

Science & Technology

REVIEW

January/February 2007

National Nuclear
Security Administration's
Lawrence Livermore
National Laboratory

Titan Leads the Way

Also in this issue:

- Countering Nuclear Terrorism
- Fast Flow through Nanotubes
- Unique Microbial Communities

EDWARD TELLER'S CENTENNIAL LEGACY



About the Cover

Livermore's Titan, the only laser in the world that couples a high-energy, petawatt short-pulse beam with a kilojoule long-pulse beam, supports Laboratory research as well as collaborations with other institutions and universities. As the article on p. 4 describes, researchers use the dual-beam laser to investigate matter under extreme conditions pertinent to astrophysics, materials science, and plasma physics. Experiments on Titan will also help determine the physics requirements for future experiments at the National Ignition Facility. Shown on the cover is physicist Pravesh Patel of the Physics and Advanced Technologies Directorate setting up an experiment in the Titan target chamber.



About the Review

Lawrence Livermore National Laboratory is operated by the University of California for the Department of Energy's National Nuclear Security Administration. At Livermore, we focus science and technology on ensuring our nation's security. We also apply that expertise to solve other important national problems in energy, bioscience, and the environment. *Science & Technology Review* is published 10 times a year to communicate, to a broad audience, the Laboratory's scientific and technological accomplishments in fulfilling its primary missions. The publication's goal is to help readers understand these accomplishments and appreciate their value to the individual citizen, the nation, and the world.

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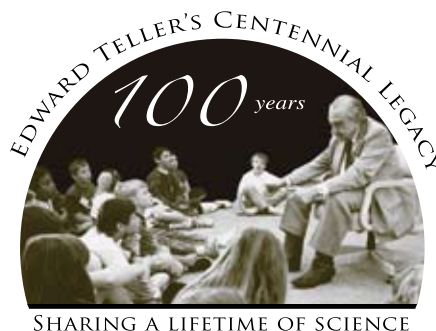
In an abandoned mine, where the pH can be even lower than zero, communities of acidophilic microbes produce hundreds of unusual proteins.

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Discovery of element 118

Scientists from the Chemistry, Materials, and Life Sciences Directorate in collaboration with researchers from the Joint Institute for Nuclear Research (JINR) in Russia have discovered element 118, the newest superheavy element. In experiments conducted at the JINR U400 cyclotron between February and June 2005, the researchers observed atomic decay patterns, or chains, that establish the existence of element 118.

The experiments yielded three atoms of element 118 by bombarding a californium target with calcium ions. The team observed the alpha decay from element 118 to element 116 and then to element 114. The Livermore–Dubna team had created the same isotope of element 116 in earlier experiments. This discovery brings the total to five new elements for the Livermore–Dubna collaboration (113, 114, 115, 116, and 118). The results were published in the October 9, 2006, edition of *Physical Review C*.

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Scientists crack open stellar evolution

Using three-dimensional models run on some of the fastest computers in the world, Laboratory physicists have created a mathematical code that cracks a mystery surrounding stellar evolution. For years, physicists have theorized that low-mass stars (about one to two times the size of the sun) produce great amounts of helium-3 (^3He). When low-mass stars exhaust the hydrogen in their cores to become red giants, most of their matter is ejected, substantially enriching the universe in this light isotope of helium. However, this enrichment when added to big bang predictions conflicts with observations. Assuming that nearly all stars were rapidly rotating, scientists theorized that stars destroy the ^3He . However, even this theory failed to bring the evolution results into agreement with the big bang and observations.

Now, by modeling a red giant with a fully three-dimensional hydrodynamic code, Livermore researchers Peter Eggleton and David Dearborn identified the mechanism of how and where low-mass stars destroy the ^3He produced during the stars' evolution.

They found that ^3He burning in a region just outside of the helium core, previously thought to be stable, creates conditions that drive a newly discovered mixing mechanism. Bubbles of material, slightly enriched in hydrogen and substantially depleted in ^3He , float to the surface of the star and are replaced by ^3He -rich material for additional burning. In this way, the stars destroy their excess ^3He , without requiring additional conditions such as rapid rotation. The research appears in the October 26, 2006, edition of *Science Express*.

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Scientists capture nanoscale images with x-ray laser

Laboratory scientists have for the first time validated the idea of using extremely short and intense x-ray pulses to capture images of objects, such as proteins, before the x rays destroy the sample. At the same time, they also established a speed record of 25 femtoseconds for flash imaging. When even more powerful x-ray lasers are available, the new method will be applicable to atomic-resolution imaging of complex biomolecules. The technique will allow scientists to gain insight into the fields of materials science, plasma physics, biology, and medicine.

Using the free-electron laser at Deutsches Elektronen-Synchrotron in Hamburg, scientists, as part of an international collaboration led by Henry Chapman of Lawrence Livermore and Janos Hajdu of Uppsala University, were able to record a single diffraction pattern of a nanostructured object before the laser destroyed the sample. A Livermore-developed computer algorithm then used the recorded diffraction pattern to re-create an image of the object. This imaging technique could be applied to atomic-resolution imaging because it does not require a high-resolution lens. The flash images could resolve features 50 nanometers in size, which is about 10 times smaller than what is achievable with an optical microscope. The research appeared in the November 12, 2006, online edition of *Nature Physics*.

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Another Step for High-Energy-Density Science and Teller's Legacy

THIS January, we begin a journey that will trace and celebrate the intellectual contributions of one of Lawrence Livermore's founders—Edward Teller. January 15, 2008, is the centennial of Teller's birth. Between now and then, we will examine the accomplishments of this man who, along with Ernest O. Lawrence and Herbert York, established in 1952 the institution now known as Lawrence Livermore National Laboratory.

Teller's scientific legacy is distinguished by an unusual blend of abstract inquiry and innovative application over three-quarters of a century. His scientific career began as the quantum physics revolution opened vast areas to detailed understanding. In later years, Teller's interest in nuclear fusion and matter at high energy density (HED) meshed naturally with his role in national defense. This enduring interest in HED science, which is key to mastery of fission and fusion science and its applications, provides the starting point for our journey.

HED science is the study of matter under extreme conditions, such as those found in the center of a star or the implosion of a nuclear weapon. The exploration of matter at HED has been a Laboratory hallmark since its founding. HED science is a critical component of Lawrence Livermore's mission responsibilities and a key strategic component of the Laboratory's future. The National Science and Technology Council identified HED science as one of seven theme areas at the intersection of physics and astronomy that are ripe for investment and show great promise.

As described in the article beginning on p. 4, Livermore has taken another major step in helping to meet this promise. In commissioning the Titan petawatt-class laser at the Jupiter Laser Facility, the Laboratory enhanced its leadership in HED science and continues its legacy of using advanced lasers to explore this field. Titan joins other Livermore laser platforms including the Janus, Callisto, Europa, and COMET lasers and associated target chambers. Titan is one of only three petawatt-class lasers worldwide and currently is the only one offering synchronized short- and long-pulse operation.

Titan's mission is to support research pertinent to Laboratory programs, to promote collaborations with research institutions and universities, and to provide a stimulating environment for training young scientists. The National Nuclear Security Administration, the Department of Energy's Office of Science, and the Laboratory Directed Research and Development Program all sponsor work at the facility. With Titan and the other Livermore lasers, scientists

are pursuing mission-based science with HED research activities to study laser-plasma interactions, fast ignition, high-pressure material science, opacity, x-ray lasers, and warm dense matter.

Titan, with both short-pulse (subpicoseconds to a few picoseconds) and long-pulse (typically nanoseconds) capabilities was completed in the summer of 2006. In September, the Jupiter Laser Facility Academic Use Program was the first to simultaneously use Titan's long- and short-pulse laser beams in experiments facilitated by the Institute for Laser Science and Applications (ILSA).

The ILSA academic team comprised faculty, postdoctoral researchers, and graduate students from the University of California at San Diego, Ohio State University, and the University of Rochester working with researchers from the Laboratory's Physics and Advanced Technologies and National Ignition Facility (NIF) Programs directorates. Many of the academic participants are university researchers associated with the Fusion Science Center for Extreme States of Matter, which is centered at the University of Rochester and funded by the Office of Science's Fusion Energy Sciences Program, or graduate students who are supported through ILSA's participation in the Laboratory's University Education Partnerships Program. The collaborative experiments included measuring hot-electron transport in compressed, shocked foams.

The value of Titan and the Jupiter Laser Facility will continue to grow as NIF—the ultimate HED user facility—begins full operation. Titan will remain crucial for developing diagnostics and targets for NIF, and Titan experiments will help advance the understanding of issues facing fast ignition for inertial confinement fusion energy. Many Titan experiments will explore new concepts—some of which will undoubtedly emerge from NIF HED experiments—and serve as a test bed for more complex tests using NIF. Titan also will help train the cadre of young scientists who will use NIF over the next 20 to 30 years.

True to Teller's legacy of applying physics to solve problems of national importance, HED experiments with high-energy, short-pulse lasers promises to lead to new discoveries and applications, some of which are poised to become reality and others which are as yet only imagined.

■ George H. Miller is director of Lawrence Livermore National Laboratory.

Titan Leads the Way in Laser–Matter Science

Titan's unique short- and long-pulse capability is being used to explore fundamental science questions and applications of high-energy-density matter.

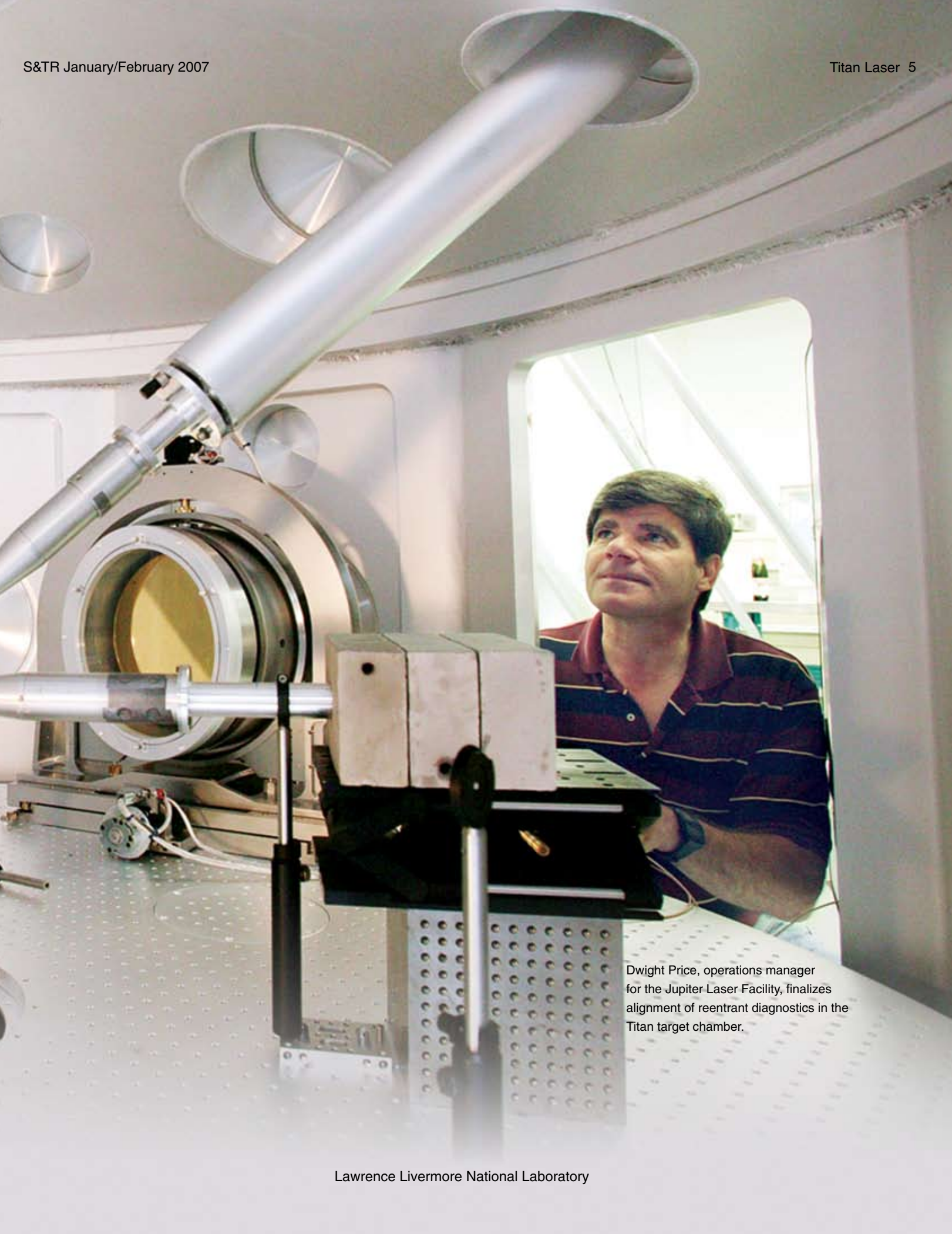
EVER since the development of the first optical laser in 1960, researchers have engineered ways to manipulate laser pulses. Today, with the Titan laser, scientists are investigating matter under extreme conditions in support of the Laboratory's mission within the fields of astrophysics, materials science, and plasma physics. Results from Titan laser experiments will expand the current understanding of different states of matter from stellar conditions to nuclear weapons. Fundamental questions surrounding the possibility of inertial fusion energy, including fast ignition, are being answered, with the ultimate demonstrations planned for the National Ignition Facility (NIF).

