device and microfabrication capability to create a prototype microfluidic fuel processor. The project will provide a new approach to microreactor technologies that promises to offer significantly longer-lasting power sources for the deployment of autonomous sensor networks for covert operations in support of homeland security and national-security missions.

FY03 Results

We exceeded, by an order of magnitude, our initial goal of producing a fuel processor to feed a fuel cell producing 500 mW of electrical power. We developed a microreformer that operates at about 300°C and produces more that 5 W equivalent of hydrogen. During the course of this work, we successfully synthesized our own catalyst and developed a method of depositing it on the microchannels of the microreactor. We have also demonstrated an integrated operation of the reformer and a PEM fuel cell. Finally, we developed an evacuated thermal package that allows high thermal gradients under a geometry that makes the device suitable for use in laptop computers.

Publications

Havstad, M. A. (2001). *Surface chemistry effects in finite element modeling*. Presented at 4th Int. Conf. on Modeling and Simulation of Microsystems, Mar. 19–21, 2001, Hilton Head, SC. UCRL-JC-140335.

Havstad, M. A. et al. (2003). "A microfluidic hydrogen generator for fuel cell applications." *Proc. Am. Inst. of Chem. Engineers Ann. Meet.* UCRL-JC-148043.

Morse, J. D. et al. (2003). "A methanol steam reforming micro reactor for proton exchange membrane micro fuel cell system." *Proc. 2003 Am. Soc. of Mech. Engineers Summer Meet.* UCRL-JC-150223.

High-Accuracy X-ray Imaging of High-Energy-Density Physics Targets

Walter W. Nederbragt 01-ERD-093

Abstract

High-energy-density experiments play an important role in corroborating the improved physics codes that underlie LLNL's stockpile stewardship mission. To conduct these experiments, state-of-the-art diagnostics for measuring experimental results, as well as improved fabrication and characterization of the target assemblies, are needed. Our project will pursue development of a capability to characterize target assemblies—specifically, an instrument capable of producing x-ray images at a micrometer resolution. This capability is important because target assembly characterization is the basis for verifying that targets were constructed correctly and because current methods for characterizing target assemblies, especially assemblies that comprise millimeter-size component parts with micrometer-size features, require radical improvements.

This project will have several phases. During the first phase, a prototype x-ray instrument that can image targets will be specified and designed. This design will be based on multilayer-coated Wölter imaging optics. The second phase will focus on building and testing the prototype system, while the final phase will analyze and design a Wölter-based imaging instrument.

Mission Relevance

Our project will develop a capability for characterizing target assemblies that will support LLNL's stockpile stewardship mission. This new capability will determine whether targets were assembled as specified and will lead to more accurate and complete target assembly models. In addition, this new imaging capability can be used to characterize other millimeter-size engineering devices that are becoming prevalent at both LLNL and within the commercial sector.

FY03 Results

In FY03, we replicated several test optics using low-cost mandrels and replicated one experimental-quality optic using our super-polished, high-accuracy mandrel. The replication technique coats a super-polished mandrel with reflective multilayer materials. The multilayers are covered with a 1- to 2-mm-thick layer of nickel to provide structural rigidity. The Wölter optic, comprising multilayers with a nickel substrate, is then separated from the mandrel, resulting in an optic with nearly the same figure and roughness as the mandrel. We also designed and partially constructed a prototype microscope that consists of the optic and target-positioning hardware, the imaging system, the x-ray source hardware, and the prototype body.

Publications

Nederbragt, W. (2003). *Fabrication of a precision mandrel for replicating Wölter x-ray optics*. Ann. Conf. of Am. Soc. for Precision Eng., Oct. 26–30, 2003, Portland, OR. UCRL-JC-155073.

Extremely High-Bandwidth Diamond Tool Axis for Weapons Physics Target Fabrication

Richard C. Montesanti
02-ERD-008

Abstract

This project advances the state-of-the-art in high-bandwidth, precise motion-control mechatronic devices, and will enable the fabrication of three-dimensional (3-D) surface features needed for high-energy-density physics (HEDP) experiments on large lasers. Currently, LLNL is limited in its ability to fabricate the surfaces needed to provide quantitative corroboration of the physics codes being developed for stockpile stewardship and inertial confinement fusion (ICF). Ultraprecision, single-point machining with the fast tool servo (FTS) to be developed in this project will make fabricating desired features possible. The project will deliver a 10-kHz-bandwidth, high-accuracy, high-stiffness servo axis designed specifically for these target-surface requirements.

If successful, this project will enable the fabrication of surfaces for HEDP experimental targets to higher accuracy requirements and faster production rates than currently available. Such high-bandwidth motion control will have application in the fabrication of precision optics, particularly applications that are nonaxisymmetric with short, spatial wavelength variations (e.g., gratings, Fresnel lens, and x-ray zone plates).

Mission Relevance

A state-of-the-art capability in high-frequency, ultraprecision fabrication at LLNL supports the Laboratory's stockpile stewardship mission and ICF by enabling the fabrication of 3-D surface features needed on target components for HEDP experiments.

FY03 Results

In FY03, we designed and built a new rotary FTS capable of following a 5-μm peak-to-valley sinusoidal surface at 2 kHz with a planned accuracy of 50 nm. We developed and integrated the necessary mechanical, electrical, electronic, and control systems needed, and experimentally demonstrated the intended small-signal bandwidth. The 2-kHz system was integrated with a diamond turning machine at the Massachusetts Institute of Technology during Spring 2003, and cutting tests were performed. Development of the 10-kHz system has begun, and we are converging on a design that merges the rotary mechanism and a higher force-density actuator into one component.

Proposed Work for FY04

FY04 work will focus on the 10-kHz system, which will be assembled by mid-FY04. Control-system research and development will take place in parallel and will be merged with the hardware to deliver a close-loop tested unit. The final step will integrate the FTS on a diamond turning machine and produce test pieces by the end of the year.

Publications

Montesanti, R. C. and D. L. Trumper. (2003). "High bandwidth short stroke rotary fast tool servo." *Proc. Am. Soc. Prec. Eng.* 2003 Meeting. UCRL-JC-155142

Montesanti, R. C. and D. L. Trumper. (2003). *High bandwidth short stroke rotary fast tool servo*. Presented to the Am. Soc. for Prec. Eng. 2003 Annual Meeting, Portland, OR. UCRL-PRES-200483.

Nanoscale Fabrication of Mesoscale Objects

Raymond P. Mariella, Jr. 02-ERD-014

Abstract

Laser ablation produces periodic roughness patterns on material surfaces. Several applications of laser ablation (e.g., fabricating laser targets for physics experiments) would benefit if surface roughness could be reduced. In this project, we plan to create a technique for fabricating and characterizing mesoscale (millimeter-size) objects with micrometer-scale contours and nanometer-scale precision by using femtosecond laser pulses and ion beam etching. This project integrates LLNL expertise in precision engineering, microtechnology, and nondestructive characterization along with special capabilities in short-pulse-laser–matter interactions and numerical simulation of material properties at multiple scales.

The immediate impact of our investigation will be a much better understanding of the chemical and physical processes that affect materials that are exposed to femtosecond laser pulses. We expect to develop the capability for fabricating and characterizing mesoscale objects using femtosecond laser pulses and ion-beam etching for applications such as fabricating laser targets and developing miniature fuels cell, remote sensors, and medical technologies.

Mission Relevance

These experiments will support the Laboratory's stockpile stewardship mission by providing data that can be used to corroborate models in the physics codes for the Advanced Simulation and Computing Program and by developing new capabilities for fabricating and characterizing mesoscale objects (e.g. laser targets) with nanoscale precision.

FY03 Results

The team successfully merged the one-dimensional (1-D) hydrodynamics code HYADES with state-of-the-art, 3-D molecular dynamics modeling, which resulted in a new capability to model the interactions of femtosecond laser pulses with solid surfaces. Calculations showed an unexpected phenomenon—the formation and expulsion of a thin, bubble-like film, at roughly 1 km/s, which occurs only for pulses that are slightly above the threshold for ablation, thus experimentally validating our model.

Proposed Work for FY04

Through both experiments and modeling, we will investigate the physical and chemical processes that occur when femtosecond laser pulses are used to sculpt micrometer-scale features in carbon aerogel substrates. The very high surface area of the carbon aerogel can be expected to facilitate the conversion of solid carbon into gas-phase species such as CO at a minimum of surface temperature. Since we expect that minimizing the surface temperature will also minimize any collateral damage to the surrounding aerogel, this could lead to an oxygen-assisted laser process for sculpting extremely friable (very-low-density) carbon aerogels

Concealed Threat Detection at Multiple Frames per Second

John T. Chang

02-ERD-061

Abstract

Personnel protecting special nuclear material and others responsible for homeland defense cannot easily detect hidden threats in real time, such as weapons under clothes or persons behind walls. A serious need exists for fast and accurate technologies to search for concealed threats. This project investigates the efficacy of real-time [10 to 30 frames per second (fps)] array imaging to rapidly detect hidden threats through clothing, smoke, fog, or walls. The project will (1) demonstrate that detection is measurably enhanced by real-time techniques, using simulated examples and statistical analyses; (2) develop methodologies of data processing, using novel array beam-forming hardware and software, that assist real-time human perception of a threat; and (3) design, assemble, and test a prototype array (radar or acoustic) that demonstrates detection capability. This research will produce important information on the feasibility of a system for the rapid, accurate, real-time detection of hidden threats.

Mission Relevance

This project supports the Laboratory's national security and homeland security missions by demonstrating the feasibility of a real-time array device can be developed to detect hidden threats.

FY03 Results

In FY03, we developed a numerical imaging algorithm and implemented it to study ultrawideband beam forming and steering. One computational simulator system, capable of generating impulse radar images of moving targets with arbitrary trajectories, was developed to evaluate the performance of the imaging system. This system facilitated the parametric study on the effects of transceiver element configurations. In collaboration with the University of California, Davis, we extracted information from noisy environments using psychometric characterizations. A laboratory system has been developed to enable laboratory imaging studies. Two records of invention were submitted; patent disclosures will be forthcoming.

Proposed Work for FY04

For FY04, our major milestones are to (1) document the evaluation of theoretical detection limits for concealed-threat detection through barriers; (2) integrate the benchtop array with field-programmable gate arrays at frame rates of up to 30 fps; (3) document the expected quantitative improvement in perception using human subject models; and (4) integrate a field-ready prototype for end-user evaluation.

Large-Aperture Diffraction Gratings, the Enabling Technology for High-Energy Petawatt Lasers

Jerald A. Britten

03-ERD-059

Abstract

The need for meter-scale, high-damage-threshold, pulse-compressor optics has been identified as the most critical hurdle in high-energy, petawatt-class (HEPW) laser technology, but the capability to produce such optics does not exist anywhere in the world. The objective of this project is to develop a state-of-the-art capability for producing meter-scale, multilayer, dielectric diffraction gratings capable of pulse compression for applications in HEPW laser systems. We plan to pattern dielectric coatings on monolithic substrates with grating lines approximately 0.2 µm wide, 0.7 µm high, and ~0.6 µm apart, uniformly over areas of 1 m × 800 mm. This effort requires the development of tooling and processes applicable to large, heavy substrates, involving laser-interference lithography, reactive ion-beam etching, wet-chemical processing, and critical-dimension metrology.

Pulses from HEPW lasers would enable the generation of critical, high-energy x-ray backlighters, access to new states of matter through isochoric heating, and studies of physics relevant to a full-scale demonstration of fast ignition. Successful completion of this project will place LLNL in the lead position worldwide for producing large-scale diffractive optics for HEPW lasers in the U.S. and abroad.

Mission Relevance

This capability will enable new and enhanced Stockpile Stewardship Program-relevant studies on the Omega laser, the Z-pinch facility, and future large-scale lasers for fusion research.

FY03 Results

The reactive ion-beam etcher became operational and was used to etch submicrometer-pitch grating structures into hard dielectric materials over apertures of 2 × 1 m. A Faraday-cup current probe was built to optimize beam current and source-emission uniformity. The process chemistries and operational modes for etching silicon dioxide, tantalum-203, and hard-baked photoresist were investigated. We determined etch rates and selectivity and found that 9 hr would be required to etch an 80-cm optic. Further tests indicated that we could achieve horizontal and vertical depth uncertainties of about 3% and 6%, respectively, over 80 cm, which is in agreement with expectations based on the ion-current distribution. We began etching multilayer gratings, and initial results were promising.

Proposed Work for FY04

We will address critical processes and technologies that either do not exist or will require modification to meet the stringent requirements of total size, uniformity over the required area, and, most importantly, resistance to laser-induced damage at HEPW power densities. These critical processes and technologies include (1) modifying the large-format holography station to enable high-aspect-ratio grating patterning at 1-m apertures; (2) qualifying the manufacture of witness grating with large-aperture multilayer coating capability (3) damage-testing parts made with large-aperture coating tools; and (4) building a tool and using it to implement processing chemistries that will safely and efficiently perform all wet-processing functions for 1-m-aperture gratings.

Publications

Britten, J. A. (2003). *Grating development at LLNL for short-pulse laser systems.* Presented at the 35th Symp. on Laser-Induced Damage in Optical Materials. UCRL-PRES-155368.

Britten, J. A. et al. (2003). "Enabling technology for fabrication of meter-scale gratings for high-energy petawatt lasers." *Proc. 3rd Intl. Conf. on Inertial Fusion Sci. and Appl.*, Sept. 7–12, 2003, Monterey, CA. UCRL-JC-153261.

Silicon Nanocrystal Laser

June Yu 03-FS-010

Abstract

The purpose of this feasibility study is to demonstrate the world's first all-silicon nanocrystal-based laser. A silicon laser, fabricated using the conventional silicon-manufacturing technologies, would bridge the crucial missing link that has made a completely silicon-based photonic system elusive so far. The fact that this laser can be both extremely small (a few tens of micrometers) and silicon based (Complimentary Metal Oxide Semiconductor compatible) enables arrays of such lasers to be integrated with standard integrated-circuit chip technology. This effort will encompass the design, fabrication, demonstration, and characterization of a silicon nanocrystal-based microlaser.

The ability to fabricate arrays of lasers on an integrated circuit chip makes possible a host of promising optical technologies and devices (e.g., optical interconnects, optical computing, and new types of optically based biological and chemical sensors). After the successful conclusion of this feasibility study, the next step will be to demonstrate an integrating silicon laser based on an electrical pumping scheme.

Mission Relevance

As the world faces increasing threats from chemical and biological weapons, advanced portable sensors offering increased sensitivity and selectivity are highly sought after. With the realization of an all-silicon-based laser, a host of new biological and chemical sensors can be integrated cheaply onto a compact silicon chip. Developing an all-silicon laser would support the Laboratory's missions in counterproliferation and homeland security.

FY03 Results

In the first few months of the project, we verified photolumincence from our implanted silicon nanocrystal materials, fabricated an array of Fabry-Perot cavities, and distributed feedback structures using the silicon nanocrystal laser material as the gain material. We performed laser gain measurements of these sample and characterized the resonant peak observed for the distributed feedback sample.

Proposed Work for FY04

In FY04, we will conclude our laser gain measurements and complete a scientific paper that documents our experimental results.

Materials Science and Technology

Material Strength at High Pressure

David H. Lassila

01-SI-004

Abstract

Several elements of LLNL's national-security mission require accurate computer code simulations of the plastic flow (i.e., strength) of metals under high-pressure conditions. This project will (1) examine the unit mechanisms associated with dislocation motion, (2) develop an experimentally validated dislocation dynamics (DD) simulation code capable of predicting the strength properties of body-centered-cubic (bcc) single crystals under high-pressure deformation conditions, and (3) develop new deformation experiments to measure material strength to validate DD simulation results. This work involves a close coupling of theory, modeling, simulations, and experimental work over a wide range of length scales and will establish a new paradigm in the understanding and development of strength properties of materials. In addition, we hope to achieve a new physical understanding of how bcc metals deform plastically under various loading conditions, including high pressure. These significant achievements will meet the need for accurate plasticity models and will be used to explore other materials phenomena.

Mission Relevance

This project builds on LLNL's core competency in high-pressure science and leverages the computing resources of the Advanced Simulation and Computing Program to extend Laboratory capabilities in materials science. This research will assist the Laboratory in performing its national-security mission by providing the needed computer code simulations of plastic flow under high-pressure conditions for the Stockpile Stewardship Program.

FY03 Results

In FY03, the effect of pressure on the mobility of screw dislocations was found to scale with the pressure dependence of the shear modulus. This rule was implemented into a dislocation dynamics code and allowed, for the first time, prediction of pressure-dependent strength properties by a DD simulation. Simulations of plastic deformation at up to 0.5% strain were achieved. Experimental techniques to validate DD simulations of deformation under high-pressure conditions were developed and used to produce high-quality information of the properties of bcc metals under high pressure. We have discovered that some long-standing plasticity laws do not to apply to molybdenum and tantalum crystals.

Publications

Crowhurst, J. C. and J. M. Zaug. (in press). "Surface acoustic waves in germanium single crystals." *App. Phys. Lett.* UCRL-JC-152926.

Crowhurst, J. C. et al. (2003). "Impulsive stimulated light scattering at high pressure—precise determination of elastic constants of opaque materials." *Intl. J. of High-Press. Res.* **23**, 373. UCRL-JC-142333.

Positrons and Positronium in Insulators

Philip A. Sterne

01-ERD-015

Abstract

A predictive theory of positrons in insulators is needed to answer questions about complicated positron behavior in materials. Our project addresses this need by using a combined theoretical and experimental approach to resolve outstanding problems in interpreting positron annihilation spectra on insulators. Our theoretical work will establish the basis for understanding positrons in insulators and will develop the tools to fully interpret these experiments. Specifically, we will determine the conditions under which positronium forms in insulators and will establish a theoretical foundation for the empirical relationship between positron lifetime and free volume that applies to a wide range of insulating materials.

Our objectives are to (1) develop new computational approaches for positrons in insulators using quantum Monte Carlo (QMC) techniques, (2) apply variational and diffusion QMC techniques to understand the behavior of positrons in insulators, (3) use path-integral Monte Carlo (PIMC) to establish a theoretical basis for positron lifetime measurements of open volumes in zeolites and other insulators, and (4) use existing LLNL positron capabilities in our experiments to provide detailed annihilation spectra on appropriate systems to compare with our developing theory. These developments will enhance our ability to utilize positron techniques in problems of importance to LLNL.

Mission Relevance

This project will enhance LLNL's capabilities in materials science and characterization. In particular, our experimental work has the potential to solve defect-related problems in insulators, with widespread applications for LLNL and DOE programs. If successful, our project will expand the power of positron spectroscopy in practical applications, including stockpile stewardship materials characterization, and an understanding of defects in optics for large-scale laser systems.

FY03 Results

We implemented variational and diffusion QMC methods for a positron in a material with an arbitrary number of electrons, validated this approach on molecular systems, and began calculations on periodic solids. We conducted PIMC simulations on faujasite to interpret our experimental results. The calculations showed that positronium is not confined to a single zeolite cage. We also developed a simplified theory for positron lifetimes in mesoscopic pores at finite temperatures, used the theory to interpret positron-based measurements of free volumes in polymers, and performed experiments on porous low-k dielectric material, demonstrating the effect of electron radiation on the positronium production in the material.

Publications

Asoka-Kumar, P. and P. A. Sterne. (2002). *Positron annihilation in insulating materials*. Presented at Intl. Conf. on Positron Studies of Defects in Semiconductors, Sept. 2002, Sendai, Japan. UCRL-JC-149558.

Bug, A. L. R. et al. (2003). *Positronium in solids: Computer simulation of pick-off and annihilation*. Presented at 13th Intl. Conf. on Positron Annihilation, Sept. 2003, Kyoto, Japan. UCRL-CONF-155634.

Hebert, K. R. et al. (in press). "Positron annihilation spectroscopy study of interfacial defects formed by anodic oxidation of aluminum." *J. Electrochermical Society.* UCRL-JC-152597.

Jenei, Z. et al. (2003). *Electron radiation effects on low-k dielectric materials.* 2003 IEEE Nuclear and Radiation Space Effects Conference, Monterey, CA, July 21–25, 2003. UCRL-CONF-155633.

Sterne, P. A. et al. (2003). "New theories for positrons in insulators." *Radiation Physics and Chemistry* **68**, 409. UCRL-JC-148530.

Shock Recovery of Organic Liquids: From the Origin of Life to the Defense of the Nation

Jennifer G. Blank 01-ERD-018

Abstract

Our nation's defense against ballistic missiles carrying chemical or biological agents, as well as investigating possible extraterrestrial origins of life's fundamental building blocks, require fundamental understanding of the fate of organic materials subjected to strong shock compression. However, little is known about the effect of strong shock on organic compounds. We are conducting experiments to determine the survivability of organic compounds delivered to Earth via cometary impact and chemical weapon (CW) agents released in ballistic-missile intercept scenarios. We are using LLNL gas guns to create the high-pressure conditions to shock small volumes of organic liquids, and are developing novel methods for recovering these shocked liquids for subsequent chemical analysis.

If successful, this project will (1) develop the capability for shocking liquids using hypervelocity gas guns at LLNL and for housing the liquids in leak-tight containers over the course of an experiment; (2) develop the capability of recovering these liquids, which will be under several atmospheres of pressure after an experiment; (3) model the pressure–temperature–time histories of the shocked liquids using two-dimensional hydrodynamical codes; (4) devise an analytical method for characterizing the organic compounds before and after a shock experiment; and (5) analyze shocked liquids, compare their chemistry with that of unshocked liquids, and evaluate the chemical change in the context of missile intercept and comet impact.

Mission Relevance

This project supports LLNL's national-security mission by investigating ways to defend the nation against ballistic missiles carrying chemical or biological agents. The chemical weapon simulant results will determine whether deadly payloads are rendered partially inert by shock and whether chemical species produced in an impact are good candidates for remote spectroscopic identification. We have observed polymerization among organic compounds in shocked aqueous liquids—in direct contradiction of the widely-held assumption that shock will reduce organic compounds to smaller, primary components. This fact has potentially important implications for models of an intercepted missile's effectiveness, i.e., kill radius.

FY03 Results

In FY03, we conducted shock experiments using two types of liquids: (1) amino acids in water, as a proxy comet, and (2) liquid diisopropyl methane phosphonate, diisoamyr ether, and tributyl phosphate, as analogs for CW-related materials. We demonstrated partial survival of all of the species of amino acids studied. Our most exciting result was the observation that the amino acids reacted to produce dipeptides and longer peptides—larger, biologically important molecules. We also observed color and viscosity changes in the CW simulants.

Direct Characterization of the Electronic Structure of Shocked and Heated Materials

Art J. Nelson

01-ERD-019

Abstract

Our project is developing and demonstrating a new pump-probe characterization capability in which x-ray-laser-induced time-of-flight photoelectron spectroscopy is used to probe the core-level and valence-band electronic structure of room-temperature bulk materials with picosecond time resolution. The LLNL Compact Multipulse Terawatt laser system, a compact tabletop x-ray laser source, provides the necessary high photon flux, high energy, monochromaticity, picosecond pulse duration, and coherence for probing ultrafast changes in chemical and electronic structures. Our novel research tool will enable studies of the surface chemical and electronic structure of semiconductor or organic materials undergoing fast laser excitation or shocking.

Direct time-resolved measurements of the chemical and electronic structure of the material will be performed to provide insight into the dynamic process and final states. This technique offers a unique diagnostic of phase changes in materials and chemical reactions with a picosecond time resolution. In addition, the x-ray probe allows us to further explore the physics of laser–matter interactions. We are already recording valence band and core-level spectra for transition-metal surfaces. In situ sputter etching with argon ions at an angle of incidence of 30 deg. is used to improve the surface purity and consequently increase core-level and valence-band photoemission intensity.

Mission Relevance

This powerful new technique makes it possible to probe reaction dynamics and changes of local order on surfaces on their fundamental time scales. The ability to characterize the chemical and electronic structure of materials under extreme conditions will further efforts in laser–material interaction and high-pressure shock physics in support of the Laboratory's stockpile stewardship.

FY03 Results

For the past 3 yr, we concentrated on developing single-shot, x-ray-laser-induced time-of-flight photoelectron emission spectroscopy in a pump-probe investigation of changes in the electronic structure of materials undergoing ultrafast laser heating. We have successfully validated this technique and have observed valence band and core-level emission from transition metal surfaces. In addition to core-level emission, d-state emission was observed at the valence band maximum corresponding to direct transitions from d-like occupied bands to unoccupied bands above the vacuum energy. This was accomplished with a University of Nevada, Las Vegas-designed electron time-of flight analyzer with a multichannel plate detector assembly having improved time resolution.

Publications

Nelson, A. J. et al. (2003). "X-ray-laser-induced time-of-flight photoelectron spectroscopy." *Proc. SPIE* **5197**, 168. UCRL-JC-150282.

Microstructural Origins of Dynamic Fracture in Ductile Metals

James F. Belak

01-ERD-022

Abstract

This project involves an integrated experimental and modeling effort on dynamic fracture in ductile metals. Our project focuses on the origins of dynamic fracture in ductile metals—in particular, the nucleation and growth of microscopic voids in crystalline and defective materials—and aims to measure with increased precision the damage produced during spallation fracture in ductile metals. Dynamic gas-gun recovery experiments were used to generate incipient spall damage, which was characterized using advanced three-dimensional (3-D) x-ray tomography and traditional 2-D microscopy. In addition to single-crystal aluminum and vanadium, copper samples with engineered inclusions were shock recovered and analyzed using transmission electron microscopy (TEM) to analyze the plastically deformed zone.

Our expected results are (1) using the LLNL light gas gun to conduct systematic shock loading and soft recovery, which are essential to controlling the stress transient and capturing the incipiently damaged state; (2) concurrent free-surface velocity wave profile and soft recovery to correlate pull-back velocity to the damaged state; (3) synchrotron-based 3-D x-ray tomography of damaged soft-recovery samples to determine the full 3-D structure of evolving damage inside the ductile metal; and (4) 2-D microscopy of sliced soft-recovery samples to correlate traditional 2-D observations of damage with the full 3-D structure and to determine the plastically deformed zone surrounding incipiently grown voids.

Mission Relevance

This project is developing a paradigm to measure the full 3-D distribution of porosity produced during spallation fracture from recovered incipiently spalled ductile metals. In addition to applying the paradigm to heavier metals, these data are being used to guide the development of physically based models of material failure and ultimately will enhance our ability to predict dynamic material failure for the Laboratory's stockpile stewardship mission.

FY03 Results

Having established our paradigm to concurrently soft-recover and measure free-surface velocity during shock loading, we conducted further experiments on single-crystal aluminum. After the 3-D distribution of porosity was measured with synchrotron-based x-ray tomography, the samples were sectioned and observed using 2-D microscopy. Etching techniques were used to reveal the dislocation structure at the surface. We have, for the first time, quantitatively measured the size of the plastically deformed zone as a function of void size. Despite several attempts, we have not been able to retrieve high-quality TEM samples from the plastic zone. We also completed a series of gas-gun recovery experiments on the body-centered-cubic metal vanadium. These samples are being analyzed for the 3-D x-ray tomography.

Publications

Belak, J. et al. (2003). T*he plastically deformed zone surrounding voids in single crystal aluminum from incipient spallation fracture.* Presented at Ann. Am. Phys. Soc. March Meet., Mar. 2003, Austin, TX. UCRL-JC-150828 Abs.

Belak, J. et al. (2003). "The structure and distribution of voids in spall fracture from 3-D x-ray tomography and 2-D microscopy." *Proc. 13th Am. Phys. Soc. Topical Conf. on Shock Compression of Condensed Matter.* UCRL-JC-149186 Abs Rev.

Belak, J. et al. (2003). *The structure and distribution of voids in spall fracture from 3-D x-ray tomography and 2d microscopy.* Presented at 132nd Ann. Meet. of the Minerals, Metals, and Matls. Soc., Mar. 2003, San Diego, CA. UCRL-JC-149186 Abs Rev.

Budil, K. S. et al. (2003). "Laser-based experiments investigating the dynamics of material failure and fracture." *Proc. 13th Am. Phys. Soc. Topical Conf. on Shock Compression of Condensed Matter.* UCRL-JC-152226 Abs.

Budil, K. S. et al. (2003). "Laser-based experiments investigating the dynamics of material failure." *Proc. 3rd Intl. Conf. Inertial Fusion Sci. and Applications.* UCRL-JC-152225 Abs.

Kalantar, D. H. et al. (in press). "High-pressure, high-strain-rate lattice response of shocked materials." *Physics of Plasmas.* UCRL-JC-149256 Rev.

Kalantar, D. H. et al. (2003). "Large-angle in situ diffraction measurements of shocked single crystals." *Proc. 13th Am. Phys. Soc. Topical Conf. on Shock Compression of Condensed Matter.* UCRL-JC-152529 Abs.

Rudd, R. E., E. T. Seppala, and J. Belak. (2003). *Plastic deformation associated with void growth: multiscale modeling.* Presented at Multiscale Modeling and Simulation of Material Behavior, Jul. 2003, Albuquerque, NM. UCRL-JC-151628 Abs.

Seppala, E. T., J. Belak, and R. E. Rudd. (2003). *Molecular dynamics study of void growth and dislocations in dynamic fracture of fcc and bcc metals.* Presented at Plasticity 2003, Jul. 2003, Quebec, Canada. UCRL-JC-151375.

Shear Localization and Fracture in Shocked Metals

Geoffrey H. Campbell 01-ERD-026

Abstract

Shocked metals have a high density of defects and are very susceptible to strain localization preceding failure, making such materials highly relevant to LLNL's national-security programs. Understanding the interplay between the damage mechanisms and local loading that results in crack growth and failure requires a fracture-mechanics approach to identify and characterize phenomena within the crack-tip process zone.

This project is studying strain localization and fracture in shocked metals. Failure in shocked metals will be characterized using interrupted mechanical tests to investigate the physical phenomena (e.g., void nucleation, growth, linkage, and shear localization) that occur in the failure-process zone. Further information will be obtained by using optical and scanning electron microscopy to quantify void-nucleation sites with growth and linkage rates and the influence of shear localization. The fracture energy will be measured with novel specimen configurations that have been modeled with simulations of the specimen dynamics. The project aims for this understanding so that predictive, physics-based failure and fracture models can be developed by incorporating microstructure and failure models into Laboratory simulation codes.

Mission Relevance

The experimental investigation, modeling, and simulation of metal dynamics in this project supports stockpile stewardship. The development of models incorporating the physical influence of material microstructure on dynamic response is a key component of this effort. The knowledge gained in this project on the changes induced in metal microstructure by shocks and how those changes affect subsequent failure will help develop models to simulate this process.

FY03 Results

To optimize the experimental setup for high-loading-rate fracture tests, three-dimensional dynamic calculations were performed with the finite element code DYNA3D. These calculations considered the planned geometry, a double-edge notch beam, under impulse-loading conditions similar to those expected in the split Hopkinson bar test facility. Specifically, the growth of a singularity-dominated field was observed, and this field was found to match that expected from asymptotic analytical solutions. Further, plastic zone development was monitored throughout the loading transient. These results were used to identify the most useful locations for strain gages needed to assess loading conditions during the fracture test.

Publications

Campbell, G. H. and M. Kumar. (2003). "Effect of laser shock processing on the microstructure and mechanical properties of pure Cu." *Zeitschrift für Metallkunde* **94**, 329. UCRL-JC-150085.

Metastability and Delta-Phase Retention in Plutonium Alloys

Adam J. Schwartz 01-ERD-029

Abstract

Understanding the martensitic phase transformation in the plutonium–gallium (Pu–Ga) system is a challenge due in large part to a density change of approximately 20% that makes stress and plastic accommodation of central importance. In addition, composition, microstructure, stress, and radiation effects all play crucial roles, with resulting technological implications in a variety of defense and energy systems. In our project, a reference alloy will be fabricated with fully homogenized or highly cored microstructures. Transmission electron microscopy (TEM), x-ray diffraction, and resistometry will then be used to study nucleation and growth of the transformation product. Three-dimensional phase field modeling will combine thermodynamics and mechanical properties to simulate the width of the transformation hysteresis. This project will provide a deeper understanding of the role of microstructure, local elastic constraint, and plastic deformation in the stabilization of the low-temperature delta-phase Pu structure.

Mission Relevance

Our project supports DOE missions in national security and energy. In particular, the project supports LLNL's stockpile stewardship mission by laying a basic-science foundation for understanding the phase stability of stockpile materials. An important additional benefit of this work is the development of the scientific expertise and workforce to support future stockpile efforts. Solving the challenging problems of radioactive actinide metals requires training beyond what is available from institutions of higher learning. This initiative will provide the skills, technology, and personnel required to meet long-term strategic and tactical needs relevant to this scientifically interesting and challenging metal system.

FY03 Results

Work completed in FY03 includes (1) continued measurements of the hysteresis of the delta–alpha phase transformation in a Pu–Ga alloy using resistometry and differential scanning calorimetry (DSC), (2) quantifying heats of transformation with DSC, (3) observation of periodic spikes in the transformation and the development of a model for these spikes, (4) establishment of annealing procedures that enable us to thermally cycle a single sample repeatedly with reproducible results, (5) detailed transmission electron microscopy characterization of the crystallography and morphology of the transformation product, and (6) completing finite-element modeling of thermal hysteresis in plutonium and in the palladium hydride–palladium system (for validation of the model). The advances made this year improved our fundamental understanding of the effects of stress and plastic accommodation on controlling the crystallography and morphology of the phase-transformation product and the width of the hysteresis.

Publications

Blobaum, K. et al. (2003). *Investigating the delta/alpha′ martensitic phase transformation in Pu–Ga alloys.* Presented at Plutonium Futures: The Science Conference, July 2003, Albuquerque, NM. UCRL-PRES-149588.

Blobaum, K. J. et al. (2003). *Investigating the delta/alpha′ phase transformation in Pu–Ga alloys.* Presented at Materials Research Society Fall Meeting, Dec. 2003, Boston, MA. UCRL-JC-153511 Abs.

Blobaum, K. et al. (2002). *Investigating the delta/alpha′ martensitic phase transformation in Pu–Ga alloys.* Presented at 2nd Intl. Workshop on Spin and Orbital Magnetism of Actinides, Oct. 2002, Berkeley, CA. UCRL-MI-149588.

Haslam, J. J. et al. (2003). *Investigation of differences in morphology and transformation mechanisms of alpha′ formed from delta plutonium at low temperatures.* Presented at TMS Annual Meeting, March 2003, San Diego, CA. UCRL-JC-149061 Abs.

Krenn, C. R. (in press). “Continuum modeling of transformation hysteresis in a metal hydride system.” *Modeling Simul. Mater. Sci. Eng.* UCRL-JC-151820.

Krenn, C. R., A. J. Schwartz, and W. G. Wolfer. (2003). *Continuum modeling of hysteresis during solid-state phase transformations.* Presented at 7th US National Congress on Computational Mechanics, July 2003, Albuquerque, NM. UCRL-JC-151820.

Krenn, C. R., M. A. Wall, and A. J. Schwartz. (2003). *Transformation crystallography and plasticity of the delta to alpha′ transformation in plutonium alloys.* Presented at Materials Research Society Fall Meeting, Dec. 2003, Boston, MA. UCRL-JC-143514 Abs.

Krenn, C. R. et al. (2003). *Effects of plasticity and local solute ordering on the delta to alpha transformation in gallium-stabilized plutonium alloys.* Presented at TMS Annual Meeting, March 2003, San Diego, CA. UCRL-JC-149178 Abs.

Krenn, C. R. et al. (2003). *Effects of local solute ordering and plasticity on the delta to alpha transformation in gallium stabilized plutonium alloys.* Presented at Plutonium Futures: The Science Conference, July 2003, Albuquerque, NM. UCRL-JC-151541.

Krenn, C. R. et al. (2003). *Phase field modeling of metal hydride growth in a plastically deforming matrix.* Presented at TMS Annual Meeting, March 2003, San Diego, CA. UCRL-JC-149179 Abs.

Krenn, C. R. et al. (2002). *Effects of local solute ordering and plasticity on the delta to alpha transformation in gallium-stabilized plutonium alloys.* Presented at 2nd Intl. Workshop on Spin and Orbital Magnetism of Actinides, Oct. 2002, Berkeley, CA. UCRL-JC-149837-ABS.

Nelson, E. et al. (2003). *Local structure and vibrational properties of (alpha)'-Pu martensite and Ga-stabilized (delta)-Pu.* Presented at Plutonium Futures: The Science Conference, July 2003, Albuquerque, NM. UCRL-JC-149837-EXT Abs.

Nelson, E. J. et al. (2003). "Local structure and vibrational properties of alpha'-Pu martensite in Ga-stabilized delta' Pu." *Phys. Rev. B* **67**, 224206. UCRL-JC-1000752.

Nelson, E. et al. (2002). *Multi-scale structural study of the delta to alpha' transformation in a Pu–Ga alloy.* Presented at 2nd Intl. Workshop on Spin and Orbital Magnetism of Actinides, Oct. 2002, Berkeley, CA. UCRL-PRES-149687.

Thermodynamics and Structure of Plutonium Alloys

Patrick G. Allen 01-ERD-030

Abstract

The role of iron (Fe) and nickel (Ni) impurities in plutonium–gallium (Pu–Ga) alloys is poorly understood. These impurities induce secondary phase formation in the nominal Pu–Ga structure, which has implications for components made from Pu–Ga alloys. The project will investigate the role of impurities on the thermodynamics, secondary phase formation, and local structural effects in Pu alloys. Our goal is to develop a robust predictive tool for binary (Pu–Ga) and ternary (Pu–Ga–Fe) alloys of Pu. Alloy syntheses and characterizations will provide structures, compositions, and energetics that can confirm locations of boundaries between stability regions in Pu–Ga binary phase diagrams.

Our approach includes a theoretical method supplemented by experiments. Theory calculations involve coupling Gibbs free-energy minimization for alloy systems with other forms of thermochemical inputs to model phase diagrams. Experimentally, we are synthesizing Pu–Ga (Fe, Ni) alloys by melting and then characterizing alloy structure by x-ray diffraction spectroscopy, electron microscopy, and scanning calorimetry. The experimental results are used to couple with and validate our theoretical simulations. The information collected in this study will then be used to model the long-term stability of Pu–Ga and other alloys. The information will also be used to construct binary and new ternary phase diagrams, which will serve as a foundation for understanding the physical and mechanical properties. The long-term benefit is development of state-of-the-art experimental and theoretical tools for studying nuclear materials.

Mission Relevance

This work supports the LLNL national-security mission area of nuclear science, including actinide (uranium and Pu) metallurgy and science-based stockpile stewardship. This research will form the basis for future predictive work to assess the impact of impurities in Pu–Ga alloys on mechanical behavior. In addition, the results of this project will provide a detailed scientific foundation for understanding important aspects of metal dynamics and aging. The resulting ability to predict thermochemical and dynamic properties of multicomponent uranium and Pu alloys associated with different fabrication processes is of considerable interest to the Stockpile Stewardship Program.

FY03 Results

During FY2003, our theoretical work yielded a comprehensive thermochemical database for the Pu–Ga–Fe and related ternary systems. We made important modifications to the binary and ternary enegetics using new calorimetry data, expanded our simulations of the ternary Pu–Ga–Fe system, and calculated Pu–Ga martensitic transformation kinetics, providing new mechanistic insight into the nature of Pu phase transformations. With our new melting capability, we synthesized our first ternary Pu–Ga–Fe alloys and initiated characterization of these alloys to validate theory. We also performed more detailed studies of the Pu_3Ga and Pu_6Fe compounds using x-ray absorption spectroscopy. The results helped us to develop a model for local bonding effects for Fe and Ga sites in these alloys.

Publications

Allen, P. G. et al. (2002). "Vibrational properties of Ga-stabilized delta-Pu by extended x-ray absorption fine structure." *Physical Review B* **65**, 214107. UCRL-JC-146597 Abs.

