

Lawrence Livermore

December 2012

# Science & Technology

REVIEW

Update on  
Plutonium  
Aging

## Detecting Neutral Particle Interplay

*Also in this issue:*

**A Faster Path to Drug Discovery**

**Diagnostic Looks inside a NIF Target**

## About the Cover

The article beginning on p. 4 describes Livermore's collaborative efforts to build detectors that can identify rare neutral particle interactions. Systems are being developed to monitor nuclear reactor output, confirm neutrino oscillation parameters, and detect dark-matter particles. The cover shows fuel rods at the core of a nuclear reactor with a rendering (above) of a dying galaxy. The blue glow in the the reactor is Cerenkov radiation produced by the fission process. Also in this issue, beginning on p. 11, is a highlight that describes recent results from the Laboratory's ongoing study of plutonium aging. The photo in the upper right shows a dilatometer used for some of those experiments. (Reactor photo courtesy of Idaho National Laboratory. Galaxy image courtesy of National Aeronautics and Space Administration, European Space Agency, and Space Science Telescope Institute.)



Cover design: Daniel S. Moore

## About S&TR

At Lawrence Livermore National Laboratory, we focus on science and technology research to ensure 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 eight 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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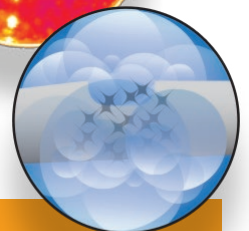
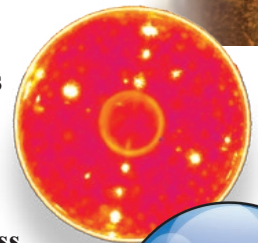
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### Inaugural Class Inducted into Entrepreneurs' Hall of Fame

On October 12, 2012, Laboratory Director Penrose (Parney) C. Albright inducted 15 scientists and engineers into Livermore's new Entrepreneurs' Hall of Fame. The researchers, all former Laboratory employees, were honored for developing technologies that have had major economic impact.

Four inductees—Bruce McWilliams, Thomas McWilliams, Jeffrey Rubin, and L. Curtis Widdoes—have been leaders in the computer and computer-aided design industries, collectively starting 13 companies in California's Silicon Valley. Mike Farmwald, an architect on the Laboratory's S1 supercomputer project, has founded six companies related to computing technologies.

M. Allen Northrup developed a micromachined silicon chip that can perform polymerase chain reaction in minutes rather than hours to more quickly analyze DNA. Robert Parker's work on liquid crystal plastics led to the Duracell battery tester and the color-changing Mood Ring, a 1970s product that sold millions of units. Walter Scott is best known for founding Digital Globe, the satellite imaging company behind Google Earth. Software written by John Hallquist enabled the first computerized automobile crash simulations, which according to the Council on Competitiveness saves the auto industry about \$14 billion a year in cost avoidance.

Brent Dane and Lloyd Hackel developed an advanced laser peening system to strengthen metal parts. To date, more than 40,000 jet engine fan blades and 1,000 discs have been treated by laser peening, saving the aviation industry hundreds of millions of dollars.

The micropower impulse radar conceived by Tom McEwan has been licensed to 42 companies. Also known as radar on a chip, this technology has been converted into about a dozen products, including fluid-level sensors, security motion systems, and back-up warning systems for heavy vehicles.

Joe Gray and Daniel Pinkel are responsible for fluorescence in situ hybridization, or chromosome painting—a key contributor to the genetics revolution. This diagnostic technique is used not only to assess radiation exposure but also to evaluate certain cancers and leukemias and their treatment options.

Engineer James Bryan is considered by many to be the founding father of modern precision engineering. One of his technologies, adapted from an old British invention, is the telescoping magnetic ball bar test gauge. Designed to evaluate the accuracy of machine tools used to produce components for

nuclear weapons, the gauge has now been adopted by hundreds of companies. In 2000, *Fortune* named Bryan one of six Heroes of U.S. Manufacturing.

Livermore established the Entrepreneurs' Hall of Fame to recognize current or former employees who have made major contributions to the U.S. through their inventiveness and entrepreneurial work in and with the private sector. More information on the 2012 inductees is available online at [www.llnl.gov/news/newsreleases/2012/Oct/attach/entrepreneurs.pdf](http://www.llnl.gov/news/newsreleases/2012/Oct/attach/entrepreneurs.pdf).  
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### JASPER Gas Gun Fires Its 100th Shot

The Joint Actinide Shock Physics Experimental Research (JASPER) Facility, a two-stage light-gas gun located at the Nevada National Security Site, fired its 100th shot on September 25, 2012. The 20-meter-long gas gun (shown below) is a key scientific tool for the National Nuclear Security Administration's Stockpile Stewardship Program, which is responsible for maintaining the nation's nuclear deterrent. Lawrence Livermore operates JASPER in conjunction with the Joint Laboratory Office—Nevada, a collaboration involving Livermore, Los Alamos National Laboratory, and National Security Technologies, the management and operating contractor for the Nevada National Security Site.

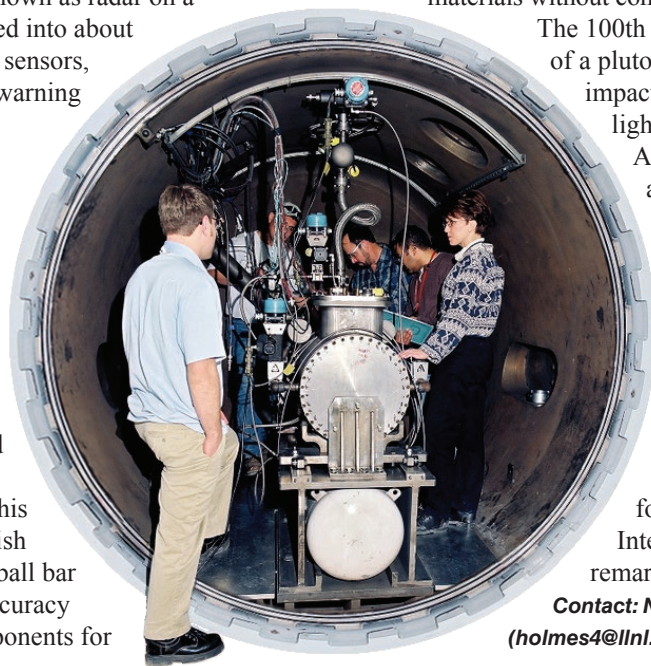
The first JASPER experiment was conducted on March 19, 2001. Of the 100 shots executed to date, 41 have investigated plutonium material properties. The remainder supported those experiments by testing target design, innovative measurement methods, and gun performance. Results from these tests are helping scientists better understand the behavior of plutonium and other special nuclear materials without conducting underground nuclear tests.

The 100th JASPER shot measured the velocity of a plutonium sample following projectile impact and how changes in velocity affected light emission from the sample's surface.

A video showing a JASPER shot is available online at [www.llnl.gov/news/newsreleases/2012/Sep/attach/JASPER.mov](http://www.llnl.gov/news/newsreleases/2012/Sep/attach/JASPER.mov).

"JASPER and the JASPER team have not only produced the largest data return on plutonium of any experimental facility, they have done it for the lowest cost per data point, by far," says Bruce Goodwin, principal associate director for Livermore's Weapons and Complex Integration Principal Directorate. "It is a remarkable facility and achievement."

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# Providing Innovative Solutions through Exceptional Science

A hallmark of the Laboratory is its ability to seek out novel approaches to solving emerging national needs and to apply cutting-edge science and engineering to address those challenges. Livermore scientists particularly excel at the intersection of basic science (the pursuit of fundamental knowledge) and applied science (with focus on addressing a mission need).

The article beginning on p. 4 exemplifies these strengths. It describes experiments aimed at identifying rare neutral particle interactions—work that has led to the development of ultrasensitive detectors for monitoring nuclear reactors. These devices measure the antineutrino flux emitted from a reactor core, and their results indicate whether plutonium is being diverted for possible clandestine activities. The successful application of such sensitive detection techniques to the difficult environments of the real world is a major accomplishment and exemplifies the value that Lawrence Livermore brings to the global security challenge of preventing the spread of nuclear materials to nonnuclear countries. For nearly a decade, the Laboratory has been a leader in demonstrating the utility and robustness of detectors for measuring weak radiation signals amid other background noise for various applications, such as screening cargo at points of entry to the country to identify possible nuclear threats.

Another example of breakthrough science that contributes to various research areas is accelerated mass spectrometry (AMS), which measures carbon-14 and other isotopes down to minute levels, indicating the age of a sample or other scientific or forensic attributes. Lawrence Livermore has more than 20 years of experience in AMS-based research. The Laboratory's Center for Accelerator Mass Spectrometry was established in 1988 to meet various programmatic needs and to serve as a user facility for University of California researchers. Early research efforts focused on forensics science in support of national security and a biomedical study to measure how rat DNA is affected by a suspected carcinogen that results from cooking meat. As explained in the highlight beginning on p. 18, researchers are now adapting AMS techniques to speed the discovery of new drugs to counter bioterrorism threats and to improve people's overall quality of life.

Our longest running mission is the continued safety, security, and reliability of the nation's nuclear weapons stockpile. The Laboratory has forged new pathways for analyzing the condition of the nuclear deterrent as part of the stockpile stewardship

mission that began in the 1990s with the moratorium on underground testing. The highlight on p. 11 discusses a continued study of naturally and artificially aged samples of plutonium to better understand the time-related changes occurring in plutonium pits. Research such as this helps the nation maintain confidence in the stockpile without nuclear testing.

The goals of stockpile stewardship also benefit from the experiments performed at the National Ignition Facility (NIF). As discussed on p. 15, a new diagnostic called RAGS (radiochemical analysis of gaseous samples) is one of several radiochemical techniques that draw on expertise gained from past nuclear experiments. RAGS allows scientists to measure key characteristics of an implosion from inside the capsule, providing insight into what happens to materials under extreme temperature and pressures.

Particle detection, AMS, materials science, and NIF neutronics all have their roots in early research performed at the Laboratory since it was founded in 1952. Over the last 60 years, Lawrence Livermore has continued to demonstrate its commitment to national security missions by applying its scientific prowess to address important national needs, ultimately making the world a safer place. We are living up to our promise as one of the foremost national security laboratories and broadening our reach into new scientific ventures. As Laboratory Director Parney Albright stated at our 60th anniversary celebration, we are "anticipating, innovating, and delivering solutions for the nation's most challenging problems."