In 1996, Livermore researchers developed a revolutionary new laser called the Petawatt, which delivered a record-setting 1.25 petawatts (1.25 quadrillion watts) of power. (See *S&TR*, December 1996, pp. 4–11.) The Petawatt laser was developed to test Livermore's fast-ignition concept for achieving inertial confinement fusion (ICF). (See the box below.) The experiments demonstrated that when tremendous amounts of energy are concentrated onto an area less than 100 micrometers in diameter, electrons could be accelerated to relativistic speeds, generating x and gamma rays. The discovery opened new opportunities for using lasers to study high-energy-density (HED) science.

Fast Ignition

Fast ignition was conceived in 1990 by a team led by Max Tabak, a scientist in Livermore's Inertial Confinement Fusion (ICF) Program. In both fast-ignitor and conventional ICF, laser or x-ray pulses rapidly heat the surface of a fusion target capsule, enveloping it in plasma. The fusion process is started in conventional ICF when a sequence of spherically converging shocks of increasing amplitude is carefully timed to stagnate simultaneously at the center of the fuel capsule and generate a hot spark, while minimizing shock heating of the fuel.

In conventional ICF, the plasma must remain highly symmetrical and spherical during implosion. This level of symmetry requires enormous energy and precision from the laser system. Tabak and colleagues proposed separating the two processes of compression and heating. For fast ignition, a petawatt laser is used to provide the hot spark. Electrons accelerated to megaelectronvolt energies slow down in the compressed material, thereby heating the material on a timescale so short that the compressed matter does not have time to disassemble. In theory, fast ignition reduces both the laser energy and precision requirements for achieving ignition. However, fast ignition presents its own challenges. The spot to be heated must be large enough to ignite all the fuel, but not so large as to waste energy. Electrons must be driven far enough to penetrate the plasma and heat the ions on the surface of the dense core, but not any farther. Scientists engaged in fast-ignition research are conducting a variety of experiments to determine the best approach.



Dwight Price, operations manager for the Jupiter Laser Facility, finalizes alignment of reentrant diagnostics in the Titan target chamber.

However, generating tremendous amounts of laser energy is only part of what is necessary to conduct HED research. Once researchers generate the laser pulse, they must be able to observe the dynamic changes that occur over a time frame of billionths of a second after a laser beam strikes its target. A key scientific focus is to advance understanding of the temperature–pressure relationship, or equation of state (EOS), for material subjected to these extreme conditions.

One method researchers use to experimentally determine a material’s EOS is shock compression. Gas guns are useful for creating high-pressure shock waves of about 500 gigapascals (5 million atmospheres of pressure). However, for some of the research to be performed at NIF, scientists need EOS data for materials subjected to pressures of 10 terapascals (100 million atmospheres) or more. High-performance lasers are the

only instruments available to researchers that have the potential to obtain EOS data for materials at those pressures. NIF and Titan will be used in complementary experiments to obtain the required EOS data.

Lasers are designed to deliver a specific range of pulse energy and duration. This range dictates the phenomena that can be studied. For example, long pulses lasting nanoseconds are effective at compressing a material, whereas short pulses lasting pico- or femtoseconds are better for probing a material. In the past, high-energy lasers have been built to deliver either long or short pulses into an experimental area, but not both.

Two Beams Pack a Punch

In 2003, the Laboratory Science and Technology Office funded a collaboration between the Physics and Advanced Technologies (PAT), NIF Programs,

Engineering, and Chemistry, Materials, and Life Sciences (CMLS) directorates to build Titan, the only laser in the world that couples a high-energy, petawatt short-pulse (subpicosecond) beam with a kilojoule long-pulse (nanosecond) beam. This unique capability is being used to explore a range of phenomena, including the acceleration of charged particles, hydrodynamics, and radiation emission and absorption in hot dense plasmas, all of which are fundamental to understanding HED materials. The research also helps to determine the physics requirements for experiments at NIF.

Titan joins the Janus, COMET, Europa, and Callisto lasers in PAT’s Jupiter Laser Facility, which supports Laboratory research and collaborations with other institutions and universities. Andrew Ng of PAT, the facility’s scientific director, says, “Every laser laboratory in the world would like to have a high-energy laser with long- and

Edward Teller and High-Energy-Density Physics

After beginning his career by brilliantly applying the new quantum mechanics to the physics of atoms and molecules, Edward Teller began to show keen interest in high-energy-density (HED) physics—the science of matter and energy above 1 million atmospheres in pressure. Two major HED physics discoveries in the late 1930s engaged his imagination. First, his friend and colleague Hans Bethe proposed the basic explanation that stars are

powered by thermonuclear reactions among light ions. Second was the discovery of uranium fission, which quickly led to the idea that a chain reaction

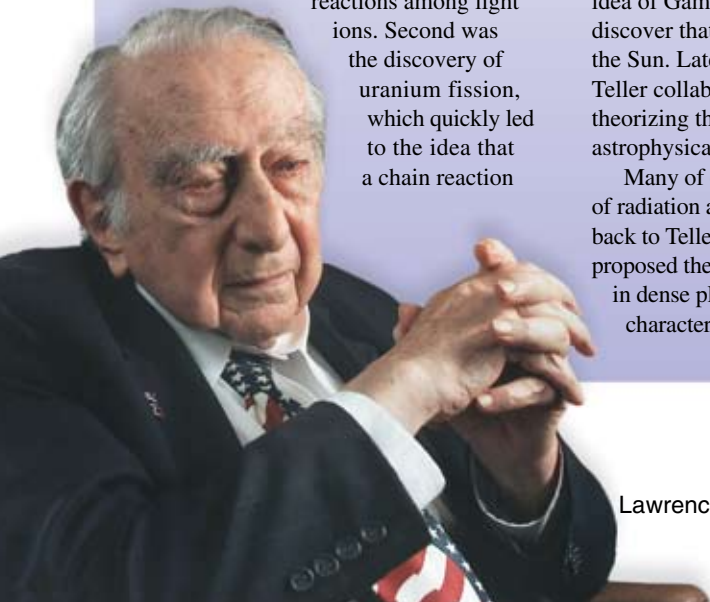
could liberate nuclear energy. Thermonuclear reactions could be studied and exploited, and fission could be developed to make explosives and power reactors.

After emigrating to the U.S., Teller worked at George Washington University with George Gamow and others on a variety of physics topics, including HED. With Gamow, Teller wrote one of the earliest papers estimating the rates of key thermonuclear reactions and applying them to red giant stars. Bethe and a student of Teller’s, Charles Critchfield, applied the new idea of Gamow–Teller nuclear beta decays to discover that the fusion of two protons powers the Sun. Later at the University of Chicago, Teller collaborated with Maria Mayer in papers theorizing the synthesis of heavy elements in astrophysical explosions.

Many of our key ideas describing the flow of radiation and energy in the HED regime date back to Teller’s papers. With collaborators, he proposed the main ideas for the behaviors of ions in dense plasmas and their spectral radiation characteristics, and he worked out the rules

for shock waves in magnetized plasmas. In his efforts to understand the laws governing the equations of state of HED plasmas for both basic science and applications, Teller drove important developments such as the famous Metropolis Method, which is essential for making statistical mechanics calculations computationally feasible. This work was done with his former student Marshall Rosenbluth, their wives Mici Teller and Arianna Rosenbluth, and Nicolas Metropolis. Teller, Metropolis, Richard Feynman, and others are responsible for developing widely used models for hot dense matter.

At Livermore, Teller was a tireless champion of innovative HED research, knowing that this important field entwined basic science and major applications. He encouraged developments that showed promise for new ways to control and exploit HED matter and plasmas, thereby leading to much of Livermore’s research on inertial fusion and lasers.



short-pulse capability. It allows researchers to study phenomena that occur in a material at different timescales, providing a more complete picture of material evolution. For example, structural changes to a material tend to occur on approximately the picosecond timescale, while electronic changes happen much faster, on approximately the femtosecond timescale.” (See *S&TR*, July/August 2006, pp. 4–10.)

Because very short pulses can overheat a laser system’s glass amplifiers and thus damage them, Titan’s short-pulse beam has an optical parametric chirped-pulse amplifier that temporarily stretches the pulse, reducing the beam intensity in the amplifier system. This stretched pulse is then compressed to its original duration using a pair of gratings in a vacuum, and a petawatt pulse of 400 joules in 400 femtoseconds is delivered to the target.

One of the major breakthroughs enabling petawatt capability was the development of multilayer dielectric optical gratings by CMLS scientist Jerry Britten in 1986 for the Laboratory’s Nova laser. Previously, ultrashort-pulse lasers using gold optical gratings to compress pulses were limited in how much energy they could generate because of the optical damage threshold of the gold layer. The multilayer dielectric gratings provide up to five times improvement in damage threshold over gold. Titan uses two gratings, each measuring 80 by 40 centimeters wide, in its vacuum compressor.

The two Titan beams, which originate from the Janus laser, can operate independently or together. At full energy, the pulses can be produced once every 30 minutes. The short-pulse beam is directed through a dedicated port into Titan’s 2.5-meter-diameter target chamber. It is focused onto a spot less than 10 micrometers in diameter (full width at half maximum). The long-pulse beam can be directed into the target chamber through different ports using an articulated beam transport. Inside the target



Titan’s long-pulse beam can be directed into the 2.5-meter-diameter target chamber through different ports. The short-pulse beam is directed through a dedicated port.

chamber, researchers mount diagnostic equipment tailored for each experiment. Physicist Pravesh Patel of PAT designed the target chamber and worked with the Titan laser team to help ensure the new facility would meet all of the requirements for experiments. The Titan facility is also equipped with an array of other instruments, including radiation detectors, particle detectors, x-ray imagers and spectrometers, and charge-coupled-device and streak cameras for measuring and recording results.

In February 2006, Titan delivered short pulses of 150 joules in 500 femtoseconds onto an 8-micrometer-diameter spot. Titan’s combined short- and long-pulse beams were commissioned for experiments in July 2006.

Focusing Electrons and Protons

PAT and the Laboratory Directed Research and Development (LDRD) Program are funding Patel to conduct experiments that will further understanding of HED plasmas, which are relevant to the Laboratory’s stockpile stewardship research. Plasmas are extremely hot states of matter that are difficult to probe when created by

a laser because the plasma is always in transition from hydrodynamic expansion. A decade ago, Livermore researchers using the Petawatt laser unexpectedly discovered that when they fired a short pulse at a thin solid foil, the resulting relativistic electrons could interact with impurities in the foil to create a proton beam. (See *S&TR*, December 2003, pp. 11–14.) A few years later, Patel and his colleagues used a proton beam generated with the 100-terawatt Callisto laser to isochorically heat a plasma. During isochoric heating, a material’s volume stays at a constant value. The proton beam heats the target to millions of degrees in a few picoseconds, which is too quick for significant hydrodynamic expansion to occur.

Patel and Andrew Mackinnon of the NIF Programs Directorate are studying the optimization of electron and proton transport for fast ignition. The Department of Energy Office of Science’s (DOE/OS’s) Fusion Energy Sciences Program and LDRD are funding the team. The team collaborates with John Pasley and Farhat Beg from the University of California (UC) at San Diego, Rich Stephens from General Atomics, and Rick Freeman

and Linn Van Woerkom from Ohio State University, who are all supported by DOE/OS's Fast Ignition Advanced Concept Exploration grant, Livermore's Institute for Laser Science and Applications (ILSA), and DOE's Fusion Science Center at the University of Rochester. An important challenge for the viability of the fast-ignition approach to ICF energy is the transport of the ignition pulse energy to the target with high efficiency.