Nelson, E. J., P. G. Allen, and C.H. Booth. (2002). *Studies of vibrational properties in actinide metals by extended x-ray absorption fine structure*. Presented at APS March Meeting, March 2002, Indianapolis, IN. UCRL-JC-146597 Abs.

Nelson, E. J. et al. (2003). "Local structure and vibrational properties of (alpha)'-Pu martensite and Ga-stabilized (delta)-Pu." *Physical Review B* **67**, 224206. UCRL-JC-151541.

High-Pressure, High-Strain-Rate Materials Effects

Daniel H. Kalantar 01-ERD-031

Abstract

This project investigated material properties at high pressures and high strain rates using dynamic x-ray diffraction (XRD) to study the response of a single-crystal silicon (Si), copper (Cu), and iron (Fe) lattice under extreme shock loading. Shocked samples were recovered to characterize the residual defect density in the material, including the competing effects of twinning and stacking faults. These different mechanisms were studied by shock-loading single-crystal Cu along different directions and by varying the atomic fraction of aluminum (Al) in a Cu–Al single-crystal alloy. Molecular dynamics (MD) simulations were carried out to study the formation and propagation of dislocations due to ramp and shock loading to demonstrate perpendicular relaxation of the lattice as shown by the XRD measurements.

As a result, we developed a large-angle diffraction technique to record the shock compression of multiple lattice planes simultaneously. Large MD simulations provided shock Hugoniot results consistent with experimental observations, as well as deformation such as twinning. Very large MD simulations with ramp pressure loading showed a relaxation of the lattice over approximately 100 ps, indicating lattice compression in the direction perpendicular to the shock. This is consistent with the experimental diffraction results that showed 3% compression in each direction in shocked Cu.

Mission Relevance

The experimental technique that was developed for recording the lattice response due to shock loading can be used to study a range of different materials under extreme conditions relevant to stockpile stewardship. These experimental results aid the development of improved models for describing the properties of shocked materials. The large MD simulations of shocked materials represent advancement of the techniques to calculate the response of materials to shock loading.

FY03 Results

We developed the large-angle diffraction technique and procedures for analyzing and interpreting the resultant data. Progress was made in using a hydrodynamics code to model both the free-surface velocity measurements and lattice compression measurements of shocked single-crystal Si. Very large MD simulations were conducted to study the effects of pre-existing defects and to observe the relaxation of the lattice perpendicular to the shock direction. This relaxation of the lattice was observed for the first time in an MD simulation by using ramp pressure loading and by conducting the simulations at late times (100 ps).

Publications

Bringa, E. M. et al. (2003). "Metals far from equilibrium: from shocks to radiation damage." *Nucl. Instr. and Meth. Phys. Res. B* **202**, 56. UCRL-JC-148559.

Kalantar, D. H. et al. (2003). "High-pressure, high-strain-rate lattice response of shocked materials." *Physics of Plasmas* **10**, 1569. UCRL-JC-149256.

Kalantar, D. H. et al. (2003). "Multiple film plane diagnostic for shocked lattice measurements." *Review of Scientific Instruments* **74**, 1929. UCRL-JC-147825.

Schneider, M. S. et al. (in press). "Laser-induced shock compression of copper: Orientation and pressure decay effects." *Met. Trans.* UCRL-JRNL-155630.

Modeling and Characterization of Recompressed Damaged Materials

Richard C. Becker

01-ERD-032

Abstract

Ductile materials subjected to impact or high-explosives loading can develop spall or significant internal damage. Additional deformation, often involving recompression, can continue after the onset of damage. Current material models do not accurately describe the behavior of recompressed damaged materials. However, an accurate description is necessary to reliably simulate the deformation, energy dissipation, and density of these materials.

The goals of this project are to (1) develop experimental techniques and an experimentally based model for the recompression of damaged materials and (2) implement this model in ALE3D, an Advanced Simulation and Computing Program code. Our project will use a gas gun and lasers to recompress samples at high strain rates and pressures. The gas-gun experiments will compress the spalled sample against an anvil and use a sabot to drive recompression. The laser experiments will employ two pulses, one on each side of the specimen. The first pulse will create spall damage, and the second, delayed pulse will close it. A model will then be synthesized to fit the data. The model will be combined with an existing ductile failure model and will be implemented in ALE3D.

Mission Relevance

The most significant tangible benefit to NNSA from this program is identification of a model describing material behavior during recompression. Improved models are necessary in high-fidelity simulations supporting NNSA's stockpile stewardship mission and in support of collaborations with the DoD. Understanding failure processes and behavior following fracture has also been a goal of the materials science community for more than half a century and is of great scientific and practical interest; this work therefore supports LLNL's basic-science mission.

FY03 Results

Our experiments previously demonstrated that spall damage could not be closed under low confining pressures obtainable in a split Hopkinson bar. Experiments in FY03 used a gas gun and lasers to provide higher stress triaxialities. Microstructures from material recompressed at a high strain rate using lasers show jetting and other signs of turbulent flow. The gas gun specimens show few signs of pre-existing damage. The material etches differently, but the specimen otherwise appears pristine. Micromechanical simulations of void growth and recompression indicate that voids shrink and collapse without leaving a residual crack. Loading histories from these simulations suggested modifications to the Gurson to account for recompression. The modified model is available in the development version of ALE3D. Appropriate parameters can be determined from experiments on relevant materials.

Double-Shell Target Design and Experiments at Omega: Nonlinear Mix Studies for Stockpile Stewardship

Peter A. Amendt

01-ERD-033

Abstract

Large laser systems, such as Omega at the University of Rochester, represent unique test beds with various applications, including science-based stockpile stewardship. Currently, many laser-target experiments at Omega employ single-shell capsules designed to have relatively low levels of hydrodynamic instability. This proposal explores the development of a double-shell capsule, which has the advantage of not requiring elaborate methods for controlling target entropy. The research value of double-shell targets lies in their utility as a robust test bed for exploring classical Rayleigh-Taylor growth. The proposed computational and experimental research intends to explore the use of double-shell targets as a potentially robust test bed for stockpile-stewardship-related issues.

We anticipate achieving a comprehensive understanding of double shell behavior under ignition conditions. Theoretical efforts to control and reduce Rayleigh-Taylor instability to levels required for demonstrating ignition will be proposed, followed by experimental studies on existing high-energy-density laser facilities for validation. Such a demonstration of instability control would provide a strong impetus for using laser-driven double-shell targets in support of future work relevant to science-based stockpile stewardship.

Mission Relevance

Our project will leverage LLNL expertise in laser-target design, weapons physics, and high-energy-density experiments in support of LLNL's stockpile-stewardship mission. In particular, our research will assess the feasibility of using double-shell capsules as a test bed for experiments relevant to stockpile stewardship.

FY03 Results

In FY03, we achieved spectacular success with double-shell research. First, experimental work with the Omega laser demonstrated unprecedented double-shell target performance under ignition-like conditions. Second, sophisticated multimode simulations were carried out and identified a previously unknown instability scenario that could have jeopardized double-shell ignition. These simulations were further implemented to dramatically improve the robustness of double shells to Rayleigh-Taylor instability growth via design modifications. Third, we developed a new design that avoids the customary onset of turbulence in double shells, thereby creating the prospect of direct numerical simulations of an igniting double shell. Based on the results of this project, LLNL is further developing double-shell ignition designs and the necessary materials-science techniques for building a prototype target by FY10.

Publications

Amendt, P. et al. (2003). *Hohlraum-driven ignition-like double-shell implosion experiments on Omega: analysis and interpretation*. Presented at Conf. on Inertial Fusion Sciences and Applications, Sept. 2003, Monterey, CA. UCRL-JC-151998.

Amendt, P. et al. (2003). "Modified Bell-Plesset effect with compressibility: Application to double-shell ignition target designs." P*hysics of Plasmas* **10**, 820. UCRL-JC-148944.

Milovich, J. L. et al. (in press). "Multi-mode short-wavelength perturbation growth studies for the National Ignition Facility double-shell ignition target designs." *Physics of Plasmas*. UCRL-JC-154498.

The Deformation-DIA: A Novel Apparatus for Measuring the Strength of Materials at High Strain to Pressures at Elevated Temperature

William B. Durham

01-ERD-035

Abstract

The primary focus of this project is to measure the yield strength of body-centered-cubic (bcc) metals at separable conditions of temperature, pressure, and plastic strain. These conditions, hitherto impossible to attain in the laboratory, have potentially important application to stockpile stewardship. Using the new apparatus that is the heart of this work, the Deformation-DIA (D-DIA), we deform samples of molybdenum (Mo) and tantalum (Ta) to strains approaching 1 at pressures to 15 GPa and temperatures to 1700 K. Measurements in the D-DIA require state-of the-art synchrotron x-ray sources, which are available through collaborators at the National Synchrotron Light Source (NSLS) at Brookhaven National Laboratory and at the Advanced Photon Source (APS) at Argonne National Laboratory.

The goal of this project is to use the D-DIA to produce actual measurements of the yield strength of bcc metals at high pressures to demonstrate that the D-DIA is a viable instrument for measuring yield properties and is therefore uniquely suited to stockpile stewardship needs.

Mission Relevance

The D-DIA will be used to measure the strength of metals such as Mo and Ta and ultimately actinides and other metals. The improved knowledge of strength of materials will be a significant step forward for the Stockpile Stewardship Program, in support of the national security mission.

FY03 Results

We reached our stated goal early in FY03 by making the first quantitative measurements of the yield strength of Ta at a controlled pressure of 6 GPa. We have since carried out a series of experiments, concentrating on Ta to refine experimental techniques and characterize our measurement uncertainties, and have produced a number of stress–strain curves for the material. Conditions have covered pressures from 2 to 8 GPa, strains to 0.7, temperatures to 1500 K, and strain rates from 1^{-6} to 3^{-5}/s. Instrument behavior has been superb. There have been no significant difficulties with mechanical control or with measurement in the x-ray beam. We have begun developing finite-element modeling as a tool for understanding measurement error. The project has been a huge success and the instrument is now ready for stockpile stewardship applications.

Designer Diamond Anvils for Novel High-Pressure Experiments: Magnetic Susceptibility Experiments on Actinides to Multimegabar Pressures

Samuel T. Weir 01-ERD-036

Abstract

The goal of this project was to develop a new, advanced, ultrahigh-pressure diagnostic technology—designer diamond anvils—and to study the physics of f-electrons in the actinides under extremely high (multimegabar) pressures. The high-pressure electronic behavior of the actinides and lanthanides presents challenges because the localized f-electrons in many of these metals can undergo pressure-induced transitions in which the f-electrons delocalize, resulting in sudden changes in unit cell volume and volume compressibility. Understanding these transitions is important for refining theoretical models and for developing more accurate high-pressure phase diagrams and equations of state for the actinides.

In this project, we expect to gain deeper insight into the nature of the electronic changes in these elements under conditions of extreme compression. In particular, we will gain experimental data that will elucidate the changing nature of the f-electrons in many of these metals as these electrons either gradually or suddenly delocalize under pressure. A better understanding of f-electron transitions will, in turn, help develop better high-pressure phase diagrams and equations of state for the actinides.

Mission Relevance

Measurements of the magnetic susceptibility of actinides will lead to an improved understanding of electronic properties and hence the high-pressure phases of these materials, which are of critical importance to the Stockpile Stewardship Program (SSP).

FY03 Results

Through this LDRD project, we have successfully developed the capability for performing sensitive magnetic susceptibility experiments to multimegabar pressures using encapsulated thin-film microcircuits fabricated on the tips of diamond anvils. This new class of multiple-loop designer anvils is in high-yield production with a 95% fabrication success rate. We have studied the magnetic properties of gadolinium to high pressures, observing the disappearance of ferromagnetic ordering at 6 GPa. We now have an apparatus that is capable of performing highly sensitive magnetic experiments on uranium (U) and plutonium (Pu) samples to megabar pressures. These designer anvils will be applied to SSP efforts to improve the accuracies of the high-pressure phase diagrams and equations of state of U and Pu.

Publications

Jackson D. D. et al. (2003). “Magnetic susceptibility measurements at high pressure using designer diamond anvils.” *Review of Scientific Instruments* **74**, 2467. UCRL-JC-149345.

Resolving Nuclear Reactor Lifetime Extension Questions: A Combined Multiscale-Modeling and Positron-Characterization Approach

Arthur B. Denison

01-ERD-042

Abstract

In deciding whether to extend the life of nuclear power plants, a key issue is the ability to predict the extent of irradiation embrittlement of the reactor pressure vessel (RPV). Our approach to predicting irradiation embrittlement combines predictive, physically based multiscale modeling with novel positron annihilation spectroscopy (PAS). By using a combination of modeling and experiment, we will identify the elemental composition and atomic structure of the nanometer copper–manganese–nickel (Cu–Mn–Ni) precipitates and the subnanometer defect cluster complexes responsible for embrittlement. Finally, we will use three-dimensional dislocation dynamics to provide quantitative understanding of the hardening and embrittlement produced by these features.

This project will demonstrate the capability of PAS to characterize the structure and chemical composition of nanometer and subnanometer features and will experimentally validate a multiscale modeling methodology for the prediction of irradiation effects on the properties of multicomponent steels. In addition, complete identification of the nanostructural features responsible for embrittlement in RPV steels will provide regulators with hard scientific data and understanding required for improved regulatory guides to determine the safe operating lifetime of RPVs.

Mission Relevance

This project will expand LLNL’s capabilities in the energy, environmental, and nuclear materials and systems areas in support of LLNL’s mission in energy security.

FY03 Results

Positron annihilation spectroscopy measurements were performed on neutron-irradiated and post-irradiation annealed Fe–Cu–Mn–Ni model alloys and surveillance specimens from nuclear power plants. The choice of materials and irradiation conditions was closely coordinated with our collaborator at the University of California, Santa Barbara, to ensure that the materials are also characterized by other methods. Using the same set of material compositions and irradiation conditions, we performed simultaneous multiscale modeling of microstructural evolution under irradiation to provide a nanostructural feature database necessary for interpreting the experiments. Dislocation-dynamics simulations provide a quantitative prediction of irradiation hardening and embrittlement.

Publications

Glade, S. C. et al. (2003). *Characterization of nanostructural features in irradiated reactor pressure vessel model alloys*. TMS Spring 2003 Meet., Mar. 2–6, 2003, San Diego, CA. UCRL-PRES-145544 Rev.

Glade, S. C. et al. "Positron annihilation spectroscopy and small-angle neutron scattering characterization of the effect of Mn on the nanostructural features formed in irradiated Fe-Cu-Mn alloys." *Philosophical Magazine A*. UCRL-JC-152567

Glade, S. C. et al. "Positron annihilation spectroscopy of nanostructural features in model reactor pressure vessel steels." *Materials Science Forum*. UCRL-JC-154520.

Glade, S. C. et al. (2003). *Positron annihilation spectroscopy and small angle neutron scattering characterization of nanostructural features in irradiated Fe-Cu-Mn alloys*. 13th Intl. Conf. on Positron Annihilation, Sept. 7–12, 2003, Kyoto, Japan. UCRL-PRES-145544 Rev.

Nanoscience and Nanotechnology in Nonproliferation Applications

John G. Reynolds

01-ERD-054

Project Description

The objective of this project is to develop new nanostructured, nonporous materials for the real-time and remote detection of chemical and biological weapons (CW and BW) of mass destruction (WMD). The intelligence community needs new, more targeted tools for detecting WMD. In particular, the need exists for organic and inorganic coatings that function at parts-per-million to parts-per-trillion detection levels and that are highly selective and specific for effluent target signatures from production facilities. These materials will be designed as compound or compound class specific through surface and chemical modification of silica sol-gels and porous silicon.

These new materials will lead to the development of new WMD detection systems that are more sensitive and flexible than current systems. The greatest impact will be the ability to target specific compounds that are either toxic or are derivatives of toxic materials in environmentally dilute conditions. This will be important for security and military applications.

Mission Relevance

This project supports LLNL's national-security mission by developing capabilities in CW and BW nonproliferation.

FY03 Results

In FY03, we tested sol-gel materials with various size-fabricated pores for increased adsorption activity on surrogate CW agents, pore-size dependence on the starting pendant organic group, and mixed-metal sol-gels with binding sites attractive to phosphonate-type compounds with fabricated pores. We also modified porous silicon by attaching the glucuronidase enzyme to the surface, constructing the flow cell, and testing the attached enzyme with an indicator surrogate.

Publications

Letant, S. E. et al. (in press). "Enzyme immobilization on porous silicon surfaces." *Advanced Materials*. UCRL-JP-200106.

Reynolds, J. G. and B. R. Hart. (in press). "Nanomaterials for security applications." *J. Minerals, Metals, & Matls. Soc*. UCRL-JP-200270.

Tillotson, T. M. and J. G. Reynolds. (in press). "Structure and characterization of sol-gel and aerogel materials and oxidation products." *MRS Proc*. UCRL-JC-149282.

Tillotson, T. M. and J. G. Reynolds. (2003). "Structure and characterization of sol-gel and aerogel materials and oxidation products from the reaction of (CH3O)4Si and C16H33Si(OCH3)3." J. *Non-Crystalline Solids* **331**, 168. UCRL-JP-200117.

Enhancement of Strength and Ductility in Bulk Nanocrystalline Metals

Tai-Gang Nieh

01-ERD-085

Abstract

This project will develop a robust scientific and technological framework for designing high-strength, -ductility, and -toughness nanocrystalline materials. Our research will couple theory and experiments with an emphasis on materials of macroscopic dimensions (a millimeter or larger) that are composed of nanoscale (<100 nm) grains. Our project has four major tasks: (1) synthesizing nanocrystalline materials with grain sizes in the 5- to 100-nm range; (2) conducting experimental studies to probe the mechanisms for mechanical deformation and failure; (3) comparing experimental observations with results obtained from atomistic, dislocation-based, crystal plasticity, and continuum simulations that cover the spectrum of mechanisms.

The results of this project can provide a framework for optimizing the properties of a nanostructured metal for maximum strength and ductility. Many well-granted theories developed in conventional polycrystalline metals will be revolutionized. For example, Hall-Petch hardening breaks down at a length scale of 10–20 nm. We are also investigating the mechanism for inverse Hall-Petch.

Mission Relevance

If successful, this project will enhance LLNL core competency in materials science by developing improved models and a better understanding of the structure, ductility, and toughness of nanocrystalline materials. Our project supports efforts within the Stockpile Stewardship Program to synthesize, understand, and predict properties of metals and will lead to the development of new high-strength, ductile nanostructured metals for large-scale laser target applications.

FY03 Results

Work accomplished for FY03 includes (1) using electrodeposition to produce nanocrystalline nickel (Ni), nickel–tungsten, and copper with grain sizes in the 5- to 100-nm range; (2) measuring the mechanical properties in tension and nanoindentation at different strain rates, with grain boundary sliding as the deformation mechanism; (3) performing simulations of Ni deformation under tension and compression, and demonstrating tension-compression asymmetry, in nanocrystalline Ni; and (4) obtaining simulated results of different types of grain boundaries on the deformation behavior of nano-Ni in good agreement with our experiments. The yielding of nanocrystalline metal obeys Coulomb-Mohr criteria instead of von Mises criteria, suggesting that nanomaterials respond differently in tension and compression. These results affect both the fundamental thinking of nanostructured materials and their design for applications.

Publications

Caturla, M.-J., J. S. Stolken, and T. G. Nieh. (in press). "Modeling the effect of texture on the deformation mechanisms of nanocrystalline materials at the atomic scale." *Appl. Phys. Lett.* UCRL-JC-148829.

Schuh, C. A. and T. G. Nieh. (2003). "A nanoindentation study of serrated flow in bulk metallic glasses." *Acta Materialia* **51**, 87. UCRL-JC-146291.

Schuh, C. A., T. G. Nieh, and H. Iwasaki. (2003). "The effect of solid solution W additions on the mechanical properties of nanocrystalline Ni." *Acta Materialia* **51**, 432. UCRL-JC-148780.

Schuh, C. A. et al. (2003). "The transition from localized to homogeneous plasticity during nanoindentation of an amorphous metal." *Philosophical Magazine* **83**, 2585. UCRL-JC-148780.

Dip-Pen Nanolithography for Controlled Protein Deposition

James J. De Yoreo 01-ERD-086

Abstract

The ability to organize macromolecules, particles, and organisms on surfaces will have a major impact on materials science, biology, and medicine. Achieving this goal requires methods for controlling deposition and attachment of organic matter to inorganic substrates. We are using a scanning-probe microscope to generate patterns of surface chemistry with feature sizes below 50 nm. These patterns can then be used as templates to order arrays of adsorbates. Our chemical nanolithography has the following goals: (1) develop chemical linkers for patterning 1- to 100-nm features, (2) use those patterns to induce organization of engineered viruses, (3) quantify the effect of pattern chemistry on virus ordering, and (4) produce a Monte Carlo code for simulating virus deposition and ordering.

If successful, this project will produce a major scientific advancement both in defining a route to inducing organization of nanostructures on surfaces and in understanding the major factors controlling order on those surfaces. Some of the specific accomplishments we expect to achieve include a method for synthesizing chemicals that will serve as linkers between inorganic substrates and specific molecular targets on the surfaces of engineered viruses, demonstration of our ability to use that chemistry to fabricate patterns, and templated deposition of genetically modified cow pea mosaic virus on those patterns.

Mission Relevance

Our project supports LLNL's nonproliferation and homeland security mission. The ability to pattern surfaces chemically at length scales below 100 nm can be used to pattern an array of ligands designed to detect specific chemical or biological agents in precise locations on micrometer-scale detection platforms. This is an important long-term goal in chemical and biological detection. More generally, our project furthers efforts in developing advanced methods for the synthesis of new materials in support of the mission in breakthrough science.

FY03 Results

In FY03, we successfully designed a chemical compound that would bind selectively to cystine groups engineered onto the surface of cow pea mosaic virus. We showed that this compound could be patterned on gold surfaces using a couple of scanned probe methods. We used these patterns for template deposition of the modified viruses. We also showed that the wild-type virus exhibited much less adsorption to the patterns. Moreover, we developed a separate chemical compound that provided resistance to virus adsorption and that could serve as a background to the chemoselective pattern. Our work was accepted for publication in journals and books and was presented in both contributed and invited talks at national meetings.

Publications

Camarero, J. A. et al. (2003). "Fabrication of assembled virus nanostructures with chemoselective linkers by scanning probe nanolithography." *Tech. Proc. of 2003 Nanotechology Conf. and Trade Show.* UCRL-JC-151507.

Cheung, C. L. et al. (2003). "Assembly of oriented 2-D protein and virus arrays by dip-pen nanolithography and nanografting." *Proc. Mat. Res. Soc. 2003 Conf.*, Apr. 2003, San Francisco, CA. UCLR-JC-149596 Abs.

Cheung, C.L. et al. (2003). "Fabrication of assembled virus nanostructures with chemoselective linkers by scanning probe nanolithography." *J. Am. Chem. Soc. Comm* **125**, 6848. UCRL-JC-152578.

Weeks, B. L. et al. (in press). "The creation of organic and biological nanostructures at surfaces using scanned probe nanolithography." *Solid-Fluid Interfaces to Nanostructural Engineering, Vol. I.* UCRL-JC-200112.

Spectroscopy of Shock-Compressed Deuterium

Neil C. Holmes

01-ERD-098

Project Description

This project performs experiments to test and challenge the predictions of first-principles simulations of the molecular vibrational spectrum and optical properties of shocked deuterium (D2). Spontaneous Raman spectroscopy, using a crossed-beam method, will be used to investigate the frequency and line shape of the D2 vibration at high pressures; absorption spectroscopy will be used to observe the opacity at pressures below 13 GPa. Once these spectroscopic techniques are demonstrated with single shocks at the gas gun, they will be transferred to laser experiments for use at higher pressures.

By developing a new capability to perform Raman and absorption spectroscopy under shock compression in fluids at high temperature and pressure, we expect to elucidate the molecular properties of such compressed fluids. This project will result in the first Raman measurements on a shocked cryogenic sample. An experimentally validated theory of compressed hydrogen and D2 is important for modeling the performance of explosives and of capsules for high-power laser experiments, and is also required for an accurate description of the interior structure and evolution of giant planets and brown dwarf stars.

Mission Relevance

The techniques developed in this project are fundamental for understanding explosives and the destruction of chemical weapons and for simulations of weapons and inertial confinement fusion experiments for stockpile stewardship. They also support LLNL's basic science mission by providing a new ability to do Raman and absorption spectroscopy under shock compression in fluid systems at high temperature and pressure.

FY03 Results

In FY03, we successfully acquired static (not shocked), single-pulse Raman spectra of the ring-breathing mode of benzene with good signal and spectral resolution. A major redesign of the input optics allowed us to achieve a higher signal-to-noise ratio and more control of the imaged volume in the shocked fluid. A test design for benzene was completed, a nitrogen target for the first shocked cryogenic Raman experiment was designed, and experiments in shocked nitrogen were performed. The high-voltage section and pulse-forming network for the white-light source needed for absorption spectroscopy was designed and will be fabricated in FY04. Absorption capability will be achieved by the end of the year.

Proposed Work for FY04

The focus of work for FY04 will be shock experiments at the two-stage gas gun. We expect to complete the initial Raman experiments at 5 and 15 GPa in shocked D2. Once the white light source is completed, the first absorption experiments will commence in shocked D2 at 10 to 13 GPa, and at ~5 GPa. We plan to undertake Hartree-Fock and configuration-interaction calculations of a variety of molecular interactions that may occur in the shocked fluid. These first-principles calculations, particularly important because they directly treat excited electronic states of the molecules, will be compared with experiments.

Use of Hydrophobic Silica Aerogel–Granular-Activated Carbon Composite to Remove Arsenic and Hexavalent Chromium from Groundwater

Sabre J. Coleman

01-ERD-106

Abstract

We propose to research the feasibility and efficiency of using functionalized hydrophobic silica aerogel and granular activated carbon (GAC) composites to adsorb hexavalent chromium [Cr(VI)] and arsenic (As) from groundwater. Our approach builds on past efforts on composites for uranium (U). Preliminary tests of functionalized composites looks promising as a means to remove Cr(VI) and As from water. The optimization process will include determining (1) the appropriate source of the functional groups in the aerogel; (2) the appropriate balance of functional groups, aerogel, and GAC; and (3) the adsorptive capacity of the aerogel composites for Cr(VI) and As in simulated groundwater.

If successful, this project will produce composites that could be used at LLNL, other DOE sites, and DoD sites in the treatment of contaminated groundwater. Aerogel–GAC composites may be used in future environmental-remediation efforts and for the treatment of drinking water. For example, it could be used to treat groundwater that is purged form a borehole as part of well installation or used in a portable water treatment facility.

Mission Relevance

By developing an aerogel–GAC composite for removing As, U, and Cr(VI) from groundwater, this work supports LLNL's mission in environment management and remediation. These composites may provide a cost-effective alternative for achieving the clean-up goals.

FY03 Results

In FY03, we produced two composites that were effective in removing Cr(VI) and As from groundwater. The composites have functional groups for hydrophobicity and for adsorption of these specific contaminants. The composites were bench tested using flow-through columns and successfully reduced Cr(VI) concentration from a range of 60 to 100 µg/L to below the detection limit (2 µg/L) of the spectrophotometer used. In testing a different composite for As adsorption with an inductively coupled plasma mass spectrophometer, As concentration was reduced from 100 µg/L to <1 µg/L, which would meet the EPA's recently promulgated maximum contaminant level of As in drinking water of 10 µg/L.

Proposed Work for FY04

In FY04 we will produce several composite formulations with one or more functional groups specific for As and test the composites by conducting comparison flow-through column testing of simulated groundwater with known concentrations of As in the range of 20 to 200 µg/L. The desired result is to confirm that a functionalized aerogel composite is a feasible technology for removing As from water.

Publications

Coleman, S. J. et al. (2003). "Granular activated carbon modified with hydrophobic silica aerogel: Potential composite materials for the removal of uranium from aqueous solutions." *Environ. Sci. Technol.* **37** (10), 2286–2290. UCRL-JC-145512.

Properties of Actinide Nanostructures

Alex V. Hamza

02-ERD-025

Project Description

We propose to obtain a detailed predictive description of the structural and electronic properties of actinide materials, such as plutonium (Pu), by performing "band mapping," a detailed experimental benchmarking of band-structure theory. Structural and electronic description of the actinides requires determining the balance of 5f electron-correlation effects, including the Coulomb energy and exchange correlation. Actinide nanostructures are important for meeting this goal. We propose to produce one- and two-dimensional (1- and 2-D) quantum-confined, crystalline actinide nanostructures with macroscopic nonconfined dimensions, and use the photoemission techniques of bandmapping and scanning tunneling microscopy (STM) to determine conventional band-structure.

One of the most complex aspects of the actinide elements is the correlated behavior of the 5f electrons. The properties of the 5f elements and compounds are quite complex, representing one of the last frontiers of condensed-matter physics. The results from this project will define the balance of the various electron-correlation effects.

Mission Relevance

Predicting Pu behavior over tens of years is key to LLNL's stockpile stewardship mission. A confirmed electronic structure for Pu will make this possible by enabling predictions of vacancy mobility and dislocation motion.

FY03 Results

In FY03, we completed the construction of our actinide nanostructure hardware, including in situ STM and ultraviolet photoelectron spectroscopy. The laser deposition of uranium (U) was performed in a vacuum suitcase used to bring the U target from the glove box. We demonstrated successful containment of the sputtered actinide, including sample transfer to the analysis ultrahigh-vacuum chamber. As the first investigation performed with the new system, U was deposited on silicon by laser ablation. In situ STM revealed 10- to 200-nm U clusters on the silicon and showed cluster-size broadening of the valance features.

Proposed Work for FY04

We plan to produce 2-D samples of Pu on silicon templates by pulsed-laser deposition; determine atomic and electronic structure by STM and in situ band mapping, respectively; calculate the band structures of the atomic arrangements produced; and, after 2-D sample production and characterization, produce 1-D nanostructures (actinide wires) on stepped surfaces. The nanostructures will be characterized by photoemission band mapping and STM.

Nanofilters for Metal Extraction

William L. Bourcier

02-ERD-032

Abstract

As water resources become depleted and populations increase over the coming decades, energy-efficient water-purification technologies will play a key role in water and waste treatment. In this project, we have developed a nanoporous membrane that selectively removes individual metals from electrolyte solutions by coating the inner surfaces of through-going nanopores with functional groups that selectively transport the targeted element. This novel extraction method is more selective and requires much less energy than existing technologies.

The development of this solid-substrate "facilitated transport" membrane supplies a new and needed technical capability to the waste-water treatment industry. The membrane can be used to concentrate elements for use in sensors, or to remove toxic, radioactive, or valuable elements from water.

Mission Relevance

This project furthers LLNL's mission in environmental assessment and management by developing element-specific filters for drinking water and DOE site-cleanup programs. In addition, applications for water treatment related to energy production support LLNL's efforts to help ensure U.S. energy independence and contribute to national water security by providing an inexpensive method of treating marginally impaired waters, thus reducing the risk of global boundary disputes over scarce water resources.

FY03 Results

In FY03, this project successfully demonstrated the applicability of functionalized nanoporous membranes for selectively separating cations from water. The project concentrated on developing methods for functionalizing pore walls of nanoporous silicon, characterizing the molecular groups using nuclear magnetic resonance (NMR), and performing transport tests of arsenate through the pores. We successfully found accelerated transport rates of arsenic through 8 to 10-nm pores that were functionalized using a copper ethylene diamine ligand. Because porous silicon of the necessary pore size turned out to be too brittle and delicate for routine handling, we switched to nanoporous alumina for the membrane substrate and developed methods for functionalizing alumina surfaces with copper EDA. Characterization and transport tests using NMR were again carried out on the alumina membranes. The membrane should allow us to develop a working arsenic sensor for use in detecting arsenic at less than 10 parts per billion.

Publications

Sawvel, A. M. et al. (2003). *Multinuclear NMR investigation of nanofilters for selective metal extraction*. Presented at the Experimental Nuclear Magnetic Resonance Conference, Pacific Grove, CA, April 18–23, 2004. UCRL-ABS-201635.

Rapid Resolidification in Metals using Dynamic Compression

Fredierick H. Streitz 02-ERD-033

Abstract

Kinetic limitations play a large role in the dynamics of solidification during freezing, but the role of kinetics in solidification at high temperatures and pressures is not at all understood. This project will investigate the effect of kinetics on the rapid resolidification of metals by designing and performing novel, time-resolved dynamic compression experiments using the LLNL light gas gun (LGG). Focusing on the rapid resolidification of molten bismuth (Bi), the project will design isentropic compression experiments with highly optimized layered impactors; develop novel, time-resolved diagnostic tools for use with the LGG facility; and perform atomic-scale simulations of the resolidification process.

The primary deliverable of this project will be two new capabilities: (1) using novel impactors to experimentally control pressure and temperature loading rates, and (2) using subnanosecond time-resolved shock diagnostics to characterize, in situ, the behavior of a metal as it follows a carefully prescribed thermodynamic path. Together, these capabilities will substantially broaden the range of phase space that can be accessed with dynamic compression experiments and will eventually lead to a better understanding of the kinetics of phase transitions in nonequilibrium situations.

Mission Relevance

Detailed knowledge of melt/refreeze/solidification transitions under highly nonequilibrium conditions is critical for the LLNL mission of stockpile stewardship. These experiments will improve our interpretation of equation-of-state experiments and our ability to predict phase behavior and material strength at high pressures and temperatures.

FY03 Results

We created a novel bullet capability that allows complete control of the impactor density from 0.1 to 15 g/cm^3. The ability to both increase and decrease density allows us to access complex paths experimentally through phase space, including virtually any combination of shock, release, isentropic compression, and static pressure. We succeeded in resolidifying a liquid water sample during isentropic compression, and have seen evidence of a phase transition in Bi by observing simultaneously the material response of a solid copper and liquid Bi sample.

Proposed Work for FY04

In FY04, we plan to (1) continue our study of rapid resolidification in molten Bi using graded-density flyers to produce near-isentropic compression with the LGG; (2) probe the time scale necessary for solidification, investigating the effect of loading rate and amplitude on the solidification process; (3) perform molecular dynamics simulations of solidification to yield insight into the kinetics of the transition; and (4) use our novel bullets to design experiments in which the sample crosses and then re-crosses the melt line. By repeating the experiments at different loading rates, we can begin to investigate the effects of hysteresis on the dynamics of solidification.

Publications

Nguyen, J. H. et al. (2003). *Dynamic compression by design at LLNL gas gun*. Presented at the Meeting of the American Physical Society, Austin, TX, March, 2003. UCRL-JC-151658-ABS.

Nguyen, J. H. et al. (2003). *Resolidification of liquid bismuth during dynamic compression*. UCRL-JC-153353.

Streitz, F. H. et al. (2003). *Resolidification of liquid bismuth durning isentropic compression*. Presented at the Meeting of the American Physical Society, Austin, TX, March, 2003. UCRL-JC-151963-ABS.

Magnetic Transition Metals and Oxides at High Pressures

Valentin Iota-Herbei 02-ERD-046

Abstract

The magnetic 3d-transition metals and their oxides have unique magnetic properties, strong correlation effects, and are abundant in the Earth's interior. Yet most studies at high pressures have been limited to phase stability, crystal structure, and mechanical properties. In experiments at Argonne National Laboratory's Advanced Photon Source (APS), this project will use two dignostic techniques—x-emission spectroscopy (XES) and x-ray magnetic circular dichroism (XMCD)—to study the electronic and magnetic properties of 3d metals and their oxides in a diamond anvil cell. Using angle-resolved x-ray diffraction in conjunction with first-principles calculations, the project will determine detailed electronic structures of iron, cobalt, nickel (Fe, Co, Ni) and their oxides at high pressures, including spin polarization of the valence electrons.

Results from our studies of the electronic structures of these 3d-transition metals and their oxides at high pressures, including spin polarization of the valence electrons, will enhance our

understanding of the high-pressure interior of the Earth, where these elements are abundant, and in special materials under weapons-relevant conditions in which correlated electron systems play a major role.

Mission Relevance

The project will develop new element-specific magnetic probes for use at high pressures. Such probes will be essential to future studies of alloys and highly correlated systems, including rare-earth metals and actinides, which are of critical importance to stockpile stewardship.

FY03 Results

Our FY03 experiments on Fe, Co, and Ni studied the electronic and magnetic transitions at high pressure. XMCD measurements on nickel at the APS confirmed the feasibility of using XMCD to monitor the magnetic properties of materials at high pressures. Using XES, we determined the existence of spin transitions in Fe and Co and found evidence for the loss of spin in high-pressure Fe. A high-spin to low-spin transition was found in Co, which may be magnetically driven (results to be published). A high-spin to low-spin, second-order electronic transition was also discovered in high-pressure Co. From XMDC measurements in Ni at high pressure, we determined that no electronic phase transitions occur up to 70 GPa.

Proposed Work for FY04

For FY04 we propose the following experiments: (1) XMCD measurements on Fe, Co, and Ni to determine the existence of a low-spin phase of Ni, the nature of the electronic changes observed in high-pressure/high-temperature Co, and the detailed mechanism for electronic and structural transitions in 3d metals; and (2) XES on simple-transition 3d metal oxides (MnO, FeO, CoO, NiO) to investigate magnetic phase boundaries in 3d oxides and volume collapse across the magnetic extinction transition in simple transition metal oxides. In addition, we plan to use in situ transport measurements to monitor the conductivity jump across half-metal normal-metal transitions in CrO_2 and Fe_3O_4 insulator-metal transitions in 3d oxides.

Exploring the Linkage between Impurities and Optical Properties in Rapid Growth of Crystals

Ruth A. Hawley-Fedder 02-ERD-070

Abstract

LLNL has an international reputation in the field of solution-growth of large potassium dihydrogen phosphate (KDP) crystals. Future advances require more sophisticated approaches to understand the link between crystal properties and growth conditions. Impurities play a key role in growth habit and properties, but other parameters are also important to successful rapid growth of these materials. Our research will focus on developing advanced, rapid-growth technologies suitable for providing high-quality optical components needed for advanced laser and photonics applications. We study the effect of key impurities (Al^{3+}) on specific properties of rapid-growth KDP crystals necessary to control growth habit and to develop a thorough understanding of these interactions as a function of solution temperature, growth rate, material composition, and the presence of competing compounds [e.g., ethylene dinitrilotetracetic acid (EDTA)].

Results from this project will further the fundamental science underlying crystal growth from solution, and enable crystals to be grown rapidly with the desired material and optical properties for a variety of applications.

Mission Relevance

This work will benefit the broad array of worldwide efforts to develop crystalline materials for laser-system applications, optical components, and photonics applications. These areas of research are particularly important to the Laboratory's stockpile stewardship mission.

FY03 Results

In FY03 we developed an improved understanding of the speciation chemistry of trivalent cations (Al^{3+}) in the presence of KDP and EDTA. Temperature and pH dependencies of the formation of various Al^{3+} complexes were completed. Molecular-scale experimentation on KDP and deuterated KDP in the presence of high levels of Al^{3+} showed no effect on prismatic growth at temperatures greater than 52°C. These results account for the previous observation that Al^{3+} addition did not affect aspect ratio as expected. Investigation of inclusion morphology revealed that morphology varies with inclusion type. Modeling of the stresses experienced under various rotation conditions and microscopic investigation of secondary nuclei suggest that one source of secondary nuclei is the primary crystal. Modifications to the growth conditions have been made to reduce the incidence of secondary nucleation.

Proposed Work for FY04

In FY04, we plan to (1) perform and document the effect of regeneration conditions (supersaturation, rotation) on aspect ratio, (2) determine the surface kinetics and Al^{3+} distribution coefficient as a function of temperature in both prismatic and pyramidal crystal faces of DKDP and KDP, and (3) complete surface–impurity interaction modeling and kinetic Monte-Carlo modeling of inclusion formation.