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■ Bruce E. Warner is principal associate director for Global Security.

# Positively Scintillating

## Neutral

*Ultrasensitive detectors reveal rare particle interactions that could help scientists improve global nuclear security and advance fundamental physics research.*

**S**INCE the 1930s, when Austrian-born physicist Wolfgang Pauli first theorized the existence of the neutrino, this tiny, nearly massless particle with no charge has made mighty contributions to scientific exploration. Researchers first thought the neutrino would be undetectable because it seldom interacts with matter and is not affected by the electromagnetic force. But its existence was proven 25 years later when its antiparticle, the antineutrino, was detected as a by-product of the fission process occurring in a nuclear reactor. Ongoing studies continue to reveal the mysterious properties and behaviors of neutrinos and other neutral particles.

For nearly a decade, the Laboratory has been a leader in demonstrating the utility and robustness of detectors that can identify the rare interactions between neutral particles. Working with research and academic institutions worldwide, Livermore scientists are applying computational resources and technological advances to improve the measurement fidelity and sensitivity of these devices.

In a nonproliferation project completed in 2008, researchers from Lawrence Livermore and Sandia national laboratories deployed a prototype detector at a nuclear

reactor and demonstrated that the detector precisely tracked the flux of antineutrinos emitted by the reactor core. “At Livermore, we’re also involved in two basic science experiments: one to better understand the oscillation properties of neutrinos and the other to directly measure, for the first time, interactions of dark-matter particles in a detector on Earth,” says Adam Bernstein, who leads Livermore’s advanced detectors group. “Rare neutral particle detection underlies nuclear security and fundamental nuclear science.”

A detector’s application influences decisions regarding its construction, cost, and operability. For example, a system built to monitor nuclear reactors for international safeguards must have a cost-effective design that is easy to replicate, has low maintenance requirements, and can be readily used in various locations. Detectors for specialized physics experiments, on the other hand, can be more elaborate and may be designed for one-time operation.

For users to have confidence in the acquired data, both types of systems must efficiently distinguish a signal of interest from background noise. Detectors also must be sensitive enough to measure particle interactions with kilo- to

megaelectronvolt energies, in particular those from electromagnetically neutral particles such as gamma rays and neutrons, dark-matter particles, and antineutrinos.

### A New Regime in Reactor Monitoring

Inside a nuclear reactor, antineutrinos are produced through the fission of plutonium and uranium isotopes within the core. (See *S&TR*, January/February 2006, pp. 21–23; July/August 2008, pp. 23–25.) During the fuel cycle of a typical reactor, the number of antineutrinos emitted from the core decreases as plutonium

Livermore physicists Nathaniel Bowden (left) and Timothy Classen calibrate a scintillator-based antineutrino detector before deployment. These massive systems can weigh up to 20 tons and measure more than 3 meters per side. The background image, taken by the Hubble Space Telescope, shows the galaxy cluster MACS J1206. Clusters such as these have enormous mass, much of which is theorized to be dark matter. The Laboratory’s expertise in neutral particle detection is also being used to directly characterize these particles. (Background image courtesy of National Aeronautics and Space Administration, European Space Agency, and Space Telescope Science Institute.)

# Particles Brighten Scientific Prospects

*“I have done a terrible thing, I have postulated a particle that cannot be detected.”*

—Wolfgang Pauli



content builds. By using this flux rate and the known thermal power of the reactor, operators can track antineutrino emissions throughout a core's one- to two-year lifespan and identify abnormal shifts.

Scintillator-based detectors, which measure inverse beta-decay events, are the most effective technology for monitoring the flux rate from a reactor. When an antineutrino collides with a proton in the scintillation liquid, the interaction produces a positron and a neutron that induce a measurable signature—two bright flashes of light occurring almost simultaneously. (See the movie at [str.llnl.gov/Dec12/images/antineutrino.mov](http://str.llnl.gov/Dec12/images/antineutrino.mov).)

The Livermore–Sandia demonstration in 2008 showed that scintillator detectors operating 10 meters underground and 25 meters from a reactor core can detect an anomalous flux in antineutrino emissions, which in turn reveals changes in fissile content. However, such detectors weigh up to 20 tons, with each side measuring 3 meters or more—a size that reduces the number of possible deployment locations at nuclear reactor sites.

In 2010, Livermore researchers began testing a water-based detector that is still rather large—it fits inside a standard cargo container—but offers several advantages

over scintillator-based systems. (See *S&TR*, September 2010, pp. 20–22.) “Water is likely the only material that can be scaled in a cost-effective and environmentally safe way to the hundreds of kiloton detector sizes required for remote monitoring,” says Bernstein. “The only other viable alternative, liquid scintillator, is far more expensive to procure and maintain, increases handling risks to workers, and is more damaging to the environment.”

In addition, water is inherently more resistant to the background signals produced by cosmic rays. As a result, a water-based detector might operate aboveground without heavy shielding, thereby greatly simplifying the deployment process at nuclear reactors. Unfortunately, tests of the Livermore prototype revealed only marginal sensitivity for aboveground operations because of residual background signals.

Bernstein notes that the basic detection concept remains viable, and his team is working on a Laboratory Directed Research and Development project to explore a revamped water-based detector. Because antineutrino flux drops as it travels away from a reactor core, a detector designed for long-distance monitoring must be large enough to provide the

required probability for particle interaction with atoms inside. By building a detector with cheaper materials, the team can increase the detector's size and thus extend the range of detection.

The team has completed a conceptual design for a 1-kiloton water-based detector to identify an antineutrino signal several kilometers from a reactor. If the new system is successful, it will be the first water-based detector used to monitor reactor antineutrinos. It would also allow researchers to gain insight into the characteristics of even larger systems for true remote operations in support of the nation's nonproliferation efforts.

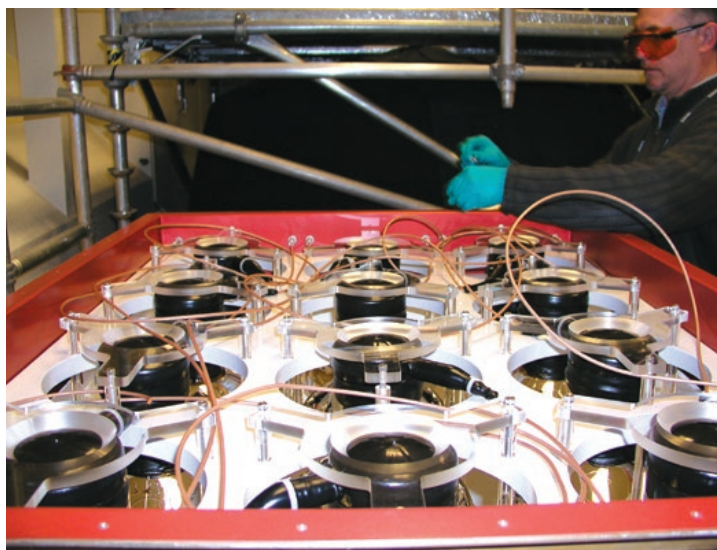
### A “Flavor”-full Mixture

Much like ice cream comes in three primary flavors—vanilla, strawberry, and chocolate—neutrinos have three flavor states: electron, muon, and tau. Scientists theorize that a neutrino can oscillate, or transform, from one flavor to another as it travels from its source. (See *S&TR*, April 2003, pp. 13–19.) Quantum mechanics dictates, however, that oscillation can occur only if neutrinos have mass, even if it is only a tiny amount. Experiments since the mid-1990s have shown that oscillation is possible, indicating that neutrinos do have mass. In fact, oscillation would account for the difference between the number of predicted and detected electron neutrinos emitted by the Sun.

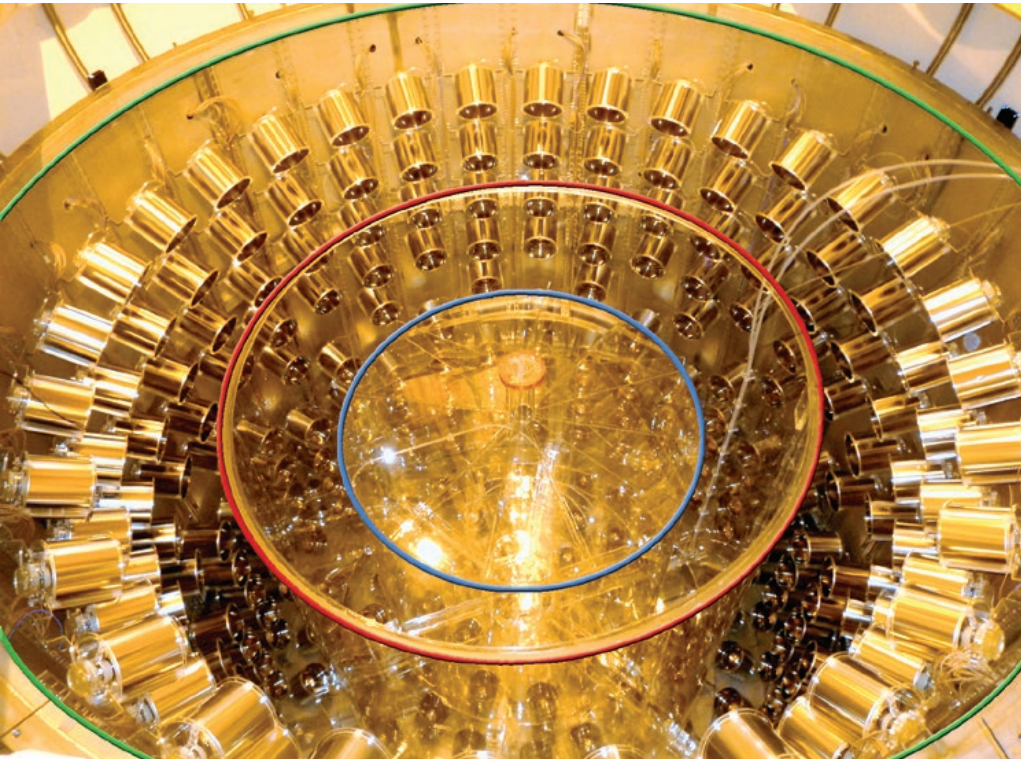
Oscillation parameters, or mixing angles, describe the probability that one type of neutrino will transform into another over a given distance. Researchers have measured two of the three mixing angles. The third, a fixed parameter called theta-13, is considerably smaller than the other two, making it more difficult to detect.