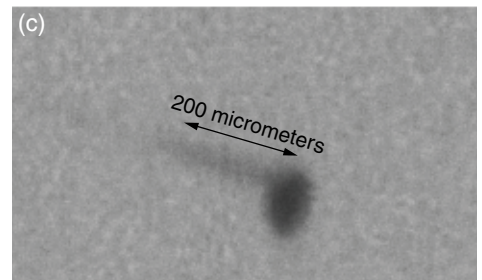
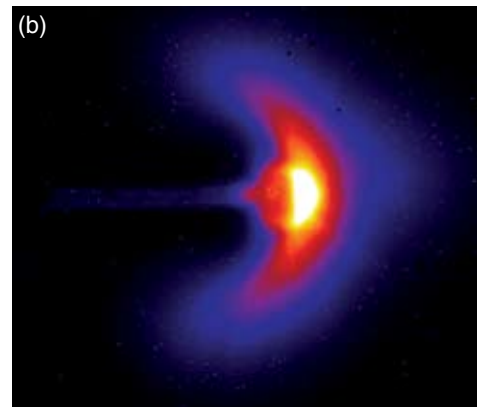
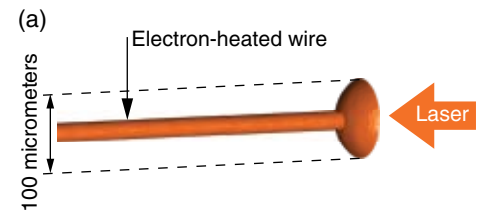
In one possible design for fast ignition, a gold cone is inserted radially into the fusion-fuel capsule, with the tip of the cone penetrating almost to the center of the capsule. Long-pulse laser beams 1 to 10 nanoseconds in duration compress the fuel capsule to a dense plasma core. A 10- to 100-picosecond petawatt laser pulse is then focused into the gold cone to generate energetic electrons at the top of the cone and ignite the fuel's plasma core. Mackinnon says, "We are using Titan

to study materials at a density of 1 gram per cubic centimeter and a temperature of 25 electronvolts, but NIF will be used to study materials 300 times that density and temperatures between 100 and 200 electronvolts. Titan experiments test the physics requirements on a small scale and produce data needed to prepare for future experiments on NIF."

A common method researchers use to examine electron transport in short-pulse laser-matter interactions is K-alpha x-ray emission. When a short pulse with intensity greater than 10^{19} watts per square centimeter strikes a target, electrons with energies up to 3 mega-electronvolts are created. These energetic electrons interact with bound electrons in an atom. In K-alpha emission, an incident electron ejects an electron from an atom's innermost shell (called a K-shell electron). Then an outer-shell electron drops in to fill the vacancy, emitting a photon

in the process. By studying K-alpha emission from the electron interactions, researchers can obtain information about the properties of electrons in a high-density plasma environment.

In one study, the team examined electron transport using a 20-micrometer-diameter copper-wire target covered with titanium. The team focused 100 joules of energy for 1 picosecond on a 10-micrometer spot at the end of the wire and measured the spatial distribution of



(a) A short laser pulse is fired onto the head of a titanium-covered copper wire to study electron transport. (b) A radiograph in the extreme ultraviolet band of 68 electronvolts shows heating by hot electrons along the wire. (c) An x-ray image of copper K-alpha fluorescence at 8 kiloelectronvolts shows hot electron penetration 200 micrometers into the wire.

University of California at San Diego researchers John Pasley and Farhat Beg participate in the Laboratory's Institute for Laser Science Applications. Pasley and Beg are collaborating on a team studying fast ignition for inertial confinement fusion energy.



the K-alpha emission for both copper and titanium. X-ray images showed the electrons' depth of penetration along the length of the wire. The images showed good agreement with simulations produced by the team using hybrid fluid-particle codes.

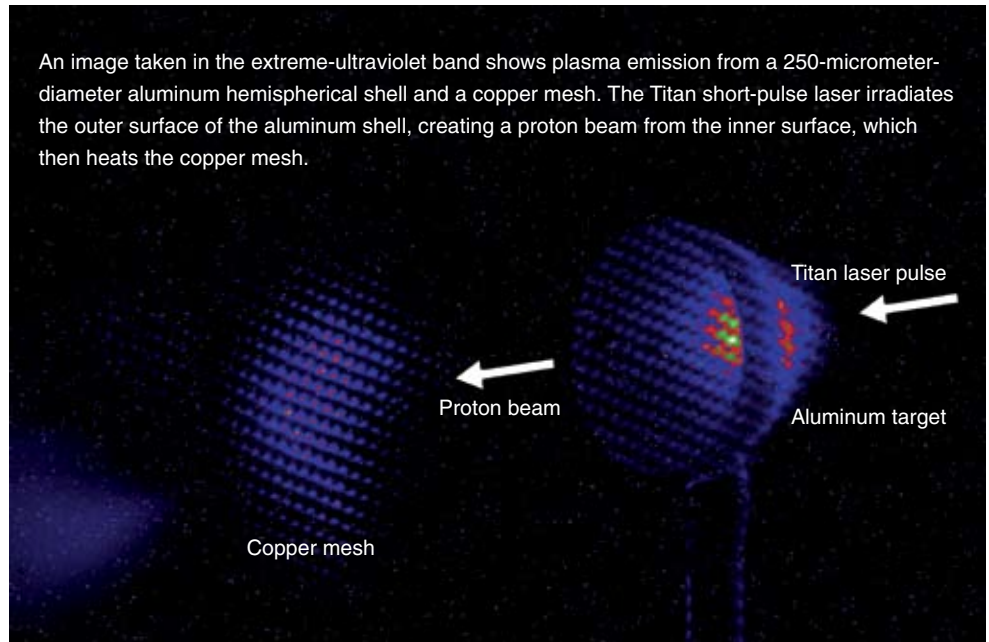
Paving the Way to Fast Ignition

The advantage of using an electron beam for fast ignition is that electrons have a conversion efficiency of 30 to 50 percent (conversion of energy in laser pulse to energy in electron beam). However, electron transport is uncertain. "Electrons have a tendency to spray in a large angle," says Mackinnon. "For fast ignition to work, electrons must be concentrated on a small spot in the center of the plasma."

While the team is mainly concentrating on electron transport for fast ignition, it is also exploring the use of a proton beam. The earlier discovery that impurities in gold could generate a proton beam prompted studies for exploiting the phenomenon for fast ignition. Mackinnon explains, "Ultrashort laser pulses produce electrons and protons, either of which could potentially be used to ignite fusion fuel."

Protons offer attractive features. With a mass about 1,800 times the mass of electrons, protons are deflected less often than electrons and can be focused with curved targets as they travel through a plasma. However, the conversion efficiency for creating protons is only about 10 percent. The team is determining whether the conversion efficiency could be increased with the use of thin, hydrogen-rich layers on the rear target surface.

Some proton-focusing experiments have been conducted using an aluminum hemispherical shell target and a copper mesh. The Titan short-pulse beam was fired onto the aluminum target, producing a focused proton beam that simultaneously heated and radiographed the copper mesh. By measuring magnification (grid size) of the mesh projected onto a proton detector, researchers can determine the location of the focal spot of the proton beam generated from the spherical shell. These experiments showed that protons



could be focused to an area less than 50 micrometers in diameter, creating solid-density plasmas with temperatures between 100 and 200 electronvolts.

These techniques will be applied at the University of Rochester's OMEGA EP (extended performance) laser facility, where the collaborative team may be able to isochorically heat plasmas to much higher temperatures and pressures. The 60-kilojoule OMEGA EP laser, which is scheduled to begin operation in 2007, is participating in the National Nuclear Security Administration's HED program. One of the facility's goals is to provide a staging ground for fast-ignition concepts that could be applied to NIF.

The Livermore results are also helping Pasley, Beg, Freeman, Stephens, and Van Woerkom with studies being conducted at UC San Diego and Ohio State University. Last year, academic collaborations at the Jupiter Laser Facility included more than 25 students and postdoctoral researchers. The students also collaborate on many other Livermore projects. Don Correll, director of Livermore's Institute for Laser Science Applications, says, "Engaging graduate students and postdoctoral researchers in HED science experiments on the Titan laser is

a cornerstone of the institute's mission to strengthen collaborations between faculty and Laboratory researchers. The scientific breakthroughs benefit both the Livermore and academic HED science communities. The collaborations are also ideal for attracting the next generation of researchers to the Lab's scientific missions."

Measuring at the Extremes

Many of the shock wave techniques for obtaining EOS data were developed originally in gas-gun experiments, in which pressures are typically less than 100 gigapascals. One method, called impedance matching, relies on driving a shock wave through a reference material whose EOS is known into a test sample in contact with it. Because the materials share a common interface, the shock pressure and particle velocity will be the same in both materials. Using the measured shock velocities in each material and the known EOS of the reference material, researchers can determine the shock pressure and density of the sample. A major goal of Livermore researchers is to obtain experimental data up to very high pressures, densities, and temperatures.

However, HED regimes place extreme demands on the conventional impedance-matching technique.

Physicist Damien Hicks of PAT is conducting studies to improve Livermore's EOS database. Hicks says, "Impedance matching works well at low pressures. The compressions are much higher at pressures of hundreds of gigapascals, so highly precise velocity measurements are needed. Small errors in velocity measurements become large errors in density measurements." Problems also arise from using reference materials whose EOSs have been tested only at lower pressures. For example, the shock data on aluminum is patchy above 500 gigapascals. Even with additional data from theoretical models or extrapolations, uncertainties increase at these higher pressures.

Hicks's team is developing a technique that eliminates these uncertainties by directly measuring the density of materials that are highly compressed by a shock wave. The team has conducted experiments in collaboration with researchers at the University of Rochester's Laboratory for Laser Energetics. It focused a nanosecond-long pulse from the OMEGA laser onto a 500-micrometer-diameter spot to generate a spherical shock wave in a polystyrene sample. Radiographic imaging using long-pulse-driven 5-kiloelectronvolt x rays produced a snapshot of the expanding shock wave. This two-dimensional (2D) image was then tomographically inverted, providing the density profile behind the

shock front. By simultaneously measuring the shock velocity with a velocity interferometer, the team obtained absolute EOS data.

Last year, the team began experiments using Titan's long- and short-pulse beams to study silicon dioxide. They fired the long pulse to generate a shock wave, then the short pulse to produce higher energy and deeper penetrating x rays from a molybdenum foil adjacent to the silicon dioxide. The molybdenum x rays produced a radiograph of the shocked region. "The 2D picture of the symmetric, expanding shock wave enabled us to infer its density from the intensity of the transmitted x rays," says Hicks. The team estimates that the method accurately measures density to within 5 to 10 percent. The pressure in the shock wave is determined by measuring the shock velocity with an interferometer. "We bounce a probe laser pulse off the shock wave and measure the Doppler shift in the reflected light," says Hicks. "Combined with the density measurement, this measurement tells us the pressure of the sample."