Determining the Microstructural Morphology of Shock-Induced Melt and Resolidification

Wayne E. King

03-ERD-018

Abstract

The principal objective of this project is to determine the factors in shock-induced melting (solid–liquid phase transformation) that affect the material's microstructure upon resolidification. Such factors include the initial crystal ordering at the microscale, the speed of solid disordering, and the length of time in the amorphous (disordered) phase. The project has four major phases: (1) computational design of laser targets and drive pulses to produce the desired shock morphologies; (2) development of specially tailored target materials that have, for example, specific initial microstructures; (3) pre- and post-shot analyses of the target samples to determine the microstructural morphology; and (4) theory and simulation at the atomistic scale. The results of this project will increase scientific understanding of the dynamics of metals by determining how a shock-melted pure metal structures itself upon refreezing at a very high cooling rate.

Mission Relevance

This project will provide data needed for the development of advanced constitutive models for hydrocodes and proven techniques to determine target specifications for planned solid-state materials strength experiments in support of stockpile stewardship.

FY03 Results

In FY03, we completed detailed design calculations for a baseline bismuth (Bi) experiment, using newly developed models in LASNEX, in which the specific phase curve for Bi was inserted. In parallel, we used detailed large-scale molecular dynamics (MD) simulations to model the kinetics of the solid–liquid and liquid–solid phase transitions. The MD code MDCASK was optimized to run more quickly and efficiently, and was used with embedded-atom potentials for copper (Cu) and silicon (Si) to explore the details of the solid–liquid transition in Cu expansion and the liquid–solid transition in Si expansion. The results showed that the phase transition is ~10 times slower in Si than in Cu. Detailed experiment planning was also begun.

Proposed Work for FY04

Using the tamped-ablation technique, we plan to shock-melt Bi with a 20- to 30-kbar pressure pulse driven by the Trident laser at Los Alamos National Laboratory. The thickness of the Bi will be selected so that the pressure pulse reaches the back surface when the laser pulse driving the pressure turns off. In a sequence of shots, we will vary the pulse duration and the Bi thickness to obtain samples that contain material near the front (drive) side that has experienced the melt transition for the full duration of the laser pulse, and material near the back side that has been in the melt phase only momentarily before release.

Hydrogen Storage in Carbon Nanotubes at High Pressures

William J. Evans 03-ERD-047

Abstract

This project undertakes experiments to address key issues of hydrogen adsorption by single-wall carbon nanotubes (SWNTs). High-pressure methodologies are used to measure interactions in the hydrogen–SWNT system. Previous claims of hydrogen storage by SWNTs at 7 wt% exceed the DOE Hydrogen Plan threshold for fielding viable hydrogen fuel systems. However, these claims are controversial, and an understanding of the mechanism of the hydrogen storage has yet to emerge.

This project will yield fundamental interaction parameters of the hydrogen–SWNT system. Theory suggests that to achieve the claimed high levels of storage, a novel adsorption mechanism intermediate between physisorption and chemisorption must exist. However, very little experimental work has addressed the adsorption mechanisms and binding energies. Our work is developing x-ray diffraction, Raman spectroscopy, and electrical-transport capabilities to study these properties. High-pressure methodologies force maximum uptake of hydrogen by the SWNTs, enabling us to probe fundamental parameters of the hydrogen–SWNT system and determine their maximum storage capacity. This work will lead to a firm understanding of the hydrogen storage by SWNTs and determine their viability as an energy storage system.

Mission Relevance

This research advances LLNL's energy-security mission by developing capabilities and data to help validate SWNTs as a hydrogen storage system that is safe and reliable and achieves the necessary storage capacity. Such a hydrogen storage system would be a crucial enabling technology for realizing the goals of the DOE Hydrogen Plan and national energy policy.

FY03 Results

A suite of capabilities was developed and utilized to study the hydrogen–SWNT system. A cryogenic capability for loading high-pressure diamond anvil cells with SWNTs and hydrogen was developed. Using this, SWNTs were immersed in liquid hydrogen and pressurized to maximize hydrogen uptake. In situ Raman and x-ray diffraction studies of the hydrogen–SWNTs under pressure determined vibrational properties and lattice parameters. A high-pressure optical cell for electrical-transport measurements of individual SWNTs, capable of 50 kpsi, was developed and used to study the electronic properties of SWNTs under pressure.

A Two-Particle Formulation of Electronic Structure

Antonios Gonis

03-ERD-064

Abstract

The treatment of correlated electrons in solids is considered to be the most important remaining problem in the theoretical and computational study of solids. The problem of correlated electrons in elemental solids and alloys figures prominently in the Laboratory's efforts to understand the behavior of plutonium (Pu) metal and its alloys, and to predict such properties as phase and dimensional stability under self-irradiation and as a function of time.

Through development of formalism and computational algorithms, we will study the electronic structure of matter based on two-particle states, as well as improve treatment of electron correlations. This research will enable us to develop codes based on two-particle states, and could provide theoretical and practical electronic structural information about important materials and systems, particularly complex systems such as Pu, for which conventional methods are currently inadequate.

Mission Relevance

Because the electronic structure and properties of materials, especially actinides, is crucial to areas such as stockpile stewardship, this project supports the national-security mission.

FY03 Results

The formal aspects of the study of correlated electrons using the two-particle approach were worked out. We developed a set of equations that allow the determination of electronic self-energy, and initiated calculations for model systems.

Proposed Work for FY04

First, we will perform tight-binding (TB) calculations of correlated electrons in model systems, both ordered and disordered, accounting both for the Coulomb interaction and disorder effects. We will also delineate the formal connection of the two-particle approach with the formalism of the dynamical mean field approximation. Next, we will perform realistic, all-band TB calculations of face-centered-cubic nickel within the two-particle framework and obtain electron self-energies and densities of states to compare with results from photoemission and other experiments.

Optics Performance at 1 omega, 2 omega, and 3 omega

John Honig

03-ERD-071

Abstract

The interaction of intense laser light with dielectric materials is a fundamental applied science problem that is becoming increasingly important with the rapid development of ever more powerful lasers. To better understand the behavior of optical components in large, fusion-class laser systems, we are systematically studying the interaction of high-fluence, high-power laser light with high-quality optical components, with particular interest in polishing/finishing and stress-induced defects and surface contamination. The project focuses on obtaining comparable measurements at three different wavelengths: 1ω (1053 nm), 2ω (527nm), and 3ω (351 nm). Modeling at both microscopic and macroscopic scales supports interpretation of the experimental results.

Materials research that enhances our understanding of the complex mechanisms describing the response of optical materials to intense laser irradiation will benefit worldwide efforts to field ever more powerful, fusion-class laser systems and allow them to operate efficiently and reliably at or above their design specifications. Our modeling efforts will allow the operators of fusion-class laser systems to predict the effects of upcoming experimental campaigns on optics lifetime and to optimize campaign planning to minimize incremental costs that result from replacing optical components.

Mission Relevance

This work will contribute substantially to the knowledge base of optical materials under the intense laser illumination created in fusion-class lasers systems and will allow us to validate our theoretical and stochastic models. This work supports NNSA and Laboratory missions in stockpile stewardship because the safety and reliability of the nuclear stockpile in the absence of testing will rely heavily on the experimental data from fusion-class laser systems to validate complex computer simulations.

FY03 Results

While the detailed mechanisms of contamination-related damage growth are still not fully understood, in FY03 we systematically completed a dataset that allows us to directly compare the effect of contaminants and drive wavelength. We found that contamination reduces the damage threshold of fused silica and that, for a given contaminant, the damage occurs at lower fluence of 3ω compared to 2ω. Studies to determine the effect of lenslets, small index variations in glass that behave as focusing lenses, on optic performance at 1ω, 2ω, and 3ω were begun. This work includes a modeling component that calculates the field intensification due to lenslets at the rear surface of the optic and on other optics downstream.

Proposed Work for FY04

In FY04 we will continue the systematic campaign to measure and model the damage initiation thresholds and the damage growth parameters for fused silica and potassium dihydrogen phosphate as a function of pulse length and fluence level at 1ω and 2ω for comparison with data obtained at 3ω. We will also develop a novel method for characterizing frequency-conversion crystals that will allow direct and accurate measurement of obscuration and pinpoint density and size for all three wavelengths, and determine crystal performance with conditioning as a function of pulse length and wavelength. Finally, we will develop and test various conditioning protocols to determine optimal conditioning performance for crystals.

Publications

Feit, M. D. and A. M. Rubenchik. (2003). *Influence of subsurface cracks on laser induced surface damage*. Presented at SPIE Damage Symp. XXXV, Boulder, CO. UCRL-CONF-153481.

Norton, M. A. et al. (2003). *Growth of laser initiated surface damage in fused silica at 527 nm*. Presented at SPIE Damage Symp. XXXV, Boulder, CO. UCRL-CONF-155364.

Nostrand, M. C. et al. (2003). *A large aperture, high energy laser system for optical component testing*. Presented at SPIE Damage Symp. XXXV, Boulder, CO. UCRL-CONF-155394.

Novel Methods for Bonding Disparate Materials

Michael W. Mcelfresh
03-ERD-074

Abstract

This project will use a systematic scientific strategy to develop new processes for bonding disparate materials to yield extremely thin, uniformly thick, strong bonds without altering the materials to be bonded. The bonded material will be used for fabricating experimental samples (e.g., laser targets). We plan to explore a set of new methods that includes hydroxide bonding, "molecular bonding agents," and a new strategy for bonding materials based on studies of how geckos attach themselves to surfaces. Bonds made with industry-standard polymer-based adhesives will be used as a reference point for this work. For the more promising bonding methods, we plan to develop reliable methods and protocols for laser-plasma target fabrication.

The expected results developed from our analysis of four very different bonding strategies will be a more scientific understanding of bonding between disparate materials. This research will result in publications in refereed journals and provide new concepts for multilayer targets.

Mission Relevance

This project supports the Stockpile Stewardship Program (SSP) by enabling multilayer targets for an important class of SSP experiments that studies materials under precisely controlled shock conditions driven by a gas gun or laser. The bonding of the multilayer targets must be sufficiently strong to not delaminate prematurely, and in many cases, the bonding must be transparent for the use of optical diagnostics.

FY03 Results

With a start date in the last two months of FY03, we obtained samples for bonding, including diamond-turned aluminum pieces, gold-coated silicon substrates, and quartz and sapphire crystals, and obtained the reagents for synthesizing molecular bonding agents. Surface analytical studies were performed to characterize the surfaces to be bonded. We began using a tool that will allow us to control the size and distribution of the load on pieces being bonded.

Proposed Work for FY04

Our goal for FY04 is to form bonds between a metal and a ceramic, starting with copper and alumina, by using commercially available crystalline ceramic, i.e. sapphire, that will allow the interface between the materials to be observed optically so that spectroscopic studies can be performed. The strength of bonding between the materials will be studied as a function of several variables, including bonding chemistry and surface roughness. A specific set of bonding methods for bonding ceramic to metal (at low temperature, i.e. room temperature) will be explored, including hydroxide bonding and molecular adhesive bonding agents.

Plutonium and Quantum Criticality

Michael J. Fluss

03-ERD-077

Abstract

Quantum criticality explains the emphasis on empirical investigations of plutonium (Pu) because the usual theory of solids, based on quantum mechanics of single electrons moving in periodic potentials, cannot explain the behavior of strongly correlated electron materials like Pu. This project explores the low-temperature phase diagram of Pu and Pu alloys near the f-electron localization–delocalization boundary in the Pu phase diagram, which may be the signature of a low-lying quantum critical point (QCP) that underlies the origin of delta-Pu. To find the QCP, we will tune the composition, pressure, and magnetic field, and search for non-Fermi liquid (NFL) characteristics in the physical properties at low temperatures expected near a QCP. A unique aspect of this work is inspired by the proximity of Pu and Pu alloys to a magnetic–nonmagnetic boundary. We plan to use a muon probe to make contact with local magnetic properties. Once the QCP is determined, we will endeavor to understand the results in terms of the projected behavior, if any, for the solid-state system.

Profound issues in condensed-matter physics surround Pu. The solution will lead to a deeper understanding of metal physics. The demonstration or refutation of quantum criticality through mapping the low-temperature phase diagram is of considerable fundamental importance to our ability to correctly predict the equation of state. The data collected in these experiments will be useful in validating the underlying principles and refining new theories concerning the origin of the high-temperature phases of Pu and Pu alloys.

Mission Relevance

Quantum criticality will be important to our fundamental understanding of how to correctly insert Pu properties into our codes for stockpile stewardship. In addition, the data obtained in this project support LLNL's mission in basic science by serving as both validation and motivation for emerging microscopic theories of highly correlated electron systems.

FY03 Results

In FY03, we prepared instruments and specimens for FY04 transport measurements. A 15-yr-old superinducting quantum interference device magnetometer was refurbished and calibrated, and Pu specimens were prepared by electrorefinement and chemically assayed. We wrote specifications for a physical properties measurement system used for determining electrical resistivity, magnetic susceptibility, and heat capacity in the temperature range of 300 mK to

500 K. A proposal for muon spin resonance experiments on Pu at the TRIUMF meson facility (Canada) was drafted, in collaboration with Los Alamos National Laboratory.

Proposed Work for FY04

Proposed work for FY04 includes (1) synthesize research-quality specimens of alpha-Pu and delta-phase stabilized Pu (Ga) and Pu (Am) with <40 ppm of magnetic impurities; (2) measure the magnetic susceptibility and conductivity as a function of field (0–9 T) and temperature (2.4–350 K); and (3) determine the spin and magnetic properties of defect-induced damage in Pu. We expect the results of these measurements will lead to the identification of magnetic and/or superconducting phases of Pu usually found near a QCP.

Publications

Fluss, M. J. et al. (in press). "Temperature dependent defect properties from ion-irradiation in Pu (Ga)." *J. Alloys & Compounds*. UCRL-JC-153046-REV 1.

Exchange-Coupling in Magnetic Nanoparticles Composites to Enhance Magnetostrictive Properties

Harry B. Radousky 02-ERI-006

Abstract

This project is investigating the properties of magnetostrictive materials, which have a variety of applications as both actuator and sensor devices. Our goal is to create coated nanoparticles and embed these particles in elastomer composites using the techniques of spark erosion and chemical synthesis. The work will include understanding the spark-erosion process, studies of the physical and magnetic properties of the particles as a function of size, and a study of the properties of the composites.

The result of this project will be a thorough understanding and characterization of the magnetostrictive response of magnetoelastomers bonded to nanoparticles. Our results will validate the concept of using nanoparticles in these materials and will give us an initial understanding of the nanoparticle physics involved. This project is a collaboration with the University of California, San Diego and Los Angeles.

Mission Relevance

This work will support LLNL's national security mission by enhancing capabilities in magnetorestrictive composites, a prime future technology for microfluidics (e.g., pumps and valves) and for fusion reactor walls. These potential nanomaterials will also enable the next generation of bioassay devices for chemical and biological national security applications, e.g., sensors and actuators, which also have application in vibration damping for automobiles, buildings, and bridges.

FY03 Results

We exceeded our expectations for FY03 in the area of understanding the spark-erosion process. The experiments on the "fixed gap" spark-erosion facility show a clear relationship between spark energy and particle size. This work was accepted for publication in *Journal of Applied Physics*. We produced hollow nickel (Ni) particles by sparking in liquid nitrogen, which was an unexpected and important result. Both hollow and solid Ni particles produced by spark erosion were incorporated in composites. Terfenol-D (a giant magnetostrictive material) particles and Terfenol-D composites were also produced. Terfenol-D x-ray measurements were made in a magnetic field at the Advanced Photon Source at Argonne National Laboratory.

Proposed Work for FY04

In FY04, we plan to complete our work with Ni as a model system. An iron–gallium composite (83% Fe and 17% Ga) will be studied as a second model system that has magnetostrictive values between Ni and Terfenol-D. The spark-erosion process for controlled production of a wide range of materials in particulate form will continue to be optimized. We will coat nanoparticles, at first with the model materials, to create a core-shell system that takes advantage of the magnetic exchange coupling between the hard and soft materials. These core-shell systems will be fully characterized and embedded in composites.

Publications

Carrey, J., H. B. Radousky, and A. E. Berkowitz. (in press). "Spark-eroded particles: Influence of processing parameters." *J. Appl. Phys.* UCRL-JC-155490.

Nersessian, N. et al. (2003). *Bulk and Composite* $Gd_5Si_2Ge_2$. Presented at the SPIE Smart Structures and Materials Symposium, San Diego, CA, March 2–6, 2003. UCRL-JC-147076-ABS.

Development of Sample Handling and Analytical Expertise for STARDUST Comet Sample-Return Mission

John P. Bradley 03-ERI-007

Abstract

In 2006, the NASA Stardust space mission, the first solid-state sample return mission since the Apollo missions of the 1970s, will return more than 1000 submicrometer- and micrometer-sized comet dust grains to Earth. Today, methods for working with nanoparticles captured and embedded in aerogel are in their infancy. It is imperative that analytical instrumentation be in place and suitable strategies for detailed studies of the captured cometary particles be developed in time to maximize the scientific yield from the Stardust mission. The focus of this project is to develop methods to extract the captured particles from aerogel capture cells and identify and develop procedures to analyze both extracted particles and in situ particles in aerogel capture cells. In collaboration with the Space Sciences Laboratory (SSL) at the University of California, Berkeley, we will use needles, microtweezers, laser-cutting devices, and focused ion beam (FIB) microscopy to extract particles and optical-, electron-, and proton-beam microscopy to analyze in situ and extracted particles.

For the particles returned to Earth in 2006, we will be able to extract grains down to submicrometer diameters using optical and ion microscopy. Detailed chemical, mineralogical, and isotopic information will be obtained using state-of-the-art instrumentation such as the NanoSIMS, the Nuclear Microprobe, and the yet-to-be-built Super-STEM that will answer some of the most significant questions in planetary science.

Mission Relevance

Since the collected samples are natural nanomaterials, the techniques and analytical procedures developed under this project are directly applicable to nanotechnology critical for key Laboratory missions in stockpile stewardship, homeland security, and basic science. The proposed work will prepare for full scientific exploitation of the Stardust samples in support of NASA's sample-return missions.

FY03 Results

In FY03, the extraction techniques used at SSL were enhanced to enable the extraction of both individual particles and entire tracks. For the first time, FIB microscopy was used to expose an embedded particle in aerogel. In addition, we used the nuclear microprobe at the Center for Accelerator Mass Spectrometry (CAMS) to characterize extraterrestrial particulates captured in low-Earth orbit (LEO). This includes the first measurement of hydrogen in such material. We also used the synchrotron infrared beamline at the Advanced Light Source to identify organic carbon in LEO extraterrestrial particulates, significant for understanding the role of dust in bringing life to early Earth and for future Mars mission.

Proposed Work for FY04

Our work in FY04 will continue to characterize space-exposed, aerogel-captured orbital debris and extraterrestrial dust grains using the CAMS nuclear-microprobe facility and Raman microprobes at LLNL. We will also evaluate a microscope laser-cutting device to assist in particle extraction, investigate the use of a new dual-beam scanning electron microscope/FIB instrument at LLNL to prepare and analyze samples, use electron microscopes at LLNL and the National Center for Electron Microscopy to characterize thin sections prepared using FIB, and continue to develop our collaborations with extensive analytical facilities in the Bay Area.

Publications

Graham, G. A. et al. (2003). "Investigation of ion beam techniques for the in situ analysis and exposure of particles encapsulated by silica aerogel: Applicability for Stardust." *Meteorit. Planet Sci.* **38**. UCRL-JC-155390.

Westphal, A. J. et al. (2003). "Multi-technique analysis of micrometeoroids captured in low-Earth orbit." *Meteorit. Planet Sci.* **38**, A144. UCRL-JC-153006-ABS.

Photoluminescent Silica Sol-Gel Nanostructured Materials Designed for Molecular Recognition

Sharon J. Shields 02-LW-038

Abstract

The objective of this proposal is to develop silica sol-gel nanostructured materials—aerogels—to function as a separation device for biomolecule detection using laser desorption/ionization mass spectrometry (LDI-MS). Detecting biological molecules is difficult, owing to the complex nature of the cellular matrix, which interferes with detection. Silica sol-gel nanostructured materials have several properties, including photoluminescence and the ability to discriminate between chemical moieties, that make them uniquely suited for the rapid separation of biomolecules with LDI-MS system. Photoluminescent properties enable them to act as energy receptacles for ultraviolet (UV) laser radiation, thus promoting desorption/ionization of biomolecules from the sample target. Their ability to discriminate between chemical moieties allows aerogels to perform molecular separations.

We expect to develop new aerogel materials, characterize the fundamental properties of these aerogels, and use them to develop a means for rapidly separating biomolecules and to understand the fundamental mechanisms in matrix-assisted laser desorption ionization. This work will produce a silica sol-gel nanostructured material that will enable the detection and analysis of biological and chemical warfare agents. This material will also advance research in functional genomics, proteomics, and molecular diagnostics.

Mission Relevance

DOE missions in functional genomics, proteomics, and molecular diagnostics for biological and chemical warfare exposure will be enhanced through the development of the silica sol-gel nanostructured material for nanoscale separations. This project supports the Laboratory's national security mission area of countering the spread and use of biological weapons of mass destruction and the homeland security mission of keeping the nation safe from bioterrorism.

FY03 Results

To address aerogel photoluminescence, synthesis routes for chemcial modification of silica aerogels with UV-absorbing molecules were constructed. Covalent modification of cinnamic acids to the silica aerogel framework red-shifted the flourescence spectrum of the cinnamic acid from 300 to 360 nm (neat) to 460 to 530 nm (aerogel modified), eliminating the material as an energy recepticle for the 337-nm radiation. These findings affect the fundamental theories of biomolecule desorption/ionization in UV matrix-assisted LDI. Sol-gels of differing chemical structures, (i.e., hydrophobic versus hydrophilic) were also synthesized, and a direct correlation between the molecular structures retained by the aerogel and molecules detected by MS was found. This work has led to further research on the fundamental properties of laser–material interactions. End-of-the-year results produced two manuscripts to be published next year.

Covalent Attachment of Metallic Nanorods to Nanocrystals

Louisa J. Hope-Weeks 03-LW-048

Abstract

This research project will investigate ways to develop a tunable synthetic pathway for the formation of Group IV nanocrystals so that their observed properties can be compared to those predicted from theoretical calculations. Our goal is to form nanocrystals with a narrow predictable size distribution. Since the size of the nanocrystals is of fundamental importance to their observed properties, the main focus of the project will be to develop an understanding of how reaction conditions can be modified to produce nanocrystals with a monodisperse size distribution from a one-pot synthesis.

The main goal to this project is to develop a tunable synthetic pathway for the formation of Group IV semiconductor nanocrystals. The fluorescent properties of the nanocrystals are strongly dependent on their size due to the quantum confinement effects. Therefore a one-pot synthesis, which can be tuned to produce a wide range of sizes, would provide nanocrystals for applications such as biological detection and tags for biological agents.

Mission Relevance

This work will be aimed at forming nanocrystals with the potential to act as sensors for biological agents and tags for biological agents, both of which have applications for LLNL mission areas of nonproliferation and homeland defense.

FY03 Results

The major accomplishment in FY03 was the formation of germanium nanocrystals with narrower size distributions than have previously been reported. The conditions necessary to control the reaction have been established with respect to reducing agent and reaction solvent. The time-dependent growth studies have resulted in the formation of halide-capped germanium nanocrystals with narrow size distributions, in which the shape and size of the nanocrystals can be varied by simple changes in reaction time.

Publications

Hope-Weeks, L. J. (2003). "Time dependent size and shape control of germanium nanocrystals." *Chem. Comms.* UCRL-AB5-200210.

A New Material for Inertial Confinement Fusion Targets: Diamond-Like Polyimide

Thomas E. Felter 03-FS-027

Abstract

Inertial confinement fusion (ICF) target specifications demand high tensile strength for the polyimide target material. To fabricate the target in house, a thin film of polyimide material is deposited uniformly by chemical vapor deposition (CVD) techniques onto a spherical mandrel. To meet the exacting tolerances we require for surface roughness, the material needs to be smoothed. However, the smoothing process reduces strength below that required for room-temperature storage of the deuterium fuel. This study focused on determining the feasibility of strengthening polyimides by irradiation with an energetic ion beam. Our work will integrate a number of techniques, including infared absorption, matrix-assisted laser-desorption ionization, nanoindentation, and tensile-strength measurements resulting in a better understanding of these important changes to the polyimide.

The irradiation experiments conducted in this study exposed both sides of 50-μm-thick samples of commercially available foils and films grown in house. Pull tests of irradiated foils were compared with each other and to pristine reference samples. Our goal was to improve the tensile strength of polyimide target material by more than a factor of two. Such a discovery would lead to further research, including building a mechanism for two-axis rotation of spherical ICF targets in the ion beam.

Mission Relevance

The ICF Program is of paramount importance to stockpile stewardship and eventual energy independence of the U.S. Stronger target shells have a number of immediate benefits to target designers including operation at higher pressures and better dimensional stability during filling and implosion.

FY03 Results

We used ion-beam irradiation of CVD polyimide and its commercial counterpart to elucidate the differences between the two forms of polyimide and to improve mechanical properties. A number of analytical techniques were integrated to address these important changes to the polyimide. For the CVD polyimide used for targets, we increased the bulk modulus by a factor of 2 to 3. We obtained indirect evidence that the density of polyimide is substantially changed by the ion beam. The ability to substantially alter the density of the polyimide can greatly reduce Rayleigh-Taylor instabilities and allow for much improved implosion behavior.

Feasibility of Proton Radiography for Mesoscale Characterization

Graham Bench 03-FS-028

Project Description

This project studied the usefulness of two- and three-dimensional proton imaging for characterizing mesoscale objects such as inertial confinement fusion (ICF) targets. Imaging with protons complements x-ray techniques and is superior in many cases. The accuracy of proton measurement is affected by lateral and longitudinal straggling, but the amount of straggling for an energetic proton in a given material is poorly known. We propose to measure lateral and longitudinal straggling for representative ICF target materials. These measurements will be used to calibrate relevant imaging and ion trajectory codes so that accurate simulations can be performed with realistic treatment of resolution, contrast, and noise.

From the calibrated codes resulting from this study, it should be possible to determine whether proton imaging can provide the required characterization for any ICF target design. Proton imaging provides better image contrast and resolution than x-ray imaging for mesoscale objects that contain both low- and high-*Z* materials.

Mission Relevance

By determining the usefulness of proton imaging for characterizing complex meso- and microsystems, such as ICF targets, the project will contribute to improved target design for ICF, a critical component of the Laboratory's and DOE's stockpile stewardship mission.

FY03 Results

We concentrated on experimentally characterizing the lateral straggling of 2.5 to 3.5-MeV protons in several thin-film targets. The experimental and computed data were found to have full width half maximum (FWHM) values that typically agreed within 20%, which is adequate for our characterization and modeling requirements. The two-dimensional straggling data have been used to extend our ion-beam modeling capabilities. The data suggest that we could typically expect to be able to resolve micrometer-scale features within a variety of mesoscale objects, given an operational high-energy (tens of MeV) proton microbeam facility. Models resulting from this work should produce accurate simulation results needed for planning and evaluating experiments with pertinent material combinations and geometries.

Mathematics and Computing Sciences

The Stochastic Engine: Improving Prediction of Behavior in Geologic Environments We Cannot Directly Observe

Roger D. Aines

01-SI-003

Abstract

Accurate prediction of complex phenomena can be greatly enhanced by using data and observations to update simulations. The ability to create these data-driven simulations is limited by error and uncertainty in the data and the simulation. A key challenge is developing representation methods that integrate the local details of data with the global physics of simulations, enabling supercomputers to efficiently solve the problem. Our generalized solution will contribute both a means to make more informed predictions and to improve those predictions using new data in real time. Current applications include evaluating groundwater contamination movement, imaging rapidly evolving systems, utilizing diverse intelligence data, and rapidly evaluating a large structure's internal flaws.

Using modern computational power, the stochastic engine rapidly chooses among a very large number of hypothesized states and selects those that are consistent (within error) with all the information at hand. Predicted measurements from a simulator are used to estimate the likelihood of actual measurements, which in turn reduces the uncertainty in the original sample space via a conditional probability method called Bayesian inferencing. A highly efficient, staged Metropolis-type search algorithm allows us to address extremely complex problems. This combination of probabilistic and deterministic approaches opens the door to solving many data-driven, nonlinear, multidimensional problems.

Mission Relevance

The computational methods developed in this initiative will (1) help increase the reliability of nonproliferation assessments and stockpile stewardship calculations, (2) increase the efficiency of fossil-energy extraction systems, (3) support more efficient cleanup of underground environmental contaminants, and (4) improve the accuracy and speed of assessment of the hazard represented by atmospheric releases due to accidents or hostile intent. All of these are core Laboratory and DOE missions. In each of these applications, the stochastic engine method will rapidly improve predictions based on existing data. This approach provides a means to quantitatively estimate the value of new data, answering the question, "If new measurements are made, how much would they change our existing prediction?"

FY03 Results

We focused this year on large-scale problems and on examining the mathematical robustness of the approach in diverse applications. Multiple data types were combined with simulations to evaluate systems with ~$10^{20,000}$ possible states, which corresponds to detecting underground leaks at the Hanford waste tanks. We assessed the probable uses of chemical process facilities using an evidence-tree representation and in-process updating. We determined preferred contaminant flow paths at the Savannah River Site, demonstrated the location of structural flaws in buildings, improved the models for seismic travel times systems used to monitor nuclear proliferation, initiated a method for characterizing the source of indistinct atmospheric plumes, and applied stochastic analysis to improving flash radiography.

Publications

Aines, R. D. et al. (2003). *The stochastic engine: improving prediction of behavior in environments we cannot directly observe*. Workshop on Sensor Fusion for CBR Defense, Sept. 23–24, 2003, Salt Lake City, UT. UCRL-JC-153920 Abs.

Binley, A. (2003). *Vadose zone tracer testing in the UK Sherwood sandstone: hydrogeophysical data report*. UCRL-TR-200172.

Carle, S. R. et al. (2003). *Environmental applications of the stochastic engine*. Workshop on Simulation and Optimization, April 28–30, 2003, Research Triangle Park, NC. UCRL-JC-153613 Abs.

Franz, G. A. et al. (2003). *Stochastic engine software development guide*. UCRL-TM-200386.

Glaser, R. E. et al. (2003). *A Markov Chain-, Monte Carlo based method for system identification*. UCRL-ID-15094.

Glaser, R. E. et al. (2003). *A Markov Chain-, Monte Carlo-based method for system identification*. Intl. Modal Analysis Conf. XXI, Feb. 3–6, 2003, Kissimee, Fl. UCRL-JC-15094.

Johnson, V. M. et al. (2003). *A stochastic approach to assessing chemical production activities at an industrial site: a national security application of the Stochastic Engine Initiative*. UCRL-TR-200691.

Levine, R. A. et al. (in press). "Adaptive scanning strategies: the Gibbs sampler." *Journal of Computational Statistics and Data Analysis*. UCRL-JC-152443.

Levine, R. A. et al. (in press). "Adaptive scanning strategies: the Hastings sampler." *Journal of Computational Statistics and Data Analysis*. UCRL-JC-152437.

Sengupta, S. K. and A. L. Ramirez. (2003). *Exploratory global inference from posterior image samples in a Markov Chain Monte Carlo simulation experiment: A high-dimensional data clustering approach with extensions to categorical data*. UCRL-JRNL-200985.

Slone, D. M. and M. B. Aufderheide III. (2003). *An MCMC approach to transmission tomography from a limited number of views*. 5th Intl. Congress on Industrial and Appl. Mathematics, July 7–11, 2003, Sidney, Australia. UCRL-JC-150592 Abs.

Strategic Initiative in Applied Biological Simulations

Michael E. Colvin 01-SI-012

Abstract

The goal of this project is to use advanced computational physics and chemistry techniques to demonstrate the value of large-scale numerical simulations for the biological sciences. Using LLNL-developed simulation methods that permit highly accurate modeling of biological processes, this project focuses on simulating the dynamics of biochemical processes. A broad set of simulations techniques will be employed, ranging from empirically based protein-structure methods to first-principles, quantum-mechanical predictions of molecular structure,

chemical binding, and reaction rates. This project leverages LLNL's core competency in computational biology.

Mission Relevance

This project builds on LLNL expertise in advanced chemical modeling to forge a new core competency in computational biology. This project also uses LLNL's advanced computing resources to make scientific advances in biological problems that have been chosen for their importance to LLNL's mission in bioscience to improve human health and in environmental management. This new competency will strengthen the Laboratory's existing experimental biology research program and will provide a key resource for new LLNL mission areas such as chemical and biological nonproliferation.

FY03 Results

During FY03 we completed several simulation projects. We performed an extensive analysis of the accuracy of ab initio molecular dynamics in reproducing the properties of water, and we have used it to simulate solvation of the Ca^{++} ion, yielding results in close agreement with experimental results. We simulated DNA encapsulated in carbon nanotubes to elucidate the role of ions in DNA structure; used advanced sequence analysis coupled with molecular modeling to study the Rad17 family of eukaryotic proteins involved in DNA damage cell cycle checkpoints; performed similar studies on the Rad51 family of proteins; and completed simulation studies to optimize the components a novel anticancer radioimmunotheraputic drug.

Publications

Grossman, J. C. et al. (in press). "Towards an assessment of the accuracy of density functional theory." *J. Chem. Phys.* UCRL-JC-155397.

Reboredo, F. A., E. Schwegler, and G. Galli. (in press). "Optically activated funcionalization reactions in Si quantum dots." *J. Am. Chem. Soc.* UCRL-JC-151240.

Gygi, F., J-L. Fattebert, and E. Schwegler. (2003). "Computation of maximally localized Wannier functions using a simultaneous diagonalization algorithm." *Comp. Phys. Comm.* **115**, 1. UCRL-JC-151591.

Miller, K. A. et al. (in press). "Domain mapping of the Rad51 paralog protein complexes." *Nucleic Acids Res.* UCRL-JC-155506.

Tran, N. L. et al. (2003). "Catalysis of decarboxylation by an adjacent negative charge: A theoretical approach." *Bioorg. Chem.* **31**, 271.
UCRL-JC-152008.

Winthrop, M. D. et al. (2003). "Selection and characterization of anti-MUC-1 scFvs intended for targeted therapy." *Clinical Cancer Res.* **9**, 3845S. UCRL-JC-151393.

Biological and Synthetic Nanostructures Controlled at the Atomistic Level

Giulia A. Galli 03-SI-001

Abstract

The successful development of nanotechnology requires tackling many challenges in nanoscience. We propose to develop new capabilities for the design of Group IV

semiconductor nanostructures with desired labeling and sensing properties, using advanced simulation and experimental techniques. We plan to focus on quantum dots and carbon nanotubes because of their biocompatibility and other technical advantages over cadmium-containing nanoparticles. We will use ab initio simulation techniques and spectrosocpic measurements to microscopically investigate interfaces between these dots and tubes and specific chemical and biological media.

The project will build expertise, computer codes, colloidal-synthesis techniques, and surface-sensitive spectroscopic characterization techniques for semiconductor nanostructures. In addition, we hope to identify novel compound semiconductor nanostructures for building semiconductor-based chemical labels and biotags.

Mission Relevance

This proposal will help predict, develop, and design semiconductor materials with tailored properties for advanced biodetection needs in support of LLNL's national security mission. This will provide a scientific foundation for the creation of next-generation technologies based on manipulating and controlling matter at the nanoscale, in support of the Laboratory's mission in breakthroughs in fundamental science and technology.

FY03 Results

We obtained several new results in the field of ab initio simulations of nanostructures, including predictions of the stability and surface properties of silicon (Si) nanoparticles, the stability and ultradispersivity of diamond at the nanoscale, and the photoinduced attachment of organic molecules to Si dots. In the experimental arena, a chemical synthesis laboratory was set up, and important new results were obtained for germanium (Ge) dots. Progress was made in characterizing Ge nanoparticles (at the Advanced Light Source) and nanodiamond. A new laboratory for the optical characterization of dots was also set up.

Proposed Work for FY04

To build the nanoscience competency in the area of chemical tags and biological sensors, we plan to (1) develop predictive, ab initio simulation techniques to model organic–inorganic interfaces between nanoparticles and organic molecules in solution; (2) use the results of classical and ab initio simulations to guide and interpret characterization and functionalization experiments and to guide synthesis experiments to search for new materials that are biocompatible and have the desired size and stoichiometry; (3) identify surface probes for specific chemicals; and (4) identify surface terminations that enable the assembly of nanoparticles with desired electronic and transport properties.

Publications

Bostedt, C. et al. (2003). "Characterization of Ge-nanocrystal films with photoelectron spectroscopy." *Nucl. Instru. Meth. B* **199**, 402–405. UCRL-JC-150516.

Bostedt, C. et al. (2003). "Evidence for cubic phase in deposited germanium nanocrystals." *J. Phys. Condens. Mat.* **15** (7), 1017–1028. UCRL-JC-151543.

Draeger, E. et al. (2003). "Influence of synthesis conditions on the stuructural and optical properties of passivated silicon nanoclusters." *Phys. Rev. Lett.* **90** (16), 167402. UCRL-JC-150588.

Draeger, E. et al. (2003). “Synthesis dynamics of passivated silicon nanoclusters.” *Phys. Status Solidi B* **239** (1), 11–18. UCRL-JC-150588.

Galli, G. et al. (2003). *Ab-initio simulations of semiconductor nanostructures*. Presented at the Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA, Sept. 10, 2003. UCRL-PRES-154795.

Martin, R. and G. Galli. (2003). “Condensed-matter physics: Hydrogen falls into line.” *Nature* **423** (6940), 595–596. UCRL-JC-154363.

Raty, J. Y. et al. (2003). “Quantum confinement and fullerene-like surface reconstructions in nanodiamonds.” *Phys. Rev. Lett.* **90** (3), 037401-1. UCRL-JC-148251.

Puzder, A. et al. (in press). “The role of surface reconstructions on quantum confinement in semiconductor nanocrystals.” *Phys. Rev. Lett.* **91** (15), 157405. UCRL-JRNL-200813.

Puzder, A. et al. (in press). “Simulation of silicon nanostructures.” *Encyclopedia of Nanoscience and Nanotechnology*. UCRL-JC-151672.

Puzder, A. et al. (2003). “Computational studies of the optical emission of silicon nanocrystals.” *J. Am. Chem. Soc.* **125** (9), 2786–2791. UCRL-JC-151084.

Raty, J. Y. and G. Galli. (2003). “Ultradispersity of diamond at the nanoscale.” *Nat. Mater.* **2** (12), 792–795. UCRL-JC-155389.

Reboredo, F., E. Schwegler, and G. Galli. (2003). “Optically activated functionalization reactions in Si quantum dots. *J. Am. Chem. Soc.* **125** (49), 15243–15249. UCRL-JC-151240.

Image Content Engine

James M. Brase 03-SI-003

Abstract

Advancements in imaging-sensor technologies have resulted in volumes of data that overwhelm human analysts. The goal of this project is to develop a framework—the Image Content Engine (ICE)—that will allow human analysts to interpret massive volumes of imagery in a timely fashion. The ICE approach focuses the attention of humans on relatively few small areas with specified attributes by capturing content extracted from images (regions and edges corresponding to possible objects) as nodes in semantic graphs. The links in these graphs define relationships between pieces of image content and information obtained from non-image sources.

The ICE framework will allow content extracted from imagery, the most voluminous data source, to be seamlessly integrated with information from other sources. ICE tools will allow analysts to query large image databases based on content and contextual information. These new tools will help break down the compartmentalized nature of analyst-based interpretation in the defense and intelligence communities. The ICE approach also applies to other areas of experimental science (e.g., physics, biology, and environmental science) by providing tools to mine massive archives of complex measurement data to discover new patterns and relationships.