An experiment at the Chooz nuclear power plant in France helped confirm the amount of oscillation between the different neutrino flavors and set lower and upper limits for theta-13. Called Double Chooz, this experiment used a 10-ton

The water-based antineutrino detector developed at Livermore contains a mixture of water and gadolinium. Physicist Steve Dazeley fits the 1-ton device with an array of 12 photomultiplier tubes, each about 25 centimeters in diameter, to record the signal of interest.







The core of the main detector for the Double Chooz experiment in France contains four concentric cylindrical tanks. The innermost 8-millimeter-diameter tank (blue) is filled with the gadolinium-doped target liquid, which is surrounded by another type of liquid scintillator (red) that captures gamma rays emitted by the target. A buffer layer (green) filled with mineral oil shields the target from background signals and radioactivity generated by the 390 photomultiplier tubes attached to the buffer tank's inner wall. (Photograph courtesy of CEA-Saclay/IFRU-SIS.)

gadolinium-doped scintillation detector to record the signals of antineutrinos emitted by the reactors, which provide a pure source of electron antineutrinos.

For the Double Chooz project, Livermore physicists developed simulations to predict the reactor output. "At any given point, the antineutrino could be in one of the three flavor states," says Greg Keefer, a physicist in the Laboratory's Physical and Life Sciences Directorate. "To have a baseline, we had to predict how many electron antineutrinos would be emitted."

Working with European collaborators, the Livermore team modeled the Chooz reactors using two neutron-transport codes, MURE and DRAGON. MURE is a three-dimensional Monte Carlo-based code that tracks individual particle motion in a reactor, from the moment each neutron appears through fission, until it is ultimately absorbed. DRAGON, a two-dimensional lattice code, solves

differential equations to provide an overall average for key parameters.

"We use these complementary models to predict the number of fissions per unit time for each fissile isotope over the running period of the reactor cores," says Keefer. "We can then assess a systematic error on the predicted number of fissions and fold this number into another code that gives us the number of neutrinos per fission of a specific isotope. The results provide us with a predicted antineutrino rate as a function of time and an estimated error on the predicted rate."

The Laboratory's experience in modeling reactor cores for nonproliferation efforts has been an asset for the Double Chooz experiment. "We have created high-fidelity simulations to accurately predict reactor output," says Keefer. "We were the first to bring nuclear engineers onboard to help establish input parameters that are more faithful to how a reactor operates." Simulation results compared

well with established benchmark data from destructive assays of fuel rods at the Takahama reactor in Japan. "A major part of our effort was validating the benchmark data and improving our understanding of diagnostic and systematic errors to obtain the precise measurements."

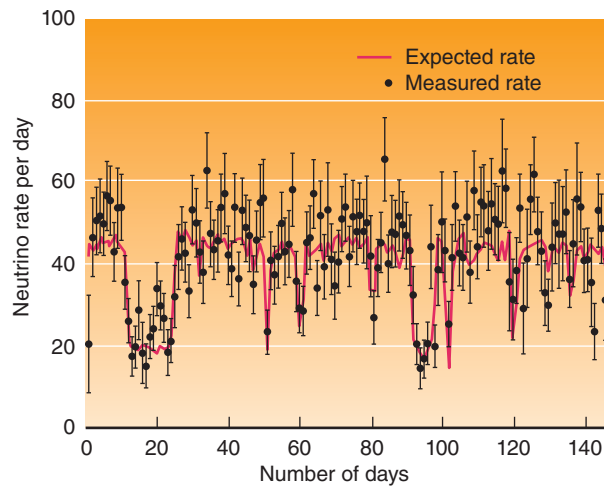
When researchers compared the number of simulated antineutrinos with the amount detected in the Double Chooz experiment, they discovered a deficit in the number detected. Bernstein notes that this finding is consistent with some of the electron antineutrinos emitted by the reactor having oscillated into other flavors. "Double Chooz is a so-called disappearance experiment," he says, "and indeed the experiment found that nearly 10 percent of the electron antineutrinos had disappeared, compared with the number that would have been expected if oscillations were not occurring."

The collaboration was the first reactor experiment to identify a deficiency consistent with a nonzero final mixing angle for theta-13. Later experiments have since confirmed these results, indicating that theta-13 is both nonzero and has a magnitude close to previous upper limits.

### Bringing Dark Particles into the Light

The putative dark-matter particle is much like the neutrino in at least one respect: Both were theorized to exist based on indirect evidence before they were ever detected. Dark matter is postulated

The observed rate of antineutrino events in the Double Chooz experiment compared well with the predicted rate generated by the DRAGON and MURE neutron-transport codes using thermal power measurements as inputs. The two drops in the rate indicate when the reactor was off or operating at reduced thermal power. The predicted event rate was continuously updated to account for reactor downtime.



to make up about 84 percent of the universe, but it does not emit or reflect electromagnetic radiation. Its composition is therefore unknown.

Astronomical observations indicate that dark matter permeates much (or all) of the known universe. The gravitational influence of this matter alters the structure of visible galaxies and other large-scale astronomical objects. Many theoretical and empirical studies point to a candidate dark-matter particle, known as the weakly interacting massive particle (WIMP).

“Dark-matter particles are expected to be heavy, weighing perhaps 10 to 1,000 times the mass of a proton, but they have no electric charge,” says Livermore physicist Peter Sorenson. As a result, they seldom interact with other particles, making them extremely difficult to observe directly. A detector that rigorously suppresses background signals from other particles, such as neutrons that mimic dark-matter signals, might record one true event in many millions.

The Large Underground Xenon (LUX) detector is designed to suppress these background signals so that dark-matter particles can be characterized. “LUX is a dual-phase time-projection chamber for detecting the nuclear recoil signal produced when a dark-matter particle collides with an atom of liquid xenon,”

says Sorenson, whose involvement with LUX predates his time at the Laboratory.

The initial collision of a dark-matter particle with a xenon atom creates a first flash of scintillation photons (S1) and also produces electrons. While the particle recoils off the atom and continues to travel in space, the electrons are tracked by the detector. An applied electric field causes the electrons to drift up the chamber through the cryogenically cooled xenon into a gas above the liquid. The electric field then accelerates the electrons, and they collide with other atoms in the gas. This collision produces many more photons and creates a second, larger flash of light (S2).

“The S1 flash occurs within about 100 nanoseconds,” says Sorenson. “The S2 flash is a factor of 10 more spread out, occurring over microseconds.” Photomultiplier tubes detect and measure the amplified light. The intensity of the S1 light and the S2 charge and the time between the two flashes allow researchers to locate the particle interaction. Says Sorenson, “The ratio of S2 to S1 also indicates the incident particle type, whether it’s a boring background gamma particle or a neutral particle such as dark matter.”

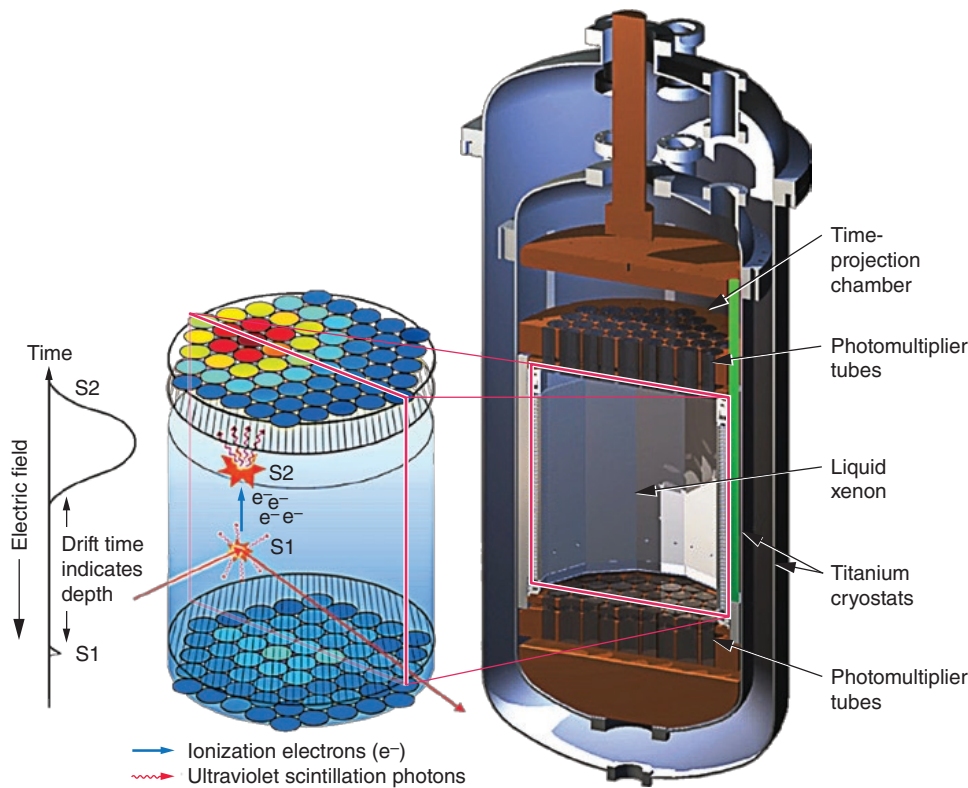
Using xenon instead of lighter elements such as hydrogen or oxygen increases the number of target nucleons with which

the “timid” WIMPs can interact. Xenon is also very dense, so LUX is more compact than gas detectors or those that use other less dense materials. As a result, the nuclear recoil event can be precisely localized within a small section of the device. Sorenson says, “We can reconstruct in three dimensions the position of the particle interaction to within a few millimeters, which helps us determine if the interaction was from dark matter or some other known particle physics process, such as a neutron recoil.”

LUX’s location and construction also protect the detector from background radiation. The detector is deployed 1.5 kilometers underground in the Sanford mine in South Dakota. The overlying rock—called overburden—is the first and best line of defense. “Aboveground, we are bombarded by extremely high-energy muons emitted by cosmic rays,” says Sorenson. “On average, one muon passes through an object the size of a human hand every second. Fortunately, rock does a great job of attenuating the muon signal, and these background signals drop exponentially with depth. Underground, one muon might pass through a LUX-size detector less than once per day.”