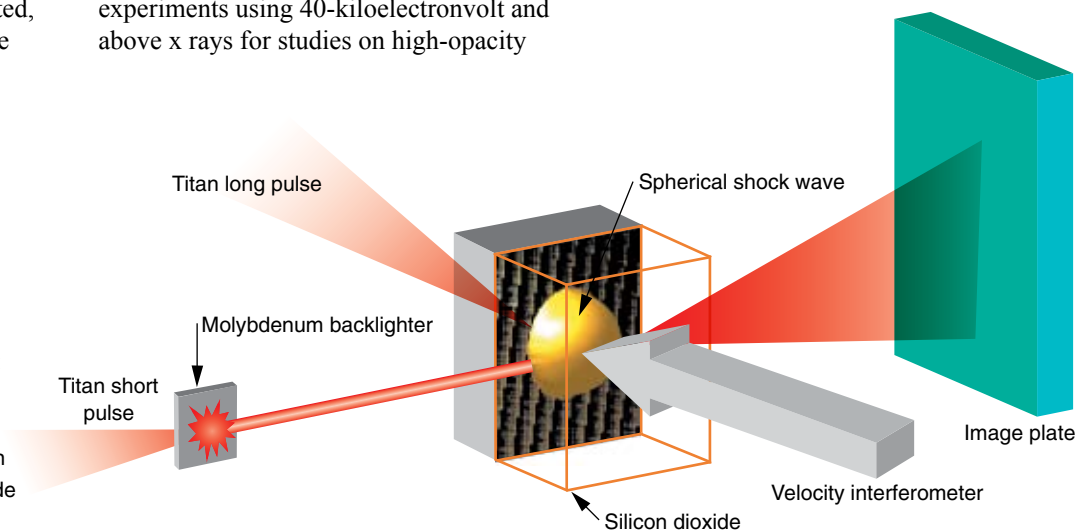
Unlike the conventional impedance-matching technique, where density is inferred from shock and particle velocities, the new direct density measurement does not demand increased precision at higher compressions. Therefore, the method readily scales to ultrahigh-pressure measurements. The team plans to conduct experiments using 40-kiloelectronvolt and above x rays for studies on high-opacity

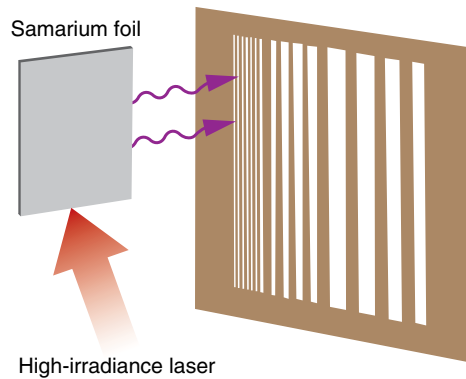
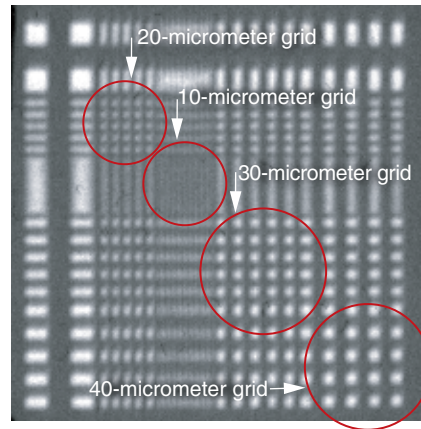
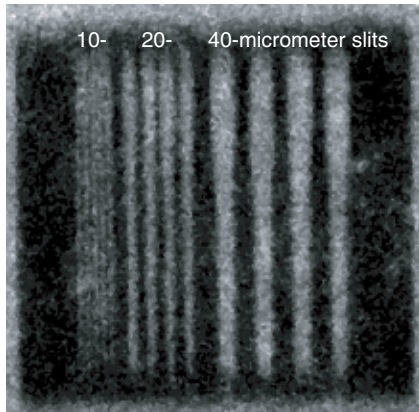
(high-Z) materials. The researchers also want to extend the EOS data for materials used as references. Hicks says, "Because so much information is known about aluminum at lower pressures, it makes sense to extend aluminum's EOS data to 1 terapascal and above so that the element can serve as a reliable reference at higher pressures."

Shedding Light on the Matter

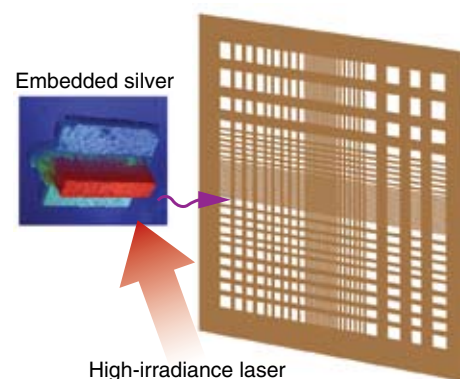
With funding from the Defense and Nuclear Technologies (DNT) Directorate and LDRD, physicist Hye-Sook Park of the NIF Programs Directorate is developing a high-energy backlighting technique to produce radiographs for x-ray energies up to 100 kiloelectronvolts. As NIF approaches completion, physicists will be conducting experiments that involve larger and denser targets than previously studied. NIF will need suitable diagnostics to ensure the dense materials are probed to specification. High-energy x-ray backlighting will be used in many HED experiments such as material strength, mid- to high-Z capsule implosions, and high-Z EOS studies. "NIF will fire single shots that are just several nanoseconds long," says Park. "The backlight needs to be bright enough to capture events occurring during that short time. We also need to differentiate small features at the surface of a material

This schematic shows a technique designed to directly measure the density in a shock wave. A long laser pulse drives a spherically expanding shock wave into silicon dioxide. Several nanoseconds later, a short laser pulse generates a burst of 17.5-kiloelectronvolt x rays from a molybdenum backlighter. The x rays project a snapshot of the spherical shock wave onto an image plate detector. A velocity interferometer measures the shock speed in the sample; shock speed and density provide data to determine the shock pressure.





Short pulses fired onto a 5-micrometer-thick sample of samarium provide a backlight for this one-dimensional radiograph of a 35-micrometer-thick tantalum grid. In this image, spatial resolution is clear at 10 micrometers.



Silver microwires are buried in a geometric-shaped material to produce two-dimensional (2D) radiographs of the x-ray emission of high-energy-density materials. In this image, 2D features at 20 micrometers are well resolved.

or within a thick and dense substrate, both of which require a high-energy x-ray backlighter with a spatial resolution less than 10 micrometers.”

Traditional laser backlighters have energies less than 9 kiloelectronvolts. Park’s team has developed 1D and 2D radiography methods with greater than 17-kiloelectronvolt x rays and a spatial resolution less than 20 micrometers. To produce 1D radiography of a 35-micrometer-thick sample of tantalum, researchers irradiated a 5-micrometer-thick samarium foil with a short-pulse laser, creating 40-kiloelectronvolt x rays. Edge-on views of the foils showed the technique could resolve features down to 10 micrometers.

The team is also testing microwire and microdot targets buried in different geometric shapes to create a small point source for 2D radiography. “Two-dimensional radiography is more difficult, because the K-alpha photon production in microwire volumes is not well understood,” says Park. So far, they have tested the K-alpha emission from microdot samples of molybdenum and silver as well as x-ray emissions from various microwire targets made of molybdenum, silver, and samarium. A radiographed test grid using silver resolved 2D features clearly to 20 micrometers. Park says the technique could be scaled by using different materials for the type of energy required.

Park’s team is also using high-energy backlighting techniques to study material strength. The NIF Programs and DNT directorates are funding research to examine material strength under high pressure and high strain rate. The experiments rely on the properties of instabilities in materials at HED conditions. “Perturbations always exist at the interface of two joining materials,” says Park. “By studying the differences of the perturbation growth rate in various materials, we can measure the strength of the materials.” The team has studied aluminum and vanadium and is conducting experiments on tantalum.

Each experiment using lasers requires specific laser parameters (wavelength, energy, and pulse duration) and configuration (illumination geometry). Titan and the other lasers in the Jupiter Laser Facility allow scientists to tailor experiments to obtain optimal results. “The parameters and configuration depend on the information we are trying to gather,” says Ng. “Titan provides a cutting-edge environment to conduct a wide range of studies. It also facilitates collaborations with other scientists in HED research.”

For scientists working on NIF, Titan experiments will help advance their understanding of issues facing fast ignition. The importance of Titan laser research will continue to grow as NIF begins full operation. Many Titan experiments will explore new concepts and serve as a test bed for more complex tests using NIF. Titan also will help train the cadre of young scientists who will use NIF over the next few decades.

—Gabriele Rennie

Key Words: backlighter, electron transport, equation of state (EOS), fast ignition, high-energy-density (HED) science, impedance matching, inertial confinement fusion (ICF), Janus laser, Jupiter Laser Facility, multilayer dielectric grating, Petawatt laser, proton transport, Titan laser.

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Identifying the Source of Stolen Nuclear Materials

*Livermore scientists
are analyzing interdicted
illicit nuclear and
radioactive materials for
clues to the materials'
origins and routes
of transit.*

NUCLEAR forensics and attribution are becoming increasingly important tools in the fight against illegal smuggling and trafficking of radiological and nuclear materials. These include materials intended for industrial and medical use (radiological), nuclear materials such as those produced in the nuclear fuel cycle, and much more dangerous weapons-usable nuclear materials—plutonium and highly enriched uranium.

Livermore scientists are among the leaders in nuclear forensics—the chemical, isotopic, and morphological analysis of interdicted illicit nuclear or radioactive materials and any associated materials. (See the box below.) They are also supporting the national effort in nuclear attribution, which is the challenging discipline of combining input from nuclear and conventional forensics to identify the source of nuclear and radiological materials and determine their points of origin and routes of transit.

Nuclear forensics and attribution go beyond determining the physical, chemical, and isotopic characteristics of intercepted nuclear or radiological materials. Simply determining that an interdicted material is, for example, enriched nuclear power plant fuel often tells law-enforcement authorities little about the origin of the material, where it has been, or who is responsible for the theft. “We want to know the exact nature of the interdicted material, who the perpetrators were, and where legitimate control was lost,” says David Smith, a geochemist in the Nonproliferation, Homeland and International Security (NHI) Directorate and a leader of the Livermore nuclear forensics team. Knowledge of the pathways involved in transporting the material is also vital to nonproliferation and counterterrorism efforts.

Nuclear forensics and attribution can be difficult because of the large amount of radioactive materials in the industrialized and developing world. For example, the nuclear fuel cycle produces a variety of materials at different processing steps, ranging from processed ore (commonly known as “yellow cake”) to uranium oxide fuel enriched in uranium-235. Opportunities exist at many points during the fuel cycle for materials to be diverted outside the channels authorized for their legitimate manufacture, handling, and protection.

Successful nuclear forensics and attribution contribute to the prosecution of those responsible and strengthen national efforts in nuclear nonproliferation and counterterrorism. According to former Livermore physicist Jay Davis, the first director of

the federal Defense Threat Reduction Agency (DTRA), “Nuclear attribution, with the accompanying possibility of prosecution and retribution, may be one of our greatest deterrent tools and hence a vital and compelling component of our defense against terrorism.”

Livermore and seven other Department of Energy (DOE) national laboratories have been tasked by the Federal Bureau of Investigation (FBI) and the Department of Homeland Security (DHS) with further developing the nation’s technical forensics capability for nuclear and radiological materials. Major U.S. government partners in addressing domestic and foreign obligations include DOE, the National Nuclear Security Administration (NNSA), the Department of State, and DTRA. Technical nuclear forensics includes

A Textbook for Nuclear Forensic Scientists

Nuclear Forensic Analysis, written by Livermore scientists Ken Moody, Ian Hutcheon, and Pat Grant, is the first primary reference source for the growing specialty of nuclear forensics. In addition to being a resource for nuclear forensic scientists, the book also serves as a textbook for traditional radiochemistry science courses that increasingly include material on nuclear forensics. The book covers the principles of the chemical, physical, and nuclear characteristics associated with the production and interrogation of a radioactive sample; protocols and procedures; and attribution. The authors discuss principles and techniques used in numerous case studies of nuclear investigations conducted at Livermore.

Hutcheon is deputy director of the Glenn T. Seaborg Institute. Moody, who studied with Nobel Laureate Glenn Seaborg (the discoverer of plutonium), is a radiochemist and co-discoverer of four heavy elements. Grant serves as the deputy director of the Forensic Science Center. “Pat is trained in classical forensics, Ken is a nuclear chemist, and I specialize in instrumentation,” says Hutcheon. “Together, we give a balanced viewpoint.”

According to Hutcheon, “The book gives one an idea of the analytic techniques we use and the types of material we are asked to investigate. It also describes case studies and the ways we assign attribution.” Hutcheon says the book was written to be understandable by nonspecialists.

the analysis of conventional evidence that is radiologically contaminated as well as the analysis of the radiological materials themselves.