Mission Relevance

The work supports LLNL’s mission in national security by developing the technology to allow content extracted from images to be seamlessly integrated with information from other sources, enabling analysts to submit sophisticated queries relevant to many topics, among them defense, intelligence, counterterrorism, deployment strategies, functions of enigma facilities, and the production of weapons of mass destruction. ICE capabilities will also support the analysis of other types of LLNL program imagery, for example, high-resolution tomography for weapons inspection, laser diagnostics images, and biological microscopy images.

FY03 Results

In FY03 we developed algorithms and a working prototype for extracting basic content from large, diverse, remotely sensed images on the fly using desktop or laptop computers. This content includes both regions that occur naturally in images and arbitrary rectangles, ellipses, and long, straight shapes. We demonstrated an initial capability to represent the spatial relationships between these objects as semantic graphs. Several key advances in content extraction were documented and prepared for publication. An initial query-by-example prototype was developed that allows queries based on local image tile features, which is useful for classifying area characteristics such as human habitation.

Proposed Work for FY04

In FY04, we expect to demonstrate fundamental advances in both image-content extraction and query capabilities. We will extend tile-based image feature work to fully characterize its application to human habitation detection. Object-level detection will be developed for more complex objects. A shape calculus for boundary matching will be developed for detecting whole regions of specified shape. We will study the effect of the segmentation-coarseness parameter on the ability to detect objects of known size as whole regions. Semantic graph creation will be extended to full images and an initial graph-query capability will be demonstrated and characterized. Performance of all these algorithms will be analyzed for prototype problems relevant to LLNL mission areas.

Publications

Meyer, A. D., W. Paglieroni, and C. Astaneh. (2003). *K-means re-clustering: Algorithmic options with quantifiable performance comparisons*. Presented at SPIE Photonics West, Optical Engineering at LLNL, Jan. 2003. UCRL-JC-151259.

Paglieroni, D. W. (in press). "Design considerations for image segmentation quality assessment measures." *Pattern Recognition*. UCRL-JC-154917.

Paglieroni, D. W. (2003). *Coarseness regulation for segmented images*. UCRL-JC-154918.

Paglieroni, D. W. (2003). *A self-calibrating multi-band region growing approach to segmentation of single and multi-band images*. Presented at SPIE Photonics West, Optical Engineering at LLNL, Jan. 2003. UCRL-JC-150988.

Generalized Methods for Finite-Element Interfaces

Michael A. Puso 01-ERD-004

Abstract

The goal of this work was to develop improved methods for interfacing (i.e., tying and contacting) dissimilar meshes formed from meshed parts whose nodes do not coincide at a common boundary. Current mesh-tying methods are inaccurate and not a viable alternative to

a conforming mesh. Current contact methods only work for low-order elements and are not robust, particularly for implicit finite-element analysis.

The "mortar method" for connecting two-dimensional (2-D) flat interfaces was extended to connect arbitrary 3-D curved meshes in such a way that dissimilar meshing will not compromise solution accuracy. Our approach required novel techniques to integrate 3-D surface integrals involving Lagrange multiplier fields for traction and displacement and extending them to large deformation contact applications.

This new technology can save a significant amount of time and money invested in mesh production. For example, a large weapons model may take a month to build and may undergo many revisions over a period of years. Analysts estimate that an accurate method for tying dissimilar meshes could save 10–25% of the time invested in meshing over the course of a project. Furthermore, analysts spend a significant amount of time dealing with problems related to contact surfaces, particularly for implicit finite-element analysis. The new, more robust approach developed in this project should make analysis with contact surfaces more accurate, more reliable, and simpler. Finally, this technology allows the application of quadratic finite elements to contact problems, whereas the previous contact algorithms precluded them.

Mission Relevance

This project enhances LLNL's computational mechanics that support the Laboratory's stockpile stewardship mission. In particular, our algorithms will reduce the effort needed for weapons modeling by simplifying the development of finite-element meshes and running the analyses.

FY03 Results

In FY03 we worked on a mortar implementation that can connect meshes with different element types (e.g., tetrahedrals and quadratic elements). Now, automatically generated tetrahedral meshes can be accurately interfaced with hexahedral meshes. Friction was also added to the mortar contact. The new mortar contact provides vastly superior nonlinear convergence behavior by effectively smoothing the behavior near the interface and eliminating the "locking" associated with the standard contact methods. Consequently, contact analyses that failed in the past can be successfully analyzed using implicit finite elements with our new mortar approach.

Publications

Puso, M. A. (in press). "A 3-D mortar method for solid mechanics." *Int. J. Numer. Meth. Eng*. UCRL-JC-148546.

Puso, M. A. and T. A. Laursen. (in press). "A mortar segment-to-segment contact method for large-deformation solid mechanics." *Com. Meth. Appl. Mech. and Eng.* UCRL-JC-153341.

Puso, M. A. and T. A. Laursen. (in press). "A mortar segment-to-segment frictional contact method for large deformations." *Com. Meth. Appl. Mech. and Eng*. UCRL-JP-200667.

Higher-Order, Mixed Finite-Element Methods for Time-Domain Electromagnetics

Niel K. Madsen

01-ERD-005

Abstract

The popular Cartesian-grid finite-difference methods that are commonly used to solve Maxwell's equations in rectangular domains are not adequate for the complicated geometries in accelerators, radar remote sensing, photonics, and weapons. As a possible solution, researchers have studied finite-volume schemes, but these schemes suffer from numerical instabilities, lack of conservation, and low accuracy. To overcome these limitations, our project investigated higher-order, mixed finite-element methods (MFEM), a new methodology for solving partial differential equations on three-dimensional (3-D) unstructured grids.

Our goal was to advance the state of the art in MFEM used in electromagnetics. The primary deliverable was a stable, conservative, higher-order-accurate simulation code for Maxwell's equations and related equations of electromagnetism. This will enable electromagnetic simulations of unprecedented fidelity on complicated geometries that cannot be modeled using Cartesian grids.

Mission Relevance

Our MFEM methodology enables electromagnetic simulations on unstructured grids that conform to complex geometry. In addition, since the MFEM methodology is similar to methods in mechanical engineering, this research will lead to a coupled electrothermomechanical modeling capability in support of the Laboratory's national-security mission.

FY03 Results

Work in FY03 focused on (1) completing our prototype parallel, higher-order electromagnetic-application code and (2) evaluating this code by comparing computed results to well-known solutions. To demonstrate the efficacy of the code, we computed resonant frequencies and modes of an accelerator induction cell for the Stanford Linear Accelerator Center and computed the attenuation of radio-frequency waves propagating in a random rough surface tunnel. As another interesting application, we simulated substrate coupling effects in microwave integrated circuits. Possible future research activities in the area of computational electromagnetics include adaptive mesh refinement, improved absorbing boundary conditions, and coupled electrothermomechanical simulations.

Publications

Castillo, P., R. Rieben, and D. A. White. *FEMSTER: An object-oriented class library of discrete differential forms.* Presented at the 2003 Institute for Electrical and Electronics Engineers American Physical International Symposium. UCRL-JC-150238-ABS.

Catsillo, P. et al. (in press). "Discrete differential forms: A novel methodology for robust computational electromagnetics." *ACM Trans. on Mathematical Software.* UCRL-ID-151522.

Rieben, R., D. A. White, and G. Rodrigue. (in press). "Discrete differential forms for high-order finite-element vector bases." *IEEE Trans. Antennas and Propagation.* UCRL-JC-152292.

Rieben, R., D. A. White, and G. Rodrigue. (in press). "High-order symplectic integration methods for finite-element solutions to time-dependent Maxwell equations." *IEEE Trans. Antennas and Propagation.* UCRL-JC-152872.

Rieben, R., D. A. White, and G. Rodrigue. (2003). *Arbitrary-order hierarchical bases for computational electromagnetics*. Presented at the 2003 Institute for Electrical and Electronics Engineers International Symposium on Antennas and Propagation. UCRL-JC-150240.

White, D. A. and J. M. Koning. (2002). *A discrete-forms-based finite-element framework for wave equations*. Presented at the Soc. for Appl. Math. 50th Anniv. and 2002 Ann. Meet., Philadelphia, PA. UCRL-JC-147162-ABS.

White, D. A. and M. Stowell. (in press). "Full-wave simulation of electromagnetic coupling." *IEEE Trans. on Microwave Theory & Techniques*. UCRL-JC-155139.

Smart Nanostructures from Computer Simulation

Jeffrey C. Grossman 01-ERD-017

Abstract

Our project developed new computational tools that combine quantum Monte Carlo (QMC) and quantum molecular dynamics simulations to study the electronic, structural, and optical properties of semiconductor nanostructures. In particular, our project focussed on (1) surface structure and passivation effects, (2) ground- and excited-state optical properties, and (3) interactions between nanoclusters to form quantum molecules. Our simulations helped to interpret spectroscopic measurements. In addition, our simulations provided information—such as the effects of charging, shape changes, and different surfactant molecules on the optical properties of nanoparticles—that is not directly accessible in experiments. Through the development and successful application of these quantum simulation tools, the project has generated a new capability that can be applied to designing smart nanostructured materials.

Mission Relevance

Utilizing the computing platforms of Advanced Simulation and Computing Program, our project has developed new simulation tools for predicting the structural and optical properties of semiconductor nanostructures. The results of our research will enable the design of smart nanostructures with tailored physical properties, a critical technology for LLNL missions in countering biowarfare and bioterrorism and in biosciences to improve human health.

FY03 Results

In FY02, we applied our recently developed (in FY01) linear-scale QMC code to understand and predict optical properties of silicon nanostructures. We showed that certain contaminants on the surface such as oxygen, which is present in silicon quantum dot experiments ongoing here at LLNL, can have an enormous impact on resulting optical properties. In FY03, we took this approach several steps further by calculating both absorption and emission properties of these nanostructures. Furthermore, using a combination of ab initio molecular dynamics to model the synthesis coupled with our linear scaling QMC code to accurately predict optical properties, we simulated the actual synthesis process of silicon quantum dots. Our results showed that, quite surprisingly, even amorphous silicon quantum dots have strong quantum confinement effects and are capable of emitting visible light.

Publications

Benedict, L. X. et al. (2003). "Calculation of optical absorption spectra of hydrogenated Si clusters: Bethe-Salpeter equation versus time-dependent LDA." *Phys. Rev. B* **68**, 085310. UCRL-JC-150403

Draeger, E. and D. M. Ceperley. (2003). "Superfluidity in a doped helium droplet." *Phys. Rev. Lett.* **90**, 065301. UCRL-JC-147902.

Draeger, E. et al. (2003). "Reconstruction and synthesis processes in silicon nanoclusters." *Phys. Rev. Lett.* **90**, 167402. UCRL-JC-151272 Abs.

Draeger, E. et al. (2003). "Synthesis dynamics of passivated silicon nanoclusters." *Phys. Stat. Sol. B* **239**, 11. UCRL-JC-150588-REV-1.

Grossman, J. C. et al. (in press). "Towards an assessment of the accuracy of density functional theory for first-principles simulations of water." *J. Chem. Phys.* UCRL-JC-155397

Grossman, J. C., E. Schwegler, and G. Galli. (in press). "Structure of hydrophobic hydration." *J. Am. Chem. Soc.* UCRL-JC-151247.

Puzder, A. et al. (2003). "Computational studies of the optical emission of silicon nanocrystals." *J. Am. Chem. Soc.* **125**, 2786. UCRL-JC-151084.

Puzder, A. et al. (2003). "Structural stability and optical properties of nanomaterials with reconstructed surfaces." *Phys. Rev. Lett.* **91**, 157405. UCRL-JC-151250.

Williamson, A. J. et al. (2003). "Quantum Monte Carlo calculations of nanostructure optical properties: application to silicon quantum dots." *Phys. Rev. Lett* **89**, 196803. UCRL-JC-148678.

First-Principles Molecular Dynamics for Terascale Computers

Francois Gygi 01-ERD-044

Abstract

First-principles molecular dynamics (FPMD) is a simulation method that provides detailed information on the microscopic and electronic structure of materials. This method relies entirely on fundamental equations of quantum physics and therefore allows for predictive simulations of materials properties. In this project, we are exploring new algorithms and numerical methods used in FPMD simulations. We are also developing parallel implementations of FPMD methods that include these new algorithms.

The algorithms and implementations developed in this project will enable large-scale FPMD simulations on the Laboratory's largest computers. In particular, linear-scaling algorithms make it possible to reduce the computational complexity of FPMD from a cubic to a linear dependence on the number of atoms involved in the simulation. For this reason, they open the way to numerical simulations of physical processes occurring on length scales of several nanometers, which was previously inaccessible. Scalable implementations of FPMD will also reduce the time needed to perform simulations, thereby extending the time scale that can be simulated on large, parallel computers.

Mission Relevance

First-principles molecular-dynamics simulations have numerous applications in areas relevant to LLNL's national-security mission: calculations of the properties of high explosives, equation of state of molecular liquids, shock simulations, biochemistry, and nanotechnology.

The codes developed in this project have enabled the largest such simulations to date and contribute to LLNL's mission in breakthroughs in basic science.

FY03 Results

In FY03, we completed the implementation of a finite-difference linear-scaling code capable of microcanonical molecular dynamics simulations and tested it on several systems of interest to Laboratory missions. In particular, we quantitatively analyzed the error caused by the localization of electronic orbitals used in linear-scaling algorithms. We also developed a new implementation, in C++ and the Message Passing Interface, of a plane-wave pseudopotential method optimized for scaling on large numbers of CPUs. Near-optimal strong scaling of this new code was demonstrated in a simulation run on 1960 CPUs of the Multiprogrammatic Capability Resource computer recently installed at LLNL. This implementation will serve as the basis for the development of FPMD codes for the future BlueGene/L platform.

Publications

Fattebert, J.-L. and M. Buongiorno Nardelli. (2003). "Finite difference methods for ab initio electronic structure and quantum transport calculations of nanostructures." *Handbook of Numerical Analysis—Volume X: Special Volume: Computational Chemistry*. UCRL-JC-147118.

Gygi, F., J.-L. Fattebert, and E. Schwegler. (2003). "Computation of maximally localized Wannier Functions using a simultaneous diagonalization algorithm." *Computer Physics Communications* **155**, 1. UCRL-JC-151591.

Computational Methods for Collisional Plasma Physics

Dennis W. Hewett 01-ERD-073

Abstract

Our goal is to develop the capability to simulate plasmas in the regimes spanning from collisionless kinetics to collisional fluids. Although existing codes model each extreme of collisionality, this important intermediate regime is neglected. We will model the physics of various mean-free paths—from the very long, as in collisionless particle in cell (PIC), to the very short, as in hydrodynamics and diffusion. Such semicollisional physics arises in very-high-energy-density systems that are small (e.g., laser targets) or in low-density systems [e.g., high-altitude nuclear events (HANEs)]. Our project uses new adaptive-resolution, smart PIC algorithms [kinetically extended hydrodynamics (KEYDRO)] and collisional extensions of plasma PIC in our massively parallel three-dimensional (3-D) code, Z3.

Mission Relevance

Our project will enhance LLNL's competency in computational plasma physics and maintain LLNL's expertise and position at the forefront of plasma modeling. The computational models developed by our project will be applied to plasma problems—e.g, semicollisional fluid behavior in hohlraums, high-energy-density physics, HANEs, and electromagnetic phenomena—relevant to stockpile stewardship, in support of the national security mission.

FY03 Results

In FY03 we incorporated finite-size KEYDRO particles in our new partially collisional algorithms and connected this to a quasi-neutral electric field algorithm; benchmarked the 1-D and 2-D KEYDRO codes with known partially and fully collisional results; and continued to develop collision algorithms in Z3 for application to laser–plasma interactions using both high-power and short-pulse lasers. We used Z3 in the largest-scale simulations for which it has ever been used. One result was the first-ever identification of intense magnetic fields associated with the electron currents generated by stimulated Raman scattering.

Publications

Hewett, D. W. (2003). *Adaptive particle simulation: The next step in simulation*. Presented at the University of Kansas, Sep. 13, 2003, Lawrence, KA. UCRL-JC-153619 Abs.

Hewett, D. W. (2003). "Fragmentation, merging, and internal dynamics for PIC simulation with finite-size particles." *J. Comp. Phys.* **189**, 390. UCRL-JC-148321.

Hewett, D. W. and D. J. Larson. (2002). "Fragmentation, merging, and internal dynamics for collisional PIC simulation with finite-size particles." *Proc. 44th Ann. Meet. of the Div. of Plasma Phys.,* Nov. 11–15, 2002, Orlando, FL. UCRL-JC-148430 Abs. Rev.

Hewett, D. W. and D. J. Larson. (2003). "Fragmentation, merging, and internal dynamics for PIC simulation with finite size particles." *Proc. 18th Intl. Conf. on Numerical Sim. of Plasmas,* Sept. 7–10, 2003, Flamouth, MA. UCRL-JC-148321 Rev.

Larson, D. J. (2003). "A Coulomb collision model for PIC plasma simulation. " *J. Comp. Phys.* **188**, 123. UCRL-JC-148993.

Larson, D. J. and D. W. Hewett. (2003). "Modeling partially-collisional plasmas using finite-size particles with internal dynamics." *Proc. 18th Intl. Conf. on Numerical Sim. of Plasmas,* Sept. 7–10, 2003, Flamouth, MA. UCRL-JC-153204.

Larson, D. J. and D. W. Hewett. (2003). "Partially-collisional plasma simulation using complex particle kinetics." *Proc. 45th Ann. Meet. of the Div. of Plasma Phys.*, Oct. 27–31, 2003, Albuquerque, NM. UCRL-JC-153963 Abs.

Larson, D. J. and D. W. Hewett. (2002). "Progress in modeling collisional plasmas using particles with internal dynamics." *Proc. 44th Ann. Meet. of the Division of Plasma Physics*, Nov. 11–15, 2002, Orlando, FL. UCRL-JC-149395 Abs.

Lasinski, B. F. (2003). *Laser–plasma interaction studies with modern particle-in-cell codes for high-energy-density applications*. April 2003 Meet. of the Am. Phys. Soc., Apr. 5–8, 2003, Philadelphia, PA. UCRL-JC-151304 Abs.

Lasinski, B. F. et al. (2002). *Magnetic fields in laser light speckles*. UCRL-PRES-149780 Rev.

Lasinski, B. F. et al. (2003). *Raman-generated magnetic fields in laser light speckles*. Third Intl. Conf. on Inertial Fusion Sci. Appl., Sep. 7–12, 2003, Monterey, CA. UCRL-JC-152017 Abs.

Lasinski, B. F. et al. (2002). "Ion beams in short-pulse, high-intensity laser–matter interactions." *Proc. 44th Ann. Meet. of the Div. of Plasma Phys.,* Nov. 11–15, 2002, Orlando, FL. UCRL-JC-149086 Abs.

Still, C. H., B. F., Lasinski, and A. B. Langdon. (2002). "LPI simulations over an entire speckle volume with the PIC code Z3." *Proc. 44th Ann. Meet. of the Div. of Plasma Phys.*, Nov. 11–15, 2002, Orlando, FL. UCRL-JC-149208 Abs.

Still, C. H., B. F. Lasinski, and A. B. Langdon. (2002). *PIC simulation of LPI over an entire speckle volume in three dimensions*. 32nd Ann. Anomalous Absorption Conf., Jul. 21–26, 2002, Oahu, HI. UCRL-JC-148399 Abs.

Radial Reflection Diffraction Tomography

Sean K. Lehman

02-ERD-011

Abstract

This project is using diffraction-tomography techniques to develop a wave-based imaging algorithm. Our work focuses on two hardware configurations, multimonostatic and multistatic. The multimonostatic mode uses a single, rotating transducer that is oriented radially outward. At successive angular locations at a fixed radius, the transducer launches a primary field and collects the backscattered field in a "pitch/catch" operation. The hardware configuration, operating mode, and data collection method are identical to those of most medical intravascular ultrasound systems (IVUS). Based upon a straight-ray model of sound propagation, IVUS form images of the medium surrounding the probe. We use a Hilbert space inverse-wave algorithm to construct images. The multistatic mode consists of a single transmitter and an aperture formed by multiple receivers. In this latter case, the entire source/receiver aperture rotates about the fixed radius. Practically, such a probe is mounted at the end of a catheter or snaking tube that can be inserted into a part or medium with the goal of forming images of the plane perpendicular to the axis of rotation.

If successful, this research will develop a new and versatile method of inverse-wave imaging for nondestructive evaluation (NDE) and an improved IVUS system.

Mission Relevance

Inverse-wave imaging for NDE can be used to determine the status of parts and materials in the stockpile, in support of the Laboratory's national security mission, and for borehole imaging for waste management and remediation. An improved IVUS system will contribute to better clinical treatment of vascular and prostate disease, in support of the Laboratory's mission in bioscience to improve human health.

FY03 Results

In FY03, we developed a Hilbert space inverse-wave reconstruction code and successfully reconstructed radial reflection data from various finite-difference time-domain simulations. We determined that radial resolution is frequency dependent and that azimuthal resolution is dependent upon the number of receivers in the multistatic subaperture.

Publications

Lehman, S.K. and S. J. Norton. (2003). *Radial reflection diffraction tomography.* UCRL-TR-200707.

Automated Imagery Data Exploitation

John A. Futterman

02-ERD-034

Abstract

We propose to develop techniques for automated broad-area search of satellite imagery. As a prototype application, we propose to develop and test algorithms for automated recognition of human settlements in satellite imagery by extracting textural features from IKONOS 1-m

panchromatic imagery and fusing them with color features extracted from IKONOS 4-m multispectral image data. To account for geocultural and seasonal variation, we will segment the world into regions according to patterns of manmade structures, for which we will develop tuned parameter sets to control our algorithms.

This project will result in tools that can be adapted to broad-area searching of image databases, as well as to automating the Counterproliferation Analysis and Planning System (CAPS) population database production and the global demography and resource management database production.

Mission Relevance

This project will directly benefit LLNL's national security and homeland security missions in the areas of nonproliferation and counterterrorism by enabling broad-area search and area monitoring of general image databases, automation of consequence analysis population map production, production of global demography and resource management databases, and monitoring for signatures of interest to intelligence and law-enforcement communities.

FY03 Results

In FY03 we developed a plan to characterize the geographic and cultural sensitivity of our feature extraction and classification techniques. As a result of our work, we presented two SPIE (International Society for Optical Engineering) conference papers and submitted one Record of Invention, which DOE is pursuing for a possible patent.

Publications

Kamath, C. et al. (2003). "On the use of machine vision techniques to detect human settlements in satellite images." *SPIE Proceedings* **5014**. UCRL-JC-150218.

Sengupta, S. K. et al. (2003). "Detecting human settlements in satellite imagery. "*SPIE Proceedings* **5001**, 76. UCRL-JC-150220.

Atomically Controlled Artificial and Biological Nanostructures

Giulia A. Galli 02-ERD-043

Abstract

Nanotechnology holds great promise for industry, medicine, and national security, but key physical science at the nanometer scale must first be understood. Using advanced simulation and experimental techniques, we are investigated the structural, optical, and transport properties of semiconductor nanostructures with desired labeling and sensing properties. We focused on silicon and germanium quantum dots, which offer technical advantages such as biocompatibility, and used ab initio simulation techniques to model interfaces between these quantum dots and specific chemical and biological media.

Ab initio simulation techniques to model interfaces between semiconductor nanoparticles and organic molecules in solution will help guide and interpret characterization and functionalization experiments, as well as experiments to synthesize new biocompatible materials with the desired size and stoichiometry. Coupling simulation with state-of-the-art experimental techniques will result in a new capability for designing semiconductor nanostructures with desired labeling and sensing properties, such as pathogen bioassays.

Mission Relevance

The scientific capabilities developed in this project enhanced the development of novel sensing and labeling devices to address specific problems in national security and environmental monitoring. This work is primarily intended for the development of rapid field analysis in support of efforts to counter chemical and biological weapons.

FY03 Results

In FY03, we set up two new laboratories, one to carry out synthesis of class II–VI semiconductor nanoparticles and the other for proof-or-principle characterization of the surfaces of silicon nanoparticles. The project resulted in several publications.

Publications

Galli, G. et al. (2003). *First-principles simulations of semiconductor nanostructure*. Presented at the ACS Meeting, New Orleans, LA, March 23–27, 2003. UCRL-JC-151198-ABS.

Galli, G. et al. (2003). *Structural and electronic properties of semiconductor nanostructure from first-principles calculations*. Presented at the APS Meeting, Austin, TX, March 3–7, 2003. UCRL-JC-151278-ABS.

Grossman, J.C. and L. Mitas. (2003). *Dynamical Quantum Monte Carlo simulations*. Presented at the ACS Meeting, New York, NY, Sept. 7–11, 2003. UCRL-PRES-200891.

Raty, J-Y. and G. Galli. (in press). "Nanodiamonds." *Encyclopedia of Nanoscience and Nanotechnology*. UCRL-JC-153266.

Wagner, L. et al. *The structure and Stokes shift of hydrogenated silicon nanoclusters*. UCRL-JC-148679.

Chemical Dynamics at Interfaces

Eric R. Schwegler
03-ERD-001

Abstract

We propose to expand the application of ab initio simulation methods to study the complex interfaces in high-pressure physics at the microscopic level. We will build on our existing expertise in first-principles molecular dynamics and path-integral simulation methods by incorporating two-phase simulation techniques. These simulation methods will be applied to a set of low-*Z* materials that are relevant to laser experiments at large-scale laser facilities and to systems that can be directly compared to new spectroscopic measurements of interfacial properties.

The primary benefit of this project will be the continued development of new computational tools and expertise for simulating interfacial systems at the microscopic level. The techniques that will be developed will be general and have widespread applicability to many scientific problems in physics, chemistry, and biology. We also expect that results obtained for all of the proposed systems will lead to high-profile publications.

Mission Relevance

The work described in this proposal will support LLNL missions in national and energy security by providing a fundamental understanding of the changes materials undergo as a function of pressure, melting, and phase transitions. This understanding will particularly benefit the Stockpile Stewardship Program.

FY03 Results

In FY03, we successfully developed the methods needed to perform two-phase simulations in a first-principles molecular dynamics context. This work represents the first-ever demonstration that the two-phase simulation approach is computationally feasible at a first-principles level. As a first application, we computed the melt curve of lithium hydride (LiH) under pressures ranging from ambient to 200 GPa and investigated dynamical and electronic properties of the fluid under high temperatures and pressures. A paper describing this work has recently been published in *Physical Review Letters*. In addition, we started to map the melt curve of hydrogen under pressure; preliminary results indicated that a maximum in the hydrogen melt curve exists at pressures near 130 GPa.

Proposed Work for FY04

We will (1) continue to develop new capabilities for investigating interfacial systems, i.e., ab initio molecular-dynamics-based two-phase techniques and coupled two-phase–path-integral molecular dynamics; (2) continue to generate new data on the equation of state of LiH under pressure and examine phase transitions in the solid, also extending the computed melt curve of LiH from 200 to 1000 GPa; (3) perform two-phase simulations of hydrogen to determine whether a maximum exists in the melt curve; (4) examine the phase diagram of diamond and liquid carbon at temperatures and pressures relevant to ongoing laser shock experiments at the Omega facility; and (5) simulate water at pressures of 50 GPa to investigate the possibility of a superionic phase upon melting.

Publications

Bonev, S. A., B. Militzer, and G. Galli. (in press). "Dense liquid deuterium: Ab initio simulations of states obtained in gas gun shock experiments." *Phys. Rev. B*. UCRL-JC-153551.

Bonev, S. A. et al. (2003). "The high-pressure molecular phases of solid carbon dioxide." *Phys. Rev. Lett.* **91**, 65501. UCRL-JC-151202.

Bonev, S. A. et al. (2003). The high-pressure molecular phases of solid carbon dioxide. Ann. Am. Phys. Soc. 2003 March Meet., Mar. 3–7, 2003, Austin, TX. UCRL-JC-151202 Abs.

Ogitsu, T. et al. (2003). First-principle study of melting curve of lithium hydride under pressure. Ann. Am. Phys. Soc. 2003 March Meet., Mar. 3–7, 2003, Austin, TX. UCRL-JC-151272 Abs.

Ogitsu, T. et al. (2003). *Melting curve and microscopic property of lithium hydride under pressure*. 2003 Study of Matter at Extreme Conditions, Mar. 24, 2003, Miami, FL. UCRL-JC-151248 Abs.

Ogitsu, T. et al. (2003). "Melting of lithium hydride under pressure." *Phys. Rev. Lett.* **91**, 175502. UCRL-JC-153550.

Correction of Distributed Optical Aberrations

Scot S. Olivier

03-ERD-006

Abstract

The objective of this project is to demonstrate the use of multiple distributed deformable mirrors (DMs) to improve the performance of optical systems with distributed aberrations. This concept is expected to provide dramatic improvement in the optical performance of systems in applications in which the aberrations are distributed along the optical path. Our approach uses multiple DMs that are distributed to match the aberration distribution. The project will simulate the performance of these multiple DM systems, develop a test bed to evaluate distributed correctors matched to the distributed aberrations, conduct experiments, and compare experimental results to the predictions of detailed modeling. With these results, we will develop the underlying theoretical and experimental capabilities for correcting distributed optical aberrations, which will significantly increase the corrected field of view for tactical or surveillance imaging scenarios over long horizontal or slant paths.

Mission Relevance

The development of advanced techniques for correcting distributed aberrations will support new programs in remote sensing and imaging applications for the Laboratory's national-security and homeland-security missions. It will also enable forefront astronomical science on the next generation of giant telescopes, furthering LLNL's mission in breakthrough science and also helping to attract the next generation of top scientists and engineers.

FY03 Results

In FY03, we completed development of the simulation tool and upgraded the microelectromechanical systems adaptive optics test bed to provide capabilities for distributed turbulence tests and installed a single DM. The initial performance test of the system showed the corrected Strehl ratio to be 0.44.

Proposed Work for FY04

We plan to upgrade the test bed with multiple DMs and distributed turbulence, use it to measure the performance of multiple-DM wavefront correction of distributed aberrations, and quantify the increase in corrected field of view and the effects of scintillation on system performance. The experimental results will be used to validate our computational models of the systems, and the validated model will be used to extend performance prediction into realistically scaled, relevant scenarios.

Techniques for Judging Intent Behind Cyber Attacks

John Allen

03-ERD-012

Project Description

The ability to detect serious, network-based cyber attacks by intruders, discriminate attacks from background chatter, and respond to attacks as quickly as possible is a key cybersecurity concern. However, research institutions like the Laboratory rely on collaborations with outside researchers. The vitality of the these scientific collaborations can be enhanced by security measures that include flexible responses to attacks rather than relying exclusively on barriers that apply to both attacks and collaborations. This project will research and develop algorithms and build a prototype expert system that can identify an attacker and engage the attacker with a controlled but unexpected response. By measuring the attacker's reactions to our response, the expert system will be able to evaluate the intent behind the attack quickly enough to deploy an effective tactical defense.

Expected Results

We expect to develop a proof-of-concept system that implements algorithms to supply attackers with controlled but unexpected responses to their probes, to measure the attacker's reactions to that specially crafted information, and to produce scaling information to estimate the feasibility and costs of a larger system capable of protecting a site with multiple Class B networks (those with up to 65,535 devices each). If successful, this project will prove our hypothesis about the effectiveness and practicality of such a system and the practicality of deploying such a system. This research could result in a new type of active cyber defense.

Mission Relevance

A secure, unclassified computing environment at the Laboratory would support all of the Laboratory's missions at an institutional level and contribute to the cybersecurity of the nation. In addition, the ability to conduct secure, open scientific collaborations would bring additional benefits to a wide range of scientific programs at the NNSA laboratories.

FY03 Results

In FY03, we implemented the most promising of the methods for determining attacker intent in our prototype system. That system scaled well past our goal of a Class C network (up to 255 devices) to a network at least 10 times as large. We tested our system with known hacker tools, and it responded to attacks that demonstrated its usefulness for a tactical defense. Our system was calibrated by testing it against intrusion-attempt data collected at LLNL. We published a report on the feasibility and potential success of a future, full-sized system based on this work. At the very end of the year, we performed a short, controlled, double-blind study that confirmed the effectiveness of our approach.

Publications

Allen, J. (2003). *Techniques for judging intent behind network-based cyber attacks*. UCRL-TR-202494.

Propagation Models for Predicting Communication System Performance in Tunnels, Caves, and Urban Canyons

Hsueh-yuan Pao 03-ERD-023

Abstract

Current wireless radio frequency (RF) communications cannot operate reliably in enclosed environments such as caves, tunnels, and urban canyons because the rough or complex

surfaces of these environments compromise signal quality. In addition, RF propagation in these environments is poorly understood. This project will address this RF communications problem by characterizing the propagation of electromagnetic (EM) fields in such adverse environments. Our overall goals are to (1) understand the physics of propagation in rough enclosures, such as tunnels and caves; (2) develop models that can use the surface statistics of these enclosures to describe the EM field statistics; and (3) investigate the possibility of extending this work to the urban-canyon environment.

Our analytical work is focusing on (1) propagation of wave-guide modes in a rough-walled quasicircular perfect electric conductor (PEC) waveguide; (2) calculation of the fields resulting from a horizontal electric dipole in a rectangular tunnel with two smooth PECs and two smooth, lossy walls; (3) propagation of waveguide modes in a rough-walled tunnel, using an equivalent surface impedance to model the wall roughness; and (4) calculation of the fields produced in a circular smooth-walled tunnel by circularly symmetric electric and magnetic current loops.

Mission Relevance

This project supports LLNL's national-security mission areas of nonproliferation and homeland security by providing the knowledge to develop the technology for wireless communications in tunnels and caves, with a possible extension to urban canyons.

FY03 Results

In our work on modal propagation in rough-walled waveguides, we found that the total modal field can be decomposed into the sum of a coherent (deterministic) part and an incoherent (zero-mean random) part. The modal cutoff frequencies are only slightly affected by the wall roughness; furthermore, evidence suggests that certain waveguide modes can become slow above a critical frequency. The auto- and cross-covariance functions that describe the incoherent part of the field can be expressed in terms of the power spectral density of the surface roughness. The work has led to two papers presented at conferences.

Proposed Work for FY04

In FY04 we will continue the analytic work on tunnels, focusing on the start-ray representation, which is good for big caves and curved tunnels, and on including the source in the modal approach. We will also begin analytical work on urban canyons by demonstrating a basic algorithm for urban canyon problems. We will start with simple, two-dimensional PEC buildings and employ plane wave and line sources. Alternative numerical approaches for tunnels will also be investigated. The best approach (in terms of tradeoffs) will be determined and compared with the models developed. Model approximation will be quantified by comparing with full wave simulations.

Publications

Pao, H.-Y. (2004). *Probability density function for waves propagating in a straight, rough wall tunnel.* UCRL-PROC-202082.

Computation of Hypersonic Flow around Maneuvering Vehicles with Changing Shapes

Fort F. Felker

03-ERD-026

Abstract

Ballistic missile re-entry vehicles and missile-defense intercept vehicles moving at hypersonic speeds play crucial roles in national-security efforts. During flight, re-entry and intercept vehicles maneuver and experience shape changes: re-entry vehicles maneuver due to flight dynamics and change shape through ablation and structural deformations; intercept vehicles maneuver to hit targets and change shape through structural deformations and possibly control-surface deflections. A robust simulation of these critical load environments requires a numerical formulation incorporating the necessary effects including mesh deformations as the fluid domain conforms to structural shape changes, and non-inertial reference frames as the local fluid domain moves with the vehicle.

We plan to develop a numerical formulation for computational analysis of the hypersonic flow around maneuvering, deforming vehicles. The subsequent computer program will be documented with theory and user's manuals and will operate on large, parallel Advanced Simulation and Computing Program-class machines. Verification and validation calculations will confirm the reliability of the completed analysis, and the new capability will be described in journal articles and conference papers.

Mission Relevance

Qualification of the nuclear experimental package within the dynamic flight loads is an NNSA responsibility. Once developed, our new computational-analysis method can be immediately applied to work in LLNL's Stockpile Stewardship Program and would allow flight loads to be computed on deforming re-entry vehicles for the first time. These flight loads would incorporate important real-world effects, such as blast-wave encounters and transient flight dynamics.

FY03 Results

Considerable numerical formulation was conceptualized, derived, and documented in a draft 50-page theory manual. This initial effort concentrated on the deforming, non-inertial mesh considerations in the context of inviscid flows. We made contacts in the academic community to seek an independent review of this work and anticipate a future publication. This has established a baseline from which to (1) augment the numerical formulation to consider effects of viscosity and other diffusive mechanisms and (2) add reactive chemistry to enable hypersonic flow simulation.

Adaptive-Mesh-Refinement Algorithms for Nonlinear Structural Deformations using Parallel, Unstructured Finite-Element Codes

Ian D. Parsons

03-ERD-027

Abstract

This project will produce algorithms and software for adaptive-mesh-refinement (AMR) methods used to solve practical solid-mechanics and electromagnetics problems on multiprocessor, parallel computers using unstructured finite-element meshes. The goal is to provide computational solutions that are accurate to a prescribed tolerance; adaptivity is

the key to this goal. The new tools will enable analysts to conduct more reliable simulations at reduced cost, in terms of both analyst and computer time. Research issues include effective error estimators for nonlinear structural mechanics and electromagnetics, local meshing at irregular geometric boundaries, and constructing efficient software for parallel-computing environments.

Our approach is to incorporate AMR tools into state-of-the-art engineering codes, such as Diablo, which is used for solid mechanics, and Eiger and EMSole for electromagnetics. All of these codes are based on unstructured-mesh, finite-element methods and will require similar algorithms for tasks such as mesh refinement and load balancing. Deliverables include the components of AMR techniques applied to unstructured finite-element discretizations, such as treatment of irregular geometries, constraint enforcement, intermesh transfer of state variables, and partitioning of adaptively refined domains.

Mission Relevance

Our project will support LLNL's national-security mission by enhancing our ability to simulate mission-relevant events, including analyses that will benefit the Stockpile Stewardship Program. Our robust, scalable AMR capabilities with simplified mesh generation and lower computational demands will increase analysts' productivity, and model verification will be significantly easier and more transparent by combining error analysis and refinement.

FY03 Results

Basic serial AMR capability was added to an object-based structural-mechanics code. This includes the data structures and algorithms required to refine a user-defined mesh and error estimators based on patch-recovery techniques and residual computations. Simple test problems were constructed and used to confirm that the AMR implementation is able to perform both isotropic and anisotropic mesh refinement, as well as mesh derefinement. Anisotropic refinement (which is based on a directional error estimator) and derefinement are crucial for solving highly transient problems and implicit analysis, in which the cost of solving equations can increase dramatically with problem size.

Proposed Work for FY04

In FY04, we will develop and implement error estimators for solving problems in the domains of interest, and continue building the necessary software infrastructure. This work is divided into two categories: solid mechanics and electromechanics. In solid mechanics, we will (1) complete the implementation of error-estimator and mesh-interpolation procedures, assess their performance, and make appropriate improvements, and (2) start the parallelization of the serial AMR capability developed in FY03. In electromagnetics, we will (1) develop error estimators in cooperation with researchers at the University of Texas at Austin and (2) build polynomial-degree-refinement procedures built on hierarchical bases.

Publications

Parsons, I. D. and J. Solberg. (2003). *Adaptive implicit multimechanics simulations with Diablo.* 5th Biennial Tri-Laboratory Engineering Conf., Oct 21–23, 2003, Santa Fe, NM. UCRL-JC-154055 Abs.

Parsons, I. D. and J. Solberg. (2003). *Adaptive implicit multimechanics simulations with Diablo.* 5th Biennial Tri-Laboratory Engineering Conf., Oct 21–23, 2003, Santa Fe, NM. UCRL-PRES-154055.