In addition, a seamless “skin” of xenon surrounds and shields the central xenon core, and the device is immersed in a water bath to further stave off neutron signals. With this design, the interactions occurring in the core are most likely to come from dark matter, if it exists.

One of a dozen dark-matter experiments worldwide, LUX was tested aboveground prior to its installation in the mine, where it is being commissioned. Once the system is online, it will operate continuously for 300 days, after which in-depth statistical analysis will be completed to determine whether a dark-matter interaction has occurred. “By effectively suppressing backgrounds to the point where ordinary neutron and gamma-ray interactions are extremely unlikely to cause a recoil, we will be able to establish with high



Inside the Large Underground Xenon (LUX) detector, putative dark-matter particles interact with xenon atoms, creating photons and electrons. LUX immediately records the photons produced in a flash of light called S1. The electrons are extracted into a gas and accelerated, producing an amplified, secondary photon signal (S2). The ratio of S2 to S1 and the time between the two flashes indicate the incident particle type.

confidence that the interaction did not result from a more common physics process,” says Bernstein. “Measuring a signal under these circumstances allows us to infer the existence of a new interaction mechanism, consistent with the WIMP hypothesis.”

The Livermore team helped design, construct, and deploy many key subsystems for LUX and will be central to analyzing the data. If the detector provides evidence for the existence of dark matter, the results will spark new areas in physics research. “We don’t yet know what dark-matter particles are made of or how they interact with ordinary matter,” says Bernstein. “It could be a whole new particle zoo. Dark matter may consist of a completely new type of particle, one that we would then have to study and characterize.”

### A Less Obvious Event

The LUX design is also being used to develop a new class of antineutrino detectors for nuclear security applications. Today’s devices rely on inverse beta decay to monitor a reactor’s fissile content, but this process is not the most frequent type of antineutrino interaction. More common, but also far more difficult to detect, is coherent neutrino nucleus scattering, in which a recoiling nucleus collides with its neighboring atoms and “shakes” loose a few electrons. Just as wind rustles the leaves on a tree in unison, through coherent scattering, an antineutrino interacts with an entire nucleus rather than with individual nucleons.

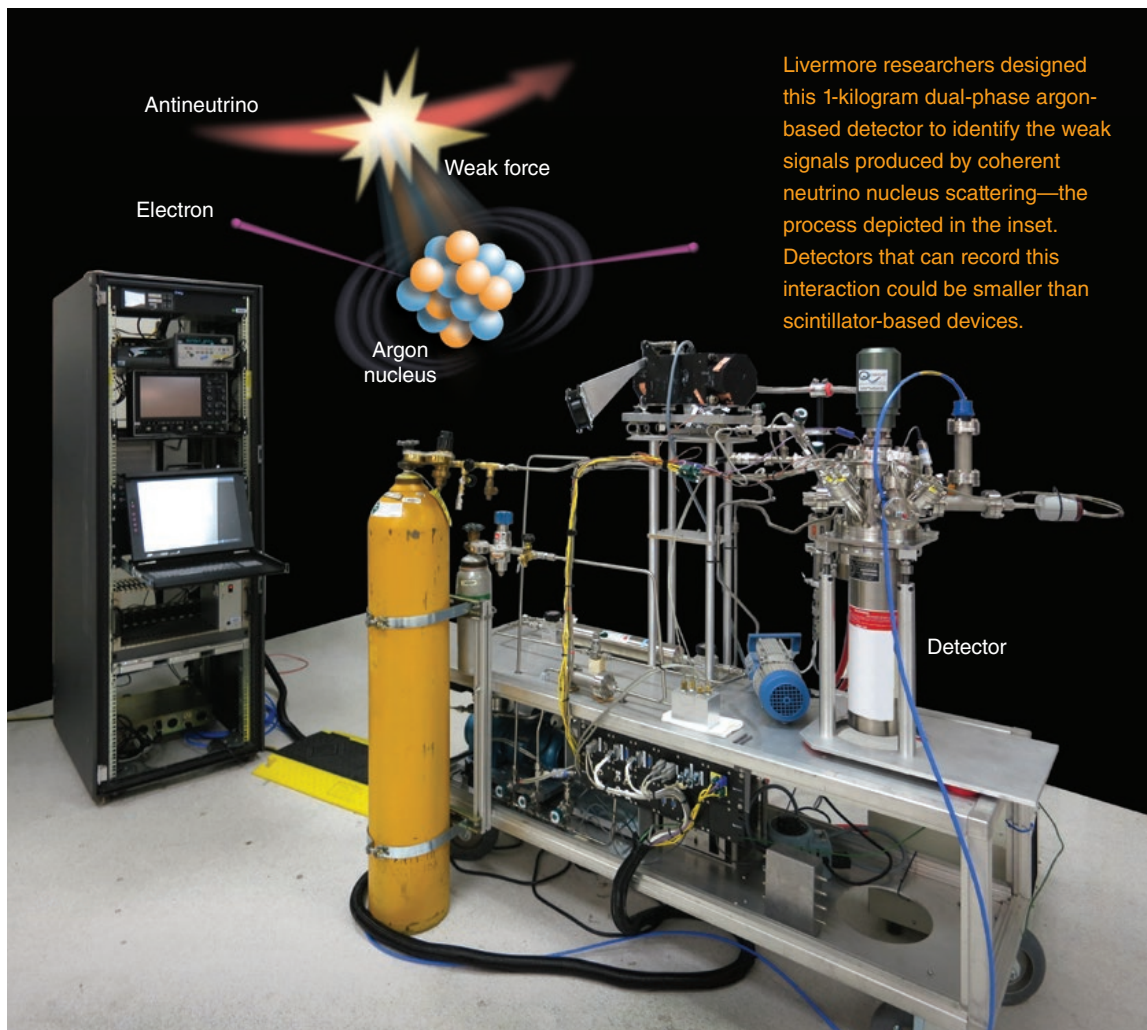
Coherent neutrino nucleus scattering has not been observed experimentally, although its detection rates have been

well calculated as part of the Standard Model of particle physics. Technology improvements made over the last decade allow much lower energy recoils to be recorded with higher reliability. Building a device to measure coherent scattering would be a breakthrough in antineutrino detector research. Coherent scattering is hundreds of times more likely to occur than inverse beta decay, so smaller devices could record these events. Bernstein says, “We could potentially build systems that are 10 to 20 kilograms as opposed to the 1,000-kilogram detectors needed for inverse beta-decay events.”

Livermore scientists have developed a 1-kilogram dual-phase argon-based detector, whose design is akin to LUX. The compact system is designed to record an electron signal in the liquid argon and an amplified signal in the argon gas blanket above the liquid. (See the movie at [str.llnl.gov/Dec12/images/scatter.wmv](http://str.llnl.gov/Dec12/images/scatter.wmv).) The detector also provides three-dimensional event reconstruction of selected signals while minimizing background noise.

Thus far, the novel detector has measured electromagnetic recoils with the lowest energy ever recorded in dual-phase argon. “Our preliminary results indicate that we have achieved sensitivities down to single ionizations,” says Bernstein.

Several important hurdles remain before coherent scatter interactions can be measured. First, the team must estimate the expected number of electrons that would be liberated by an antineutrino during an event. To do so, the team will use a neutron beam to impart the same amount of energy to the nucleus as an antineutrino, thereby establishing sensitivity to the few-electron signal arising from antineutrino interactions. Next, the detector must be reconfigured to better suppress background signals. It will then undergo testing at a nuclear reactor. A successful demonstration would be the first-ever measurement of coherent scattering. “Eventually the device might be optimized to fit in the back of a pickup truck,” says Sorensen.



Livermore researchers designed this 1-kilogram dual-phase argon-based detector to identify the weak signals produced by coherent neutrino nucleus scattering—the process depicted in the inset. Detectors that can record this interaction could be smaller than scintillator-based devices.

A mobile device for reactor monitoring could be extremely valuable for improving nuclear safeguards.

### Work Sparks Growing Interest

According to Bernstein, neutrino research, particularly in the area of reactor monitoring, has gained momentum in the last decade, and an international conference on applied antineutrino physics has been held nearly every year since 2004. “Russian researchers first demonstrated the concept of antineutrino-based reactor monitoring in the mid-1980s,” says Bernstein. “Because of the Cold War, this first-rate work was originally available only in Russian journals

and thus unknown to us and many other Western researchers. Our 2002 theoretical paper and 2006 deployment drew wider attention. As a result, the topic is now a focus of many neutrino physics groups and conferences worldwide.”

By participating in projects such as LUX and Double Chooz, the Livermore team has demonstrated how fusing basic science research with applied physics can yield benefits to both fields of study. Working with experts at the Laboratory provides external collaborators with the types of resources—from experimental hardware to computational capability to unique nuclear physics analyses—that

are available only at a national laboratory. In turn, Livermore physicists have been able to adapt new technology for nonproliferation purposes.

“Investments in basic science have led to improved detector designs for reactor monitoring, and our participation in these efforts has helped further

fundamental physics understanding,” says Bernstein. “Together, we are solving critical global nuclear security problems and promoting high-quality science.”

—Caryn Meissner

**Key Words:** antineutrino detection, coherent neutrino nucleus scattering, Double Chooz, Large Underground Xenon (LUX) detector, neutral particle, neutrino, nonproliferation, nuclear reactor monitoring, nuclear security, particle physics, scintillator, weakly interacting massive particle (WIMP).