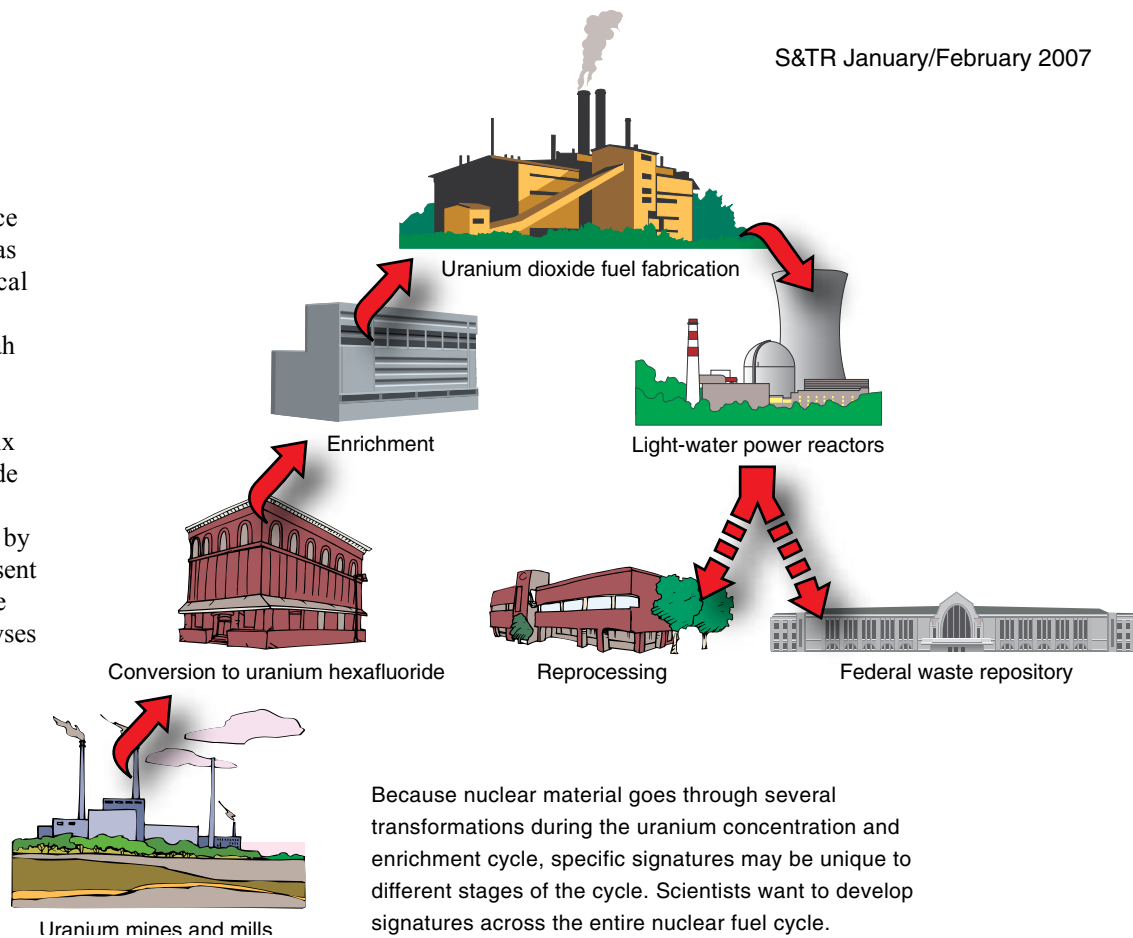
Lawrence Livermore and Savannah River national laboratories serve as a “hub” for nuclear forensics, with the “spokes” of the hub represented by six other national laboratories that provide specialized analyses and serve in supporting roles. A sample identified by one of the hub laboratories could be sent to experts at one or more of the spoke laboratories for complementary analyses and further characterization.

“We’re part of a national nuclear forensic and attribution capability that ties nuclear and radiological materials to people, places, and events,” says Smith. The effort is also part of a national strategy to counter nuclear terrorism. Other activities at the Laboratory that support the national strategy include developing more capable radiation detectors, designing detection systems for ports and other venues, and working with Russia and nations of the former Soviet Union to secure their nuclear materials.

Focus on Forensics

Nuclear forensics began at the Laboratory under the leadership of scientist Sid Niemeyer in the mid-1990s, and Livermore has maintained a leading role because of a collective group effort. The Laboratory has been involved in nuclear forensics and attribution for more than 15 years, since the collapse of the Soviet Union sparked concerns about the diversion of nuclear materials from former Soviet nuclear laboratories and other sites. Livermore’s capabilities in radiochemistry and nuclear physics, originally developed for the nation’s underground nuclear testing program, were adapted for use in nuclear forensics and attribution.

The strength of the national capability rests on the ability of Livermore to work in concert with the other laboratories, drawing on a core group of 30 to 50 scientists



from diverse backgrounds to work collaboratively on nuclear forensics and attribution cases. This depth of knowledge and breadth of experience are essential to tackling current issues in nuclear forensics.

Technical nuclear forensics can be performed on a broad spectrum of possible substances. Some interdicted samples are stolen containers of uranium diverted during one of the mining, milling, conversion, enrichment, or fuel fabrication steps used to convert uranium ore to enriched fuel for nuclear power plants. Alternatively, the interdicted sample could well be a commercial radioactive material such as cesium-137, strontium-90, cobalt-60, or americium-241. These isotopes and others are used in applications such as medical diagnostics, nondestructive analysis, food sterilization, and thermoelectric generators.

“We support investigations with the extraction of evidence that can be used by law-enforcement agencies,” says Smith. “Our casework validates and verifies our technical approaches.” Livermore and the other national laboratories

subject an interdicted sample to a host of extremely sensitive and accurate measurement techniques. Researchers analyze the material’s chemical and isotopic composition, which includes measuring the amounts of trace elements as well as the ratio of parent isotopes to daughter isotopes. These measurements help to determine the source location and sample’s age. They also examine the material’s morphological characteristics such as shape, size, and texture. Together, these characteristics are indicative of the specific processes used to produce the material.

Analytical methods include electron microscopy, x-ray diffraction, and mass spectrometry. (See the box on p. 17.) “As we obtain new tools, we open new frontiers of technical nuclear forensics,” says Ian Hutcheon, deputy director of the Glenn T. Seaborg Institute and senior scientist in the Forensic Science Center. For example, Livermore’s NanoSIMS, a secondary-ion mass spectrometer, provides a 50-nanometer (a billionth of a meter) spatial resolution. This capability enables researchers to analyze subsamples

of less than 1 microgram and perform particle-by-particle characterization to elucidate additional signatures beyond those gained through dissolving the bulk sample. “We are advancing from the micro to the nano level in analyzing samples to obtain forensic clues,” says Hutcheon.

Contaminated Evidence

An important element of nuclear forensics is the analysis of nonnuclear materials found with the radioactive material. Livermore scientists have been involved in developing forensics on radiologically contaminated evidence, which enables conventional law-enforcement forensics to be performed on materials that are radioactive. In addition, as a sample is moved from place to place, it picks up clues such as pollen, cloth fibers, and organic compounds. These so-called route materials provide information about who has handled a sample and the path it has traveled.

Nuclear forensics takes advantage of capabilities in Livermore’s Forensic

Science Center to tease out fingerprints, paper, hair, fibers, pollens, dust, plant DNA, and chemical explosives from contaminated substrates for the FBI and other government agencies. Established in 1991, the center supports DOE in verifying compliance with international treaties. It has also assisted federal, state, and local law enforcement on a wide range of criminal cases, including the 1993 World Trade Center bombing and the Unabomber investigations. The Forensic Science Center incorporates subject-matter experts from the Laboratory’s NHI and Chemistry, Materials, and Life Sciences (CMLS) directorates to build a complete profile of a sample.

Livermore’s nuclear forensics program uses laboratories equipped to handle large objects that are contaminated with radioactive materials. These laboratories make it possible, for example, to analyze a contaminated truck axle while still maintaining the evidentiary chain of custody required in a court of law. In addition, the program operates a

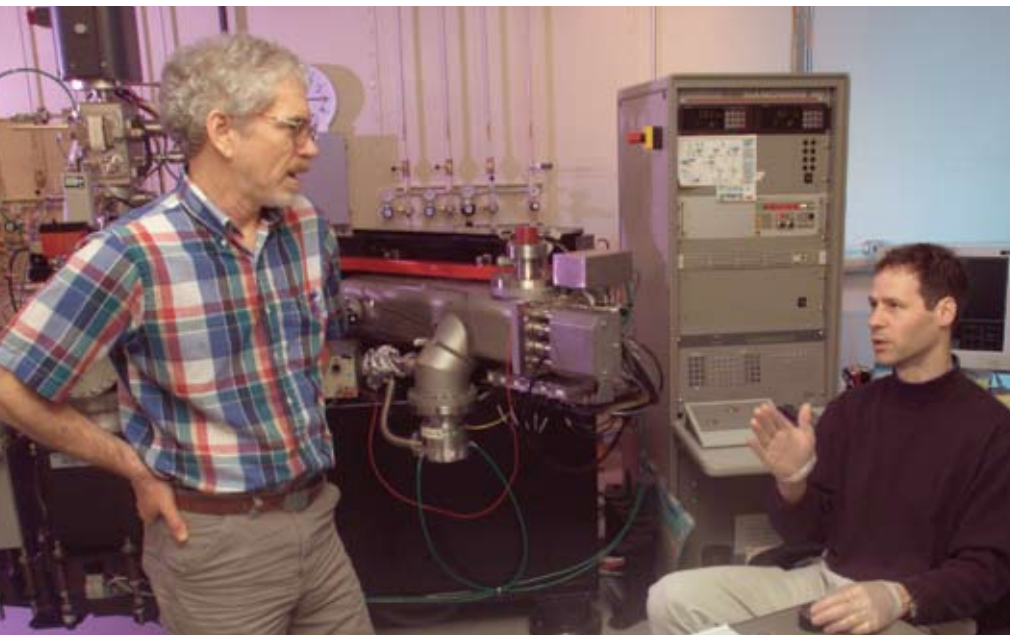
mobile van outfitted to ferry samples around the site or, if necessary, transport material between the Laboratory and offsite locations.

Developing Signatures

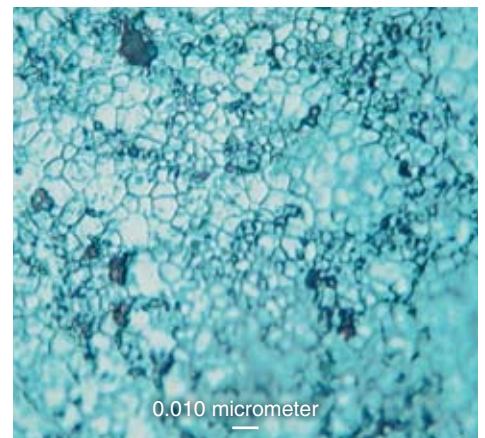
A major focus of nuclear forensics is identifying signatures, which are the physical, chemical, and isotopic characteristics that distinguish one nuclear or radiological material from another. Signatures enable researchers to identify the processes used to initially create a material.

Livermore scientists are developing signatures for a variety of nuclear materials. Enriched uranium is of particular concern. The goal is to develop validated signatures across the entire nuclear fuel cycle by experimental measurement or by simulation. Because nuclear material is transformed at different points during uranium concentration and enrichment processing, clues may be unique to different stages.

For example, unirradiated uranium reactor fuel pellets have inherent elemental oxygen content. Because the ratio of naturally occurring isotopes of oxygen-18 to oxygen-16 varies worldwide, these ratios could correlate with the locations of production sites. Similarly, the



Livermore researchers use techniques such as electron microscopy, x-ray diffraction, and mass spectrometry to analyze interdicted radiological and nuclear materials. In this photo, Ian Hutcheon (left), a senior scientist working with Livermore’s Forensic Science Center, and analytical chemist Peter Weber use NanoSIMS, a secondary-ion mass spectrometer with nanometer-scale resolution.



The microscopic shape and size of uranium particles offer clues about the material’s origin. This optical image of a nuclear fuel pellet shows the morphology of the grains.

variations of lead isotopes could provide clues about where a uranium compound was produced. Age, color, density, trace elements, and surface characteristics of the uranium compound are other important characteristics. A related investigation is studying the effects of different uranium manufacturing processes on the grain size and microstructure of the finished product.

When comparing a sample's signature against known signatures from uranium mines and fabrication plants, researchers can benefit by assembling a library of nuclear materials of known origin from around the world. Livermore scientists, with the help of the Office of Laboratory Counsel, have developed relationships with domestic suppliers of nuclear materials (uranium hexafluoride and uranium oxide reactor fuel) to assemble such a library. Contracts with major U.S. uranium fuel suppliers have provided researchers with samples and manufacturing data. Livermore scientists are using statistics to identify distinguishing signatures in nuclear

materials using samples and data obtained from manufacturers.

Livermore forensic scientists are also seeking to obtain samples of uranium products worldwide to analyze the products' isotopic and trace-element content, grain size, and microstructure. Nations with nuclear capabilities are beginning to share information about their nuclear fuel processes and materials. (See the box on p. 18.) For example, Livermore representatives signed a five-year agreement in January 2006 with KazAtomProm, the national atomic energy enterprise in Kazakhstan. KazAtomProm has long provided uranium ores to Russia and other nations of the former Soviet Union. With legal and technical support from NNSA, DHS funded the Laboratory to contract with KazAtomProm to provide Livermore with uranium ore samples and data. In addition, KazAtomProm officials have identified experts available for consultation on nuclear forensic issues.

A similar agreement was signed in

2006 with nuclear power officials in Tajikistan. This agreement provides samples to enable comparison between known and questioned sources. "We want to expand our agreements to all the Central Asian nations that process uranium," Smith says.