Entity-Based Simulation of Biological Systems

Andrew J. Cleary

03-ERD-030

Abstract

This project will develop tools and expertise in scalable entity-based modeling (EBM) of complex population-based systems. Our initial application will be national-security problems such as the spread of infectious disease and bioterrorism response models. In the academic community, discrete simulation technology based on individual entities has shown promise for these kinds of complex systems, but the technology has not been scaled to the problem sizes or computational resources of LLNL, and overall the technology is immature. Our emphasis will be on showing that this technology can be extended to parallel computers and to larger problems, as well as overall maturation of the technology from an academic to a more applied setting.

This project represents one of the few viable technologies for understanding highly nonlinear, complex, chaotic systems. In many ways, the Lab's computational groups have only tackled the easiest of real-world phenomena: those that behave regularly and predictably. Recent events show us, however, that there are many important national security-related phenomena that are not so well behaved. If we are successful, we will provide the Lab a powerful tool for understanding, predicting, and controlling such phenomena. We expect this kind of model to be used as an "in silico" laboratory in which analysts conduct "what if" experiments to assess, for instance, potential hostile tactics, U.S. responses to and strategies for those tactics, and the efficacies of each.

Mission Relevance

Our project is directly relevant to LLNL's national-security mission, because EBM is a potential core computational capability with application to a range of multidisciplinary problems, such as simulating disease transmission in populations. We are tailoring our technology for homeland-security applications such as bioterrorism-response models.

FY03 Results

In FY03 we produced a sequential epidemiological model and extended this via message-passing interface (MPI) to a modest level of parallelism. Based on this result, we have developed an innovative, scalable architecture design based on three new advances: hierarchical multiresolution modeling, time windowing, and a new EBM abstraction layer. At present, we have demonstrated scalability an order of magnitude greater than generic EBS tools, and exceeding even specific EBS tools. We also conducted an analysis of existing technologies and research-community building. Lastly, we added the development of new biological models for disease transmission and immune systems.

Proposed Work for FY04

We will conduct performance analysis on, and improve, our epidemiological model, documenting the moderate-level scalability that we predicted from our first generation of tools. We will also produce several technical reports detailing the architectural and conceptual concepts that form the basis of our second-generation, massively scalable toolkit.

Detection and Tracking in Video

Chandrika Kamath

03-ERD-031

Abstract

Detecting and tracking moving objects in video is difficult because of changes in illumination, moving trees, and shadows. Further problems arise if the video is taken with a moving camera, in adverse conditions such as fog, or at a low resolution or frame rate. In this project, we are developing robust, real-time techniques for detecting and tracking moving objects in low- to moderate-resolution video taken with a stationary camera. This allows us to model the interactions of objects, identify normal patterns, and detect unusual events. Our algorithms and software will include techniques to separate the moving foreground from the background, extract features representing the foreground objects, and track these features from frame to frame, followed by postprocessing to smooth the tracks. The capability developed in this project will also help determine the limitations of current algorithms and the requirements for data collection, and can also be applied to spatio-temporal data from computer simulations.

Mission Relevance

The capability to detect and track in video supports LLNL's national-security mission by enabling new monitoring and surveillance applications relevant to homeland security. By analyzing tracks, we can model the activity at a scene, as in counter-terrorist applications, for instance. This work can also be used to track objects in simulation data and to mine text, audio, image, and video data simultaneously, including newer forms of media.

FY03 Results

In FY03, we identified realistic videos of traffic intersections taken under different conditions, designed a software infrastructure to handle color video, implemented several techniques for background subtraction, and evaluated their performance on our test videos. We found that a simple approximate median was an accurate and efficient background model. Our research on block-matching techniques showed them to be effective and that they work well with moderate-resolution, moderate-frame-rate video. By keeping a frame history, we were able to handle small camera motion. We implemented simple schemes to calculate the velocity and extract features for moving objects. We also collaborated with the University of Colorado, Boulder, on a capability for tracking people.

Proposed Work for FY04

In FY04, we will develop more accurate techniques to extract features such as velocity and address the problem of tracking the objects from frame to frame. This will enable us to apply the first three steps of the tracking process to all our videos and determine the combination of methods that works best. We will also investigate how the techniques carry over to lower-resolution, lower-frame-rate data.

Scalable, Multiple-Sequence Alignment for Pathogen Signatures

David A. Hysom

03-ERD-032

Abstract

This project proposes to investigate and develop scalable parallel algorithms for multiple-sequence alignment (MSA), the preferred method for analyzing multiple instances of genome data to determine which DNA fragments are potential signatures for pathogen detection. However, current MSA algorithms fail for large viral pathogens or bacterial pathogens. Laboratory biodefense efforts need MSA tools that scale to tens of millions of DNA base pairs, which is more than two orders of magnitude larger than current state-of-the-art approaches.

The MSA algorithms developed in this project will address problems of this size through improved scalable algorithms and the Laboratory's high-performance parallel computers. The project will research and implement MSA algorithms that preserve robustness in the presence of noisy incomplete genomic data while still scaling to large sequences on massively parallel computers. In addition, we plan to investigate MSA algorithms that address the unique genome rearrangements in bacteria and implement our parallel MSA algorithms as a software suite of DNA sequence-analysis tools.

Mission Relevance

Laboratory pathogen-detection efforts need sequence-alignment tools that scale to tens of millions of base pairs and larger. Completion of our proposed parallel software tools would allow laboratory researchers to perform highly complex analyses not only of related organisms, but also across multiple pathogens whose only connection might be a set of shared virulence-mechanism genes. Development of a scalable MSA tool for genome analysis will support the Laboratory's growing mission in biodefense.

FY03 Results

We have developed novel algorithms for aligning multiple drafts (i.e., isolating contig sets) with a reference genome. These algorithms have been implemented within our Python and C++ infrastructure and have been incorporated into the KPATH DNA signature pipeline. We also made improvements in an existing multiple genome alignment code, permitting an order-of-magnitude increase in speed.

Scalable Discretization-Enhanced Solvers

Jim E. Jones

03-ERD-033

Abstract

We propose to develop scalable, optimal linear system solvers for discretizations of an important class of partial-differential equations (PDEs) that are handled poorly by existing linear system solvers. These PDEs are used in accurate modeling of many phenomena studied at the Laboratory, particularly electromagnetics. The performance of codes for these applications can be adversely affected by poor performance of the linear system solver. Our development of scalable, optimal linear system solvers for this class of PDEs will enable faster, more realistic

simulations. To develop novel multigrid algorithms, we will exploit discrete representations of the large, oscillatory near nullspaces that characterize these PDEs.

Our goal is to extend this linear solver capability to a whole new class of PDEs, which in turn will impact a large set of the Lab's physics codes. As a specific example, a scalable algorithm for implicit Maxwell's equations will enable the application code EMSolve to solve problems that are orders of magnitude larger—involving greater accuracy in the computed solution—than can currently be solved, significantly enhancing the Lab's capability in electromagnetics design and analysis.

Mission Relevance

These PDEs arise in several LLNL applications. Helmholtz equations arise in light-propagation models for laser–plasma applications and in single-frequency source models for accelerator design. Maxwell's equations arise in broadband-frequency sources for accelerator design and optical scattering. Extending this linear-solver capability to linear systems that result from discretization of these equations will impact physics codes that will support several LLNL missions, including stockpile stewardship.

FY03 Results

We developed a two-dimensional (2-D) protoype Maxwell code using structured Nedelec finite elements, the elements used in the LLNL application codes EMSolve and EIGER. For time-domain Maxwell, we implemented a multigrid solver with overlapping box relaxation to eliminate troublesome gradient-error components. In testing, this algorithm scaled well. For frequency-domain Maxwell's equations, we added overlapping box Kaczmarz relaxation with additional processing to eliminate troublesome plane-wave error components. We began work to improve the performance of this method. In addition, we have formulated an algebraic multigrid algorithm for the time-domain Maxwell equation discretized on unstructured meshes. Early testing on structured model problems yielded results as good as those obtained with the geometric multigrid method. All implementation and testing in FY03 was for unstructured grids.

Proposed Work for FY04

Our goals for FY04 are to implement an algebraic multigrid (AMG) solver for the time-domain Maxwell's equation; apply compatible relaxation and the generalized AMG framework to the time-domain Maxwell's equation. These two goals will require testing and development of unstructured, algebraic analogues of the techniques explored in FY03 for time-domain Maxwell on structured grids. In addition, we expect to develop operator-based interpolation approaches for the variable-coefficient Helmholtz equation on structured grids and investigate 3-D extensions of existing multigrid methods for the Helmholtz equation on structured grids.

Publications

Falgout, R. F. and P. S. Vassilevski. (2003). *On generalizing the AMG framework.* UCRL-JC-150807.

Surveillance, Prediction, and Insight for Decision Making for Earliest Response (SPIDER)

William D. Wilson

03-ERD-046

Abstract

This project focuses on developing the System for Prediction and Insight for Decision Making for Earliest Possible Response (SPIDER), an integrated system for responding to a national crisis such as an outbreak of smallpox in the human population or foot-and-mouth disease among animals. The scope of this project is very broad: development of computational methods to handle the complexities of large amounts of data that are disparate in nature. A working example will be studied here in detail and applied to data from Lackland Air Force Base (AFB), which has an anomalously high rate of a respiratory flu. The example will demonstrate a methodology for learning the fundamental science necessary to understand and control disease outbreaks. Graph techniques and entity-based modeling are coupled to Markov chain Monte Carlo (MCMC) methods to form an integrated system.

The primary scientific achievements of this project will be the development and integration of three classes of algorithms: (1) tractable predictive models; (2) methodologies for estimating the corresponding model parameters; and (3) model-validation techniques. The resulting SPIDER system would then be employed to characterize fundamental underlying principles such as disease transmission mechanisms. This knowledge exemplifies the value added by the approach and demonstrates that these three classes of algorithms can be combined seamlessly, each bringing its own strengths to bear on a single challenge.

Mission Relevance

This work supports the national-security mission of the Laboratory by providing the fundamental scientific basis for an Advanced Simulation and Computing Program-level system capable of surveillance, prediction, and optimized response to a national crisis. Such a capability can be used to prevent such a crisis and protect our national resources.

FY03 Results

We demonstrated that three algorithms (SGraphs, EbM, and MCMC) could work together synergistically to find the path of transmission of an infectious disease and predict its future behavior. An initial inversion was employed to construct a location set of reduced dimensionality for the forward model. This method directly addresses scalability to large systems by reducing the forward model presented to the MCMC. For a three-location problem, we were able to invert using MCMC to obtain the most probable disease-transmission coefficient, including confidence levels. We applied SPIDER to actual disease outbreak data from Lackland AFB, inverting for the disease path using MCMC, which agreed with the data.

Publications

Wilson, W. D. (2003). *SPIDER fact sheet.* UCRL-MI-200305.

Threat Analysis and Optimal Countermeasure Allocation

Thomas A. Edmunds

03-ERD-052

Project Description

Infrastructure networks are at risk of disruption by well-engineered and coordinated terrorist attacks. Countermeasures such as hardening targets, acquiring spare critical components, and conducting surveillance can detect, deter, and counteract these attacks. Allocation of available countermeasure resources to sites or activities in a manner that maximizes their effectiveness is a challenging problem. This allocation must take into account for adversary response after countermeasure assets are in place—the adversary may simply switch strategies to avoid countermeasures. Stockpiling spares of critical infrastructure components has been identified as a key element of a grid infrastructure defense strategy in a recent National Academy of Sciences report.

We propose to build a three-level game-theory optimization model of an attacker–defender interplay in which the target is an infrastructure network. This three-level opimization problem is an extension of bilevel models described in game theory. At the first level, the network defender acquires spares and deploys countermeasures to deter or mitigate consequences of the attack. At the second level, the attacker observes the defender's strategy and executes an optimized attack that maximizes impact. At the third level, the defender reroutes commodities on the network to minimize consequences of the attack. The technology developed in this project will interface with semantic graph models to extend in-house expertise and software to achieve more comprehensive threat-analysis capability than that provided by currently available techniques.

Mission Relevance

These technologies will support the Department of Homeland Security's mission to identify threats to U. S. assets and the DOE's mission to monitor development of weapons of mass destruction technologies and materials. The work will further develop Markov chain Monte Carlo technology to solve a new class of problems involving network structures. It will also interface with semantic graph models for a comprehensive analysis of threats.

FY03 Results

Code was developed to accept input describing the network topology and loads, countermeasure costs, and budget constraints on the defender and attacker. The code formulates the equivalent mixed-integer linear programming problem and executes a sequence of set-covering algorithms to find optimal attack and defense strategies in conjunction with a commercial linear optimization package to derive an optimal consequence-mitigation strategy. The code was demonstrated on small-scale problems involving electrical grids.

The algorithm was developed and demonstrated on small-scale electricl networks (33 generators and 38 lines.) The algorithm would need to be scaled up to a least hundreds of lines and generators to provide a practical tool for analysis of electrical networks at a useful level of aggregation. A Record of Invention has been filed.

ViSUS: Visualization Streams for Ultimate Scalability

Valerio Pascucci

02-ERI-003

Abstract

This project is developing a suite of progressive visualization algorithms and a streaming infrastructure for interactive exploration of large scientific data sets. The methodology optimizes the data flow in a pipeline of processing modules. Each module reads and writes a multiresolution representation of a geometric model, providing the flexibility to trade speed for accuracy, as needed. The data flow is streamlined with progressive algorithms that map local geometric updates of the input into immediate updates of the output. A prototype streaming infrastructure will demonstrate the flexibility and scalability of this approach for visualizing large data sets on a single desktop computer, a cluster of personal computers, and heterogeneous computing resources.

The ViSUS project will benefit scientific computing on two levels. At the deployment level, the improved efficiency in the use of hardware resources will reduce the cost of visualization-hardware infrastructures. At the scientific level, the developed technology will reduce the overall time required for the design, simulation, and visualization cycle. The ability to monitor large and expensive simulations remotely will save computing resources through early termination and restart of erroneous test simulations, for instance. Runtime steering will be possible for simulation codes with mechanisms for dynamic modification of running conditions.

Mission Relevance

Use of our innovative, high-performance visualization techniques will allow interactive display of very large data sets on simple desktop workstations and the monitoring (or steering) of large parallel simulations. This will have valuable applications to several LLNL missions that use large-scale modeling and simulations, including stockpile stewardship, nonproliferation, energy security, and environmental management.

FY03 Results

Achievements for FY03 include completing an end-to-end prototype of the streaming infrastructure for progressive monitoring of a computational fluid dynamics simulation (JEEP code); developing an external-memory technique that allows interactive traversal of multiresolution surface meshes of arbitrary size; nearly completing the definition and prototype implementation of a wavelet model for our volumetric subdivision scheme, which will be used to extend our approach to nonrectilinear meshes; and nearly completing a prototype image-cache engine for maintaining interactive data exploration even with a slow rendering algorithm.

Proposed Work for FY04

In FY04 we will bring ViSUS to a level of maturity and robustness so that it can be deployed for a few targeted users, and will demonstrate the practical advantages of the ViSUS tools. The main tasks include developing new techniques that accelerate the isosurface construction process with occlusion culling, graphics hardware, and simple view-dependent adaptive refinements; developing a new approach for multiresolution streaming large, triangulated meshes; incorporating full slicing, isocontouring, and volume-rendering capabilities in the ViSUS Progressive Viewer; deploying the ViSUS streaming technology in the MIRANDA, IMPACT, and HYDRA simulation codes; and providing easy installation procedures and Lab-wide access for our software.

Publications

Bremer, P.-T. et al. (2003). "A multi-resolution data structure for two-dimensional Morse-Smale functions." *Proc. IEEE Conf. Visualiz.* 139–146. UCRL-JC-151932.

Cole-McLaughlin, K. et al. (2003). "Loops in reeb graphs of 2-manifolds." *Proc. 19th Conf. Comp. Geom.,* 344–350. UCRL-JC-151933.

Edelsbrunner, H. et al. (2003). "Morse-Smale complexes for piecewise linear 3-manifolds." *Proc. 10th Pacific Conf. Comp. Graphics & Appl.*, 361–370. UCRL-PRES-154074.

Ibarria, L. et al. (in press). "Out-of-core compression and decompression of large n-dimensional scalar fields." *Proc. Eurographics* **2003**. UCRL-JC-151934.

Isenburg, M. et al. (2003). "Large mesh simplification using processing sequences." *Proc. IEEE Conf. Visualiz.* **2003**, 465–472. UCRL-CONF-200016.

LaMar, E. and V. Pascucci. (2003). "A multi-layered image cache for scientific visualization." *Proc. IEEE Symp. Parallel & Large-Data Visualiz. & Graphics*, 61–68. UCRL-JC-151932.

Lindstrom, P. (2003). "Out-of-core construction and visualization of multiresolution surfaces." *Proc. ACM SIGGRAPH 2003 Symp. Interact. 3D Graphics*, 93–102. UCRL-JC-150863.

Linsen, L. et al. (in press). "Hierarchical large-scale volume representation with 3rd-root-of-2 subdivision and trivariate B-spline wavelets." *Hierarchical and Geometrical Methods for Scientific Visualization. (Springer-Verlag)*. UCRL-JC-146824.

Linsen, L. et al. (2003). "Hierarchical representation of time-varying volume data with 4th-root-of-2 subdivision and quadrilinear B-spline wavelets." *Proc. 10th Pacific Conf. Comp. Graphics & Appl.,* 346–355. UCRL-JC-151063.

Nuber, C. et al. (in press). "Using graphs for fast error term approximation of time-varying datasets." *Proc. Eurographics VisSym'03.* UCRL-JC-151965.

Pascucci, V. (in press). "Topology diagrams in scientific visualization." *Surface Topological Data Structures: An Introduction for Geographical Information Science.* UCRL-BOOK-200013.

Pascucci, V. et al. (2003). "Real-time monitoring of large scientific simulations." *Proc. 18th Ann. ACM Symp. Appl. Comp.,* 194–198. UCRL-JC-151094.

van Kreveld, M. J. et al. (in press). "Efficient contour tree and minimum seed set construction." *Surface Topological Data Structures: An Introduction for Geographical Information Science*. UCRL-BOOK-200018.

Enabling Large-Scale Data Access

Terence J. Critchlow 02-ERI-007

Abstract

The goal of this project is to develop a metadata infrastructure capable of providing scientists with access to large numbers of data sources through a single, intuitive interface. This interface will simplify scientists' interaction with data and enable them to answer more complex questions than currently possible. Our infrastructure makes extensive use of a novel metadata infrastructure to identify and describe web-based interfaces to complex, scientific data sources. The resulting descriptions are then passed to an extended version of XWrap, a wrapper-generation program.

We have developed an initial prototype of this infrastructure and have performed initial testing of the system. We plan to demonstrate the feasibility of this approach by automatically identifying sequence similarity search interfaces across a variety of Web sites on genomics.

This project will contribute to several technology areas, including data integration, bioinformatics, metadata, web-aware agents, and wrapper generators by developing an infrastructure for accessing extensive data sources through a single interface that combines local data with related information publicly available over the Internet, such as scientific publications, chemistry databases, urban planning information, and census data.

Mission Relevance

The infrastructure developed in this project will support almost every LLNL mission. Most scientists would benefit from easy access to external data such as scientific publications, chemistry databases, material descriptions, production techniques, and weather data. For example, climate modeling and other research that produce and locally store large amounts of simulation data and associated metadata could benefit from connecting this information to external sources.

FY03 Results

In FY03, we wrote the initial specification of the service-class description format; defined a new version of the interface description format; completed the initial version of the web spider and used it to successfully identify 17 of 25 randomly selected blast sites; extended XWrap Composer to accept complex interface description as input and to generate wrappers based on these descriptions; used Composer to generate wrappers for two complex blast interfaces; extended the DataFoundry interface to interact with blast wrappers; and incorporated two of the wrappers into this infrastructure.

Proposed Work for FY04

Our goals for FY04 are to (1) implement complex interface-identification capabilities that will support automatic identification of simple indirection pages; (2) finalize interface-description and service-class-description formats; (3) extend the web spider to support automatic generation of interface descriptions; (4) refine Composer to accept the final interface-description format and use it to generate complex wrappers; and (5) demonstrate end-to-end automatic wrapper to interface generation capabilities for both simple and complex interfaces.

Publications

Buttler, D. et al. (2003). "Querying multiple bioinformatics information sources: Can semantic web research help?" *SIGMOD Record* **31** (4), 59–64. UCRL-JC-150542-Rev-1.

Lacroix, Z. and T. Critchlow. (2003). *Bioinformatics: Managing scientific data.* (Morgan Kaufmann Publishing). UCRL-JC-148204-REV-1.

Liu, L. et al. (2003). *BioZoom: Exploiting source-capability information for integrated access to multiple bioinformatics data sources.* Presented at the 3rd IEEE Symp. Bioinformatics and Bioengineering (BIBE), Bethesda, MD, March 2003. UCRL-JC-150297.

Rocco, D. and T. Critchlow. (2003). "Automatic discovery and classification of bioinformatics Web sources." *Proc. Georgia Tech. Conf. on Bioinformatics.* UCRL-JC-152980.

A Computational Design Tool for an Integrated Micromixer and Components Used in Pathogen-Detection Systems

David P. Trebotich

03-ERI-003

Project Description

We plan to develop new computational models to simulate complex biological flows in integrated microsensors, and to validate these models experimentally. Microfluidic designs are central to many next-generation pathogen-detection devices for chemical and biological security. Our computational tool will provide critical understanding of the fluid dynamics involved in microdevices, shorten the design and fabrication process to reduce costs, optimize prototypes, and provide a prediction capability for new, more advanced designs. Computational work will be performed in collaboration with the University of California (UC), Davis and experimental work for computational model validation in collaboration with UC Berkeley.

A computational tool that simulates complex biological flow in microdevices will eliminate trial-and-error design processes for microelectromechanical system devices such as biodetection devices for homeland security, thus shortening the design and fabrication cycle by months. This tool will optimize current prototypes and will help predict new, more advanced designs. A successful project will also produce several articles in the scientific literature.

Mission Relevance

The proposed work furthers LLNL's mission in countering the proliferation and use of weapons of mass destruction by supporting the microfluidic design strategy for next-generation pathogen-detection and monitoring devices. This project will provide a design tool to help reduce reagent consumption and to increase reaction rates in these microfluidic biodetection devices.

FY03 Results

In FY03 we researched and developed a numerical method for incompressible, viscoelastic flow in contraction microchannels [two dimensional (2-D)] and microducts (3-D). Our method, a capability that does not currently exist elsewhere, was tailored to be valid for the full range of elastic flows using a single stability condition, including the high Weissenberg problem for strong elastic flows in biological fluids. We also validated our method with preliminary experimental data for DNA flow in a contraction microchannel and developed numerical operators to treat Stokes flow in complex geometries using the embedded-boundary method.

Proposed Work for FY04

In FY04, we plan to extend our Stokes-flow capability in complex geometry to incompressible Navier-Stokes, which includes advection, using a Bell, Colella, and Glaz- (BCG-) type projection; couple our hyperbolic-enhanced viscoelastic model to BCG in complex geometry; couple a passive trace-particle model to the continuum model in complex geometry; continue to develop the particle model to include more advanced constitutive modeling; and extend the complex geometry capability to the geometries of real device components and compare to experiments.

Publications

Trebotich, D., P. Colella, and G. H. Miller. (in press). "A projection method for viscoelastic flow in microchannels." *J. Comp. Phys.* UCRL-JC-153079.

Trebotich, D. et al. (2003). "A numerical model of viscoelastic flow in microchannels." *Proc. 6th Intl. Conf. Modeling & Simulation in Microsystems.* **2**, 520–523. UCRL-JC-150969.

Quantum Vibrations in Molecules: A New Frontier in Computational Chemistry

Kurt R. Glaesemann

02-LW-022

Abstract

This project will combine a quantum treatment of electrons via ab initio methods with a quantum treatment of nuclei via path-integral Monte Carlo (PIMC) to provide a realistic treatment of molecular vibrations. Ours will be the first to use a high-level treatment of electron correlation through methods such as Möller-Plesset perturbation theory, which will allow us to perform fully ab initio simulations of van der Waals clusters and other problems involving long-range electron correlation. The PIMC code will be used to explore quantum effects in a variety of chemical systems for the first time, e.g., the behavior of hydrogen within the interiors of a carbon nanotube, proton-transfer reactions, rotational barriers, and ultracold molecular clusters.

This research will eventually lead to a methodology of calculating thermochemical data without any experimental input. Examples of calculable properties include heat capacities, enthalpies of formation, entrophies, and total free energies. This information is often needed as input to much larger chemical calculations.

Mission Relevance

The PIMC code can be used to explore quantum effects in a variety of chemical systems. This fully quantum picture will provide new insights into problems regarding the chemical reactions of energetic materials. Such problems are directly relevant to LLNL's stockpile stewardship mission. For example, our work on chemical systems, such as carbene and nitrogen hydride, is potentially useful in calculating chemical kinetic rates, which is applicable to understanding conventional explosives for stockpile stewardship.

FY03 Results

The PIMC code and theoretical methods developed in FY02 were applied to a variety of molecules in FY03. This study elucidated the need for a balanced basis set for the ab initio portion of the calculation. Results, published in the *Journal of Chemical Physics,* allowed the calculation of enthalpies of formation without any experimental input. The PIMC code was also linked to a condensed phase code for exploring larger chemical systems.

Publications

Glaesemann, K. R. and L. E. Fried. (2003). "A path integral approach to molecular thermochemistry." *J. Chem. Phys.* **118**, 1596. UCRL-JC-149046.

Parallel Graph Algorithms for Complex Networks

Edmond T. Chow

03-ERD-061

Abstract

Many scientific, technological, and social networks, such as metabolic networks, the World-Wide Web, and human interaction graphs, are being studied as complex networks: graph topologies that appear random, but have very specific clustering and vertex-degree properties. A particular complex network called a "semantic graph" is being developed to organize and search intelligence information gathered by counterintelligence agencies. Such graph data structures will contain vast amounts of information and must be stored and queried on distributed-memory parallel computers. Our objective is to develop algorithms and research software that exploit the properties of complex networks and enable fast and efficient use of semantic graphs on parallel computers. This software will be capable of partitioning complex graphs and semantic networks of up to 1 billion nodes, using one thousand or more processors. We expect that this work will impact how very-large-scale semantic graphs are developed.

Mission Relevance

Semantic graphs are used to encode relationship data gathered by intelligence agencies for counterterrorism purposes. By exploiting the large aggregate memory of parallel computers, this research will enable the scaling to unprecedented sizes of complex network applications relevant to LLNL's national-security mission. In addition, such parallel algorithms will also have application in complex networks in other disciplines, particularly the analysis of metabolic networks, in support of LLNL's mission in bioscience to improve human health.

FY03 Results

In FY03, we began developing a software infrastructure for semantic graphs. In particular, we developed (1) generators for ontologies and for semantic graphs that are consistent with these ontologies; (2) heuristic algorithms that use ontologies to accelerate point-to-point searches in semantic graphs; and (3) a preliminary algorithm for parallelizing these searches. We also (4) gained an understanding of what objectives must be optimized in partitioning semantic graphs; (5) began applying linear algebraic techniques for clustering vertices in graphs, based on their topology; and (6) began investigating a partitioning approach based on classifying relationships as long range and short range.

Proposed Work for FY04

In FY04, we will develop partitioning algorithms that utilize augmented ontologies—ontologies that are annotated with the frequencies of edge and vertex types in the semantic graph. The simplest approach is to partition the augmented ontology as a weighted graph and induce this partitioning on the semantic graph. However, we expect better results to be achieved if the augmented ontology is refined to have some of the major topological properties of the semantic graph. This refinement can be accomplished by clustering similar vertices or edges. We will also continue to pursue promising approaches discovered in the previous year, such as dynamic partitions based on analysts' queries.

Space–Time Secure Communications for Hostile Environments

James V. Candy

03-LW-005

Abstract

When communicating critical information between sensors in hostile environments such as a battlefield or in clandestine operations relevant to national security, the data must be reliably received and extracted. We propose to investigate—using theory, simulation, and experiments—the feasibility of using time-reversal- (TR-) based communications systems that contain sensor arrays equipped with multichannel processors for both acoustic and electromagnetic (EM) cases in hostile environments contaminated by multipath arrivals and multiple scatterers. The TR processor is capable of unraveling multipath returns using its acquired knowledge of the medium.

Using TR concepts and theory, we expect to develop a TR receiver that will enable communications in complex environments such as caves, and for sensor-to-sensor communications in critical applications such as water pipes, nuclear plant systems, and border-protection systems. The commercial applications of such technology also abound. For mobile communications between cooperative transmitter–receiver pairs it may be possible to establish fade-free links, which would be a major breakthrough in this area.

Mission Relevance

Development of the TR receiver will greatly benefit national-security and military applications. Channel reliability and vulnerability in complex and hostile environments are a prime concern of emergency response teams, security forces, and the intelligence community.

FY03 Results

In FY03 we developed theory for TR receivers; pilot signal, code signals, and procedures for experimental environments; acoustic simulation for experimental design and analysis of the proposed TR receiver; and controlled acoustic experiments in a multipath, multiple-scatterer environment. In addition, data from the experiments were analyzed; the performance of TR receivers was demonstrated; and an EM simulation was also developed to demonstrate that the TR receivers could function reasonably well in a hostile EM multipath, multiple-scatterer environment. This work was point-to-point communications; however, array results were synthesized using both the simulator and the controlled experimental environment to demonstrate feasibility. We met all of our proposed milestones.

Proposed Work for FY04

In FY04 our major tasks will be to (1) continue theoretical development for receiver design for transmitting arrays and multiple receiving stations and investigate the deconvolution of multichannel communications systems, incorporating space–time arrays for both acoustics and EM; (2) develop multichannel simulations for a noisy, highly reverberant, multiple-scatterer environment; (3) design (through simulation) and conduct controlled laboratory experiments on acoustics using a TR array; (4) design controlled laboratory experiments using an existing TR ultrasonic array; and (5) begin acquiring the equipment required to implement the EM design.

Publications

Burke, G. and A. Poggio. (2003). *Simulation of time-reversal processing for electromagnetic communication*. UCRL-ID-154698.

Candy, J. V. (2003). *Reversing time: A way to unravel undistorted communications.* Presented at the 144th Mtg. Acous. Soc. Am., Cancun, Mexico, Dec. 2–6, 2003. UCRL-CONF-200292.

Candy, J. V. et al. (in press). "Time-reversal processing for an acoustics communications experiment in a highly reverberant environment." *J. Acous. Soc. Am.* UCRL-JC-154219.

Candy, J. V. et al. (2003). *Time-reversal communications in a hostile, reverberative environment.* Presented at the 144th Mtg. Acous. Soc. Am., Cancun, Mexico, Dec. 2–6, 2003. UCRL-CONF-201023.

Guidry, B. L. et al. (2003). *Experimental design and processing for time-reversal communications in a highly reverberative environment.* Presented at the 144th Mtg. Acous. Soc. Am., Cancun, Mexico, Dec. 2–6, 2003. UCRL-CONF-200793.

Sullivan, E. J. and J. V. Candy. (2003). *Acoustic propagation in a water-filled cylindrical pipe.* UCRL-ID-153887.

Long-Time-Scale Atomistic Simulations

Wei Cai 03-LW-027

Abstract

The limit of present-day atomistic simulations to nanosecond time scales precludes the study of many interesting phenomena, such as glass relaxation and protein folding, that occur on time scales of milliseconds or longer. Our project promises to remove this constraint and open ways to heretofore impossible studies of scientific problems. We propose to develop a novel theoretical framework and its robust numerical implementation, combining importance-sampling formalism and high-dimensional optimization techniques for efficient simulations of rare, yet fundamental, processes that control the macroscopic behavior of complex systems. Our effort will be to extend our new method, which has been successfully applied to small systems, to realistically complex materials-science problems.

This research will establish a solid theoretical foundation and a set of efficient numerical tools for long-time-scale simulations of complex processes—a major advance in our ability to model and understand fundamental processes in condensed matter systems. Combining our methodology with the unique computational facilities at the Laboratory will significantly enhance our predictive atomistic modeling capabilities. In particular, our method should be of great value to applications involving long-time-scale biological processes, such as protein reaction and folding.

Mission Relevance

This project will provide deeper insight into the atomistic processes controlling dislocation mobility and solid–solid phase transitions, which are of critical importance to several large research programs at LLNL, such as the Advanced Simulation and Computing Program's dynamics-of-metals effort, in support of stockpile stewardship.

FY03 Results

In FY03, we made a key step toward the successful completion of our project: developing an importance-sampling method that does not require the explicit calculation of the importance function on all accessible system configurations. This is an essential step for applying our method to realistic systems with many degrees of freedom. We have realized this step by adopting an approach in which the importance function is cast into a selected analytical form with a set of undetermined parameters. These parameters are subsequently optimized by a genetic algorithm to maximize simulation efficiency. This importance-sampled Monte Carlo simulation is implemented for the process of dislocation motion in the diamond cubic lattice of silicon described by the Stillinger-Weber potential model.

Proposed Work for FY04

Our task in FY04 involves the design of sampling and optimization algorithms that take advantage of existing parallel-computing resources to study the atomic mechanisms of nucleation and growth phenomena in the face-centered cubic/body-centered cubic martensitic transformation in iron. In addition to serving as a benchmark for the applicability of the methodology to realistic materials-science problems, this application will provide a deeper insight into the physics of martensitic phase transformations, which play a crucial role in the behavior of various structural materials and “smart” shape-memory alloys.

Publications

Cai, W. et al. (2003). “Importance sampling of rare transition events in Markov processes.” *Phys. Rev. E* **66**, 046703. UCRL-JC-148441.

Nuclear Science and Engineering

Study of Chemical Reactions Controlling the Mobility of Uranium and Related Contaminants in Ground and Surface Water Systems with Emphasis on Apatite

Michael J. Taffet

01-ERD-105

Abstract

Our objective is to define the mechanisms, equilibria, kinetics, and extent of sorption of aqueous uranium (U) onto hydroxyapatite (HA, [$Ca_5(PO_4)_3(OH)$]) for ranges of pH, ionic strength, U concentration, dissolved carbon (C) [i.e., carbon dioxide (CO_2)], surface area, and other parameters. We are conducting chemical modeling, batch and flow-through experiments, chemical analysis, x-ray absorption and diffraction, and electron microscopy. Our motivation is the need to immobilize U in water and soil to prevent its entry into water supplies and, ultimately, biological systems. Applying HA to the in situ treatment of U-bearing groundwater could be an effective, low-cost technology.

We are finding that HA quickly, effectively, and reversibly sorbs U at a high capacity by inner-sphere complexation over a wide range of conditions. Specifically, our results indicate that (1) equilibrium sorption of U to HA occurs in hours, regardless of pH; (2) in ambient and CO_2-free atmospheres, over 97% of initial U is sorbed to HA; (3) the extent of sorption is reduced by higher concentrations of dissolved C but is always greater than 85%; (4) U sorption is reversible and is not affected by ionic strength; and (5) under ambient conditions, sorption is the only mode of U sequestration below 50 ppb U—above 100 ppb U, precipitation of uranyl phosphate minerals dominates. We verified that 1-m^2 surface area of HA (12.2 mg for 8-μm-diam granules) can sorb over 2.9 mg of U, which is in agreement with calculations based on phosphate (PO_4) and calcium oxide (CaO) sites on the HA unit cell. Our work is significant because small masses of HA sorb appreciable masses of U quickly over a wide range of chemistries.

Mission Relevance

This work supports NNSA and LLNL missions in homeland security and environmental management. By providing an understanding of the mechanisms that control aqueous U sorption to HA, we can optimize in situ and ex situ removal of U from groundwater, enhance filtration of U from drinking water, and fixate U in soil and solid wastes. Inexpensive sources of HA (mammal and fish bones) exist all over the world and can be used to remove U, other radionuclides, and metals from drinking water and other liquids. Use of HA can mitigate terrorist attacks on water supplies. Because HA is such a strong sorbent, it can be used to concentrate radionuclides in surface waters for forensic isotopic analysis to determine whether radionuclides are being processed for use in weapons applications.

FY03 Results

Because the imaging of U coordination to HA was difficult with transmission electron microscopy, indirect measurements with x-ray sorption spectroscopy were used and were effective in defining mono- and bidentate sorption of U to CaO and PO_4 groups. Results show that C complexation slightly retards sorption of U to HA. We used pure HA to study sorption of U in a controlled setting, and after running several column experiments, we placed cow bone char (an inexpensive HA-rich material) in a U-contaminated aquifer at LLNL's Site 300 and monitored chemistry and hydraulics there to define the effectiveness of this in situ technique. U concentrations declined from over 220 ppb to 0.3 ppb in the HA-emplacement zone.

Mononitride Fuel Development for SSTAR and Space Applications

Jor-Shan Choi

03-ERD-019

Abstract

In this project, modified nitride-based fuels will be developed and fabricated. These fuels are of interest in compact, long-life reactor cores, a key feature in proliferation-resistant reactor designs [such as the small, secure, transportable, autonomous reactor (SSTAR) concept] and in space reactors. Computational methods will be used to analyze fuel performance. Fuel pellets will be fabricated and characterized at LLNL. We plan to irradiate several fuel pellets at the McClellan Nuclear Radiation Center (MNRC) to verify the models and demonstrate feasibility of use. This project is also an important stepping stone to help LLNL participate fully in the emerging Advanced Fuel Cycle and Generation IV initiatives.

To develop the fuel, the proper amount of selected chemical additives will be added to uranium mononitride (UN) fuel to enable compactness, long life, ease of chemical processing and waste management, and proliferation resistance. Uranium nitride is more suitable for compact reactors than UO_2 because of its high actinide density and high thermal conductivity. Modification of the composition with hafnium nitride (HfN), zirconium nitride (ZrN), or other materials may significantly increase the stability and lengthen the life of the reactor core—HfN as a burnable neutron poison; ZrN by displacing ^{238}UN, limiting the buildup of plutonium in the reactor, and increasing the stability and life of the fuel. Uranium-235 enrichment will be tailored to optimize the fuel life. Modified UN-based fuels have the potential to make compact, long-life reactors much more attractive for development, testing, and licensing.

Mission Relevance

This project supports LLNL's national-security mission by advancing work in proliferation-resistant reactor designs. Such reactors would be fabricated and their cores sealed by a trusted nation and shipped to other nations for up to 30 yr of use. At the end of life, the sealed reactor core with the spent fuel inside would be returned to the trusted nation for processing and disposition. Nations operating these reactors would not need to develop their own fuel manufacturing and spent-fuel processing technologies. As a result, the spread of sensitive dual-use nuclear technologies can be minimized or eliminated. An essential feature for such a reactor design is a long-life fuel that can last for the entire 30 yr of reactor operation.

FY03 Results

In FY03 we acquired the computer codes LIFE4Rev1, SIEX3, and Monteburn from DOE's Nuclear Fuel Cycle Software Center for use in modeling fuel performance. In addition, we acquired the SPACEPIN subroutine (with DOE approval) and used it to analyze UN fuel performance for the space reactor SP-100. We ran sample problems and verified that these computer codes are now operational. After obtaining authorization to conduct uranium-based nitride fuel manufacturing, we installed a glove box with a high-purity atmosphere and proper equipment. By the end of FY03, we had completed the first step of a three-step carbo-thermal process for manufacturing uranium nitride fuel.

Proposed Work for FY04

In FY04, computational codes will be modified to evaluate the performance of the modified nitride fuel composition (containing ZrN, HfN, or other materials) for an optimized long-life reactor core. We will fabricate the modified uranium-based nitride fuel compositions at LLNL and make plans for irradiation experiments at MNRC in collaboration with the University of California, Davis.

Publications

Ebbinghaus, B., J.-S. Choi, and T. Meier. (2003). *A modified nitride-based fuel for long core life and proliferation resistance.* Am. Nuc. Soc./Euro. Nuc. Soc. Intl. Winter Meet. and Global 2003, Nov. 16–20 2003, New Orleans, LA. UCRL-CONF-200563.