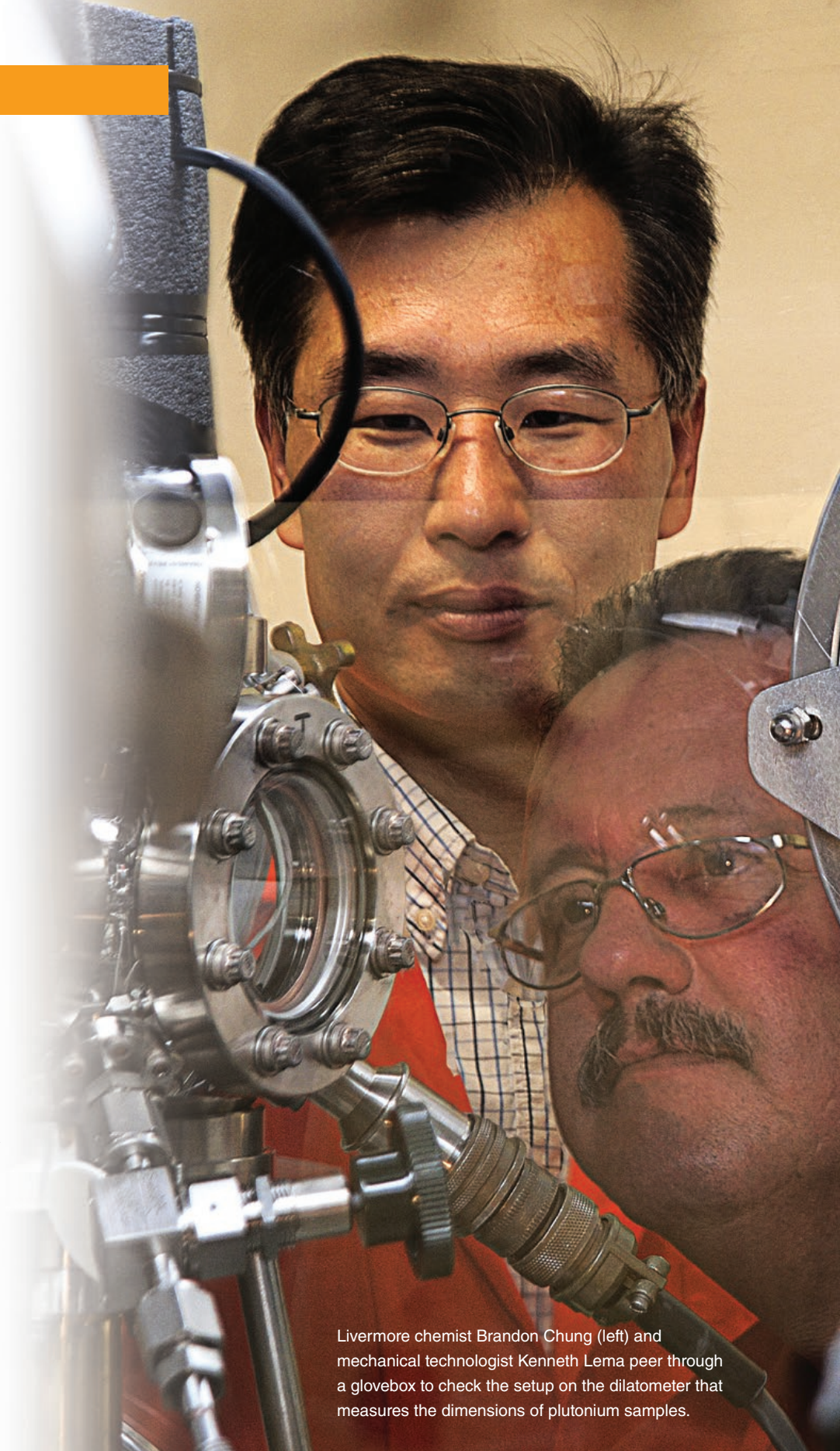
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# Plutonium at 150 Years: Going Strong and Aging Gracefully

**P**LANNING the future needs of the U.S. nuclear weapons stockpile as well as the nuclear weapons complex depends in part on maintaining confidence in the long-term stability of the pit, or core, of plutonium-239 residing inside every weapon. Scientists and engineers who ensure the safety and reliability of the nation's stockpile had long been concerned that the damage accumulated over decades as plutonium-239 self-irradiates could eventually compromise weapon performance.

In 1997, the National Nuclear Security Administration (NNSA) launched a comprehensive study at Lawrence Livermore and Los Alamos national laboratories to examine in detail how plutonium pits age and provide a firmer scientific basis for estimating the service life of these components. The study's results, announced in late 2006, showed that the slow degradation of plutonium in U.S. nuclear weapons would not affect warhead reliability for decades.

Independent research teams at the two laboratories performed extensive mechanical testing and laboratory-based experiments on aged samples of a plutonium-239 alloy—plutonium mixed with a small amount of gallium to stabilize the material in its delta phase at room temperature. Alloy samples were taken from 15- to 44-year-old plutonium pits and from plutonium that was artificially



Livermore chemist Brandon Chung (left) and mechanical technologist Kenneth Lema peer through a glovebox to check the setup on the dilatometer that measures the dimensions of plutonium samples.

aged to 65 years. These tests showed no significant changes in important physical properties such as density and strength. In analyzing the test results, the research teams determined that the minimum lifetime for plutonium pits was at least 85 years—25 to 40 years longer than previously estimated.

Now, six years later, these same naturally aged samples are 50 years old, and the accelerated alloy samples have reached an equivalent age of 150 years. Both sample lots continue to age gracefully, and extremely sensitive tests and high-resolution electron microscope images by Livermore chemists validate the confidence-building conclusions of the earlier study.

“The 2006 report was a work in progress,” says chemist Pat Allen, the deputy program leader for enhanced surveillance and leader of the plutonium aging study. “The 2006 report and recent work continue to show no alarming trends and serve to validate our theories about how plutonium ages. However, we need to keep running tests on naturally aged specimens to see further into the future and make sure there are no surprises.” Allen says that ongoing monitoring of the accelerated alloys is useful to establish the location of any aging “cliff,” where changes would severely affect performance, even if the cliff did not occur in the near term.

Bruce Goodwin, principal associate director for Weapons and Complex Integration, says he is “extremely pleased” at the continuing positive results. “Both the original study by Livermore

and Los Alamos and the ongoing long-term study by Livermore represent a tour-de-force accomplishment for stockpile stewardship.” The results, he says, are highly positive for the safety and reliability of the stockpile and for avoiding the costs associated with remanufacturing pits.

### Most Interesting Element Around

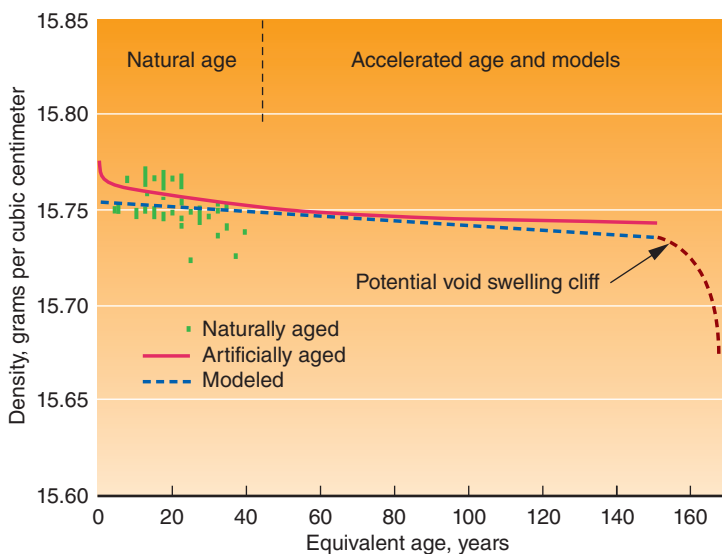
Plutonium is often called the most interesting and perplexing element in the periodic table. The radioactive metal has seven distinct crystallographic phases; its dimensions change with temperature, pressure, and impurities; and it has many different oxidation states. In addition, the material’s properties do not always change in linear fashion. Over the past two decades, researchers at Livermore and Los Alamos have greatly improved scientific understanding of plutonium, in particular how its aging mechanisms might degrade pit performance.

Livermore chemist Brandon Chung has been conducting tests on plutonium-239 alloys for more than a decade, measuring property changes as a function of age for the various samples. Changes arise principally from the alpha-decay process, in which a plutonium atom spontaneously emits a high-energy alpha particle (helium nucleus) and transforms, or decays, into a uranium-235 atom.

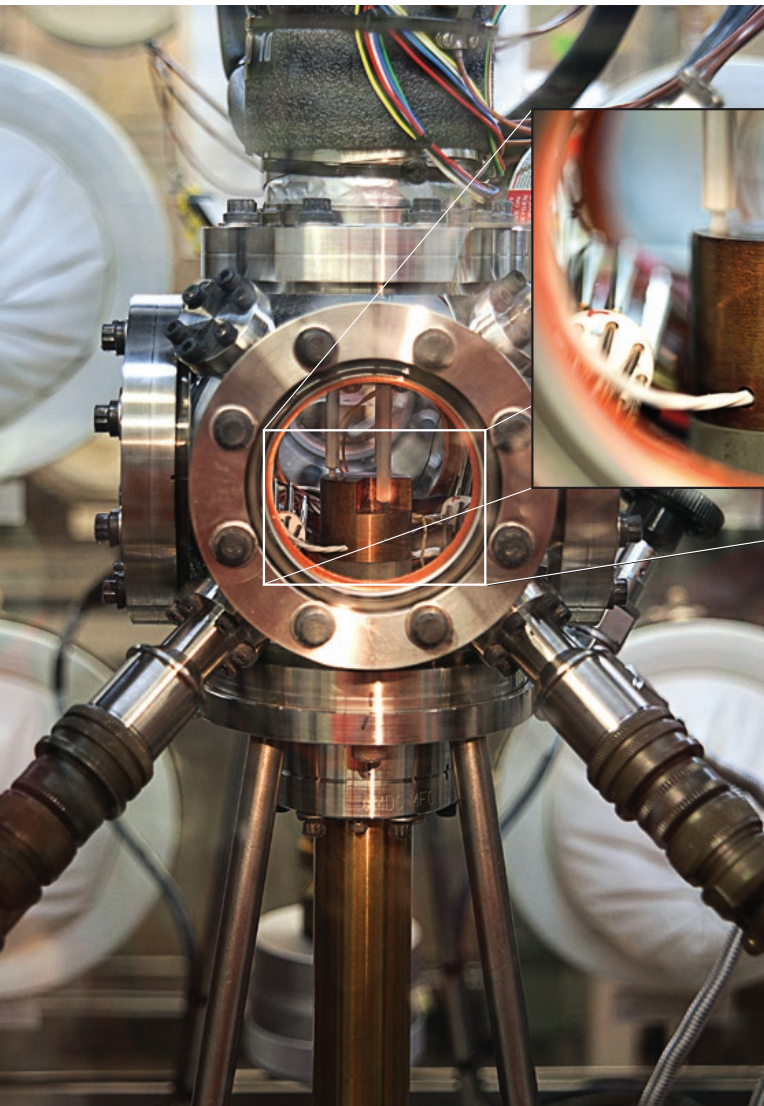
“Self-irradiation of plutonium has all the requirements for potentially catastrophic damage,” says Chung. As a result, scientists have sought greater assurance that a pit would retain its size, shape, and strength as the damage from alpha decay continued to increase.