The importance of international cooperation was underscored in July 2006, when U.S. President George W. Bush and Russian President Vladimir Putin announced the creation of a Global Initiative to Combat Nuclear Terrorism. The initiative's goal is to strengthen worldwide cooperation in making nuclear materials more secure and preventing terrorist acts that involve nuclear or radioactive substances.

Building a Knowledge Base

The data obtained from U.S. and Central Asian uranium producers are part of a growing technical nuclear forensics knowledge base involving Livermore and seven other DOE national laboratories.

Under an agreement signed in 2006, KazAtomProm is providing Livermore scientists with uranium ore samples and data and has identified experts available for consultation on nuclear forensic issues. Front row, left to right: Dave Herr from Livermore's Procurement Office and Sergei Yashin, vice president of KazAtomProm. Back row, left to right: Mike Kristo and David Smith from Livermore and representatives from KazAtomProm. (background) KazAtomProm is the leading uranium mining and fuel production facility in Kazakhstan.



“We are establishing a knowledge management system for nuclear signatures, processes, origins, and pathways,” says Livermore nuclear engineer Frank Wong, who is leading the knowledge base effort, which now includes about 30 experts nationwide.

Rapid and credible interpretation of nuclear forensics data is predicated on a knowledge base that accesses and analyzes information derived from measurements or simulation of the full spectrum of the nuclear fuel cycle. Knowledge management allows for the ready comparison of analytical signatures obtained from suspect nuclear samples against known signatures from nuclear production, reprocessing, manufacturing, and storage. It also allows for the analysis of existing knowledge and data with the goal of identifying the most diagnostic nuclear forensic signatures.

The knowledge base is being designed so that it can be accessed by a simple query on a computer. “As an example,



The Lawrence Livermore technical team that addresses the technical nuclear forensics of interdicted materials includes (left to right) in the front row: Ian Hutcheon, Erick Ramon, and Michael Kristo; second row: Brett Isselhardt, Giles Graham, and Lars Borg; third row: Ross Williams and Michael Singleton; and fourth row: David Smith and Lee Davison. Not pictured: Glenn Fox, Pat Grant, Leonard Gray, Steven Kreek, Ken Moody, Sid Niemeyer, Martin Robel, Steve Steward, Louann Tung, Alan Volpe, Philip Wilk, Nathan Wimer, and Frank Wong.

The Cosmic Connection to Nuclear Forensics

Scientific research in cosmochemistry and isotope geochemistry continues to technically validate Livermore’s nuclear forensics capabilities. Cosmochemistry is the study of the origin and development of the elements and their isotopes in the universe and the formation of our solar system. Livermore cosmochemistry projects, funded by the Laboratory Directed Research and Development (LDRD) Program and the National Aeronautics and Space Administration, allow researchers to use the same instruments and procedures that nuclear forensic scientists use when analyzing interdicted nuclear and radiological samples.

One LDRD effort is studying inclusions in Australian zircons more than 4 billion years old to determine when conditions suitable for life first emerged on Earth. The work, led by scientist Ian Hutcheon, involves dating and isotopically and chemically analyzing the zircons

and the mineral inclusions in them. The effort focuses on measuring the abundance of trace elements, notably uranium and other actinides in these ancient zircons. This analysis will help researchers to understand the evolution of the atmosphere and hydrosphere during the earliest epoch of Earth’s history and to evaluate the evidence for volcanic activity as far back as 4.4 billion years.

Hutcheon says this LDRD effort develops and enhances microanalytical capabilities needed for nuclear forensics. The investigation uses tools that also support Livermore’s nuclear forensics work, such as the Laboratory’s nanometer-scale secondary-ion mass spectrometer (NanoSIMS) and a new, ultrahigh-resolution scanning electron microscope. “Whether we’re measuring oxygen isotopes in Australian zircons or in interdicted uranium yellow cake, the techniques are similar,” he says. “Our ancient zircon research is published in peer-reviewed journals, and the suggestions and criticism made in response to these published papers strengthen our nuclear forensics work.”

Hutcheon was involved in another LDRD cosmochemistry effort headed by John Bradley of Livermore’s Institute of Geophysics and Planetary Physics. Using a transmission electron microscope, Laboratory researchers detected a 2,175-angstrom extinction feature (or bump) in interstellar grains embedded within interplanetary dust particles. The Livermore team identified organic carbon and amorphous silica-rich material as responsible for the bump. (See *S&TR*, September 2005, pp. 24–27.)

These measurements may help explain how interstellar organic matter was incorporated into the solar system. Interplanetary dust particles are roughly the same size scales of interdicted radiological and nuclear materials, such as powdered nuclear enriched uranium. By studying these dust particles, scientists gain confidence in using advanced tools to characterize other materials of interest to nuclear forensics work.

when key words are entered, the system will find the relevant information,” Wong says. He notes that the knowledge base will use a distributed architecture. That is, the information will not be centralized in one location. Instead, information will be distributed across several government sites, such as DHS, FBI, and DOE, with several levels of controlled access. The knowledge base will include a list of subject-matter experts worldwide who could be called upon to assist in specific nuclear forensics casework. Another proposed element is a U.S. evidence archive, where nuclear materials from past cases would be stored as references.

Growing Capabilities

Nuclear forensics and attribution are two important elements among several DHS and NNSA efforts in border protection, radiation monitoring, emergency response, and consequence management to provide the nation with the best protection against nuclear and radiological threats. Nuclear chemist Michael Kristo says that the nuclear forensics and attribution effort has gained strength with the launching of the Global Nuclear Energy Partnership (GNEP) in February 2006.

As part of President Bush’s Advanced Energy Initiative, GNEP is a strategy to expand nuclear energy worldwide

by demonstrating and deploying new technologies to recycle nuclear fuel, minimize waste, and improve the ability to keep nuclear technologies and materials out of the hands of terrorists. GNEP would build recycling technologies that enhance energy security in a safe and environmentally responsible manner. A basic goal of GNEP is to make it impossible to divert nuclear materials or modify systems without immediate detection. One option is to incorporate tags into nuclear materials at different stages of the fuel cycle so that the materials could be tracked and traced.

Looking to the future, Smith notes that existing Laboratory technical efforts define an emerging Nuclear Forensics Analysis Center funded primarily by DHS. This center would build on Livermore’s experience in nuclear assessment, nuclear weapons and materials, and isotope and trace-element science as well as on the capabilities and expertise of the Forensic Science Center and the NHI and CMLS directorates. “Livermore is uniquely positioned,” he says. “We have science-based signatures, expertise, and facilities. All the pieces are here for such a center and for making an even greater contribution to the prevention of nuclear materials trafficking and nuclear terrorism.”

—Arnie Heller

Strengthening the Worldwide Effort

The Nuclear Smuggling International Technical Working Group (ITWG) was chartered in 1996 to foster international cooperation in combating illicit trafficking of nuclear materials. “The ITWG was formed with the recognition that nations must work together,” says geochemist David Smith of the Nonproliferation, Homeland and International Security Directorate. The ITWG was cofounded by Livermore scientist Sid Niemeyer and has been cochaired by Lawrence Livermore since its inception.

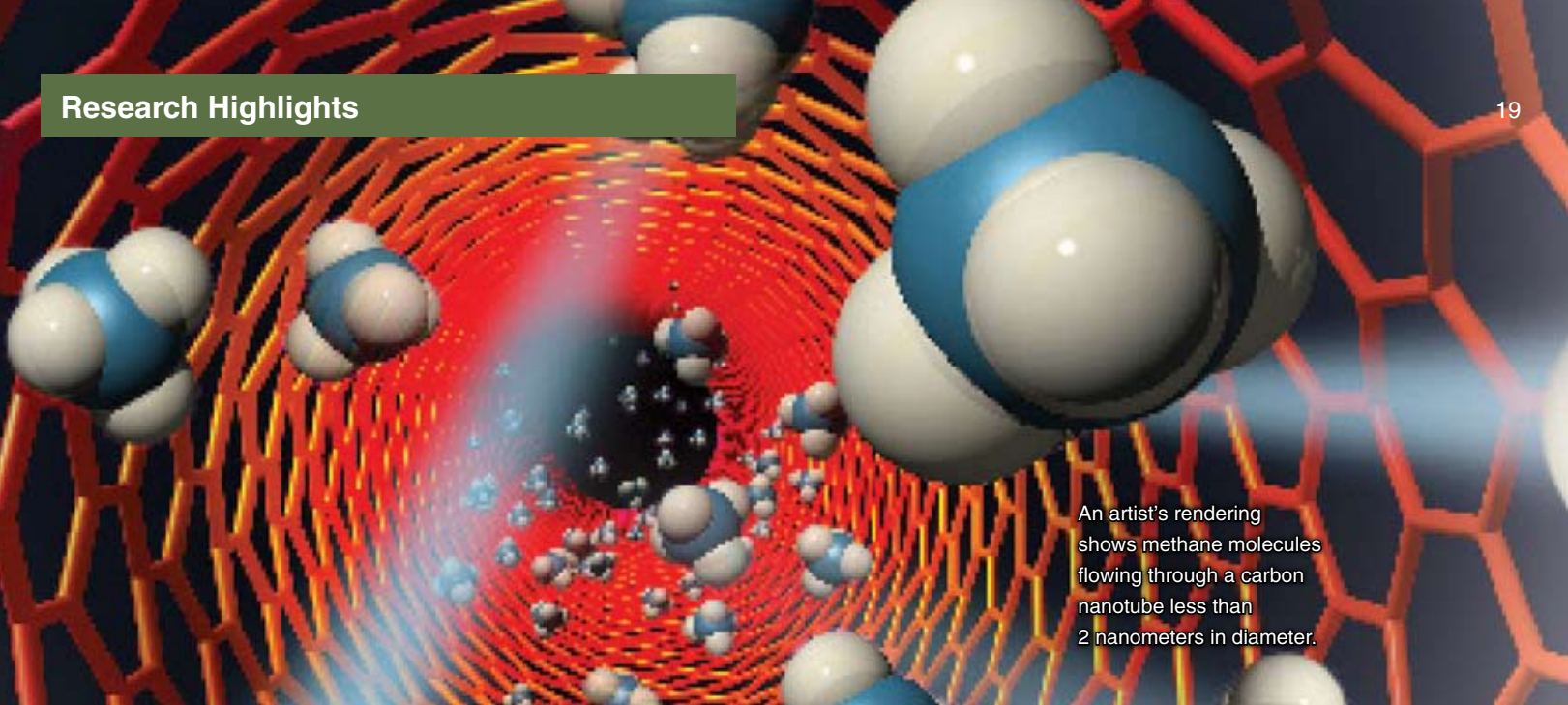
The ITWG works closely with the International Atomic Energy Agency (IAEA) to provide member countries with support for forensic analyses. Priorities include the development of common protocols for the collection of evidence and laboratory investigations, organization of forensic exercises, and technical assistance to requesting nations. Experts from participating nations and organizations meet annually to work on issues concerning illicit trafficking of nuclear materials. The 2006 meeting was sponsored by the European Commission’s Institute for Transuranium Elements in Karlsruhe, Germany.

To promote the science of nuclear forensics within the ITWG, the Nuclear Forensics Laboratory Group was organized in 2004. In that year, Livermore scientists wrote a comprehensive description of a model action plan to guide member states in their own nuclear forensic investigations. The plan provides recommendations governing incident response, sampling and distribution of materials, radioactive materials analysis, traditional forensic analysis, and nuclear forensic interpretation of signatures. In 2006, the IAEA published the model action plan as a Nuclear Security Series Technical Document. Participating countries have adopted the plan and used it in their own nuclear forensics investigations.

Exercises are critical to operational readiness within ITWG participants. An international exercise is planned for later this year, when participants will receive identical radiological samples supplied by Pacific Northwest National Laboratory. “It’s essential for laboratories to discuss the results from their findings and evaluate their capabilities,” says Smith.

Key Words: cosmochemistry, Forensic Science Center, Global Nuclear Energy Partnership (GNEP), International Atomic Energy Agency (IAEA), nuclear attribution, nuclear forensics, Nuclear Forensics Analysis Center, nuclear material, nuclear terrorism.

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An artist's rendering shows methane molecules flowing through a carbon nanotube less than 2 nanometers in diameter.

Tiny Tubes Make the Flow Go

IMAGINE a garden hose that can deliver as much water in the same amount of time as a fire hose 10 times larger. You've entered the realm of carbon nanotubes, where flow rates are enhanced many times over.