Meier, T., B. Ebbinghaus, and J.-S. Choi. (2003). *Facilities for development of modified nitride-based fuel pellets.* Am. Nuc. Soc./Euro. Nuc. Soc. Intl. Winter Meet. and Global 2003, Nov. 16–20 2003, New Orleans, LA. UCRL-CONF-200564.

Single-Crystal Chemical-Vapor-Deposition Diamond: A Novel Material for Neutron Detection

Gregory J. Schmid

03-FS-012

Abstract

This project demonstrated the feasibility of developing a solid-state neutron spectrometer based on single-crystal chemical vapor deposition (CVD) diamond, a novel material. Such a spectrometer could be very useful in the field of fast (megaelectronvolt) neutron detection, and could offer much higher efficiency and energy resolution than the leading plastic scintillators. We purchased wafers of raw, single-crystal CVD diamond and fabricated them on-site

into metal-insulator-metal neutron detectors. The performance of the detectors, along with commercially available front-end electronics and data-acquisition system, was tested at the LLNL neutron generator, and results were compared to those of a leading plastic scintillator.

Demonstrating the feasibility of fabricating and testing single-crystal CVD diamond radiation detectors could lead to fast neutron sensors that are in many ways better than the latest generation of plastic scintillators. The results of this study could pave the way for further studies that would more fully explore and understand the material properties (electronic, nuclear, and mechanical) of single-crystal CVD diamond.

Mission Relevance

A single-crystal CVD diamond is a new material that shows exceptional promise as a neutron sensor for applications in neutron spectroscopy and radiography for stockpile-stewardship experiments, and for detection of nuclear materials via fission neutrons in support of the Laboratory's nonproliferation and homeland security missions.

FY03 Results

This study met all its milestones in FY03. The efficiency of single-crystal CVD diamond to neutrons was measured at 2.5 and 14 MeV and compared to theoretical calculation. The experimental results, 2.7 and 2.3 %/mm respectively, were within 15% of theoretical calculations. Furthermore, efficiency data acquired with a BC400 plastic scintillator (1.0 and 0.7 %/mm) came in well below the diamond results, demonstrating the superiority of diamond. Energy-resolution measurements were estimated from alpha-particle data (for diamond) and gamma-ray data (for scintillator), and the results (at 14 MeV) are 0.3% and 3.0% full width half maximum for the diamond and scintillator respectively, in reasonable agreement with expectation.

Publications

Schmid, G. J. et al. (2003). *A neutron sensor based on synthetic single crystal diamond.* UCRL-CONF-200447.

Schmid, G. J. et al. (2003). *Neutron spectrometers based on single crystal CVD diamond.* Presented at the 45th Ann. Mtg. APS Div. Plasma Phys., Albuquerque, NM, Oct.28, 2003. UCRL-PRES-154198.

Physics

Ultrafast Materials Probing with the Falcon/Linac Thomson X-Ray Source

Paul T. Springer

01-SI-007

Abstract

The goal of this project is to develop Picosecond Laser–Electron Interaction for Dynamic Evaluation of Structures (PLEIADES), a high-brightness, subpicosencond hard x-ray source enabling unprecedented dynamic measurements in matter. Based on Thomson scattering of the 35-fs Falcon short-pulse laser and a cosynchronous, tightly focused electron beam from LLNL's 100-MeV linear accelerator, PLEIADES will produce tunable, ultrafast (100 fs to 1 ps), hard x-ray pulses in the 10- to 200-keV energy region. This x-ray source would greatly exceed existing

third-generation synchrotron sources in speed, brightness, and simplicity. Our research will demonstrate this new Thomson x-ray source as an enabling technology for ultrafast x-ray probes of material dynamics.

In the near term, the use of the bright, ultrafast, high-energy x-rays will enable pump-probe experiments using radiography, dynamic diffraction, and spectroscopy to address the equation of state and dynamics of phase transitions and structure in laser heated and compressed metals of interest to stockpile stewardship. Longer-term development of compact laser and accelerators could lead to ultra-bright, portable, tabletop x-ray sources with applications such as protein crystallography or probes for materials experiments on large-scale lasers. Finally, the scaling of these sources to the nonlinear intensity regime of high-energy petawatt lasers offers large (1000-fold) improvements in probe resolution compared with the conventional K-alpha sources planned for future large-scale laser facilities .

Mission Relevance

The short time duration and high x-ray flux of PLEIADES will provide new techniques and technologies for materials science experiments, making possible experiments relevant to LLNL's Stockpile Stewardship Program (SSP). For example, the x-rays produced by PLEIADES will have sufficient intensity to enable pump-probe experiments to examine the time-dependent behavior (e.g., equations of state, phase-transition dynamics, and structure) of laser-excited high-*Z* metals of interest to SSP.

FY03 Results

In FY03, we achieved a major goal of the project: a well-characterized x-ray beam with record brightness in the short-pulse, hard-x-ray regime. "First light" at PLEIADES produced a 70-keV, 2-ps, 30-μm-spot-size x-ray beam. The peak flux of 2×10^6 x rays per pulse showed quantitative agreement with three-dimensional electron–laser interaction predictions, although the electron beam focusing was poorer than predicted. The installation of strong focusing quadrapoles should reduce spot size and increase yield to 2×10^8 x rays per pulse. We demonstrated beam parameters needed for initial experiments: single-shot radiographic images of high-*Z* foils, source tunability from 40 to 80 keV, and pulse compression to 300 fs. We designed, and showed the feasibility of, dynamic diffraction experiments with bismuth.

Publications

Anderson, S. G. et al. (2003). *Pulse compression via velocity bunching with the LLNL Thomson x-ray source photoinjector.* 2003 Particle Accelerator Conf., May 12–16, 2003, Portland, OR. UCRL-JC-154348.

Brown, W. J. et al. (2003). *Generation of high-brightness x-rays with the PLEIADES Thomson x-ray source.* 2003 Particle Accelerator Conf., May 12–16, 2003, Portland, OR. UCRL-JC-153476

Fittinghoff, D. N. et al. (2003). *PLEIADES, a subpicosecond x-ray source for ultrafast materials probing.* APS invited talk. UCRL-JC-151594.

Hartemann, F. V. et al. (2003). *Compton scattering and its applications: the PLEIADES femtosecond x-ray source at LLNL.* Intl. Workshop on Quantum Aspects of Beam Phys., Jan. 7–10, 2003, Hiroshima, Japan. UCRL-JC-153189.

Hartemann, F. V. et al. (2003). *Time and frequency domain calculations of x-ray production for the PLEIADES Thomson x-ray source*. 2003 Particle Accelerator Conf., May 12–16, 2003, Portland OR. UCRL-JC-149373 Abs.

Kuba, J. et al. (2003). *PLEIADES: High-peak-brightness, subpicosecond hard Thomson x-ray source*. SPIE Intl. Symp. on Optical Sci. and Tech., Aug. 3–8, 2003, San Diego, CA. UCRL-JC-151689 Abs.

Short Pulse: An Initiative to Enable Relativistic Applications for Advanced ICF and Stockpile Stewardship

Christopher P. Barty

02-SI-004

Abstract

This project is researching and developing next-generation laser technology for high-energy, ultrashort-pulse scientific applications. The applications include fast ignition, hard x-ray radiography, and studies of high-energy-density (HED) matter. These topics have recently become the subject of a national initiative of NNSA. Our work has four main facets: (1) developing an advanced laser front end for high-energy, chirped-pulse generation; (2) developing the key enabling technologies for compression and focusing of high-energy petawatt pulses; (3) evaluating system concepts for high-energy petawatt (HEPW) lasers based on existing long-pulse (nanosecond) Nd:glass laser architectures; and (4) coordinating research and development for conceptualizing high-energy, short-pulse science applications.

We will develop chirped-pulse amplification and other technologies that will enable advanced, hard x-ray and proton radiography on the nation's HED science facilities to explore advanced concepts for inertial-confinement fusion, and creation of unique, HED states of matter.

Mission Relevance

This research will investigate the use of high-energy lasers for stockpile stewardship and fusion energy experiments that require a combination of focused, ultrashort pulses in the relativistic regime with high energy in long pulses. This research is directly relevant to LLNL's national-security and energy-security missions.

FY03 Results

FY03 accomplishments include (1) developing designs for gratings and high-energy petawatt laser compressor systems based on multilayer dialectic gratings; (2) developing a damage test facility for absolute damage fluence measurements and determining the absolute damage fluence of a small test grating, which was consistent with design figures; (3) developing front-end concepts based on fiber lasers and Bragg fiber-grating pulse stretching; (4) developing the world's largest aperture ion-beam etching facility for the fabrication of meter-scale multilayer dielectric gratings; and (5) production of a coordinated research program for the development of the science applications of HEPW lasers.

Proposed Work for FY04

In FY04 we plan to deliver validated technology solutions for first deployment of 2-kJ, 10-ps pulses at future large laser systems, including far-field damage studies of subscale gratings;

develop a fiber chirped-pulse amplification-pulse generator and couple it to an existing preamplifier to produce an integrated testbed; use this testbed for near-field damage studies on prototype gratings; explore concepts for phased array gratings; and coordinate work on HEPW science directed towards establishing the feasibility of conceptual applications.

First Physics from BaBar

Douglas M. Wright 01-ERD-014

Project Description

Cosmological models of the early universe require charge–parity (*CP*) violation to produce our current matter-dominated universe. However, a plausible source for this *CP*-violating effect has yet to be discovered. The Standard Model of particle physics may contain a source of *CP* violation, but this hypothesis has not been experimentally tested. The broad physics program for the BaBar experiment at the B Factory accelerator—a collaborative effort by the Stanford Linear Accelerator Center, Lawrence Berkeley National Laboratory, and LLNL—is aimed at unraveling the mystery of *CP* violation.

This project will focus on observing a *CP*-violating effect in B mesons by analyzing the large data sets being produced by the BaBar experiment. The expected result of this project is the measurement of a possibly null *CP*-violating effect in B mesons. We plan to extract a measurement from the so-called "golden" K_S decay channel of the B meson as well as the K_L channel, which is the only other channel where a conclusive *CP*-violating effect is expected from the first data sets of the BaBar experiment. This would be an important discovery in particle physics and would contribute to the understanding of *CP* violation in the framework of the Standard Model of particle physics. This project builds on past accomplishments at LLNL in detector hardware and software development and will establish LLNL's leadership at the forefront of particle physics experiments.

Mission Relevance

This research will aid in unraveling the mystery of *CP* violation by searching for the *CP*-violating effect in B mesons. Observation of *CP* violation in the BaBar experiments would be a major scientific discovery at the forefront of high-energy physics, a mission area of DOE's Office of Science. This project will also (1) extend LLNL capabilities for handling and analyzing massive scientific data sets, as required in surveillance and nonproliferation applications, and (2) directly benefit LLNL programs in accelerator-related science, such as proton radiography, a critical aspect of stockpile stewardship.

FY03 Results

Efforts in FY03 continued to work toward extracting a statistically meaningful measurement of *CP* violation in the K_L channel. We focused on improving the B-meson-tagging algorithm and K_L background measurements, significantly improving the algorithm both by using the most advanced numerical techniques and by incorporating additional physical observables. We also enhanced the operational procedures for, and monitoring of, the instrumented flux return subdetector, which provides roughly one-half of the K_L measurements.

Publications

Aubert, B. et al. (in press). "Measurement of the inclusive charmless semileptonic branching ration of B mesons and determination of $|V_{ub}|$." *Phys. Rev. Lett.* UCRL-JC-154655.

Aubert, B. et al. (2003). "Measurement of *CP*-violating asymmetries and branching fractions in B meson decays to $\eta' K$." *Phys. Rev. Lett.* **91**, 161801. UCRL-JC-152515.

Aubert, B. et al. (2003). "Measurement of time-dependent *CP* asymmetries and the *CP*-odd fraction in the decay $B^0 \rightarrow D^{*+}D^{*-}$." *Phys. Rev. Lett.* **91**, 131801. UCRL-JC-154649.

Aubert, B. et al. (2003). "Observation of the decay $B^\pm \rightarrow \pi^\pm\pi^0$, study of $B^\pm \rightarrow K^\pm\pi^0$, and search for $B^0 \rightarrow \pi^0\pi^0$." *Phys. Rev. Lett.* **91**, 121801. UCRL-JC-152694.

Aubert, B. et al. (2003). "Searches for D^0–$\bar{D}^0$ mixing and a measurement of the doubly Cabibbo-suppressed decay rate in D0 K decays." Phys. Rev. Lett. 91, 171801. UCRL-JC-152867

Wright, D. (2002). "Measurement of Sin 2β with BaBar." Proc. Intl. Conf. on High-Energy Phys., Jul. 24–Jul. 31, 2002, Amsterdam, The Netherlands. UCRL-JC-150088.

Developing a Radiative-Shock Test Bed

Jeffrey A. Greenough 01-ERD-075

Abstract

Radiative shocks are of specific interest to the Stockpile Stewardship Program and of broad scientific interest, particularly in astrophysics. One dramatic example of radiative shocks can be seen in the localized emissions emerging from the impact of the ejecta from Supernova 1987A with its circumstellar ring nebula. To interpret these events, a quantitative understanding of radiative shocks is needed. Yet this understanding cannot be gained by comparing models and simulations with experimental data because such data do not exist. The goals of this project are to (1) develop a planar, radiative-shock test bed; (2) demonstrate a planar, radiatively collapsed shock in a gas-cell target; and (3) establish the appropriate theoretical scaling for these radiative shocks.

This project will experimentally demonstrate, on the Omega laser, that under controlled conditions in planar geometry, a strongly radiative shock can be created and its dynamics and characteristics measured. This new experimental test bed can then serve as a testing ground for models and simulations needed to describe radiative shock conditions, whether in astrophysics or in laboratory research.

Mission Relevance

Our project will develop a test bed, which will enable laboratory studies of hydrodynamically driven, radiative shocks. The results of this research will support LLNL's stockpile stewardship mission. This project also forms a very high-visibility academic outreach endeavor that attracts high quality young scientists to LLNL and DOE-DP research programs.

FY03 Results

Work conducted in FY03 includes (1) launching a strong shock into a low-density, high-*Z* gas; (2) observing the formation of a radiative collapse using radiography; (3) observing the absolute position of the shock, and hence its velocity, using a novel new technique based on the velocity interferometer system for any reflector (VISAR); and (4) establishing the theoretical scaling that will relate our results to astrophysical settings. We have achieved all of the experimental objectives, and the results will be published in the open, peer-reviewed literature.

Scaling to astrophysical settings was also investigated by the astrophysics collaborators who have been participating in this project.

Publications

Drake, R. P. et al. (in press). "Experimental astrophysics with intense lasers." *Reviews of Modern Physics*. UCRL-JC-153579.

High-Average-Power, Frequency-Agile Fiber Lasers

Deanna M. Pennington

01-ERD-083

Abstract

The goal of this project is to develop compact, high-power laser technologies based on diode-pumped fiber lasers, which can be frequency doubled or mixed in periodically poled (PP) materials to provide a spectrum of visible wavelengths. Specifically, we are developing a 938-nm neodymium-doped fiber amplifier (NDFA) and a 1583-nm erbium-doped fiber amplifier (EDFA). The NDFA provides a useful source for global climate monitoring, whereas the longer-wavelength EDFA can be used to expand the bandwidth of existing telecommunications networks. In addition, a 589-nm wavelength can be generated by combining the EDFA and NDFA via sum-frequency mixing in a PP crystal. When used in laser-guided adaptive optics (LGAO), this wavelength has both military and astronomical applications.

Mission Relevance

This research establishes a core competence at LLNL in fiber lasers and PP materials, which have been identified as strategic, next-generation laser technologies. The NDFA can be used to detect atmospheric water vapor, assisting researchers in understanding the global climatic impact of energy use. It can also be frequency doubled to produce 469-nm-wavelength light that may enhance the ability of LLNL-developed portable devices to detect environmental biopathogens. The 589-nm wavelength can be used for LGAO, enabling remote imaging by correcting atmospheric turbulence, and medically for photodynamic therapy. The EDFA may assist in the deployment of a free-space network topology, as well as secure field communications for military applications.

FY03 Results

In FY03 we successfully delivered an 11-W NDFA. This was combined with an EDFA we built in FY02 to demonstrate a proof-of-concept 589-nm source. Characterizing this 589-nm prototype and its component technologies allowed us to assess scalability to higher average powers. A preliminary systems design has been produced for the laser; the next step is to continue scaling the power up to 50 W. We also frequency-doubled the NDFA laser using PP materials, successfully producing a blue light source. A poling station was constructed to provide an internal source for PP materials, and a test station was constructed to perform extended lifetime testing. This station was used to assess the damage mechanisms limiting PP materials.

This research has successfully demonstrated an 11-W EDFA and an 11-W NDFA. The EDFA components are now commercially available. The NDFA required development of a novel double-clad fiber amplifier, scaling previously reported results at this wavelength by 100 times. The NDFA can be used to efficiently core-pump other fiber lasers. This research has established a core competence in fiber design, which is now being applied to new applications, such as the high-energy petawatt project for the National Ignition Facility. In addition, this project has established a competence in PP materials.

Publications

Dawson, J. et all. (2003). "938-nm Nd-doped high power cladding pumped fiber amplifier." *Tech. Digest Advanced Solid-State Photonics 18th Topical Meet.*, Feb. 2–5, 2003, San Antonio, TX. UCRL-JC-150354.

Dawson, J. et al. (2003). "High-power 938-nm cladding pumped fiber amplifier." *Proc. SPIE* **4974**, 75. UCRL-JC-148985.

Pennington, D. et al. (2003). "Compact fiber laser approach to generating 589 nm laser guide stars." *Tech. Digest European Conf. on Lasers and Electro-Optics, Jun. 24,* 2003, Munich, Germany. UCRL-PRES-143873 Rev.

Ultrahigh-Power Inorganic Liquid Laser

Earl R. Ault 01-ERD-095

Abstract

Solid-state lasers are the focus of DoD studies for achieving power levels exceeding 100 kW with good beam quality. This project investigated diode-pumped, flowed-neodymium (Nd) solutions. Such media has the gain properties of glass lasers but none of the limiting features of a solid-state gain medium (e.g., fracture, thermal focusing, and birefringence). Using the atomic vapor laser isotope separation (AVLIS) dye system, a flowing laser gain medium with high optical quality, even under high optical pumping, is possible.

The most critical demonstration in this project occurred in FY02, when interferometry showed that optical pump heat-loaded flow conditioned cells of Nd solutions could be made to convert the bulk of the induced thermal optical distortions into optically benign tilt, and that two cells in counter-flow negate pump pulsations. Literature already established the cross sections and lasibilty of the solutions. What was next needed was a small working test bed to operationally demonstrate the principles. Continuous-wave (CW) operation had never been achieved due to high optical distortions. Diode pumping, key for high efficiency, had never been attempted. Heat capacity operation was of particular relevance for military platforms. Specific solvent and hardware materials compatibility and handling issues needed to be resolved.

Mission Relevance

High-average-power lasers have many DoD applications for battlefield, theater, and ballistic missile defense, in support of LLNL's national security mission. In this field, even current solid-state technology is tending toward moving media. Previous work proposed using a moving slab of glass to obtain kilowatt-class laser action with beam quality. Today, the LLNL heat capacity laser is planned to operate with mechanically interchanged discs. This work in flowing Nd solution gain media is a more sophisticated next-generation approach to moving the gain media.

FY03 Results

We developed a solvent that exhibited low toxicity, is readily available (i.e., used in industrial products), and is self-drying (which is critical to avoid quenching). It did not exhibit concentration quenching. Solutions with total absorption of 808-nm pump light over 1 mm had the same fluorescent lifetime (350 μs) as very dilute solutions. Common materials compatible with the lasing media were found. We constructed a laser test bed and achieved the first-ever CW operation of a Nd solution using a Ti-sapphire laser (350-mW out, 1.5-W pumping), as well as the first-ever diode pumping for a Nd liquid media (1.2 W, CW). Prior to our work, CW operation was not possible due to thermal optical distortions in the liquid. Finally, the first operation of a liquid heat-capacity Nd laser was performed.

Publications

Page, R. H. et al. (2003). “Renaissance of inorganic liquid-host neodymium lasers.” *Proc. OSA Ann. Meet.* UCRL-JC-153312 Abs.

Focusing Hard X-Rays at Current and Future Light Sources for Microscopy and High-Power Applications

Richard M. Bionta

01-ERD-097

Abstract

The objective of this project is to develop and demonstrate transmissive x-ray lenses suitable for use at future fourth-generation light sources. Our primary focus is to develop and demonstrate focusing lenses for the Warm, Dense Matter experiment, a Stockpile Stewardship Program (SSP) experiment proposed for the fourth-generation Linac (linear accelerator) Coherent Light Source (LCLS), a DOE Basic Energy Sciences initiative to develop an x-ray free-electron laser (XFEL) at the Stanford Synchrotron Radiation Laboratory (SSRL). LLNL has unique facilities for manufacturing these lenses through single-point diamond turning of beryllium (Be) and other toxic and hazardous materials whose use is necessitated by the large power loads expected at the LCLS. We plan to manufacture lenses and test them at the x-ray beamline at SSRL.

In this project, we expect to show how diamond turning techniques can be used to manufacture blazed Fresnel structures that will focus 8-keV x rays. These lenses will enable researchers to concentrate light from the LCLS onto solid samples, instantly turning them into a dense plasma whose properties can be studied in the laboratory environment. If successful, this project will give scientists an important tool to utilize the LCLS and may also lead to broader and even more significant roles for LLNL in the development and exploitation of this advanced light source.

Mission Relevance

The overall interest in the capabilities of the LCLS to do novel plasma, biological, and atomic physics experiments that support DOE mission in national security (specifically, the SSP) and in basic science. The availability of high-performance x-ray lenses will have direct application to the development of new x-ray sources through laser or electron interactions, including laser systems used in support of the SSP. These include both linac-driven XFELs and laser–electron beam Thomson sources.

FY03 Results

Both a diamond-turned aluminum-blazed phase plate and a very high-aspect-ratio Be paraboloid lens were constructed and tested along with a scintillation camera for imaging the x-ray patterns. The lenses and camera were tested at the Stanford Positron Electron Accelerating Ring beamline in four separate runs through March 2003. The camera sensitivity and resolution were within 10% of predictions. The aluminum lens had an efficiency that was 68% of the prediction, while the Be lens had only 9% of its predicted efficiency. Nevertheless, the demonstration was sufficient enough for LCLS to include such lenses in its work plan as part of the construction phase.

Nonlinear Saturation of Parametric Laser–Plasma Instabilities

Siegfried Glenzer 01-ERD-107

Abstract

An improved fundamental understanding of ion and electron plasma-wave saturation is important for high-energy laser experiments that require improved coupling of laser energy to targets. This project will directly measure the amplitudes of plasma waves to obtain quantitative information about laser–plasma instabilities. Experiments will be conducted at the Omega laser facility at the University of Rochester. Both the spatial and temporal evolution of the plasma waves will be measured and correlated with the time-resolved backscattering. The behavior of the plasma waves as the electron temperature, plasma density, material, and laser intensity are varied will sort out the viable mechanisms for saturation and establish the physics basis for target designs for large-scale laser facilities. We will develop a reduced model for quantitative predictions of stimulated Brillouin scattering in low-Z plasmas. The model will be tested against experiments and particle-in-cell simulations and implemented into the laser–plasma interactions code pF3D.

Mission Relevance

Direct measurements of the amplitudes of plasma waves that scatter the laser light in plasma nonlinearly will provide definitive quantitative results of laser–plasma instabilities in laser target designs. These results have direct application to inertial confinement fusion, high-energy-density physics, and nuclear weapons effects in support of the Laboratory's stockpile stewardship mission.

FY03 Results

In FY03, we performed a Thomson-scattering experiment at the Omega laser facility to provide high-quality spectral data on a single shot. For this purpose, an ultraviolet probe laser was deployed to measure both temperature and plasma-wave amplitude. Thomson-scattering diagnostics were used to independently measure the plasma electron and ion temperature, plasma flow velocity, and electron distribution function, as well as to measure the primary plasma wave and secondary nonlinear decay wave products. The measurements were particularly successful for stimulated Raman scattering, and the primary electron plasmas wave measurements for 3ω beams have now been adopted for future work.

Publications

Divol, L. et al. (2003). "Modeling the nonlinear saturation of stimulated Brillouin backscatter in laser-heated plasmas." *Phys. Plasmas* **10**, 1822. UCRL-JC-148980.

Froula, D. H. et al. (2003). "Observation of ion heating by stimulated-Brillouin-scattering-driven ion-acoustic waves using Thomson Scattering." *Phys. Plasmas* **9**, 4709. UCRL-JC-145885.

Froula, D. H. et al. (2003). "Stimulated Brillouin scattering in the saturated regime." *Phys. Plasmas* **10**, 1846. UCRL-JC-150869.

Froula, D. H. et al. (2003). "Direct observation of stimulated-Brillouin-scattering detuning by a velocity gradient." *Phys. Rev. Lett.* **90**, 155003. UCRL-JC-150767.

Kirkwood, R. K. et al. (2003). "Observation of saturation of energy transfer between copropagating beams in a flowing plasma." *Phys. Rev. Lett.* **89**, 215003. UCRL-JC-148550.

Structure and Spectroscopy of Black-Hole Accretion Disks

Duane A. Liedahl

02-ERD-004

Abstract

In this project, we are designing a computer model of accretion-disk atmospheres, Computer Models of Accretion Disk Structure and Spectra (COMPASS), with the goal of elucidating the high-radiation-density environments associated with mass flows in the curved spacetime near black holes. By combining our expertise in spectral modeling, LLNL's computational resources, and models of irradiated accretion disks developed by the project team, we will evolve the capability to generate realistic, theoretical x-ray spectra of accretion disks to explore the behavior of matter in the strong-field limit of gravitation. COMPASS will provide dramatically improved modeling capabilities of black-hole and neutron-star accretion disks.

Mission Relevance

This project maintains and extends LLNL's core competency in atomic physics modeling and computational radiation transport, which is vital to the Laboratory's stockpile stewardship mission. It will yield important new data sets useful for benchmarking our atomic physics calculations and to exercising our spectral synthesis and radiative transfer models, and will be an excellent recruiting magnet to attract talented astronomers and astrophysicists with skills already closely allied with weapons physics.

FY03 Results

Our team, in the process of refining COMPASS, discovered a number of useful applications of its capability. One result, which we wrote up for publication in *The Astrophysical Journal*, involves radiation reprocessing in accretion disk atmospheres in the vicinity of a spinning black hole. We have assessed quantitatively the importance of converting line radiation from highly-charged mid-*Z* elements into line emission from low-Z elements, in particular, oxygen and carbon, based upon our calculations of accretion disk structure and taking advantage of our Monte Carlo transfer code. This effect has been invoked in the past to explain x-ray line spectra from supermassive black holes. Similar projects, involving detailed radiation transport, are now underway.

Publications

Mauche, C. et al. (in press). "Reprocessing of soft x-ray emission lines in black hole accretion disks." *Astrophys. J.* UCRL-JP-200529.

Reaching Isochoric States of Matter by Ultrashort-Pulse Proton Heating

Pravesh K. Patel

02-ERD-006

Abstract

This project is developing a method to obtain experimental equation-of-state (EOS) data of high-pressure, off-Hugoniot material states in the warm, dense matter (WDM) regime (solid-density plasmas, 1 to 10 eV) for the first time. To accomplish this, we will use an intense, ultrashort pulse of protons produced by LLNL's Janus ultrashort-pulse laser to volumetrically and nearly instantaneously heat a material to high pressure (1 to 10 Mbar) and temperature (1 to 10 eV). In addition, the project will use a second proton beam to measure the rarefaction wave velocity in the plasma by two-dimensional spatial and time-resolved radiographs. This measurement will provide the first EOS data of any material on the isochore.

We expect to obtain, for the first time, experimental EOS data of high-pressure, off-Hugoniot material states, a previously inaccessible regime. The major result of this project will be a full understanding and optimization of the proton-production mechanism in laser–foil interactions.

Mission Relevance

Our project supports LLNL's national-security mission by investigating WDM, a regime that is relevant to the Stockpile Stewardship Program. In addition, successful collection of EOS data in a highly complex and previously inaccessible regime will impact many core elements of LLNL's scientific program in support of national security, including inertial confinement fusion target design, high-power laser experiments, planetary core physics, and high-pressure theory and molecular dynamics modeling.

FY03 Results

In FY03 we made considerable progress in the two main thrusts of the project: proton heating and proton radiography. The first successful experimental demonstration of isochoric heating was performed with an ultrashort-pulse beam of protons by heating a 10-µm-thick aluminum foil to a temperature of 4 eV. By focusing the protons, we were able to increase the temperature fivefold, to over 20 eV, within a small, 50-µm localized volume. In a separate experiment, at the LULI facility in Paris, we obtained the first proton radiograph of a release front from a laser-induced shock. This result demonstrated the potential for using the proton radiography technique for diagnosing the high-density release wave in either a laser-shocked or isochorically heated sample.

Proposed Work for FY04

In FY04 we will complete the remaining steps towards performing an integrated proton-heating and proton-radiography experiment. Our new multibeam interaction chamber, commissioned in FY03, will enable us to perform more detailed characterization measurements of the temperature and spatial uniformity of the proton-heated targets. In addition we will further develop the proton-radiography technique by probing highly transient phenomena in laser-produced plasmas. By the end of FY04, we will use both techniques simultaneously to first heat and then probe a material in a WDM state. The data obtained in these experiments will be used to benchmark LLNL EOS models.

Publications

Patel, P. K. et al. (2003). "Isochoric heating of solid-density matter with an ultrafast proton beam." *Phys. Rev. Lett.* **91**, 125004. UCRL-JC-201759.

Proton Radiography of Laser–Plasma Interactions with Picosecond Time Resolution

Andrew J. Mackinnon

02-ERD-012

Abstract

The project is developing laser-driven, megaelectronvolt (MeV) proton beams with picosecond time resolution as a diagnostic of density perturbations and electromagnetic fields in plasmas. Because proton beams with 10 to 100 MeV can penetrate through hundreds of micrometers of compressed material, they are useful in inertial confinement fusion (ICF) experiments for measuring density conditions in laser-driven capsule implosions. Proton deflectrometry can be used to directly measure transient electromagnetic fields in plasmas. The project will carry out proof-of-principle measurements of these proton beams as density and electromagnetic field diagnostics and will use the experiments to develop a predictive modeling capability for scaling the experiments to large laser systems such as the National Ignition Facility (NIF).

A proton probe beam with picosecond time resolution will provide entirely new information on the generation of extremely high electromagnetic fields inside a wide range of plasmas, from those occurring inside imploding shells to ignition conditions inside laser-driven fusion target capsules. Each stage in this process will produce publication-quality material that will benchmark progress.

Mission Relevance

The new diagnostic capability provided by proton probe beams with picosecond time resolution will contribute crucial information to the Stockpile Stewardship Program and the ICF Program in support of LLNL's national-security mission.

FY03 Results

In FY03, we determined experimentally that the proton beam arises from a virtual source located hundreds of micrometers away from the target. This work has been accepted for publication in *Physical Review Letters*. Proton deflectometry was successfully used to diagnose fields in a laser-produced plasma. We also used proton radiography to diagnose a laser-driven implosion on the Vulcan laser, and produced high-resolution proton radiographs of implosions. These were compared to picosecond x-ray K-alpha radiography.

Proposed Work for FY04

In FY04, experiments will be carried out on the JANUSP laser to investigate using proton deflectometry techniques for detecting laser-produced B fields. Our work in FY02/03 paved the way for us to begin developing the predictive capability of proton probing for diagnosing fields and density inhomogeneities using the LSP and COG codes. By the end of FY04, we plan to use these predictive capabilities to assess the suitability of proton radiography as a diagnostic of hot, dense plasmas relevant to the NIF ignition program.

Publications

Mackinnon, A. J. et al. (2003). "Proton moiré fringes for diagnosing electromagnetic fields in opaque materials and plasmas." *Appl. Phys. Lett.* **82**, 3188. UCRL-JC-149713.

Patel, P. K. et al. (in press). "Isochoric heating of solid-density matter with an ultrafast proton beam." *Phys. Rev. Lett.* **91**, 125004. UCRL-JRNL-20159.

Dense-Plasma Characterization by X-Ray Thomson Scattering

Otto L. Landen 02-ERD-013

Abstract

Our goal is to demonstrate x-ray Thomson scattering as a robust and unique technique for microscopic characterization of dense plasmas. Such information could be used to interpret laboratory measurements of material properties such as thermal and electrical conductivity, equation of state, and opacity. For example, the plasma ionization balance predicted by various theoretical models in the warm, dense plasma regime varies considerably and has never been tested against experiments. A series of x-ray Thomson scattering experiments will be completed at the Omega and upgraded Janus laser facilities, and the results compared to ionization-balance models and integrated radiation–hydrodynamic code predictions.

We expect the scattering experiments to distinguish between various existing ionization-balance models of low-*Z*, solid-density plasmas. The overall goal is to demonstrate x-ray Thomson scattering as relevant to high-energy-density physics (HEDP) and physical data research, for eventual application to the National Ignition Facility and other HEDP facilities. We hope to demonstrate the technique's applicability to kilojoule (not just >10 kJ) laser facilities. If the project is fully successful, we expect the technique to eventually be used at planned facilities such as the Linac Coherent Light Source and the Z Machine.

Mission Relevance

This work provides new insight into dense-plasma statistical mechanics, a key competency at LLNL in support of the Laboratory's stockpile stewardship mission. For HEDP, which also supports stockpile stewardship, we plan to demonstrate that x-ray scattering can provide a noninvasive, time-dependent measurement of the internal temperature in radiation-dominated flow in foams and gases. For inertial-confinement fusion, which supports LLNL's energy-security mission, x-ray scattering could be used to measure the fuel adiabat in both planar- and converging-capsule geometries.

FY03 Results

From our FY03 experiments, we obtained an improved and expanded scattered-spectra database from volumetrically heated beryllium (Be) and carbon (C) solids and foams, which provided ionization state vs. temperature data that are now beginning to distinguish between various proposed ionization-balance models. Carbon results show that high-density foam transitions to an insulating state at low electron temperature, contrary to the predictions of one model. We also began demonstrating the technique in hohlraum-driven foams for radiation-transport studies, which greatly improved our theoretical modeling of scattered spectra. Our work was documented in four published papers, one submitted manuscript, and five invited talks.

Proposed Work for FY04

In FY04, we plan to finish the demonstration and code comparisons of our scattering results from hohlraum-driven foams at Omega and extend the diagnosis to fully ionized solid-density plasmas by switching from Be and C target material to existing lower-*Z* lithium hydride (LiH) samples. We will also complete theoretical work and submit calculated spectra for publication, including weakly bound electrons and collective scattering off solid-density plasma waves. In parallel, we will design and field a higher-spectral-resolution focusing spectrometer for further LiH-probing studies at various scattering angles, allowing us potential access to the collective scattering regime. We hope to attract another postdoctoral candidate.

Publications

Glenzer, S. H. and G. Gregori. (2002). *X-ray scattering as a microscopic probe for solid density plasmas*. Presented at the APS DPP Meeting, Orlando, FL, Nov. 2002. UCRL-JC-150411-ABS.

Glenzer, S. H. et al. (2003). "Demonstration of spectrally resolved x-ray scattering in dense plasmas." *Phys. Rev. Lett.* **90**, 175002. UCRL-JC-149694-REV-1.

Glenzer, S. H. et al. (2003). "X-ray Thomson scattering from solid density plasmas." *Phys. Plasm.* **10**, 2433. UCRL-JC-151045.

Gregori, G. et al. (2003). *Calculation of x-ray scattering spectra for a carbon plasma*. Presented at the Intl. Workshop on LPI Physics, Banff, Canada, Feb. 2003. UCRL-JC-151531-ABS.

Gregori, G. et al. (2003). *Electronic structure measurements of solid density plasmas using x-ray scattering*. Presented at IFSA, Monterey, CA, Sept. 2003. UCRL-JC-152388-ABS.

Gregori, G. et al. (2003). "Strong coupling corrections in the analysis of x-ray scattering measurements." *J. Phys. A*. **36**, 5971. UCRL-JC-150760.

Gregori, G. et al. (2003). *Warm dense matter characterization by spectrally resolved x-ray scattering*. Presented at the Intl. Workshop on LPI Physics, Banff, Canada, Feb. 2003. UCRL-JC-151530-ABS.

Landen, O. L. (2003). *Energy transport and ionization balance in isochorically heated dense plasmas*. Presented at the APS April Meeting, Philadelphia, PA. April 2003. UCRL-JC-150199-ABS.

Lee, R. W. et al. (2003). "Finite temperature dense matter studies on next generation light sources." *JOSA* **20**, 770. UCRL-JC-147883.

Gaseous Laser Targets and Optical Diagnostics for Studying Compressible Turbulent Hydrodynamics

Michael J. Edwards 02-ERD-023

Abstract

This research, conducted in collaboration with the University of Texas at Austin, uses gas targets and optical diagnostics in laser-driven experiments to provide high-quality data on compressible turbulent hydrodynamics driven by high-Mach-number blast waves. High-Mach-number data cannot be obtained with traditional x-ray diagnostics and solid targets. In these experiments, the Janus laser is being used to drive a blast wave to interact with a gas

jet, producing a turbulent flow, which will be diagnosed by powerful optical methods. The experimental data will be used to develop and validate turbulence models.

If successful we expect to (1) obtain high-quality images resolving 2 to 3 orders of magnitude in spatial scale in compressible shock-driven turbulent flows; (2) obtain crude velocity information; (3) implement a laser-sheet diagnostic, which is the first step towards advanced diagnostic techniques; and (4) examine other potential advanced diagnostics. Our goal is to provide experimental data in the compressible regime that are needed to construct and validate turbulence models.

Mission Relevance

Compressible turbulent hydrodynamics cannot be simulated directly but must be calculated using models, which are necessarily based on experiment. However, virtually all existing high-quality data are from low-Mach-number experiments. This work supports LLNL's stockpile stewardship mission by providing improved data from high-Mach-number experiments for developing and validating turbulence models.

FY03 Results

In FY03, we conducted three Janus experiments, implementing greatly improved optics for high-resolution, wide-field imaging. Extensive blast-wave characterization was performed using Schlieren techniques, interferometry, and spectroscopy, demonstrating a Mach number of ~10 at a radial distance of ~ 3 to 5 cm. We also mapped out the operable background pressure range (0.01 to 0.1 atm); conducted shock–jet interactions with gas, low-density foam, and miniplasma balls; designed and tested a new gas-column target, a minichamber for operation at ~1 atm (allowing diagnostics to be incorporated more easily and with better results); and designed a laser-sheet diagnostic that will be fielded during FY04.

Proposed Work for FY04

Our two main objectives for FY04 are developing the gas target and the laser-sheet diagnostic. The first should allow us to acquire high-quality data on shock-driven turbulent flows using Schlieren doubled-pulsed imaging to provide velocity information. The second is the first step towards implementing an advanced diagnostic. We plan to acquire data using the above setup and the Schlieren and laser-sheet diagnostics, and to explore (and bench-test, if possible) additional advanced diagnostics. Particular emphasis will be on velocity spectra and correlations needed for turbulence models.

Publications

Hansen, J. F. et al. (2003). *Laboratory simulation of supernova shockwave propagation and ISM interaction*. Presented at IFSA, Monterey, CA, Sept. 8–12, 2003. UCRL-JC-152369-REV-1.

Hansen, J. F. et al. (2002). *Laboratory simulation of supernova shockwave propagation*. Presented at the APS DPP Mtg., Orlando, FL, Nov. 11–15, 2002. UCRL-JC-149200-ABS.

Hansen, J. F., M. J. Edwards, and A. Miles. (2003). *Laboratory simulation of supernova shockwave propagation and ISM interaction*. Presented at the IAU Colloq. on Supernovae, Valencia, Spain, April 22–26, 2003. UCRL-JC-152369-ABS.