One concern was that the accumulating damage might induce a change from the delta phase used in nuclear weapons to the denser and more brittle alpha phase. The alpha particle and the uranium-235 atom fly off in opposite directions, disrupting nearby atoms. The uranium-235 atom dislodges thousands of plutonium atoms from their positions in the crystal lattice. On average, each plutonium atom is displaced once every 10 years. However, in a fraction of a second, about 90 percent of these displaced atoms return to their normal lattice position, a process called self-healing. If a defect does not self-heal, the lattice may have vacancies where atoms are missing as well as interstitial atoms wedged between others.

After the alpha particle comes to rest in the lattice, it attracts two electrons to become a helium atom. Helium atoms slowly aggregate over time to form bubbles. If these bubbles coalesce into larger bubbles, they could begin to weaken a weapon component or otherwise change its volume or behavior. In addition, helium buildup combined with vacancies in plutonium’s crystalline lattice could produce voids that swell, potentially causing the material to lose its shape and strength. This so-called void swelling typically happens suddenly once a critical threshold is reached,



Small changes in density in naturally and artificially aged samples indicate that pits made from a plutonium-239 alloy will retain their size, shape, and strength despite increasing damage from self-irradiation. Results from long-term dilatometry experiments have increased confidence that the pits will function as designed up to 150 years after they are manufactured.



Sample holder  
Plutonium sample

Samples of naturally and artificially aged plutonium, each only 2 or 3 centimeters long, are measured in dilatometry experiments to check for changes in a sample's volume over time.

a process documented in other irradiated metals. The resultant swelling would dramatically increase the pit's volume and severely compromise its performance.

### Spiking a Sample to Age It

To simulate the properties of the plutonium alloy decades into the future, Livermore researchers pioneered a technique to accelerate the aging process in samples by mixing plutonium-239 with isotopes that have shorter half-lives. When plutonium-239 (which has a 24,000-year half-life) is mixed with 7.5 percent by weight of plutonium-238 (with an 87-year half-life), the alloy will accumulate radiation damage at a rate 16 times faster

than weapons-grade plutonium alone. (See *S&TR*, May 2007, pp. 12–20.)

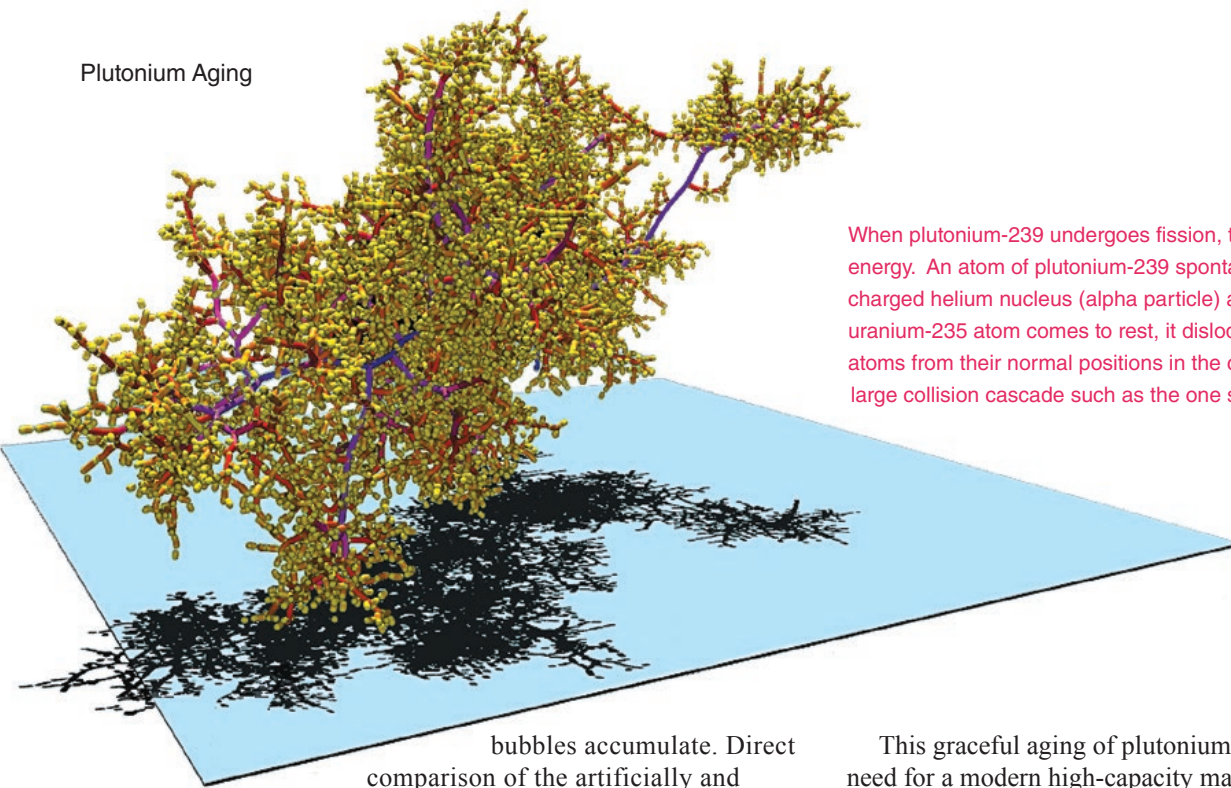
The oldest accelerated samples, manufactured about 11 years ago, are now equivalent to 150-year-old plutonium alloy. Because the damage rate is much higher in artificially aged plutonium than in naturally aged alloys, the accelerated samples are maintained at a higher temperature, to ensure that the self-healing rate is appropriately accelerated as well.

Chung subjects the two sample types to a host of tests that record changes in dimension, density, and strength. For dilatometry experiments, he uses both 2- and 3-centimeter-long samples to ensure that oxidation is not affecting results and maintains them for years in a small chamber with a vacuum-controlled atmosphere. A dilatometer periodically measures the sample dimensions to an accuracy of 0.1 micrometers or less as a check for changes in volume.

Immersion density tests compare the expansion occurring in a parallel set of artificially and naturally aged alloys. These experiments record the amount of liquid displaced when samples are immersed in a fluid bath. A third set is tested for tensile strength (a material's ability to resist being pulled apart) and ductility (its ability to bend without breaking).

Dilatometry and immersion density measurements on both types of alloys consistently show slight volume swelling as samples age but no signs of void swelling. Overall, the dimensional tests indicate that scientists should expect only a 0.25-percent expansion in volume (and a concurrent decrease in density) over a 100-year period.

Static tensile strength tests demonstrate an increase in strength with age, with the rate of change slowing considerably after about 20 years. The results also indicate that ductility decreases (brittleness increases) as lattice damage and helium



When plutonium-239 undergoes fission, the nucleus releases enormous energy. An atom of plutonium-239 spontaneously decays into a doubly charged helium nucleus (alpha particle) and a uranium-235 ion. Before the uranium-235 atom comes to rest, it dislodges thousands of other plutonium atoms from their normal positions in the crystal lattice, thereby creating a large collision cascade such as the one shown in this simulation snapshot.

bubbles accumulate. Direct comparison of the artificially and naturally aged samples combined with results from age-aware strength modeling confirms both the strengthening behavior of plutonium and the accelerated-alloy approach to evaluating damage over time.

### No Indication of Worrisome Voids

Chung found no indication of void swelling in the tests he conducted. Transmission electron microscope analyses performed by Livermore microscopist Mark Wall validated Chung's results, revealing that helium bubbles are distributed uniformly throughout the plutonium alloys. Although helium buildup is substantial after many decades, bubbles stop growing once their diameter reaches 1 to 2 nanometers, and bubble density increases with age. As shown with the accelerated samples, helium accumulation should not dramatically affect the dimensional properties of plutonium for up to 150 years after pits are manufactured. In addition, the age-induced changes do not pose any concern for the metal's strength based on current theory and simulation results.

"Clearly, the findings indicate that lattice damage and helium in-growth are not leading to catastrophic aging effects such as void swelling," says Chung. "Because we have improved the scientific understanding of plutonium aging, we have greater confidence in the reliability of pits in the stockpile." Allen adds that the consistent results produced by Livermore's continued study of plutonium aging validate NNSA's strategy of reusing pits in life-extension programs, which prolong the service life of an aging warhead or bomb and enhance its safety and security. (See *S&TR*, March 2012, pp. 6–13.)

This graceful aging of plutonium also reduces the immediate need for a modern high-capacity manufacturing facility to replace pits in the stockpile. "In the near term, the nation can save tens of billions of dollars that might be required to build a new production facility," says Allen. He notes, however, that the nation must retain a certain level of manufacturing capability to enable a speedy response should an unexpected problem arise.

Plutonium pits in the stockpile were manufactured at the Department of Energy's Rocky Flats Plant, which ended production in 1989. Rocky Flats used a cast-and-wrought approach, in which melted plutonium was poured into a mold, rolled, and pressed into the desired shape. A new facility designed to meet current environmental and safety standards would adapt a simplified manufacturing process such as cast-in-place, in which melted plutonium is poured into a mold that is near the final product shape. Because most of the tests to date have been performed on alloys produced at Rocky Flats, scientists may need to begin systematic studies on those newly manufactured plutonium alloys to ensure that the new pits will age just as gracefully as the current ones.

The Livermore team will continue to assess the plutonium-239 alloy indefinitely, carefully monitoring the properties of naturally and artificially aged samples. Says Chung, "We plan to obtain data for as long as possible to give us still greater confidence."

—Arnie Heller

**Key Words:** accelerated aging, life-extension program, nuclear weapon, plutonium pit, Rocky Flats Plant, stockpile stewardship.