These tiny tubes have extremely smooth interior walls that allow liquids and gases to rapidly flow through them, while their tiny pore diameters block larger molecules. A Livermore team led by physicist Olga Bakajin and chemist Aleksandr Noy has created a membrane with millions of carbon nanotubes—each 50,000 times thinner than a human hair—aligned on a silicon chip. Carbon nanotube membranes offer some enticing possible uses, ranging from a more energy-efficient method to filter salt from seawater to dialysis applications.

The principal contributors to the work are staff scientist Jason Holt (a former postdoctoral researcher) and Hyung Gyu Park, a participant in the Student Employee Graduate Research Fellowship (SEGRF) Program. Other Laboratory team members include staff scientist Yinmin Wang, postdoctoral researcher Michael Stadermann, and SEGRF participant Alexander Artyukhin. The team collaborated with Costas P. Grigoropoulos, a professor at the University of California at Berkeley.

Making Nanotube Fast Flow a Reality

Carbon nanotube membranes were first modeled in computer simulations. Only recently has the technology been developed to study their behavior in experiments. "A number of molecular dynamics simulations have appeared in the literature over the years, predicting fast transport of gases and fluids in very small nanotubes," says Noy. "However, no physical experiments had

been performed to prove or disprove flow-rate predictions. Here, at Livermore, we had the people, facilities, expertise, and resources to explore this phenomenon."

With funding from Livermore's Laboratory Directed Research and Development Program and support from the Chemistry, Materials, and Life Sciences Directorate, the team tackled the task of creating an array of carbon nanotubes. One challenge was to develop a method for growing nanotubes with one or, at most, two walls. "Previously, the only nanotubes grown had four to six layers of walls," says Noy. With so many walls, the possibility was high that at least one tube layer would flop over when growing and form a cap, sealing the pore.

The team successfully grew nanotubes just one or two walls thick that had a more likely chance of remaining open. Nanotubes with fewer walls also have the benefit of smaller pore sizes, allowing them to filter out even smaller molecules. Multiwalled nanotubes have pores ranging from 5 to 10 nanometers in diameter, but double-walled tubes have pores measuring just 1 to 2 nanometers in diameter—about the width of six water molecules.

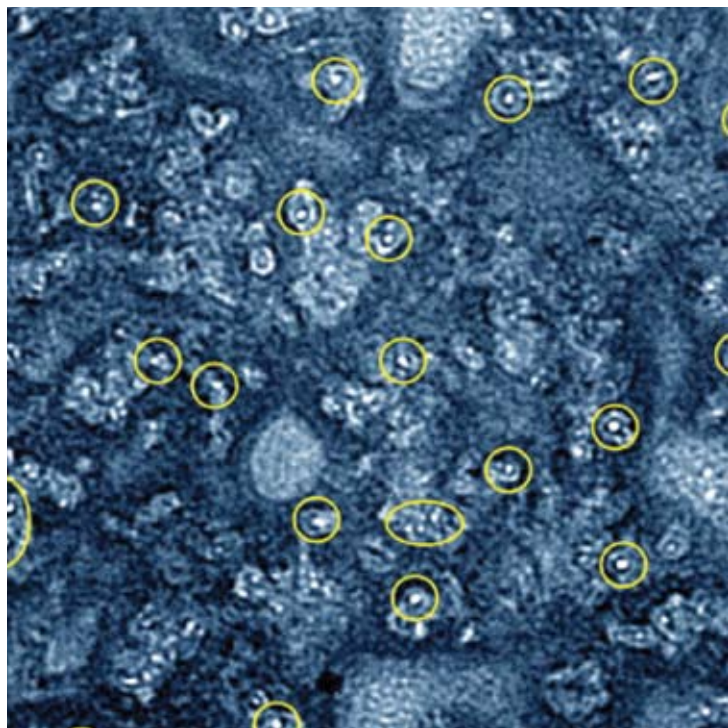
To create these membranes, the team developed a fabrication process compatible with microelectromechanical systems. The process uses catalytic chemical vapor deposition to grow a dense "forest" of double-walled tubes on the surface of a silicon chip. The next challenge was to fill the gaps between the nanotubes without leaving microcracks that would allow fluids or gases to seep through the membrane.

"We designed a deposition process that coats the outside walls of the tubes and the spaces between them with silicon nitride,"

explains Bakajin. Subsequent transmission electron microscopy images showed that this process produces gap-free membranes. The excess silicon nitride is removed from both sides of the membrane, and the ends of the nanotubes are re-opened with reactive ion etching. “The membranes are impermeable to both liquids and gases until this last etching step,” says Bakajin.

The first time the scientists set up an experiment, they covered the top of the membrane with water in which 2-nanometer-diameter gold particles were suspended and then left the experiment overnight. “When we returned the next morning, we were surprised to find a small puddle on the floor under the membrane,” says Park. Holt adds, “We at first thought the membrane had broken, but it evidently allowed the water through, while blocking the gold nanoparticles that were just a bit larger than the nanotube pores. The experiment was a success.”

The team repeated the experiment several times using different membranes to verify the flow rates through the double-walled carbon nanotube membranes. The team calculated the flow rate per tube from the total flow through the membrane, assuming every tube remained open. The measured water flow was comparable to flow rates extrapolated from molecular dynamics simulations. Because at least some of the tubes were closed, the actual flow rate was higher than the calculated rate.



Transmission electron microscopy is used to capture a high-quality image of a carbon nanotube membrane. Several of the nanotubes are circled.

Tiny Tubes Have Big Applications

The Livermore team envisions many uses for such tiny membranes. One of the main applications is to purify, demineralize, and desalinate water. The worldwide need for simple, energy-efficient ways to produce clean water is urgent. Approximately 1 billion people do not have access to clean water. In addition, more than 2 billion people now live in water-stressed areas, a number that is expected to climb to 3.5 billion by 2025. Desalination—the process of removing salts and suspended solids from brackish water and seawater—is one way to alleviate this problem.

Membranes are key to a desalination process known as reverse osmosis, in which water is pushed through a semipermeable membrane that blocks dissolved salts. One drawback of this process is the amount of energy it requires. A typical seawater reverse-osmosis plant requires 1.5 to 2.5 kilowatt-hours of electricity to produce 1 cubic meter of fresh water. The unusually fast flow rates of water through the carbon nanotubes are thus encouraging. If the tiny nanotubes created at Livermore could be scaled up and designed to exclude salts, they could enable desalination facilities to sharply reduce the amount of energy needed to purify water. In much the same way, nanotubes could also someday be used in kidney dialysis to filter waste products such as potassium, acid, and urea from blood.

The team’s research also has the potential to enhance fundamental knowledge of how fluids and gases flow at extremely tiny scales. Such understanding could impact the design of microfluidic chips now under development for genetic analysis. It could also elucidate how ions and water flow through cellular pores.

“Our nanoscale experiments have given us insight into unique and useful material properties,” said Noy. “The unusually fast flows that were predicted in simulations have now been proven experimentally. The reality of this phenomenon opens a number of doors.”

Bakajin adds, “We succeeded in exploring this newly discovered phenomenon as well as we did because of the resources at the Laboratory: the capability for in-house nanotube synthesis that was pivotal for the development of the double-walled nanotube arrays, the Center for Micro- and Nanotechnology’s clean room facility, and most importantly, the people who joined the team. These people, with their skills, expertise, and enthusiasm, and their willingness to come together and find ways to combine their narrow specialties into something new and never seen before, made the project succeed.”

—Ann Parker

Key Words: carbon nanotube membrane, Center for Micro- and Nanotechnology, filtration, microfluidics.

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Acidic Microbe Community Fosters the Unique

Mount Shasta, California

WEST of Mount Shasta in northern California, some of the most acidic water found on Earth lies deep underground. Rich in dissolved metals, the water is leaching from the old Richmond Mine at Iron Mountain. The pH of the mine's water has been found as low as -3.6 in some places, far more corrosive than battery acid.

An environment so inhospitable to humans makes quite a cozy home for certain microbes, called extremophiles for their ability to survive in harsh conditions. Studying extremophiles in the laboratory has been challenging because of the difficulty of re-creating their natural environments. In fact, most of what is known about microbes has come from just a few that can be successfully cultured in the laboratory, such as *Escherichia coli* and yeast. Microbial systems account for a huge percentage of Earth's biomass and play key roles in Earth's biogeochemical cycles, yet they are poorly understood.

The genomic revolution, beginning with the completion of the Human Genome Project, has changed the study of microbes entirely. In the last two years, researchers have learned how to tease genetic information directly from an environmental sample of microbes. As it turns out, subsurface acid mine drainage ecosystems are excellent models for genomic studies of microbial ecology and evolution. These acidophilic (acid-loving) communities are self-sufficient, physically isolated, and relatively simple both geochemically and biologically.

"Now that we can examine whole communities—not just cultured samples—we are finding far more diversity among microbes than anyone had suspected," says Livermore biochemist Michael Thelen. He is leading a team of Livermore scientists and collaborating with researchers from the University of California (UC) at Berkeley and Oak Ridge National Laboratory to take microbial studies beyond sequencing and gene analysis. Their examination of microbial communities in the Richmond Mine has revealed hundreds of unique and unusual proteins.

Down in the Mine

At a junction in the Richmond Mine referred to as the "5-way," an access tunnel intersects four tunnels within the ore deposit, about 1 kilometer underground. Virtually all drainage from the mine moves through the 5-way, where the pink film on the water's surface teems with acidophilic microbes. The pH of the water at the 5-way is about 1, and the water's temperature is about 42°C (107°F).

Jillian Banfield of UC Berkeley has been studying the Iron Mountain site and its biofilms for 10 years. In 2003, a Richmond Mine biofilm from the 5-way was analyzed at the Joint Genome Institute (JGI) in Walnut Creek, California, in the first-ever sequencing of DNA from an environmental microbial



In Iron Mountain, pyrite (also known as fool's gold) interacts with microbes, oxygen, and water to create hot sulfuric acid laden with heavy metals.

community. Sequencing revealed a relatively simple consortium of three bacteria and three archaea (microorganisms of harsh, hot acidic environments).

Each microbial community in the Richmond Mine contains a consistent set of organisms but with varying numbers and strains of bacteria and archaea. “Each strain has adapted to the particular conditions of its site,” notes Thelen. But the causes for the differences—such as a dependence on a certain mineral, competition, or cooperation—are not yet known.

In 2004, Thelen spent a sabbatical year at UC Berkeley working with Banfield. Since then, he and his collaborators have analyzed hundreds of proteins in Richmond Mine biofilms using computational biology, mass spectrometry-based proteomics, and biochemical methods. They are not only identifying novel proteins but are also defining the essential functions of each protein. Some proteins are important for signaling, defense, or transport. Others, known as cytochromes, transfer the electrons gained from iron oxidation. At Iron Mountain, the team has discovered several cytochromes containing novel heme groups that specifically form a coordination complex with iron.

Proteins at Work

A sequence database of over 12,000 proteins was created from the biofilm genomic data set. From this database, the Oak Ridge collaborators identified protein spectra in biofilm extracts using nanoliquid chromatography tandem mass spectrometry. In most cases, proteins could be tracked to specific organisms, because the genes that encode them are on DNA fragments that have already been assigned to organism types. More than 2,000 proteins were determined to be from the five most abundant biofilm organisms.

Analysis of abundant known proteins revealed the importance of protein refolding, perhaps to achieve stability in a highly acidic environment. Other abundant proteins defend against oxidative stress, an important challenge in the acid mine drainage environment.

Overall, however, the biofilm was dominated by proteins that are products of genes originally annotated as “hypothetical” when the genomic data set was established (42 percent of the genes in the original data set). These genes have no known function, and their protein products have not been confirmed in any way. Among the abundant hypothetical, or novel, proteins, 15 percent were unique (not significantly similar to any known protein) and

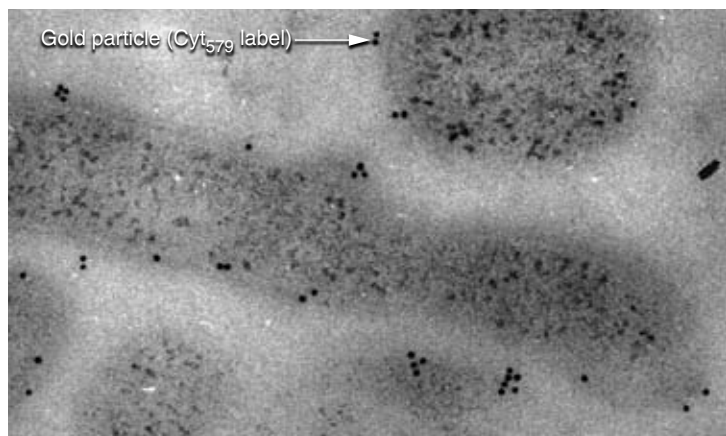
2 percent were conserved (similar to other predicted but not yet characterized proteins).