Remote-Sensing Signatures for Ballistic Target Interceptions

Glen T. Nakafuji 02-ERD-035

Abstract

The objective of this project is to identify and characterize remote-sensing signatures for assessing the destruction of intercepted nuclear, chemical, or biological missiles. We will investigate candidate signatures using analysis, modeling, and experiments. Our initial focus is on nuclear signatures generated after a successful missile intercept, because identifying the type of incoming warhead is vital for determining operational, policy, and response options. We plan to validate nuclear signatures using explosively driven experiments to examine the behavior and spectral emission of low- and high-*Z* surrogate materials in conditions analogous to those during a successful missile intercept. We also plan to investigate the spectral signatures of organophosphate compounds. Successful results will significantly reduce uncertainty in signature calculations for nuclear targets and enhance capabilities in characterizing the signatures from devices with weak emission.

Mission Relevance

This work supports LLNL's national-security mission by characterizing signatures used for missile intercept, extending capabilities for identifying proliferant signatures, and providing a novel opportunity to further validate codes used in stockpile stewardship in modeling the temperature of metals in release and expansion.

FY03 Results

In FY03 we conducted five molten-metal jet experiments to benchmark LLNL code capabilities in modeling the release and expansion of molten metal. The measured jet tip speed and arrival time were in excellent agreement with pre-shot calculations. Molten debris temperature was measured with a four-color radiometer for all shots. The radiometric data were found to be reproducible over this series of shots. However, aerodynamic interaction of the jet with ambient conditions in the test chamber produced behavior that complicates accurate calculation of the surface emissivity. Reducing chamber pressure and selecting a suitable high-*Z* surrogate is expected to reduce uncertainty in the temperature measurements.

Proposed Work for FY04

In FY04, we plan to continue jet experiments using gold as a high-*Z* surrogate, conduct scaled surrogate experiments for signature validation, conduct tin-emissivity measurements, design experiments with high-*Z* surrogates, and explore the option of using laser-driven jet experiments to examine uranium.

Investigation of the Shores of the Island of Stability

Kenton Moody 02-ERD-038

Abstract

In this project, we explored the topography of the southwest edge of the "Island of Stability" by bombarding americium-243 (^{243}Am) (half-life = 7370 y) with calcium-48 (^{48}Ca) using the U400 cyclotron, at the Joint Institute for Nuclear Research (JINR) in Dubna, Russia. The goal of

this experiment was to synthesize the as-yet undiscovered element 115; should the element 115 nuclides decay via alpha-decay, as expected, element 113 would also be discovered. Chains of alpha decays are expected to lead away from the area of enhanced nuclear stability, terminated by spontaneous fission. The experiments were performed with the Dubna Gas-filled Separator, which is designed to observe these rare signatures.

Nuclear decay properties of the two elements we hoped to discover were expected to help verify that the closed proton shell that defines the center of the Island of Stability is at $Z = 114$. The spontaneous fission and alpha decay hindrance factors for odd-Z nuclides suggest that the signature for the decay of an element 115 isotope would be a 2- or 3-member alpha-decay chain, eventually terminated by spontaneous fission, thus helping define the extent of the region of enhanced nuclear stability.

Mission Relevance

This work supports national security programs by providing state-of-the-art training in nuclear physics and chemistry to staff members who provide radiochemical expertise in the areas of diagnosing nuclear-explosive device performance, identifying and diagnosing possible proliferant activities involving nuclear materials, and understanding issues related to nuclear power production and the safe disposal of radioactive materials. This high-profile science research is also a powerful recruitment tool.

FY03 Results

Between July 14 and August 10, 2003, in collaboration with the Heavy Element Group at JINR, we delivered a large number of ^{48}Ca ions to the ^{243}Am targets at two different irradiation energies, hoping to produce isotopes of element 115 through the evaporation of 3 or 4 neutrons, 288[115] and 287[115], respectively; we produced both isotopes. Both element-115 isotopes decay by alpha emission with half lives of approximately 40 ms, producing isotopes of element 113, which in turn decay by alpha emission with half lives of approximately 100 ms. We have discovered elements 115 and 113 (four atoms of each), further delineated the extent of the Island of Stability, and submitted a paper describing our results to *Physical Review C*.

Publications

Oganessian, Yu. et al. (in press). “Experiments on the synthesis of element 115 in the reaction $^{243}Am(^{48}Ca,xn)^{291}$-x115.” *Phys. Rev. C*. UCRL-JC-200047.

Development of a Predicive Computational Tool for Short-Pulse, High-Intensity Laser–Target Interactions

Max Tabak 02-ERD-041

Project Description

We propose to develop the theoretical understanding and computational capability to make credible predictions about the short-pulse laser design requirements and likely performance of proposed high-energy, short-pulse laser facilities. The code developed through this effort would be used to help define the requirements for these laser capabilities and develop target designs for short-pulse experiments. We plan to investigate the physics of high-intensity, short-pulse lasers with plasmas, focusing on the key issue of supercurrent electron transport in dense plasmas. We will use a hybrid particle-in-cell (PIC) method as the basis for the new computational tool, which will be validated against the results of recent short-pulse laser experiments at LLNL and elsewhere.

If successful, the project will be able to model the transport of electrons and protons for a large number of applications for the stockpile and the ignition and fusion-energy programs: fast ignition, isochoric heating of matter with protons for equation-of-state measurements, design of high-temperature hohlraums to drive opacity measurements, use of protons to radiograph imploded objects and to measure electric and magnetic fields in hohlraums, and transport of electrons to produce K-alpha and bremsstrahlung backlighters of optically thick objects. In addition, this capability will be used for modeling pair plasmas and neutron star atmospheres within the laboratory.

Mission Relevance

This work will support both the stockpile stewardship and fusion energy missions of DOE and the Laboratory.

FY03 Results

In FY03, we continued to benchmark the hybrid PIC code, LSP, against both theoretical models and experiments. We performed two- and three-dimensional calculations using LSP to help understand a number of recent experiments: K-alpha electron transmission measurements and plasma heating experiments on various short-pulse lasers. We improved the electrical conductivities used in the code and discovered that electron runaway could be important for experiments performed at longer pulse lengths. The code was used to model experiments measuring electric and magnetic fields with proton beams.

Publications

Kruer, W. L. et al. (in press). "Ingredients for improved modeling of MeV electron generation and transport for fast ignition studies." *Bull. Am. Phys. Soc*. UCRL-JC-153932-ABS.

Kruer, W. L. et.al. (2003). *Interaction of ultra-intense laser light with overdense plasmas*. UCRL-BOOK-200411.

Kruer, W. L. (2002). "Laser plasma interactions at intensities from 10^{12}–10^{21} W/cm^2." *Phys. Plasmas* **10**, 2087. UCRL-JC-149143-ABS.

Tabak, M. et al. (2003). *Models of gain curves for fast ignition*. Presented at the Third International Conference on Inertial Fusion Sciences and Applications, Monterey, CA, Sept. 2003. UCRL-PRES-151974.

Tabak, M. et al. (2002). *Electron transport for fast ignition*. Presented at the 6th Workshop on Fast Ignition of Fusion Targets, St. Petes Beach, FL, Nov. 16–19, 2002. UCRL-PRES-150144.

Tabak, M. (2002). *Fast ignition optimization depends on relative cost of short and long pulse energy.* Presented at the 6th Workshop on Fast Ignition of Fusion Targets, St. Petes Beach, FL, Nov. 16–19, 2002. UCRL-PRES-150145.

Tabak, M. (2002). *Physics of fast ignition*. Presented at the 6th Workshop on Fast Ignition of Fusion Targets, St. Petes Beach, FL, Nov. 16–19, 2002. UCRL-JC-151374-ABS.

Town, R. J. P. et al. (2003). *Calculations of proton radiography of magnetic fields in hohlraums*. Presented at the Third International Conference on Inertial Fusion Sciences and Applications, Monterey, CA, Sept. 2003. UCRL-PRES-152014.

Town, R. J. P. et al. (2003). *Recent LSP calculations of electron transport experiments*. Presented at the US-Japan workshop on the Theory, Simulation and Target Design for Fast Ignition, San Diego, CA, Sept. 2003. UCRL-PRES-155431.

Town, R. J. P. et al. (2002). *Calculations of electron transport in fast ignition targets*. Presented at the 44th Annual Meeting of the American Physical Society Division of Plasma Physics, Orlando, FL, Nov. 2002. UCRL-PRES-150143.

A Revolution in Biological Imaging

Henry N. Chapman 02-ERD-047

Abstract

This project will develop the science to enable ultrahigh-resolution imaging of biological materials with x-ray free-electron lasers (XFELs), now under development. We plan to solve several critical problems in XFEL imaging, including modeling the atomic motion of the sample and image reconstruction. Our calculations will be used to determine limits to single-molecule imaging as a function of fluence and pulse length. We will assess the importance of XFELs and existing light sources to life-science programs, and experimentally test x-ray imaging techniques on a three-dimensional (3-D), micrometer-sized test object.

Early results from our models and experiments indicate that atomic-resolution imaging on XFELs should be feasible. The pioneering work that will be performed in this project, such as the hydrodynamic model of the Coulomb explosion and the first lensless three-dimensional x-ray imaging, will establish the feasibility of atomic-level imaging of macromolecules. The single-molecule imaging will allow the structure of virtually any macromolecule, protein, or virus to be determined, and will have an enormous impact on structural biology and medicine. Our work will also determine a plan of research to develop the required technologies.

Mission Relevance

Single-molecule imaging furthers LLNL's missions in homeland security and in bioscience and technology to improve human health. Better tomography algorithms developed in this project will benefit stockpile stewardship. Our research also enhances the capabilities of the Linac Coherent Light Source, in support of LLNL's mission in breakthrough science and technology.

FY03 Results

The hydrodynamic model of the molecular Coulomb explosion, a major improvement over molecular dynamics studies, was completed. In addition to the Coulomb force, we added electron pressure, Debye shielding of electrons, trapping of photo- and Auger electrons, and rate equations for Auger production. With the insight from our analytic model, we proposed using a water tamper to delay the Coulomb explosion, thereby allowing longer and higher-intensity XFEL pulses.

We performed the first truly lensless experimental x-ray imaging—using coherent x-ray diffraction patterns we recorded at the Advanced Light Source—and developed a novel algorithm to reconstruct a 2-D image from the diffraction data, with a resolution of 10 nm.

Proposed Work for FY04

In FY04, we will extend our hydrodynamic model by adding photoelectron trapping and use the model to determine image resolutions as a function of sample size, pulse fluence, and duration. This will help determine the required parameters of XFEL sources and the work needed prior to any XFEL experiment. We plan to investigate methods to reduce damage, such as orienting molecules in a laser field and using a water tamper. We will extend our x-ray image-reconstruction algorithms and perform the first true 3-D x-ray imaging and, finally, design experiments to be conducted at the Tesla Test Facility (in Hamburg, Germany) in preparation for the world's first experiments with a XFEL source.

Publications

Bajt, S. et al. *Normal incidence multilayer coatings for 6 to 12-nm wavelength region.* Presented at the SPIE Conference on Advances in Mirror Technology for X-Rays, San Diego, August 2003. UCRL-JC-151205-ABS.

Chapman, H. N. et al. (2003). *Compression of x-ray FEL pulses with volume diffraction element. Presented at the SPIE Conference on X-Ray Optics for Fourth Generation X-Ray Sources*, San Diego, August 4, 2003. UCRL-PRES-151222.

Chapman, H. N. et al. (2003). *Prospects for single-particle imaging with XFELs*. Presented at the Second International Workshop on Noncrystallographic Phase Retrieval, Palm Cove, Australia, July 2003. UCRL-PRES-154182.

Hau-Riege, S. P. (2003). *Dynamics of x-ray irradiated molecules*. Presented at the Workshop on Plasma Physics, DESY, Hamburg, March 4, 2003. UCRL-PRES-151688.

Hau-Riege, S. P., R. A. London, and A. Szoke. (2003). *Hydrodynamic model of x-ray irradiated biological molecules.* Presented at X-Ray Science with Coherent Radiation, Berkeley, CA, August 2003. UCRL-PRES-151328.

Hau-Riege, S. P. et al. (in press) "SPEDEN: Reconstructing single particles from their diffraction patterns." *Acta Crystallographica A.* UCRL-JC-154574.

He, H. et al. (2003). "Inversion of x-ray diffuse scattering to images using prepared objects." *Phys. Rev. B* **67**, 174114. UCRL-JC-151664.

Howells, M. et al. (2003). "X-ray microscopy by phase-retrieval methods at the Advanced Light Source." *J. Physique IV* **104**, 557. UCRL-JC-151355.

Huldt, G., A. Szoke, and J. Hajdu. (in press). "Single particle diffraction imaging: image classification." *J. Struct. Bio.* UCRL-JC-154036.

London, R. A. (2002). *Hydrodynamic model of short pulse x-ray imaging.* Presented at the Imaging workshop, Uppsala University, Sweden. UCRL-PRES-149846.

London, R., S. P. Hau-Riege, and A. Szoke. (in press). "Dynamics of x-ray irradiated biological molecules." *Phys. Rev. E.* UCRL-JC-155565.

London, R. A., S. P. Hau-Riege, and A. Szoke. (2003). *Hydrodynamic model of x-ray irradiated nanoparticles.* Presented at the SPIE Conference on X-Ray Optics for Fourth Generation X-Ray Sources, San Diego, August 4, 2003. UCRL-PRES-151328.

Marchesini, S. et al. (2003). "Coherent X-ray diffractive imaging: applications and limitations." *Optics Express* **11**, 2344. UCRL-JC-155105.

Marchesini, S. et al. (2003). *Imaging without lenses.* Presented at the Second International Workshop on Noncrystallographic Phase Retrieval, Palm Cove, Australia, July 2003. UCRL-PRES-153586.

Marchesini, S. et al. (2003). “X-ray image reconstruction from a diffraction pattern alone.” *Phys. Rev. B* **68**, 140101(R). UCRL-JC-153571.

Miao, J. et al. (in press). “Taking x-ray diffraction to the limit: Macromolecular structures from femtosecond x-ray pulses and diffraction microscopy of cells with synchrotron radiation.” *Ann. Rev. Biophys. & Biomolecular Struct.* UCRL-JP-200184.

Nilsen, J. et al. (in press). “Mo:Y multilayer mirror technology utilized to image the near-field output of a Ni-like Sn laser at 11.9 nm.” *Opt. Lett.* UCRL-JC-152269.

Nugent, K. A. et al. (in press). “Unique phase recovery for nonperiodic objects.” *Phys. Rev. Lett.* UCRL-JC-152722.

Single-Particle Nanotracking for Genomes-to-Life Applications

Klaus Widmann

02-ERD-054

Abstract

This project is developing real-time, single-particle imaging techniques based on darkfield microscopy to observe the behavior of individual polymerase enzymes (molecular motors). Investigation of single polymerase behavior will allow direct measurements of enzyme velocity, DNA replication rate, residence time, processivity, and statistical variation, all as a function of the environmental parameters such as temperature, pH level, and polymerase concentration. Besides providing information adding to fundamental biological understanding, these measurements relate to rolling-circle DNA amplification (RCA), which may become a key process for the next-generation, real-time detectors for genetically coded material such as biological agents.

This project will result in a new capability for real-time imaging of labeled single biomolecules. Experiments on the RCA will generate fundamental information regarding the mechanisms that control the mobility of the enzyme.

Mission Relevance

The project will support LLNL’s national-security mission by enhancing competence in RCA, which has potential applications in bioterrorism-agent detection. In addition to ongoing and expected national-security-related projects, the experiments are relevant to the new DOE Genomes-to-Life initiative and will position LLNL to support existing and future DOE needs, e.g., pathogen detection, atmospheric warming, environmental cleanup, and energy production.

FY03 Results

Most of the work in FY03 focused on sample preparation, i.e., the characterization of functionalized polymerase with respect to its mobility and stability, and the optimization of the buffer and plasmid concentration with respect to the ss DNA loops and functionalized enzymes, respectively. We successfully linked biotinylated polymerase to functionalized silver-enhanced gold plasmon resonant particles (PRPs) without immobilizing the polymerase. The functionality of the biotinylated polymerase was measured as a bulk quantity. In particular, it was deduced from the amount of RCA-produced DNA material, and we could determine that the “output” of the biotinylated polymerase was more than 80% in comparison to the unlinked polymerase. Another important achievement was the preparation of the sample such that the ss DNA loops remain localized while providing unlimited mobility to the tagged polymerase enzyme. The concentration of the ss DNA loops has to be low enough to ensure the ability to observe single DNA loops while maintaining a high interaction. For the real-time imaging of our samples, we put together a darkfield microscope and image-intensified charge-coupled device camera system to study the motion of a single PRP-tagged enzyme with only tens of nanometers spatial resolution and a frame rate of up to 1 kHz. For studying the RCA process, we used frame rates of only 5 Hz or less by inserting a time delay between the individual frames. In our samples, we observed three different types of "movement" for the PRP-tagged polymerase: no motion at all, drift motion, and localized motion where the PRP moves but remains within a micrometer-size boundary. Utilizing a software algorithm developed during FY03, we demonstrated that this localized motion is consistent with the RCA process.

Proposed Work for FY04

Our proposed FY04 experimental program will have three major components: confirming previously observed polymerase activity, measuring the effect of environmental factors on polymerase trajectories, and generalizing tracking to other labels and bioorganisms.

Program of Simulations and Experiments for Assessment of Rapid Multipurpose Cargo-Scanning Technologies

Arden D. Dougan 02-ERD-064

Abstract

Cargo containers entering the U.S. by truck, rail, ship or aircraft can hide weapons of mass destruction (WMD), but inspecting the millions of cargo containers that enter the US is a daunting task. An effective detection method must be able to inspect containers quickly and to detect and identify contraband cargo with acceptable error rates, i.e. low rates both of false positives that interrupt commerce and false negatives that undermine confidence in the system.

This project focuses on developing a concept for a neutron interrogation system that can detect small targets of special nuclear materials (SNM) even when well shielded by thick cargo. The project will map out the limits of the method using carefully chosen experiments together with extensive high-fidelity modeling. This project will assess the usefulness of variable-energy neutron sources, larger and more capable neutron and gamma-ray detectors, efficient collimation and shielding, background-suppression techniques, and sophisticated data-analysis algorithms.

Mission Relevance

This project supports LLNL's national security and homeland security missions by providing enhanced capabilities for detecting WMD materials entering U.S. ports in cargo containers.

FY03 Results

A viable concept for cargo interrogation has been developed, and its components have been evaluated experimentally. A new radiation signature unique to SNM has been identified that utilizes high-energy, fission-product gamma-rays. That signature due to radiation in the range 3-6 MeV is distinct from normal background radioactivity, which does not extend above 2.6 MeV. Its short half-life of 20 to 55 s makes it distinct from neutron activation due to interrogation, which typically much longer lived. The signature flux, while small, is 5 decades more intense than the delayed neutron signals used historically and facilitates the detection of SNM even when shielded by thick cargo. The actual benefit is highly dependent on the type and thickness of cargo, with modest benefit in the case of metallic cargos of iron, lead, or aluminum, but maximum benefit in the case of hydrogenous cargo. In addition, unwanted collateral effects of the interrogation, such as neutron activation of the cargo, were analyzed, and one significant interference due to oxygen activation was characterized. This interference can be eliminated by lowering the energy of interrogating neutrons. No other collateral effects have yet been identified. The neutron source technology required exists commercially.

Publications

Norman, E. B. et al. (2003). “Signatures of special nuclear material: High-energy gamma rays following fission.” *Nucl. Instr. and Meth. A.* UCRL-JC-153259.

Slaughter, D. R. et al. (2003). *Detection of special nuclear material in cargo containers using neutron interrogation*. UCRL-ID-155315.

Slaughter, D. R. et al. (2004). *The “nuclear car wash”: A Scanner to detect illicit special nuclear material in cargo containers.* UCRL-JRNL-202106.

Photon Collider Physics

Jeffrey B. Gronberg
03-ERD-003

Abstract

This project will develop the physics case and run plan for photon collider experiments at high and low energies. A photon collider involves backscattering short-pulse laser photons (~1 eV) from incident electron beams just a few millimeters before collision. This upshifts the photons to nearly the full energy of the electrons, producing collisions of hundreds of gigaelectronvolts. A photon collider experiment has no precedents, and a detailed simulation of the signal and background is critical to quantifying its physics potential. A signal possibly representing new physics and well suited to study at a photon collider will be analyzed at the proton collider at Fermilab. Moreover, this project, if successful, will create a suite of Monte Carlo tools that will allow the evaluation of physics analyses possible at a photon collider proposed at the Stanford Linear Collider (SLC), in which LLNL would have technical and scientific leadership. This project will also produce publishable estimates of the reach of new physics and spin-zero heavy-quark physics at a photon collider and a publishable search for new physics at the Collider Detector at Fermilab (CDF).

Mission Relevance

Photon colliders open an entirely new field of particle physics and are enabled by LLNL’s unique competencies in high-average-power, short-pulse lasers and laser–electron beam interactions. Both of these are central to innovative concepts for future brilliant light sources to be used in dynamic materials studies in the Stockpile Stewardship and Inertial Confinement Fusion Programs, in support of the national and energy security missions.

FY03 Results

The luminosity of an SLC-based photon collider experiment was simulated, and the yield of eta (η) and chi (χ) events was determined. The Monte Carlo generator Pandora was modified to generate η-particle events, and the Monte Carlo generator Pythia was tuned to match the production rate of two-photon background events. The reconstruction of η and χ decays to two protons was simulated for a representative detector along with the expected background from direct production of proton–antiproton pairs. The bottom-squark meson decay signatures were studied, and the lambda baryon–hadron decay mode was chosen for reconstruction. The optics required to time-format the laser pulse to match the time structure of the electron bunch was simulated, revealing effects from differential path length in the optics.

Proposed Work for FY04

The Monte Carlo tools created in FY03 will be used to analyze the ability to reconstruct and observe physics beyond the Standard Model in the SLC photon collider experiment. This experiment will produce thousands of Higgs bosons for some parameters of general supersymmetric models, for which a low-mass Higgs boson has not yet been ruled out. The ability to observe low-mass spin-zero supersymmetric particles will also be quantified at CDF, and the results published. The effects of radiation damage in the optics will be quantified by irradiating test blanks at Stanford Linear Accelerator Center.

QED and Electron Collisions in the Super Strong Fields of K-Shell Actinide Ions

Peter Beiersdorfer

03-ERD-004

Abstract

This project studies novel physics in the ultrahigh fields of the heaviest long-lived elements available—uranium, curium, and californium. Our objective is to record the world's first high-resolution K-shell x-ray spectra of these elements. To access new physics missing in modeling codes used at LLNL and elsewhere, the elements will be stripped of all but a few electrons in collisions with energetic electrons. The resulting spectra will allow us to determine electron-impact excitation and dielectronic recombination cross sections, as well as ionic energy levels. The project combines several LLNL capabilities: the world's only source of stationary high-*Z* ions, access to actinide isotopes, high-resolution x-ray detectors developed in collaboration with the National Aeronautics and Space Administration, and unique theory capabilities.

The project will (1) provide the most definitive test of high-field quantum electrodynamics ever undertaken, (2) identify the role of new forces active in high-*Z* collisions, and (3) quantify their significance for spectral modeling of high-temperature plasmas. The project will also develop a new class of hard x-ray detector—a high-resolution microcalorimeter array operating near absolute zero—and a new, ultrahigh-resolution extreme ultraviolet spectrometer, creating unique experimental capabilities.

Mission Relevance

This effort to investigate new physics in the ultrastrong fields of highly charged actinide ions test the validity range of collisional theory important to stockpile stewardship. It also builds capabilities in astrophysics and atomic physics in accordance with the Laboratory's mission in basic science. In addition, this work will also attract talented young scientists to the Laboratory.

FY03 Results

In FY03, we brought online at Livermore the Super Electron Beam Ion Trap (SuperEBIT), the world's only source of stationary, highly charged ions, and carried out low-resolution measurements of K-shell x-ray spectra of highly charged uranium. The measurements showed spectral line intensity ratios indicative of the importance of the generalized Breit interaction, resulting in differences of a factor of 2 over standard calculations. We also brought online a new grating spectrometer for determining the 2s Lamb shift, began construction of a system for injecting rare isotopes, and started developing a high-resolution, large-area, hard x-ray detector. A 2-pixel prototype calorimeter has shown excellent performance during testing.

Proposed Work for FY04

The second year of the project will concentrate on characterizing the 2-pixel prototype hard x-ray calorimeter, then implementing a full 16-pixel detector array. The instruments will first be used to measure high-resolution K-shell spectra of xenon, then hydrogen-like and helium-like uranium. The latter will yield a measurement of the 1s Lamb shift in uranium. The uranium K-shell excitation rates will be determined and compared with calculations utilizing the generalized Breit interaction. In addition, we plan to measure the uranium dielectronic recombination and bring the actinide and rare isotope injection system online.

Publications

Chen, M. H. et al. (2003). "Transition energies of the 3s-3p3/2 resonance lines in sodiumlike to phospheruslike uranium." *Phys. Rev. A* **68**, 022507. UCRL-JC-152236.

Trabert, E. et al. (2003). "EUV spectra of highly charged Xe ions." *Phys. Rev. A* **68**, 042501. UCRL-JC-153669.

Exploring Properties of Quantum Chromodynamics with Proton–Nucleus and Deuteron–Nucleus Collisions

Stephen C. Johnson 03-ERD-005

Abstract

The standard model of strong interactions predicts a phase transition in which nucleons dissolve into a plasma of quarks and gluons. Relativistic heavy-ion collisions are the only laboratory-controlled method available for producing and studying this phase of matter and is the primary goal of the Relativistic Heavy Ion Collider (RHIC) at Brookhaven National Laboratory. Experimental proof of the production of this phase relies on a set of observations that are currently inconclusive. The key to their resolution lies in studying baseline proton–nucleus collisions. We will upgrade the Pioneering High Energy Ion Experiment (PHENIX) experiment at RHIC to characterize the fragmentation of the nucleus in proton–nucleus collisions. This will aid in the systematic study of bulk nuclear matter at high energy densities and help resolve a long-standing question: Can we create a new phase of matter, the quark–gluon plasma, in the laboratory and study its properties? We expect to gain a much improved understanding of physics in this regime.

Mission Relevance

This work further the Lab's national-security mission by advancing cutting-edge techniques in nuclear radiation detection and state-of-the-art computer simulation of complex events. This project will also help recruit top scientists in these fields through forefront research in nuclear and particle physics.

FY03 Results

In FY03, this project's work was used in the PHENIX collaboration to help resolve the plasma puzzle. We tested all components of the calorimeter and installed them in the PHENIX experiment. This undertaking involved testing all calorimeter modules and phototubes and overseeing design of the sophisticated detector stand, electronics, power distribution, and readout. The detector was completely installed and used to collect data in RHIC experimentation. We performed a first-pass calibration and began analyzing signals from our detector, which, in conjunction with complementary measurements, was used to characterize the global properties of proton–nucleus collisions.

Proposed Work for FY04

The installed detector has produced a wealth of data that need to be systematically studied over the following 1 to 2 yr. In FY04, following global characterization we will begin the analysis of specific predicted observables in these collisions with the hope of resolving the question of the existence of a quark–gluon plasma. To accomplish this work, we will hire two new postdocs who are highly ranked in this field.

Electron Production and Collective Field Generation in Intense Particle Beams

Arthur Molvik

03-ERD-008

Abstract

The future of heavy-ion fusion could depend on predicting and controlling electron cloud effects (ECEs), which limit the performance of present electron and ion storage rings. To better understand the phenomena and develop mitigating mechanisms, this project is conducting theoretical and experimental studies of the generation, trapping, and transport of electron clouds and their effects on ion beams. Experiments will be carried out at the Heavy-Ion Fusion Virtual National Laboratory. Theoretical studies will coordinate with Lawrence Berkeley National Laboratory collaborators who are combining a code that models electron clouds with a particle-in-cell code, WARP, for modeling self-consistent beams in accelerators. The project will validate the models and tools through comparison with experimental data.

Results from modeling and experiment will lead to the development of definitive, validated tools for studying, and mitigating the effects of, ECEs in advanced accelerators to enable higher performance. Energy and particle sources leading to gas desorption and electron emission by ion impact on surfaces will be studied and identified to improve mitigation measures, which would have a large impact on the accelerator community.

Mission Relevance

By addressing electron effects in ion beams, the models and diagnostic techniques developed in this project will be applicable to high-intensity accelerator technology for stockpile stewardship and fusion energy, in support of LLNL's national and energy security missions.

FY03 Results

In FY03 we formulated and began implementing the drift-kinetic electron model in WARP to enable simulation on the longer ion time scale. Ion-beam simulations with WARP showed that random electron clouds exceeding a few percent of beam density were deleterious to beam quality. Simulations also showed electron detrapping at acceleration gaps. We measured electron emission and gas desorption coefficients, identified models, and began commissioning diagnostics inside the bores of magnetic quadrupole magnets. During the year, collaborations were established with the University of California, Berkeley, and private companies.

Proposed Work for FY04

In FY04, we plan to describe electron clouds and resultant field modifications, averaged over fast electron time scales; compare the model in detail with experiments; and evaluate selected future accelerators in light of these results. Work will include experiments to understand the energy and particle sources driving gas desorption and electron emission; integrating gas desorption, transport, and ion scattering at the accelerator wall into the model; applying codes to understanding ECEs in accelerator elements; applying and improving present diagnostics; working with the accelerator community to prioritize diagnostics for development; and evaluating and developing further diagnostics to more completely characterize electron clouds.

Publications

Cohen, R. H. et al. (2003). Stray-electron accumulation and effects in HIF accelerators." *Proc. 2003 Particle Accelerator Conf.*, May 12–16, 2003, Portland, OR. UCRL-JC-153199.

Molvik, A. W. et al. (2003). "Initial experimental studies of electron accumulation in a heavy-ion beam." *Proc. 2003 Particle Accelerator Conf.*, May 12–16, 2003, Portland, OR. UCRL-JC-151178.

Coupled Turbulence-Transport Model for Edge Plasmas

Thomas D. Rognlien 03-ERD-009

Abstract

This project is developing the first predictive simulation model of the edge-plasma characteristics in tokamak fusion devices. Experiments have shown that edge-plasma properties are key to producing high fusion-energy output, and predictive simulations are needed to reliably design large devices. This project uses parallel computer codes to solve magnetized-fluid equations for both the fast-timescale turbulence in three dimensions (3-D) and the much slower toroidally averaged transport (profile evolution) in 2-D. Because the turbulence is driven by the plasma profiles, and profile transport is determined by turbulent plasma fluxes, multiple-timescale coupling techniques are developed to obtain self-consistent solutions, including plasma and neutrals from recycling and sputtering.

The model developed in this project will be used to help design and guide operation of future devices and power plants and will also be an important precursor of a more comprehensive computer model of a fusion device being planned under DOE's Integrated Simulation and Optimization of Fusion Systems initiative; both the physics models and the coupling techniques will be directly applicable to this planned computing initiative. The coupling techniques should be applicable to other complex fluid systems, such as climate and combustion.

Mission Relevance

This project advances fusion-energy research in support of LLNL's energy-security mission and develops numerical algorithms for large, coupled physics simulations on LLNL's high-performance parallel computers in support of stockpile stewardship. The project also provides a postdoc position to train a new LLNL scientist in a mission-critical discipline.

FY03 Results

In FY03 we imported the full 2-D plasma profiles from the transport code UEDGE into the turbulence code BOUT and found a new turbulence regime with high fluctuations near the divertor material surfaces; developed and tested coupling methods for nonlinear and noisy turbulent fluxes to the transport code, and found that the numerical performance of our robust transport solver is acceptable even with this added complexity; performed initial UEDGE simulations with turbulent density fluxes from BOUT; completed 2-D domain message-passing modifications for BOUT; worked on a final tridiagonal solver; and demonstrated the first profile evolution on a turbulence timescale within BOUT in the presence of power and particle sources.

Proposed Work for FY04

We will benchmark turbulence–transport coupling by comparing results from UEDGE and BOUT with background mode evolution for moderate times; extend coupling to the full set of five variables, including electrostatic potential, which gives rise to the key shearing electric field; parameterize residual collisional radial transport with a guiding-center code to describe edge plasma with shear-suppressed turbulence; validate the coupled edge plasma model in collaboration with experimentalists from the DIII-D and Alcator C-Mod tokamaks; delineate how experimental benchmarks scale with collisionality to begin quantification of kinetic effects; and assess useable kinetic corrections for fluid models to be implemented.

Publications

Rognlien, T. D. et al. (2003). *Coupled transport and turbulence modeling for tokamak edge-barriers.* 9th IAEA Technical Meeting on H-mode Physics and Transport Barriers, Sept. 24–26, 2003, San Diego, CA. UCRL-JC-155467 Abs.

Rognlien, T. D. et al. (2003). *Coupled turbulence and transport evolution for magnetized edge plasmas.* 2003 Intl. Sherwood Theory Conf., Apr. 28–30, 2003, Corpus Christi, TX. UCRL-JC-152125 Abs.

Rognlien, T. D. et al. (in press). "Self-consistent simulation of turbulence and transport in tokamak edge plasmas." *Contributions to Plasma Phys.* UCRL-JC-153197.

Umansky, M. V. et al. (in press). "Numerical solution of strongly anisotropic diffusion equation on misaligned grids." *J. Comp. Phys.* UCRL-JC-153575.

Umansky, M. V. et al. (2003). "Modeling of turbulence and transport in tokamak edge." *Bull. Am. Phys. Soc.* **48**, 228. UCRL-JC-154188.

Umansky, M. V. et al. (in press). "Turbulence in the divertor region of tokamak edge plasma." *Contributions to Plasma Phys.* UCRL-JC-155308.

Xu, X. Q. et al. (in press). "Correlation of density pedestal width and neutral penetration length." *Contributions to Plasma Phys.* UCRL-JC-153196.

Strain-Rate Scaling of Deformation Mechanisms

Bruce A. Remington

03-ERD-015

Abstract

Our objective is to determine material-deformation mechanisms at ultrahigh pressures and strain rates. Further, we intend to assess the scalability of the results across a wide range of very high strain rates to determine whether what we learn about deformation mechanisms at very high strain rates is applicable to other regimes. To accomplish our objectives, we will use controlled, shockless loading along with soft-capture recovery on four experimental facilities: the Omega laser, the LLNL gas gun, a high-explosives facility, and the National Ignition Facility Early Light laser. Post-shot characterization will allow us to determine deformation mechanisms. Multiscale modeling simulations will guide experimental planning and mechanistic interpretation.

We expect to demonstrate the methodology for determining deformation mechanisms over a wide range of strain rates and ultrahigh pressures. This methodology uses multiple facilities to produce the relevant test conditions along with recovery and post-shot characterization to infer the operative deformation mechanisms. The results from this project will provide basic scientific information in unexplored regions of pressure–strain rate space, as well as providing a demonstrable scientific underpinning for the high-pressure strength measurements critical to the Stockpile Stewardship Program. Additional benefits include initiating an effort to investigate solid-state material behavior under extreme conditions and recruiting young scientists.

Mission Relevance

Deformation mechanisms provide the scientific underpinning of strength measurements at ultrahigh pressures. This project supports LLNL's national-security mission by developing a methodology for both assessing these deformation mechanisms and determining the scalability of high-strain-rate results, which will be critical input for long-range, science-based stockpile stewardship.

FY03 Results

In FY03 we focused on demonstrating our techniques at a single facility using a single material. We verified shockless drive and developed recovery on the Omega laser and recovered single-crystal and polycrystalline samples driven to peak pressures between 100 and 400 kbar; these samples were prepared for characterization. We also used molecular-dynamics simulations to tentatively identify the slip-twin transition in shock-driven single-crystal copper at about 370 kbar.

Proposed Work for FY04

In FY04, we propose to (1) determine deformation mechanisms in single-crystal and polycrystalline copper over a pressure range of 100 to 400 kbar; (2) deploy soft-recovery capability on the LLNL gas gun; and (3) conduct a scaling study on copper using the LLNL gas gun, the Omega Laser, and a high-explosives facility. Use of these facilities will allow us to obtain results on a single material over four orders of magnitude in strain rate.

Publications

McNaney, J. M. et al. (in press). “High pressure, laser driven deformation of an aluminum alloy.” *Met. Trans A.* UCRL-JC-152750.

Determining Phonon-Dispersion Curves in Delta-Phase Plutonium–Gallium Alloys

Joe Wong

03-ERD-017

Abstract

The goal of this project is to measure the phonon-dispersion curves (PDCs) of face-centered-cubic (fcc) plutonium–gallium (Pu–Ga) alloys, along with their temperature, composition, and pressure dependencies. Pure Pu exhibits six solid-state phase transitions, with large volume changes, along the path to melting: alpha, beta, gamma, delta, delta′, epsilon, and liquid. The delta phase has desirable engineering properties, and alloying Pu with Ga expands the delta-phase field from high temperature to below room temperature.

To measure the PDCs of Pu–Ga alloys, we will use x-ray scattering methods employing high-brightness synchrotron sources to eliminate both the neutron absorption problem of ^{239}Pu and single-crystal requirements for conventional PDC measurements with neutron scattering.

We expect to determine (1) complete PDCs for a 0.6-wt% Ga–Pu alloy at room temperature; (2) the temperature dependence of the T[111] mode; (3) the Ga content dependence of the T[111] mode and the Kohn anomaly in the T1[110] branch; and (4) the Gruneisen’s parameters from pressure-dependent measurements. The significance of this project is that, if successful, it will (1) obtain the first full phonon dispersion for any Pu material ever determined and provide bona fide data to test first-principle theories for Pu and other 5f-electron systems; (2) help understand the role of T[111] softening and the Kohn anomaly with respect to the delta-to-alpha′ phase transformation in fcc Pu–Ga alloys, which will help elucidate their phase stability; and (3) provide materials data for design code that simulates extreme environmental conditions.

Mission Relevance

This project will provide much-needed basic lattice dynamical data for phase-stability and property simulations of Pu materials relevant to science-based stockpile stewardship, in support of the national-security mission.

FY03 Results

In FY03 we (1) fabricated large-grained, single-domain fcc Pu–Ga specimens; (2) designed scattering geometries for high-resolution inelastic x-ray scattering (HRIXS) measurements at the European Synchrotron Radiation Facility (ESRF), in Grenoble, France; (3) were awarded beam time at ESRF via competitive proposal; and (4) determined the first full phonon dispersions for an fcc Pu–Ga alloy. The results were published in *Science*.

Proposed Work for FY04

In FY04 we will fabricate and characterize large-grained fcc Pu–Ga alloys with various Ga content; refine sample configuration and scattering geometry and instrumentation; use HRIXS to investigate the Ga content dependence of T[111] softening; perform thermal diffuse scattering (TDS) measurements at room temperature for delta-Pu–Ga alloys; and analyze the IXS and TDS data.

Colliding Nanometer Beams

Jeffrey B. Gronberg 03-ERD-044

Abstract

The goal of this project is to develop and demonstrate an alignment and metrology frame that will allow beam-position monitors (BPMs) to reach nanometer resolution in measuring the transverse position of an electron beam required for a linear collider. Future linear colliders will achieve high event rates by focusing electron beams down to spot sizes of 3 nm. Stabilization systems to hold the final magnetic optics steady at that level are under development, but definitive tests would require construction of a linear collider. Stabilization system failure would be a showstopper. A stabilization system test using a single beam is possible with nanometer-resolution BPMs.

The project will determine whether existing cavity BPMs can achieve nanometer resolution. This would be a more than an order-of-magnitude improvement in current beam diagnostics and would provide sufficient signal sensitivity to monitor bunch tilt, which is expected to have a significant effect on the luminosity of future accelerators. We will design and build a metrology and alignment frame for a set of BPMs to demonstrate nanometer resolution. A working nanometer-resolution BPM would allow the stabilization scheme required for colliding beams at a linear collider to be tested, greatly improving our confidence in the chosen linear collider design.