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# A New Detector for Analyzing NIF Experiments

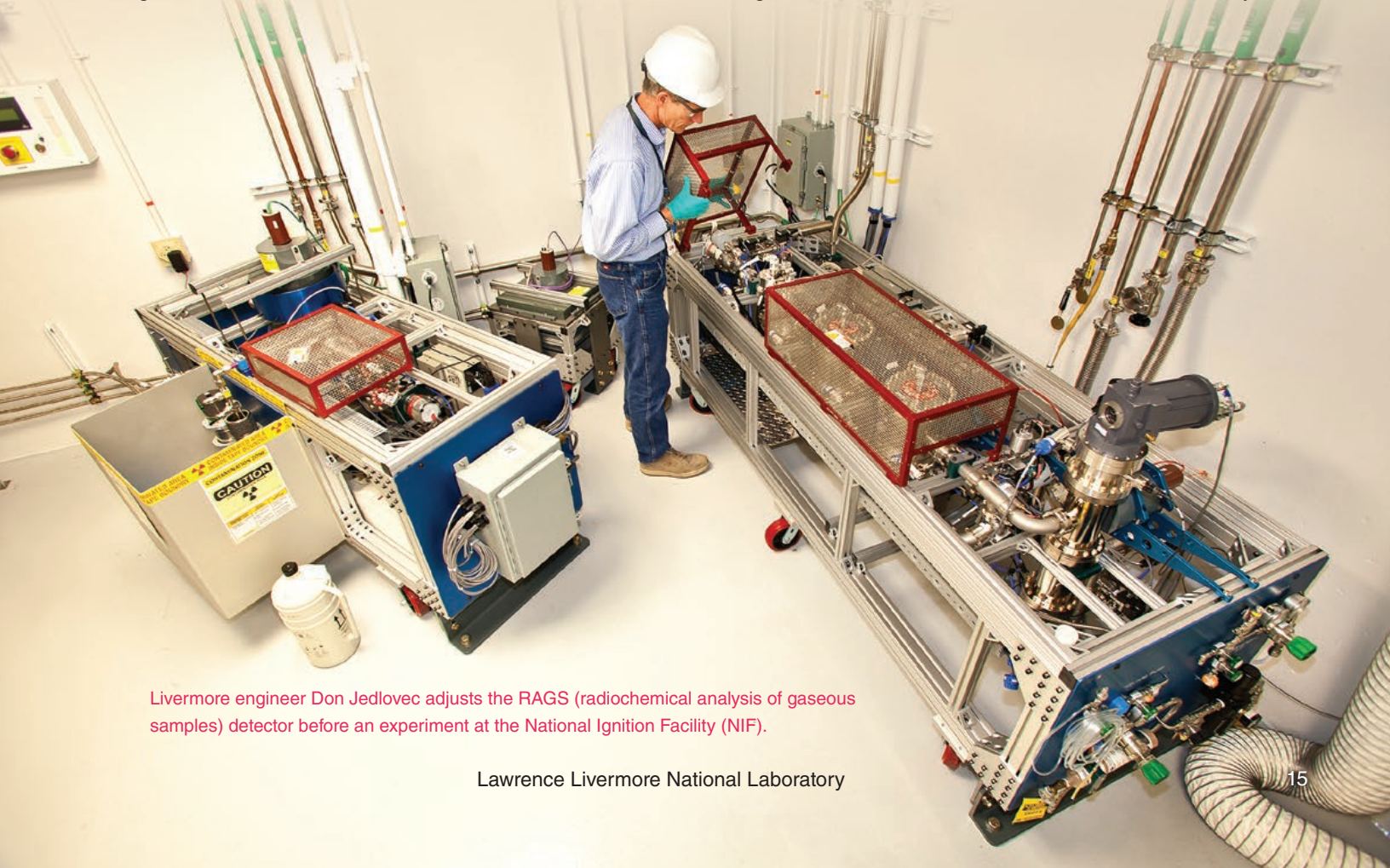
**W**HEN the National Ignition Facility's (NIF's) 192 laser beams deliver an enormous jolt of energy and power to a fusion fuel capsule, extremely sensitive, fast, and high-resolution instruments record the data needed to analyze an experiment. A new detector, called RAGS (radiochemical analysis of gaseous samples), collects and analyzes gases produced during an experiment to characterize the results with new data and in greater detail than was previously possible.

The principle behind RAGS is “doping,” or implanting, regions of the fuel capsule with a noble gas, a chemically nonreactive element such as xenon, krypton, or argon. During a NIF ignition experiment, atoms of tritium and deuterium (the heavy isotopes of hydrogen) fuse to form a helium nucleus—a reaction that has as a by-product a high-energy neutron. When an atom of a stable noble isotope absorbs an emitted fusion neutron, it transmutes

to a different gaseous radioactive isotope. The transmutation can be studied and provides a signature of the fusion reactions in the fuel capsule.

These kinds of nuclear reactions occur during the normal life cycle of stars, offering researchers information about our universe and its history. Livermore physicist Richard Fortner also notes that RAGS mimics radiochemical techniques used in underground nuclear tests before they were discontinued in 1992. In those tests, tracer elements were activated by neutrons generated in the nuclear detonation. Researchers then analyzed the newly synthesized isotopes to determine the temporal and spatial history of the device conditions. “If you want to know what’s going on in a particular location, tracer amounts of radioactive materials will tell you,” says Fortner, who was the Laboratory’s last associate director for nuclear testing.

RAGS joins the close to 60 optical, x-ray, neutron, and gamma-ray diagnostics on NIF designed to provide a virtually complete picture of the fleeting events that occur in an ignition experiment. (See *S&TR*, December 2010, pp. 12–18.) The new detector is one of several neutron diagnostics that together allow researchers to measure neutron yield and temperature, bang time (the interval from laser pulse initiation to maximum neutron emission), and reaction history so they can evaluate how well the target performed. According to Livermore radiation chemist Dawn Shaughnessy, RAGS is particularly useful because it provides information on two important areas for NIF scientists. The first is areal density, a



Livermore engineer Don Jedlovec adjusts the RAGS (radiochemical analysis of gaseous samples) detector before an experiment at the National Ignition Facility (NIF).

measure of the combined thickness and density of the imploding frozen fuel shell. The second is mix, a potentially undesirable condition during which spikes of the plastic rocket shell penetrate to the core of the hot fuel and cool it, decreasing the probability of igniting a sustained fusion reaction with energy gain.

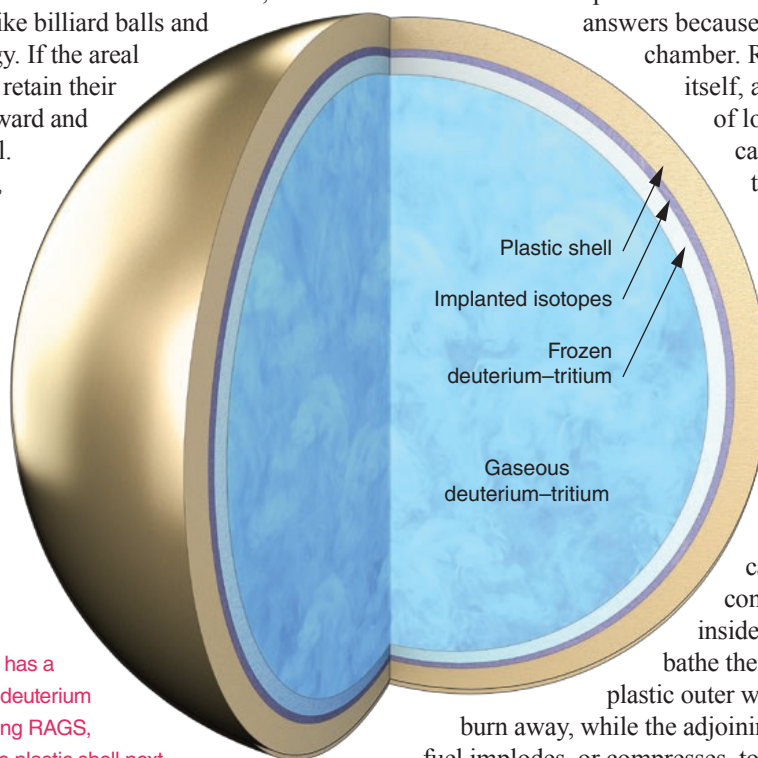
### Probing the Density of the Fuel Layer

Lee Bernstein, a physicist in Livermore's Physical and Life Sciences Directorate, says, "For ignition to occur, we must prepare the fusion fuel with a high enough areal density and temperature as well as a symmetric shape at the time of peak compression." Over the past year, NIF ignition experiments have demonstrated a steady increase in the areal density by improving the timing and shape of the laser pulse and the symmetry of the compressed fuel.

RAGS is key to accurately measuring the fuel's overall areal density, which has proven to be particularly challenging. When the hydrogen isotopes deuterium and tritium fuse, they generate energetic neutrons along with helium nuclei, which are often called alpha particles. If the areal density is high, many of these neutrons collide with the highly compressed deuterium–tritium fuel, scattering and bouncing around like billiard balls and losing some of their kinetic energy. If the areal density is too low, most neutrons retain their original energy as they travel outward and pass through the thin layer of fuel.

For experiments using RAGS, the innermost 5 to 6 micrometers of the plastic shell next to the frozen deuterium–tritium fuel layer is implanted with atoms of xenon-124. During compression, this isotope will undergo one of two possible transmutations, depending on the energy of the neutron. When a high-energy (about 14.1-megaelectronvolt) neutron is absorbed by an atom

A fuel capsule for ignition experiments has a plastic shell enclosing layers of frozen deuterium and tritium atoms. For experiments using RAGS, the innermost 5 to 6 micrometers of the plastic shell next to the frozen fuel is implanted with atoms of the stable noble gas xenon-124 isotope. When NIF laser beams strike the internal walls of the hohlraum, they are converted to x rays that irradiate the capsule inside. The plastic outer wall rapidly ablates, or burns away, while the adjoining fuel layer implodes and compresses the capsule core. (Rendering by Kwei-Yu Chu.)



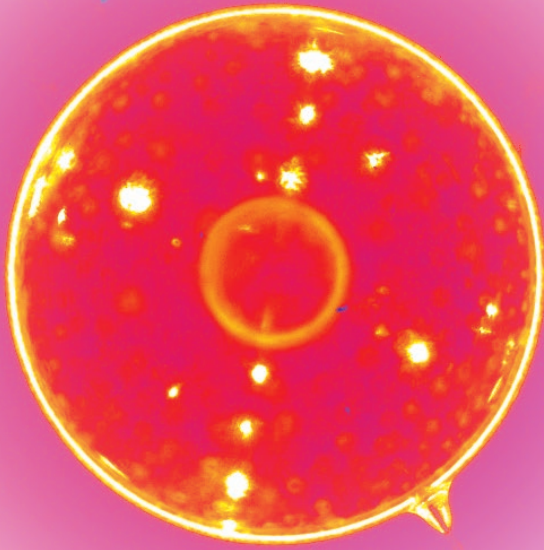
of xenon-124, the isotope immediately ejects two neutrons, yielding xenon-123 (which has a 2-hour half-life). When xenon-124 captures a lower-energy (less than 5-megaelectronvolt) neutron, it produces xenon-125 (with a 17-hour half-life).