In the extracellular portion of the biofilm, 52 percent of proteins were unique and about 14 percent were conserved novel proteins. These proteins may be important for adaptation to the extremely acidic and metal-rich conditions.

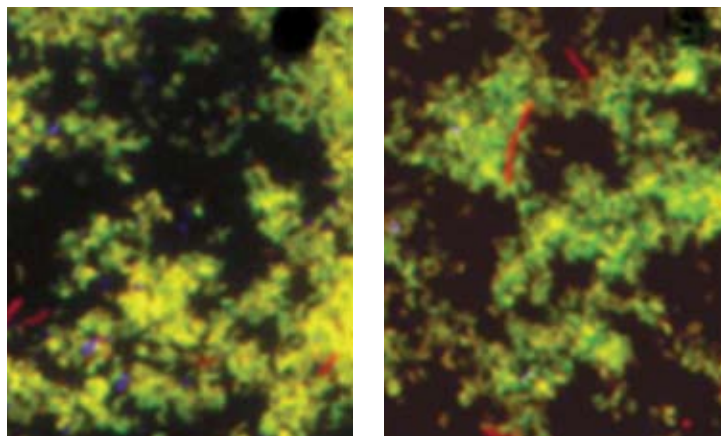
Two of the new extracellular proteins were identified as cytochromes, which contain the heme-iron complex. The spectra



In January 2004, samples of biofilm were collected from the Richmond Mine at Iron Mountain, near Mount Shasta in California. (a) The biofilm occurs as a continuous sheet over the surface of the acid mine drainage pool; wrinkles form because of movement in the solution. (b) In this close-up photograph taken during sample collection, the biofilm is thick and appears to be homogeneous.



Cyt₅₇₉ is a protein actively involved in iron oxidation. The protein's location outside the *Leptospirillum* bacteria is shown here, where an antibody labeled with gold binds to its target cytochrome protein.



Fluorescence in situ hybridization analyses of the collected Iron Mountain biofilm show the *Leptospirillum* protein group II (yellow) and other bacteria (red).

of these two proteins contain peaks at 572 and 579 nanometers—unique among the hundreds of known cytochromes—so they have been dubbed Cyt₅₇₂ and Cyt₅₇₉. Thelen's laboratory analyses showed that both have unusual heme characteristics and are actively involved in iron oxidation. Because *Leptospirillum* group II dominates most biofilms from the Richmond Mine and is frequently detected at other mining sites, the team predicts that these cytochromes are two of the key enzymes connecting the biology and geochemistry of metal-rich acidic environments.

The team was able to assign a probable function to 69 percent of the detected *Leptospirillum* group II proteins on the basis of sequence similarity. It turns out that, as in so many communities, individuals or groups of individuals take responsibility for various activities on behalf of the entire group. In the Richmond Mine biofilms, nitrogen fixation for all members of a community is handled by proteins in *Leptospirillum* group III, the less abundant bacterium. Similarly, carbohydrate metabolism is considerably greater in *Leptospirillum* group III than in *Leptospirillum* group II.

Whether this division of labor has always been this way or has evolved over time is unknown. "We're interested in how these species have adapted to the extreme geochemical conditions in the iron mine ecosystem," says Thelen. "We want to know whether these microbes have been selected as a community by the environment as well as how they evolve with the changes caused by seasonal and other geochemical fluctuations."

Other Extremophiles, Too

Team members are now working with Everett Schock, a geochemist from Arizona State University, to examine mats of thermophilic microbes in the boiling hot springs of Yellowstone National Park. They hope to isolate, identify, and begin to understand the proteins of microbes that interact with silicate, the most abundant mineral in the hot springs environment.

A long-term goal of Thelen's work is to apply new information about microbial decomposition processes to the creation of new sources of energy from plant matter. The hope is that microbes could someday speed the transformation of the lignin in biomass to sugar and then to usable ethanol.

Such studies are part of the Department of Energy's Genomes to Life Program, an effort to better understand the mechanisms of life, beginning with microbes. Microbes are already being used to clean up contaminated soil, and their usefulness for energy production, carbon sequestration, and cleaner industrial processes may eventually be established. Better living through microbes.

—Katie Walter

Key Words: acid mine drainage, archaea, bacteria, genomics, iron oxidation, *Leptospirillum*, proteomics.

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Each month in this space, we report on the patents issued to and/or the awards received by Laboratory employees. Our goal is to showcase the distinguished scientific and technical achievements of our employees as well as to indicate the scale and scope of the work done at the Laboratory.

Patents

Nanolaminate Microfluidic Device for Mobility Selection of Particles

Michael P. Surh, William D. Wilson, Troy W. Barbee, Jr., Stephen M. Lane
U.S. Patent 7,118,661 B2
October 10, 2006

A microfluidic device is made from nanolaminate materials that are capable of electrophoretic selection of particles based on their mobility. In general, nanolaminate materials consist of alternate layers of two materials (one conducting, one insulating) that are made by sputter coating a flat substrate with a large number of layers. Specific subsets of the conducting layers are coupled to form a single, extended electrode, interleaved with other similar electrodes. Thereby, the subsets of conducting layers may be dynamically charged to create time-dependent potential fields that can trap or transport charged colloidal particles. The addition of time dependence is applicable to all geometries of nanolaminate electrophoretic and electrochemical designs from sinusoidal to nearly steplike.

Phasing Surface Emitting Diode Laser Outputs into a Coherent Laser Beam

John F. Holzrichter
U.S. Patent 7,120,184 B2
October 10, 2006

A system for generating a powerful laser beam includes a first laser element and at least one additional laser element having a rear laser mirror, an output mirror that is 100 percent reflective at normal incidence and less than 5 percent reflective at an input beam angle, and laser material between the rear laser mirror and the output mirror. The system includes an injector, a reference laser-beam source, an amplifier and phase conjugator, and a combiner.

Metal Hydride Fuel Storage and Method Thereof

Jeffrey D. Morse, Alan F. Jankowski, Conrad Yu
U.S. Patent 7,122,261 B2
October 17, 2006

A metal hydride fuel-storage cartridge has integrated resistive heaters that can be used in conjunction with fuel cells such as microelectromechanical systems-based fuel cells. The cartridge is fabricated using micromachining methods and thin- and thick-film materials synthesis techniques.

Method for Preparing a Solid-Phase Microextraction Device Using Aerogel

Fred S. Miller, Brian D. Andresen
U.S. Patent 7,125,580 B2
October 24, 2006

A sample collection substrate of aerogel or xerogel materials bound to a support structure is used as a solid-phase microextraction (SPME) device. The xerogels and aerogels may be organic or inorganic and doped with metals or other compounds to target specific chemical analytes. The support structure is typically formed of a glass fiber or a metal wire (stainless steel or Kovar). The devices are made by applying gel solution to the support structures and drying the solution to form aerogel or xerogel. Aerogel particles may be attached to the wet layer before drying to increase sample collection surface area. These devices are robust, stable in fields of high radiation, and highly effective at collecting gas and liquid samples while maintaining superior mechanical and thermal stability during routine use. Aerogel SPME devices are advantageous for use in gas chromatograph-mass spectrometer (GC-MS) analyses because of their lack of interfering background and tolerance of GC thermal cycling.

Awards

James Wilson of the Laboratory's Defense and Nuclear Technologies Directorate is the recipient of the **American Physical Society's 2006 Hans Bethe Award**. Throughout his career, Wilson has made substantial contributions in both aerophysics and astrophysics. He is best known for his supernova calculations, proposing how one works and why it explodes, and for his work on neutron-star binaries.

The Hans Bethe Award recognizes outstanding work in theory, experiment, or observation. This award is presented annually to one individual for outstanding accomplishments in astrophysics, nuclear physics, nuclear astrophysics, or closely related fields. The award commemorates Hans Bethe, a German-American physicist who won the 1967 Nobel Prize in Physics.

Masaru Takagi of the National Ignition Facility (NIF) Programs Directorate received the **2006 Larry Foreman Award for Excellence and Innovation in Inertial Confinement Fusion (ICF) Target Fabrication**.

The award is given every two years in memory of Edward Teller Award recipient Larry Foreman (formerly of Los Alamos National Laboratory) and his innovative work in target fabrication for ICF. Takagi invented the chemical processes used to make extremely

round and smooth plastic shells that are the starting point for ICF capsule fabrication. His work enables the production of both plastic and beryllium shells that meet the stringent specifications required for ignition experiments with NIF.

A team of scientists led by **Francois Gygi**, formerly of the Laboratory and currently at the University of California at Davis, received the **2006 Gordon Bell Prize** for a large-scale electronic structure simulation of the heavy metal molybdenum, conducted on the world's fastest supercomputer—the IBM BlueGene/L at Livermore. Other team members included researchers from IBM's T. J. Watson Research Center, Carnegie Mellon University, and Institute of Analysis and Scientific Computing at the Vienna University of Technology, Vienna, Austria.

Named for C. Gordon Bell, one of the founders of supercomputing, the Gordon Bell Prize is awarded to innovators who advance high-performance computing. Bell established the prize in 1987 to encourage innovation that would further develop parallel processing—the computer design philosophy that has driven high-performance computing since the 1980s. The prize, one of the most coveted awards in high-performance computing, is administered by the **Institute of Electrical and Electronics Engineering**.

Titan Leads the Way in Laser–Matter Science

Last July, Lawrence Livermore commissioned experiments on Titan, the only laser in the world that couples a high-energy, petawatt short-pulse (subpicosecond) beam with a kilojoule long-pulse (nanosecond) beam. With this capability, scientists are exploring a range of phenomena from the acceleration of charged particles to hydrodynamics to the emission and absorption of radiation in hot dense plasmas. Research using Titan will help determine the physics requirements for future experiments at the National Ignition Facility. One team is focusing on the optimization of electron and proton transport for fast ignition. Studies are also being conducted to extend equation-of-state information for materials at very high pressures, densities, and temperatures. In addition, high-energy backlighting techniques are being developed for use in high-energy-density experiments.

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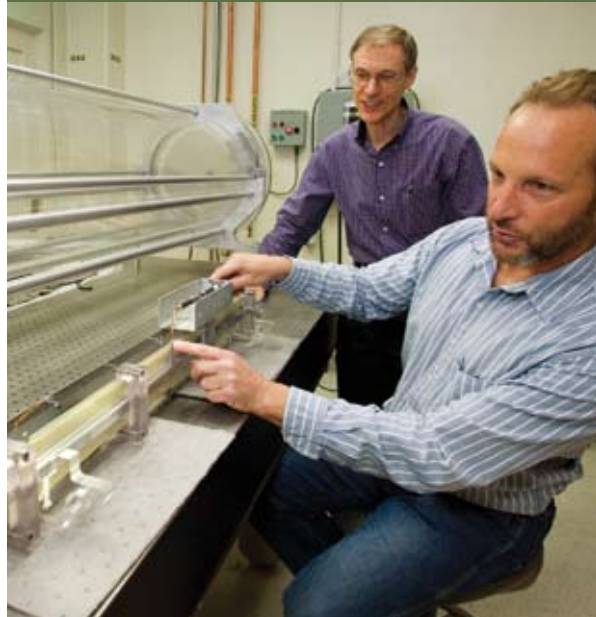
Identifying the Source of Stolen Nuclear Materials

Lawrence Livermore is supporting a national nuclear forensics and attribution capability that ties nuclear and radiological materials to people, places, and events. The effort is part of an overall national strategy to counter nuclear terrorism. Livermore scientists subject an interdicted sample to a host of extraordinarily sensitive and accurate measurement techniques to determine the source location and age. They are obtaining samples of uranium products from around the world and analyzing isotope and trace-element content, grain size, and microstructure. The information obtained from uranium producers forms one part of a growing technical nuclear forensics knowledge base effort led by Livermore scientists. Nuclear forensics and attribution is an essential component of a comprehensive national program to deter illicit trafficking.

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A Collaborative Fight against Cancer



Lawrence Livermore is working with the University of California at Davis Cancer Center to develop technologies that better diagnose and treat cancer.

Also in March

- *At the National Atmospheric Release Advisory Center, researchers are on call to predict the distribution pathways of a toxic airborne release.*
- *A Livermore computer model shows how a warmer climate could affect California's agricultural industry.*
- *Gas-gun experiments using functionally graded material impactors allow researchers to study unexplored regions of phase space during shock compression.*

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