Mission Relevance

The DOE advisory panel on high-energy physics has identified the Next Linear Collider (NLC)—a teraelectronvolt-scale electron–positron linear collider—as the next big project in high-energy physics. LLNL is a founding member of the NLC consortium, and this project will allow a potential showstopper to be tested before NLC construction and advance our capability in nanometer metrology, which will support LLNL's mission in breakthroughs in fundamental science and help recruit young talent.

FY03 Results

The conceptual design of the alignment frame was finished and its vibrational characteristics fully simulated, ensuring that the BPMs will move in lockstep for the relevant ground-motion excitations. All parts for the alignment frame were procured, and metrology frame designs were created. We evaluated sensor technologies and selected an optical nanogrid as the only viable alternative. We analyzed use of the Final Focus Test Beam at Stanford but rejected it due to concerns that higher-order modes would contaminate the measurement.

Proposed Work for FY04

In FY04 we will install the BPM alignment frame at the Accelerator Test Facility in Japan, then begin gathering data. The resolution of the system will be measured and published. We will also procure, construct, and install the metrology frame for this system. Thermal drift of the BPM positions will allow the metrology frame measurements to be correlated with BPM measurements to validate the absolute position measurement accuracy of the system.

Laser–Matter Interactions with a 527-nm Drive

Warren W. Hsing

03-ERD-070

Abstract

The primary goal of this research is to develop an understanding of laser–matter interactions with 527-nm light (2ω), which is relevant to future large lasers for fusion research. The potential of significantly greater energy delivered onto targets at 2ω allows us to access a wider range of parameter space for ignition (higher yield or more robust ignition), probe increasingly higher pressures in matter, and access a wider parameter space for hohlraum-driven experiments. By reducing unconverted light in the target chamber, it also provides significant simplification of experiments due to the. This project includes developing the technology and prototype instrumentation to diagnose a high-fluence laser beam for energy, power, and near-field intensity profile at 2ω.

As a result of this work we will resolve important scientific questions relating to laser–plasma interactions, laser-target coupling, preheat, and hohlraum filling, and gain a better understanding of the advantages and limitations of 2ω illumination relative to 3ω for high-energy-density (HED) and inertial confinement fusion (ICF) applications. The database of 2ω light coupling to matter is much less developed compared to 3ω. This project will make an important contribution toward assessing the utility of 2ω light for stockpile stewardship experiments on future large laser systems such as NIF.

Mission Relevance

Five key areas within the Stockpile Stewardship program would significantly benefit from the ability to conduct experiments with a 2ω drive: ignition, material dynamics, potential experiments using special nuclear materials, nuclear weapons effects testing, and radiation-hydrodynamics. This project has applications for cutting-edge research in other LLNL mission areas such as inertial confinement fusion (ICF) for energy security and high-energy-density (HED) physics for basic science.

FY03 Results

Our FY03 work made significant progress in all areas of 2ω laser–plasma interaction physics and diagnostics: (1) 2ω transmitted beam diagnostics were installed on Omega and have provided measurements of transmitted light through a large, under-dense plasma with beam smoothing at 2ω. (2) A Thomson scattering experiment measured electron plasma wave amplitudes locally and a new 4ω Thomson scattering diagnostic was commissioned, which provides the first accurate temperature measurements in gasbags at Omega. (3) 2ω stimulated Raman scattering and stimulated Brillioun scattering reflectivity into the lens were measured for smoothed 2ω beams. The design work for diagnostics to measure energy, power, near-field intensity profile, and full-aperture backscatter at 2ω was nearly completed.

Proposed Work for FY04

In FY04 we plan to compare data from Omega experiments with theory for 2ω propagation in filled, thin-wall hohlraums; obtain scaling of x-ray conversion efficiency and hot-electron production as a function of incident laser intensity with 2ω light incident on well-characterized plasmas; compare experiment with theory for both 2ω and 3ω coupling in well-characterized hohlraum-relevant, high-temperature, low-Z plasmas; compare experiment and theory for a hohlraum filling mitigation scheme that allows greater utility of the energy available at 2ω; and build prototype diagnostics and make hardware modifications needed for 2ω.

Characterization and Optimization of High-Energy K-alpha X-ray Sources

Jeffrey A. Koch 03-ERD-072

Abstract

The use of x-ray sources as backlights for radiography experiments is an established technique for studying hydrodynamics, equations-of-state, and other properties of materials under extreme conditions of high temperature and density. This project is exploring the generation, characterization, and optimization of high-energy (20- to 100-keV) K-alpha x-ray backlight sources for high-energy petawatt (HEPW) radiography experiments in future large laser systems. Our goal is to develop the capability to predict source parameters in HEPW experiments and develop experimental hardware appropriate for these experiments. We plan to characterize K-alpha source parameters as functions of laser and target parameters in experiments at the Janus ultrashort-pulse laser facility and (if possible) at higher-energy petawatt-scale laser facilities. We will also investigate high-energy x-ray detection and x-ray imaging technology relevant to this energy range and determine the relative utility of various approaches.

Accomplishing the above goals will provide a baseline understanding of how to produce 22-keV x-ray sources for various planned HEPW experiments, as well as specific measurements of various important source characteristics such as yield, spectral bandwidth, time duration, and size. We will also understand how to detect and focus these x-rays most efficiently. Our characterization will be the first comprehensive study. Work in future years will move to higher x-ray energies, up to 100 keV.

Mission Relevance

This project investigates how to most effectively perform stockpile assessment experiments at existing and future large laser systems that use high-energy x-ray backlights to study large-volume, high-atomic-number materials.

FY03 Results

In FY03, we performed experiments at the JanUSP laser facility to measure silver (Ag) K-alpha yield for various laser intensities, laser durations, laser spot sizes, and laser energies and compared the results to existing data in the literature with good agreement. We also performed a preliminary measurement of the Ag K-alpha source size. Preliminary measurements were obtained of Ag foil K-alpha yield in experiments at the Vulcan laser facility at Rutherford Appleton Lab, and a more thorough experimental investigation at Vulcan has been planned for

for early FY04. We also obtained preliminary data on the usefulness of overcoating the Ag foils with plastic, with unclear results. Finally, we performed Monte Carlo simulations of various target geometries to help guide future experiments.

Proposed Work for FY04

In FY04, we plan to measure Ag K-alpha spectral bandwidth using crystal spectroscopy; measure Ag K-alpha time duration using a fast-streak camera; demonstrate an Ag K-alpha radiograph using apertured foils, edge-on foils, or embedded dots and wires; test a refractive bubble lens for focusing Ag K-alpha x rays; and perform comparison tests of a microchannel plate, imaging plate, and film against a scintillator charge-coupled device for Ag radiographs.

Publications

Park, H. -S. (2003). *High energy K-alpha x-ray sources for HEDES experiments.* Presented at APS/DPP 03 Meeting, Albuquerque, NM, Oct. 27–31, 2003. UCRL-JC-154249-ABS.

Park, H. -S. et al. (2003). *High energy K-alpha x-ray source generation by short pulse high intensity lasers.* Presented at IFSA 03 Conference, Monterey, CA, Sept. 8–12, 2003. UCRL-JC-152380-ABS.

Park, H. -S. et al. (2003). *High energy x-ray source generation pulse high intensity lasers*. Presented at SPIE 03 Conference, San Diego, CA, August 3–8, 2003. UCRL-JC-151565-ABS.

Wilks, S. C. et al. (2003). *Modeling K-alpha images produced in short-pulse laser driven electron transport experiments*. Presented at IFSA 03 Conference, Monterey, CA, Sept. 8–12, 2003. UCRL-JC-152290-ABS.

A Compact Accelerator for Proton Therapy

George J. Caporaso

03-ERD-073

Abstract

High-gradient insulator technology developed at LLNL, coupled with the dielectric-wall accelerator (DWA) concept, has produced the potential for a compact, high-voltage, short-pulse accelerator, which is being developed as a flash x-ray radiography source for stockpile stewardship. We plan to exploit this compact accelerator technology to build small proton accelerators for cancer radiation therapy and make this effective treatment widely available. Proton therapy is superior to x-ray therapy yet is only practiced in a few locations in the U.S. due to the large size and high cost of these facilities.

This project has three major objectives. The first is to examine the feasibility of a DWA architecture that meets the requirements for proton therapy. This architecture will be verified by simulation and by construction and testing of scalable prototypes. Second, we plan to develop an understanding of the beam parameters needed for standard and novel proton radiotherapies. Third, we will evaluate the use of this short-pulse accelerator to produce Compton sources and advanced light sources. At the conclusion of this effort, we will know whether DWA can be applied to proton therapy. In addition, this work may lead to compact accelerators for other applications such as high energy physics, Compton scattering, and free-electron laser drivers.

Mission Relevance

The project focuses on using DWA in a regime that has the potential to produce the highest accelerating gradient of any non-plasma-based acceleration scheme. These accelerators could be used for medical treatment, Compton scattering sources, free-elecctgron laser drivers, and high-energy physics, in support of LLNL missions in national security and biotechnology to improve human health. Furthermore, the project will enhance capabilities in induction linac technology for stockpile stewardship.

FY03 Results

We began developing a DWA pulse format (repetition rate, pulse width, and charge per pulse) in FY03 that would match the parameters required for proton therapy. We also began designing a prototype voltage-transformation system to achieve high gradients. The system consists of a symmetric Blumlein gallium arsenide photoconductive switch and a dispersive nonlinear transmission line to compress and amplify pulses for acceleration. We began design of a silicon carbonate photoconductive switch.

Proposed Work for FY04

In FY04, we will test our prototype voltage-transformation system and investigate the use of this accelerator architecture for electrons to produce a Compton scattering source and an advanced light source.

A Tunable, Monochromatic, 1-Angstrom, Compton-Scattering X-Ray Microfocus for Multiwavelength Anomalous Diffraction Experiments

Frederic V. Hartemann

02-ERI-004

Abstract

The main goal of this project is to achieve the first demonstration of the microfocus multiwavelength anomalous diffraction (micro-MAD) concept, whereby tunable, monochromatic x rays produced by a Compton-scattering microfocus source are imaged on a small protein crystal to yield the full three-dimensional (3-D) structure of the macromolecule, with atomic resolution. The demonstration will be performed on a simpler crystal, such as a mercury salt, to limit data collection to a manageable time. The x rays are produced at LLNL's Picosecond Laser-Electron InterAction for the Dynamical Evaluation of Structures (PLEIADES) facility.

The demonstration of relatively inexpensive, compact, tunable Compton-scattering x-ray sources operating near 1 Å, and capable of producing diffraction patterns revealing the full 3-D structure of macromolecular proteins with atomic resolution using the MAD phasing technique, has the potential to revolutionize x-ray protein crystallography by allowing access to high-throughput structural genomics at small university laboratories. Such novel, compact x-ray sources are also essential for advanced backlighting applications at large laser systems and for in situ material studies, including the time-resolved dynamics of high-Z elements.

Mission Relevance

This work supports the Stockpile Stewardship Program by developing techniques for in situ studies of materials under extreme conditions. In addition, these techniques strengthen to the Laboratory's homeland security mission by enabling the systematic search for viral and pathogenic protein structures, and has broad implications for LLNL's biotechnology mission to improve human health.

FY03 Results

FY03, we successfully produced 70-keV x-rays and characterized them in detail, which showed excellent agreement with our 3-D theory. Our work produced a static radiograph of tantalum and demonstrated K-edge imaging. We completed electron beamline simulations and experimental characterization and developed a 3-D time- and frequency-domain Compton scattering code. A new x-ray window/infrared mirror, described in a previous Record of Invention, was implemented, along with the micro-MAD interaction setup at PLEIADES. We also presented several invited talks at major international conferences and began writing a number of papers describing these important results and the new technologies involved.

Proposed Work for FY04

In FY04, two key experiments will be performed. The x-ray absorption K-edge of selenium will be scanned to show the required tunability and monochromaticity of the Compton x-ray source and its relevance to MAD phasing; we have already shown that PLEIADES can produce static radiographs of high-*Z* materials, and that the K-edge absorption can readily be measured and imaged. Second, anomalous diffraction will be measured on a small, well-diffracting crystal to validate the micro-MAD concept. We also plan to collaborate on measurements of the high average x-ray flux produced by Compton scattering in the Jefferson Laboratory's infrared free-electron laser. Jefferson Laboratory's 75-MHz superconducting linac technology, coupled with LLNL's high-average power lasers, provides a direct path to novel, compact, tunable x-ray sources.

Publications

Brown, W. J. et al. (2003). *Generation of high brightness x-rays with the PLEIADES Thomson x-ray source*. Presented at the 2003 Particle Accelerator Conf., Portland, OR, May 12–16, 2003. UCRL-JC-153476.

Fittinghoff, D. et al. (2003). *PLEIADES: A subpicosecond Thomson x-ray source for ultrafast materials probing*. Presented at the Commercial and Biomedical Applications of Ultrafast Lasers V, San Jose, CA, Jan. 25–31, 2003. UCRL-JC-151594.

Hartemann, F. V. et al. (2003). *Basic concepts in advanced x-ray sources*. Presented at the Ultrashort High-Energy Radiation and Matter Conf,, Varenna, Italy, Oct.–10, 2003. UCRL-JC-155311-ABS.

Hartemann, F. V. et al. (2003). *Compton scattering and its applications: The PLEIADES femtosecond x-ray source at LLNL*. Presented at the Intl. Wkshp. Quantum Aspects of Beam Physics, Hiroshima, Japan, Jan. 7–10, 2003.UCRL-JC-153189.

Hartemann, F. V. et al. (2003). *PLEIADES: A picosecond, high peak brightness Compton scattering x-ray source for advanced backlighting and time-resolved material studies*. Presented a the 3rd Intl. Conf. on Inertial Fusion Sciences and Applications, IFSA 2003, Monterey, CA. UCRL-JC-152294-ABS.

Kuba, J. et al. (2003). *First light at PLEIADES: High peak brightness, femtosecond Thomson x-rays*. Presented at the 29th Inertial Confinement Fusion Assoc. Advanced Beam Dynamics Wkshp. on Laser-Beam Interactions, Oxford, England, July 7–11, 2003. UCRL-JC-152658-ABS.

Le Sage, G. P. et al. (2003). *The LLNL Thomson x-ray source.* Presented at the 27th ICFA Advanced Beam Dynamics Wkshp.: The Physics and Application of High Brightness Electron Beams, Sardinia, Italy, July 1–6, 2003. UCRL-JC-153890.

Springer, P. T. et al. (2002). *Ultrafast materials probing with the LLNL Thomson x-ray source.* Presented at the 21st Intl. LINAC Conf., Gyeonju, Korea, Aug. 19–23, 2002. UCRL-JC-150440.

Fermion Monte Carlo

Malvin H. Kalos

01-LW-040

Abstract

This research aims for a major advance in solving quantum-many-body problems. The Schrödinger equation for many-fermion systems has been under study for six decades, but no accurate methods that are without uncontrolled approximations have emerged. Our aim is to develop and demonstrate a new class of methods capable of overcoming this difficulty—practical Monte Carlo methods that solve the nonrelativistic Schrödinger equation without relying on uncontrolled approximations. This is a formidable task: No such numerical method currently scales to large numbers of electrons. Monte Carlo methods are beset with the "Fermion sign problem." Overcoming these obstacles will be a major scientific accomplishment and, among other results, will move quantum chemistry into a realm of objectively predictive science. Our project has three general themes: (1) improving computational efficiency by several orders of magnitude; (2) developing robust computer programs for different applications; and (3) predicting the physical and chemical properties of molecules, including benchmark computations. In addition, we should be able to compute atomic structure, equations of state, properties of condensed matter, nuclear structure, and chemical binding.

Mission Relevance

Our new methods will provide LLNL with advanced computational tools to treat many scientific challenges relevant to LLNL's stockpile stewardship mission, including atomic structure, equations of state, properties of condensed matter, nuclear structure, and chemical binding.

FY03 Results

We continued work on improving the efficiency for extensive and molecular systems. Numerical results for the Boron dimer were robust and agreed with experimental results. Progress continued on the helium-3 system. We began a study of the two-dimensional electron gas and have been able to treat successfully a system of 58 electrons—the largest number studied so far. Promising new analytic forms of the importance functions were introduced and studied. Our work has been presented at three international scientific conferences and workshops.

Beta-Decay Experiments and the Unitarity of the Cabibbo-Kobayashi-Maskawa Matrix

Paul E. Garrett

02-LW-026

Abstract

A study of super-allowed beta decay in nuclei, governed by the weak force, provides a unique way to test the Standard Model predictions of electroweak interactions. This project will test the unitarity of the quark-mixing Cabibbo-Kobayashi-Maskawa (CKM) matrix required by the Standard Model. Beta-decay tests of unitarity require extreme precision in measuring

beta-decay half lives and branching ratios, and calculated corrections must also be applied to the data. The project will test and constrain these calculated correction factors by performing accurate half-life and branching-ratio measurements at Canada's TRIUMF radioactive beam, ISAC, for nuclei where different sets of calculated corrections give divergent results.

The ultimate goal of this project is a definitive conclusion regarding the unitarity of the quark-mixing matrix and adequacy of the Standard Model of particle physics. This will be achieved by testing the unitarity condition for the first row of the CKM matrix, where the nuclear physics contribution is largest. Findings suggest that the sum of squares of the matrix's first row differs from unity at the 98% confidence level. If the results of our testing support these findings, it would be the first indication of physics beyond the Standard Model, a result of major scientific importance. This significant and high-profile research in nuclear and particle physics will attract talented young physicists to the Laboratory.

Mission Relevance

Tests of the Standard Model have been identified as a high priority in the DOE and National Science Foundation Long-Range Plan for Nuclear Science. These studies complement and support the experimental program at DOE Office of Science laboratories such as the Stanford Linear Accelerator Center and Fermilab. Since the work demands extremely precise measurements, techniques developed for the experimental program are being transferred to other areas, such as cross-section measurements on radio-chemical detectors performed at the Los Alamos Neutron Science Center's, Weapons Neutron Research facility with the GEANIE spectrometer.

FY03 Results

Accomplishments in FY03 include completing and debugging the new data-acquisition system and testing it with the sodium-26 radioactive beam. The data acquisition system was further expanded to incorporate various detector types, and has grown into one of the most sophisticated presently in use for nuclear physics experiments. A wide variety of supporting experiments have been performed, including studies of lithium-11 decay, isomer searches in mass 172, 178, and 179 nuclei, and gamma-ray decays from the 16-yr ^{178m2}Hf isomer. The LLNL team is also the spokesman on the experimental effort to search for a mass or charge dependence on the extracted superallowed ft values [a product of the Fermi integral function (f) and the half-life (t)].

Proposed Work for FY04

In FY04, we plan to perform branching-ratio and half life measurements on argon-34, analyze the data, and extract the superallowed ft value. We will then compare our experimental results to the different theoretical calculations to determine which theoretical approach gives the correct description. Preparatory work for the heavier decay studies, in 66-As and bromium-70, will also be performed.

Publications

Garrett, P. E. (2003). *A ≥62 super-allowed Fermi beta decays: constraining unknown corrections with 66-As and 70-Ar decay.* UCRL-PROP-151366.

Smith, M. B. et al. (2003). "Gamma rays emitted in the decay of 31-year ^{178m2}Hf." *Phys. Rev. C* **68**, 031302(R). UCRL-JP-200315.

Smith, M. B. et al. (2003). "Studies of high-K isomers at TRIUMF-ISAC." *Proc. 6th Intl. Conf. Radioactive Nuclear Beams (RNB6)*. UCRL-ABS-200314.

A High-Efficiency Grazing-Incidence-Pumped X-Ray Laser

James Dunn

03-LW-001

Abstract

Our objective is to demonstrate a new type of high-efficiency, short-wavelength (<20-nm) x-ray laser operating at 10-Hz rates and scalable to 1 kHz, with a pulse duration between 100 fs and a few picoseconds. This high-average-power x-ray laser would complement the waveband of third- and future fourth-generation light sources like the Linac Coherent Light Source. The latter are large, expensive, synchrotron-based facilities, whereas our laser-driven tabletop source will be compact and inexpensive. This research will improve the laser-pumping efficiency by a factor of 10 to produce a proof-of-principle grazing-incidence-pumped (GRIP) 10-Hz x-ray laser. Plasma density characterization and simulations of Compact Multipulse Terawatt (COMET) laser coupling, x-ray laser production and propagation will be performed using the LLNL JanUSP and high-power, short-pulse lasers.

With our new concept for improving x-ray laser efficiency, we expect to achieve near-synchrotron-level average brightness on a tabletop but with orders-of-magnitude higher peak brightness. The goal is to produce a proof-of-principle demonstration with sufficient output energy for applications relevant to LLNL programs and the scientific and high-technology communities. In addition, by using the single-shot, high-energy capability of the JanUSP laser, we will create the shortest wavelength (<4.5-nm) laboratory x-ray laser operating in the water window, important for cellular imaging and holography.

Mission Relevance

X-ray probing of ultrafast processes in materials such as actinides is relevant to stockpile stewardship. A tabletop source at-wavelength ~13-nm would be ideal for metrology of extreme ultraviolet lithography optics for microchip development, in support of LLNL's mission in breakthroughs in fundamental science and technology. This x-ray laser could operate below 4.5 nm, giving a single-shot capability lasting 100 fs for microscopy and holography of biological cells, in support of LLNL's mission in bioscience to improve human health.

FY03 Results

All our FY03 milestones were accomplished, and we demonstrated the new GRIP x-ray laser. We designed a low-cost laser-beam-focusing geometry for a high-quality, traveling-wave-line focus necessary for the novel pumping geometry. A dedicated x-ray laser target chamber was constructed and an x-ray spectrometer installed. We performed a detailed study, with precise density-profile measurements, of a transient nickel-like protactinium and molybdenum (Mo) x-ray laser plasma using the unique COMET picosecond x-ray laser interferometer. Simulations were performed to determine the initial laser pumping for the GRIP inversion. The first x-ray laser experiments were carried out in June and August. We successfully demonstrated a nickel-like Mo x-ray laser at 18.9-nm wavelength, operating at 10 Hz with only 130-mJ pump energy.

Proposed Work for FY04

We will continue to develop the <20-nm, 10-Hz x-ray laser in FY04. A detailed understanding of the laser-coupling process is essential since the scheme relies on selectively pumping a chosen plasma gain region. This will be achieved through direct x-ray laser experiments and density-profile measurements of different flat- and confined-plasma target geometries with the COMET x-ray laser interferometer. Detailed simulations of the actual experimental geometry will help us to determine the optimum laser–plasma conditions and geometry for creating the high-efficiency 10-Hz x-ray laser. The aim is to achieve operation in the maximum-output gain-saturation regime and implement an x-ray laser beamline with x-ray optics. High-power single shots towards a 4.5-nm x-ray laser will be performed.

Publications

Keenan, R. et al. (2003). “Efficient pumping schemes for high average brightness collisional x-ray lasers.” *SPIE Proc.*: Soft X-Ray Lasers and Applications V, E.E. Fill (ed.). 5197. UCRL-PROC-200323.

Shlyaptsev, V. N. (2003). *Modeling of grazing incidence pumping scheme.* UCRL-SR-201464.

Diode-Pumped Alkali Atom Lasers

Raymond J. Beach

03-LW-024

Abstract

The objective of our project is the first-ever scientific demonstration of a diode-pumpable laser using atomic alkali vapors as the gain media, a new class of laser that has the potential for extreme power scaling and offers definite advantages over competing diode-pumped solid-state lasers for machining special materials. The project will demonstrate lasing to the ground electronic level of an alkali atom using a titanium:sapphire laser as a surrogate diode pump.

If successful, the project will enable efficient, visible laser transitions on the first and second D-line series of the alkali atoms for the first time. The project will demonstrate the viability of diode-pumpable alkali-based lasers in the blue region of the spectrum, where there are significant materials-processing, bioanalysis, and commercial applications.

Mission Relevance

High-average-power, high-beam-quality laser systems have particular relevance to Laboratory stockpile stewardship mission involving the machining of special materials, as well as scientific studies focused on the interaction of matter with intense resonant fields. The success of the alkali laser endeavor could lead to significant commercial applications in the field of material processing such as disk mastering and in diagnostic bioanalysis.

FY03 Results

We successfully achieved our FY03 milestones by demonstrating for, the first time, a diode-pumpable rubidium laser operating at 795 nm. We also demonstrated a convincing understanding of the underlying physics associated with this class of lasers via the excellent agreement between our experiments and our model-predicted performance.

Proposed Work for FY04

Using a cascaded two-pump excitation sequence, we plan to demonstrate visible lasing in the blue spectrum with an alkali gain medium. The design of this laser builds on our work in the first year, using our experimentally anchored modeling codes. Specifically, using atomic cesium we propose to demonstrate blue lasing at 455.7 nm, with surrogate diode pumps at 852.3 nm and 876.4 nm.

Publications

Beach, R. J. et al. (2003). *End-pumped 895-nm Cs laser.* Presented at the 2004 Adv. Solid State Phot. Mtg. UCRL-ABS-155602.

Krupke, W. F. et al. (2003). “Diode pumpable rubidium laser.” *OSA TOPS* **83**. UCRL-JC-151484.

Krupke, W. F. et al. (2003). “Resonance-transition, 795-nm rubidium laser.” *Opt. Lett.* **28** (23), 2336–2338. UCRL-JC-152977.

A Next-Generation Compton Imager

Lucian Mihailescu 03-LW-031

Abstract

Imaging gamma rays efficiently and accurately is a very important goal in many applications involving radioactive materials. The purpose of this work is to demonstrate a compact gamma-ray imager of unprecedented sensitivity with a 4-π field of view by using Compton camera concepts. For this, we employ large-volume planar germanium (Ge) detectors accompanied by the use of new concepts for signal processing and data analysis. This type of camera, unlike the collimator-based imagers, can be used reliably for objects situated at infinity and for 3-D objects in close proximity.

This system will be used as a demonstration unit for nuclear nonproliferation and environmental monitoring applications. We plan to test the Compton camera in a single-photon-emission computer tomography (SPECT) system for imaging small animals for biomedical research. Compton-enhanced SPECT could potentially increase imaging sensitivity by an order of magnitude. In this respect, lighter radionuclides that emit gamma rays of energies higher than 1 MeV can be investigated as new SPECT tracers. The developments made on Compton imaging will help advance the science for an Advanced Compton Telescope mission planned for the next decade.

Mission Relevance

By introducing a new, highly sensitive monitoring system, the project will contribute to mission-related applications in nuclear nonproliferation and nuclear-waste monitoring. Advanced gamma-ray imaging systems based on this design will be also find application in nuclear medicine for the Laboratory’s mission in bioscience to improve human health, and in gamma-ray astrophysics for the Laboratory’s basic science mission.

FY03 Results

In the first year of the project, a preliminary design of the imaging system was completed, a first prototype of a gamma-ray imager based on Compton camera concepts was assembled, and a complete first version of a data-analysis chain was developed, which brought the data from the level of detector-signal waveforms to final gamma-ray images. Completely new concepts for data processing were found and implemented, including new filters for digital signal processing, pulse-shape analyses for superior detection granularity, and new image-reconstruction methods. The prototype demonstrated good sensitivity. A 360° lens photo camera was used in conjunction with the imaging system to visually identify radioactive sources in the environment.

Proposed Work for FY04

Development and refinement of algorithms for the reconstruction of the gamma-ray scattering sequence and algorithms for image reconstruction from Compton camera data will be the main tasks for FY04. Dedicated image-reconstruction algorithms will be developed for far-field sources, and imaging of extended sources and close-field sources will be also addressed. Finally, measurements will be performed with the newly developed dedicated detectors to verify the system performance for typical cases.

Publications

Burks, M. et al. (2003). *A 4-π field of view Compton imager based on a single planar germanium detector*. Presented at the IEEE Nucl. Sci. Symp. UCRL-POST-200368.

Mihailescu, L. et al. (2003). *A high-sensitivity 4-π gamma-ray imager*. Presented at the IEEE Nucl. Sci. Symp. UCRL-POST-200717.

Mihailescu, L. et al. (2003). *Data analysis for Compton scatter imaging*. Presented at the IEEE Nucl. Sci. Sym. Compton Wkshp. UCRL-PRES-200716.

Optical Parametric Amplification in Photonic Crystals

Igor Jovanovic 03-LW-040

Abstract

Optical parametric chirped-pulse amplification (OPCPA) is a technique for generating high-energy, high-average-power, ultrashort pulses. This project studies quasi-phase-matched (QPM) nonlinear crystals for use with OPCPA. QPM has many advantages over birefringent phase matching: higher gain, better beam quality, and greater wavelength flexibility. The project will develop a theoretical framework for extending QPM OPCPA to a broad range of important wavelengths, and, for the first time, scale QPM experimentally to multimillijoule energies by increasing the crystal aperture. The poling facility developed in this project will be used to design, fabricate, and test a novel, multilayer photonic structure based on a nonlinear material that has favorable properties at shorter wavelengths. Developing a multilayer photonic structure for OPCPA will result in much improved beam quality and stability of the front-end, high-gain preamplifier for petawatt (PW) laser systems, and will enhance research in the field of ultraviolet-light amplification.

Mission Relevance

This work is of critical importance for developing broad-bandwidth, short-pulse technology for petawatt (PW) high-peak-power lasers. The use of PW lasers in high-energy-density facilities will enable high-resolution x-ray backlighting and fast ignition studies for the Stockpile Stewardship Program.

FY03 Results

In FY03, we studied angular acceptance in QPM theoretically, using a new mathematical model that evaluates spectral, angular, and energetics properties of an arbitrary QPM structure. We designed and experimentally demonstrated, for the first time, a 1053-nm high-gain, broad-bandwidth preamplifier that uses periodically poled potassium titanyl phosphate. Our poling station for lithographic manufacturing of periodically poled structures was in the final phase of commissioning at the end of the year.

Proposed Work for FY04

In FY04, we plan to develop a general code to optimize the spectral bandwidth in any nonlinear crystal configured for QPM. The result of our modeling will provide the first complete framework for amplifying broad-bandwidth pulses centered at an arbitrary center wavelength. We plan to identify the limits for scaling crystals to large apertures and produce first ultrabroad bandwidth in QPM OPCPA operating far from wavelength degeneracy.

Publications

Jovanovic, I., J. R. Schmidt, and C. A. Ebbers. (in press). "Optical parametric chirped-pulse amplification in periodically poled $KTiOPO_4$ at 1053 nm." *Appl. Phys. Lett.* UCRL-JC-153251-REV-1.

Jovanovic, I., J. R. Schmidt, and C. A. Ebbers. (2003). *Optical parametric chirped-pulse amplification in periodically poled $KTiOPO_4$ at 1053 nm.* Presented at Ultrafast Optics IV, Vienna, Austria, June 29–July 3, 2003. UCRL-JC-152595-ABS.

The Creation of Neutron Star Atmospheres on a Petawatt Laser

Richard I. Klein 03-FS-005

Abstract

Extreme conditions of density and temperature that are relevant to stockpile stewardship and inertial confinement fusion are similar to those found in low-altitude atmospheres of magnetized neutron stars. This project focuses on developing a preliminary design for a petawatt laser experiment that reproduces the dynamics and thermodynamics of the atmospheres of a magnetized accreting neutron star's atmosphere and on investigating the possible existence of the photon bubble instability. We performed a detailed analytic and numerical study of the feasible densities, temperatures, and magnetic fields achievable with a petawatt laser experiment. This was accomplished by performing radiation-hydrodynamic simulations to estimate thermal temperatures and radiation fields, particle-in-cell (PIC) simulations to characterize magnetic fields and hot electrons, and analytic studies of instability timescales.

Preliminary estimates of the temperatures, densities, and magnetic-field strengths achievable with current petawatt laser interactions with solid-density material address the feasibility of scaling the conditions on a neutron star to a laboratory experiment.

Mission Relevance

This project has direct relevance to understanding the extreme conditions of high-energy-density and ultrashort-pulse laser–matter interactions that are relevant to LLNL's stockpile stewardship and energy security missions, and will be a driver for advanced scientific applications at future petawatt lasers. This world-class science project will also enhance recruiting by attracting talented scientists to LLNL, and will forge strong links with the U.S. astrophysical community.

FY03 Results

In FY03, we obtained preliminary analytic estimates for physical timescales of the photon bubble instability, performed preliminary PIC and laser-shock processing simulations to characterize the magnetic field spatiotemporal distribution, characterized the energy and angular distribution of the hot electrons, and performed preliminary radiation-hydrodynamic simulations to estimate the electron radiation coupling and the expected acceleration of the plasma package.

The Search for Extra Dimensions: Probing Spacetime with Positronium

Leslie J. Rosenberg

03-FS-018

Abstract

Unifying gravity with the strong, weak, and electromagnetic interactions requires that spacetime have additional, undetected dimensions. Positronium may provide a way to probe these extra dimensions. A small probability exists that positronium annihilation “disappears” into the unseen spacetime dimensions. This research studied the feasibility of conducting experiments to detect positron production from an enclosed radioactive source within a large-volume, liquid-scintillator detector. Steps included identifying feasible source and detector configurations, evaluating and optimizing the positron “mis-tag” rate, evaluating the gamma-ray interactions in the liquid scintillator, developing a tagger design, and determining whether the experiment would need to operate underground.

Mission Relevance

The modeling conducted in this study will provide valuable information for detectors now being constructed for cargo-container active interrogation for the Laboratory’s nonproliferation and homeland security missions. Furthermore, both the basic science goal of this experiment and the cutting-edge technology and modeling capabilities employed make this project a good recruiting opportunity for the Laboratory.

FY03 Results

In FY03, we evaluated several potential detector concepts, beginning with a sodium-22 positron source at the center of a tank of liquid scintillator. When we realized that this design failed to reject rare Compton processes from electron capture (EC) decays in the source, the original concept was augmented with a germanium detector. While this detector has impressive rejection of EC backgrounds, it was less impressive at rejecting inner bremsstrahlung backgrounds associated with the EC branch. Although this baseline detector did not reach the target branching-ratio sensitivity of 10 ppb, it laid the groundwork for future research.

Diode-Laser Phase Conjugation

Ralph H. Page

03-FS-030

Abstract

This project assesses the feasibility of using optical phase conjugation to achieve coherent beam combination by using a plurality of high-power semiconductor laser diodes. This technique, if successful, would constitute an improved method of constructing a high-power, high-brightness, efficient, compact laser system. Based on our previous experience with solid-state-laser rate-equation modeling, we are adapting theoretical models from low-power, single-mode diodes to high-power, broad-area (multimode) diodes and designing a laboratory experiment to validate the concept.

If feasible, large-scale coherent multiplexing of powerful, compact, efficient, inexpensive laser diodes would be a major development in laser technology. In principle, it could displace alternative means of “radiance conditioning” (diode-pumping of solid-state lasers, for example) and lead to high-power laser applications in materials processing, missile defense and power beaming, for instance. Diode-laser phase conjugation may enable powerful systems to be more easily deployed in remote locations. In addition, the project will identify key issues and obstacles to developing the technology.

Mission Relevance

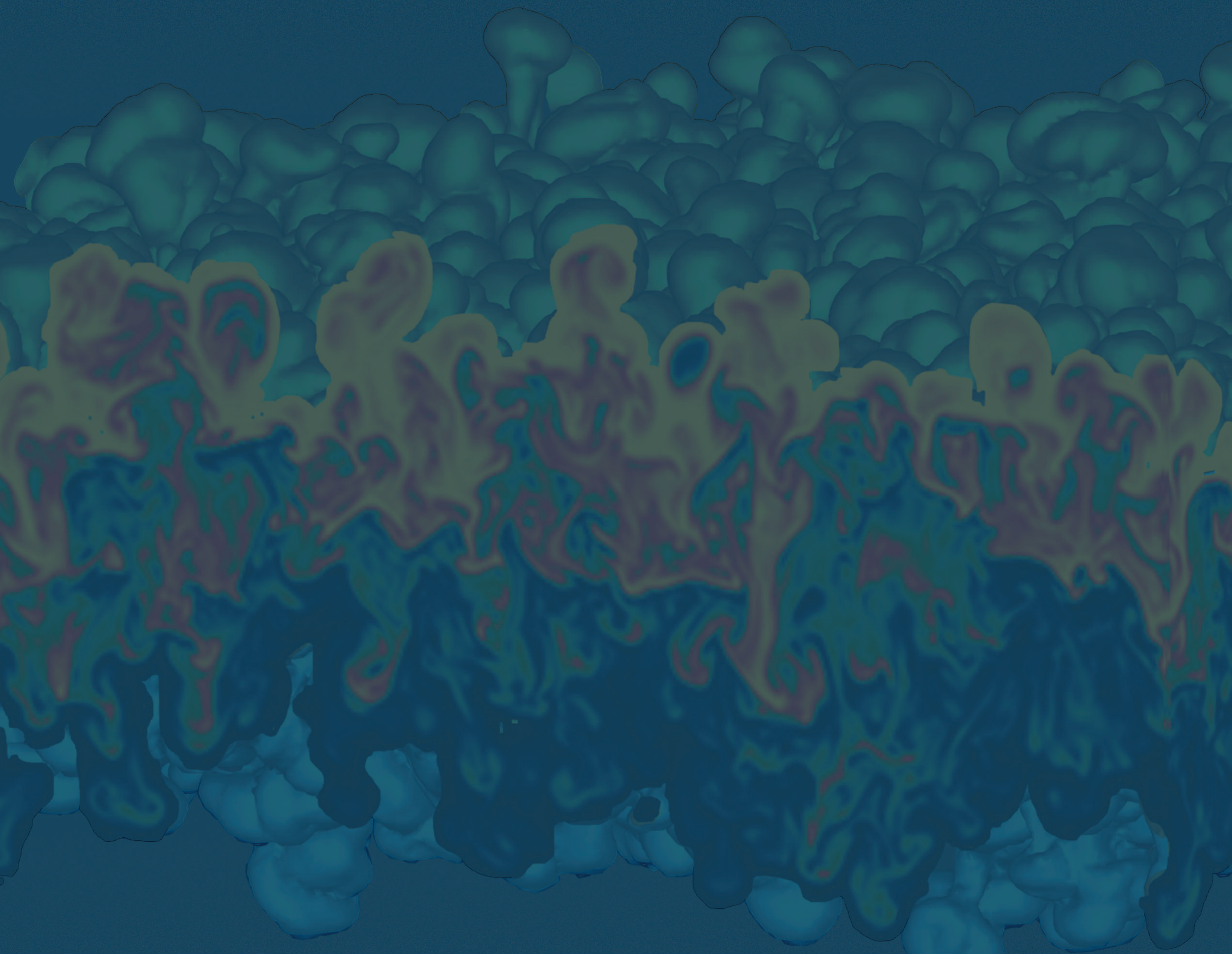
High-power, high-brightness laser systems are applicable to missile defense and materials processing for the Laboratory’s stockpile stewardship mission in support of the Laboratory’s national security missions.

FY03 Results

We reviewed and evaluated previous research experience (through literature searches and consultation with experts) on optical phase conjugation, laser diodes, injection locking, external-cavity diode lasers, and “chirp.” Initial contacts were made with key contributors in this field at the University of Rochester and the Massachusetts Institute of Technology. Based on this research, we devised a “strawman” architecture for an experiment on phase conjugation in broad-area laser diodes.

Proposed Work for FY04

In FY04, we will continue our study on phase conjugation and perform theoretical modeling specific to our architecture, comprising many modules and a large (meter-size) spatial extent. We plan to develop scaling rules in addition to numerical estimates of wavefront-reversal gain and efficiency, for instance. Eventually, we should be able to use laser-diode parameter values in various expressions to determine the level of scaling (multiplexing) possible. A laboratory experiment is planned to demonstrate phase conjugation in a broad-area diode to help check our modeling results and identify other phenomena of interest and key issues.



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