RAGS collects the xenon gas produced by the experiment, separates xenon-123 and xenon-125, and cryogenically processes the gaseous species in several stages. The first stage removes water vapor, particulates, and reactive gases such as nitrogen, oxygen, and carbon dioxide. The noble gases flow through this precleaner and into the second stage, where a collector cryogenically fractionates (separates) the remaining gases on the basis of their vapor pressures under vacuum and at temperatures near 80 kelvins. Radioactive samples are then transferred to the Laboratory's Nuclear Counting Facility, which measures the gamma rays emitted as the gases decay. The ratio of the two isotopes gives researchers a quantity that is related to the capsule's areal density.

"We want a large ratio of xenon-125 to xenon-123 because that indicates a large areal density," says Shaughnessy. "Because our tracer is inside the capsule, we gain an average areal density from the entire capsule. Some diagnostics appear to give different answers because of their position around the chamber. RAGS is insensitive to the chamber itself, and its performance is independent of location or line of sight into the capsule. In addition, RAGS collects the postshot gas load, so it does not compete for real estate with other diagnostic instruments inside the chamber."

### Measuring Mix

Researchers at NIF also plan to use RAGS to determine the extent of fuel–ablator mix, a critical measure of an experiment's performance. The ablator is the plastic outer layer of the fuel capsule. In ignition experiments, converging laser beams illuminate the inside of the hohlraum, creating x rays that bathe the fuel capsule inside. The capsule's plastic outer wall is designed to rapidly ablate, or burn away, while the adjoining layer of frozen deuterium–tritium fuel implodes, or compresses, to extraordinarily high temperatures, pressures, and density. Mix occurs when spikes of the plastic ablator shell penetrate the hot burning fuel, thereby cooling it and lowering fusion performance overall. "When ablator material gets into the center of the burning fuel, it is equivalent to dropping ice cubes into a hot drink," says Bernstein.



Evaluating the extent of mix is difficult because it occurs over a very small area. RAGS offers an elegant way to measure mix because of its sensitivity. In addition, it can reveal the distance over which mix occurs.

Determining how much of the shell penetrates the center depends on detecting certain nuclear reactions. For these measurements, the intersection of the fuel layer and the ablator shell is doped with atoms of iodine-127. A mixture of iodine-127 plus a molecule of deuterium yields xenon-127 plus two neutrons. A high concentration of xenon-127 would thus indicate an undesirable level of ablator–fuel mix.

### **Conceived at Livermore, Built at Sandia**

Several researchers at Lawrence Livermore conceived of the RAGS detector. Physicist Wolfgang Stoeffl worked out the operating design, and Allen Riddle from Sandia National Laboratories (now at Livermore) created the final design. The instrument was built at Sandia’s New Mexico site and shipped to Livermore for installation.

In February 2012, RAGS was commissioned during an exploding pusher shot, in which NIF lasers fired 375 kilojoules of energy directly into a microballoon—a 2.1-millimeter-diameter spherical glass shell. The microballoon (shown above) was filled with a 50/50 mixture of deuterium and tritium and a small amount of stable xenon-124. RAGS successfully measured the xenon-123 and xenon-125 created in the implosion.

Shaughnessy notes that data from RAGS complement other NIF instruments such as the neutron time-of-flight (nTOF) detector, which records the neutron energy spectrum, fuel temperature, bang time, and areal density. The nTOF diagnostic measures the energy

This microballoon, only 2.1 millimeters in diameter, was used in the exploding pusher shot to commission the RAGS detector. The spherical glass shell was filled with a 50/50 mixture of deuterium and tritium and a small amount of stable xenon-124. RAGS successfully measured the xenon-123 and xenon-125 that were generated when NIF’s laser beams hit the target.

of the neutron signal based on the time a neutron originates in the capsule to when it arrives at the detector. The travel time is a function of temperature, which is directly related to how fast the fuel capsule implodes. Temperature is critical because, without the right temperature, ignition will not occur.

Three nTOF detectors are installed outside the NIF target chamber at different locations, each about 20 meters from the target. The instruments were calibrated on the OMEGA laser, a 30-kilojoule, 60-beam system operated by the University of Rochester’s Laboratory for Laser Energetics. Although nTOF instruments help scientists determine areal density, each one has a limited line of sight. Physicist Jim Knauer from the University of Rochester is combining data from the three instruments to map neutron scattering within the target chamber and determine the detectors’ uniformity.

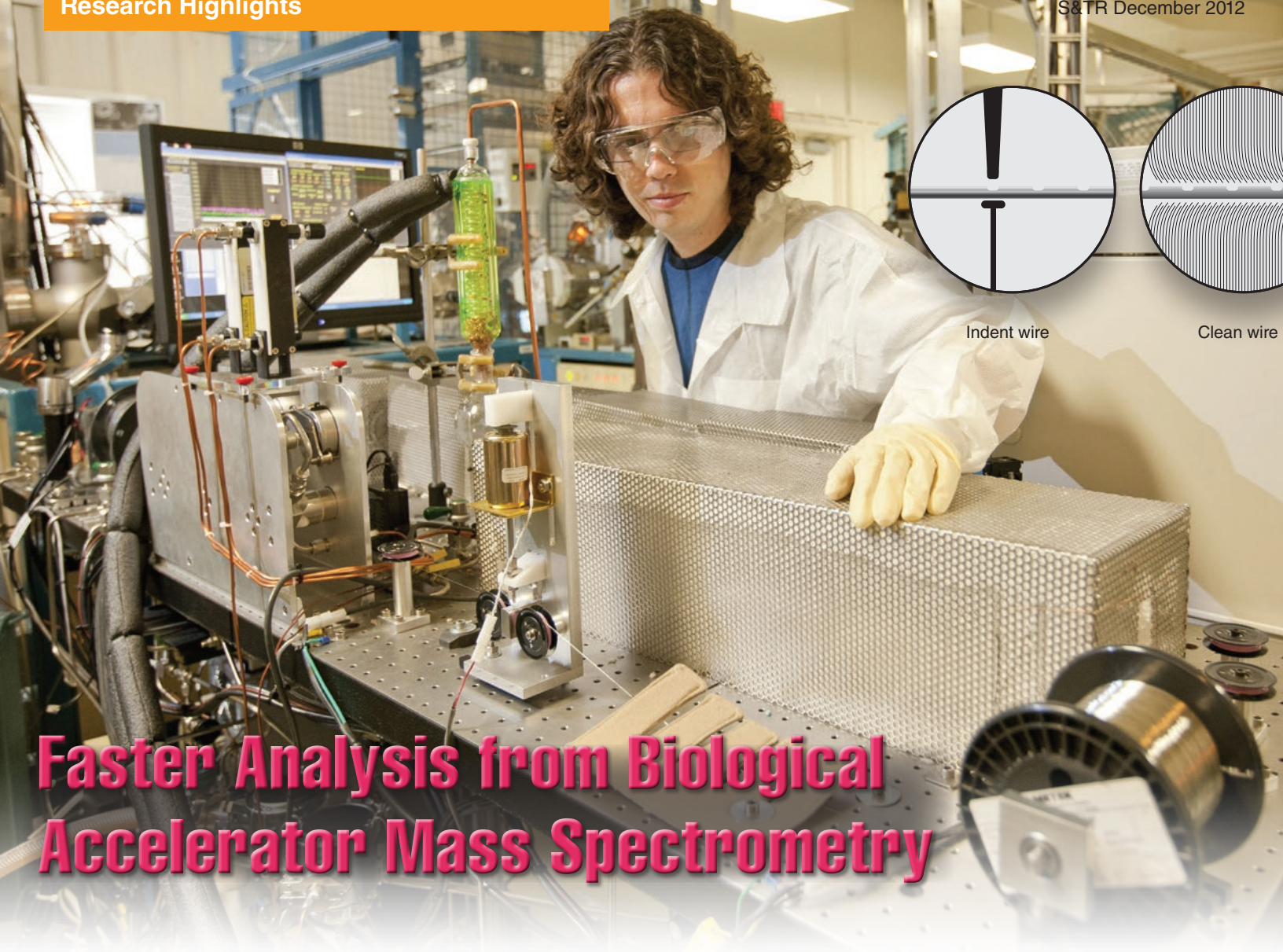
Another device, the neutron activation diagnostic, uses zirconium metal foils, each about 7 centimeters in diameter, positioned on 17 ports around the target chamber to measure the relative distribution of neutrons produced in an ignition experiment. During a shot, neutrons hit and activate the metal foils. The foils are removed and transferred to the Livermore Nuclear Counting Facility, where gamma rays are recorded as the reacted nuclei decay. Using this information, physicist Darren Bleuel and postdoctoral researcher Charles Yeamans create fuel density maps to determine the symmetry of the imploding fuel layer and thereby help physicists plan future experiments.

Combining RAGS data with those from other neutron-sensitive diagnostics provides an in-depth picture of performance within the fusion environment. Together, the NIF diagnostics are yielding unprecedented data about ignition experiments. “With RAGS, we have a new view into the fusion reactions occurring inside the capsule,” says Shaughnessy, “and we can obtain information about the extent of mix from the imploding shell into the burn region.”

—Arnie Heller

**Key Words:** alpha particle, areal density, ignition, iodine-127, National Ignition Facility (NIF), mix, neutron time-of-flight (nTOF) detector, radiochemical analysis of gaseous samples (RAGS) detector, stable noble gas, xenon-124.

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## Faster Analysis from Biological Accelerator Mass Spectrometry

**T**HE search for a new drug—whether to treat cancer or a flu pandemic—is time consuming and costly, requiring up to 15 years and hundreds of millions of dollars to turn an idea into an effective product. Tom Baillie, the dean of pharmacy at the University of Washington, adds that “Just one of the 10,000 molecules studied early on by chemists will make the grade. There are lots of expensive failures along the way.”

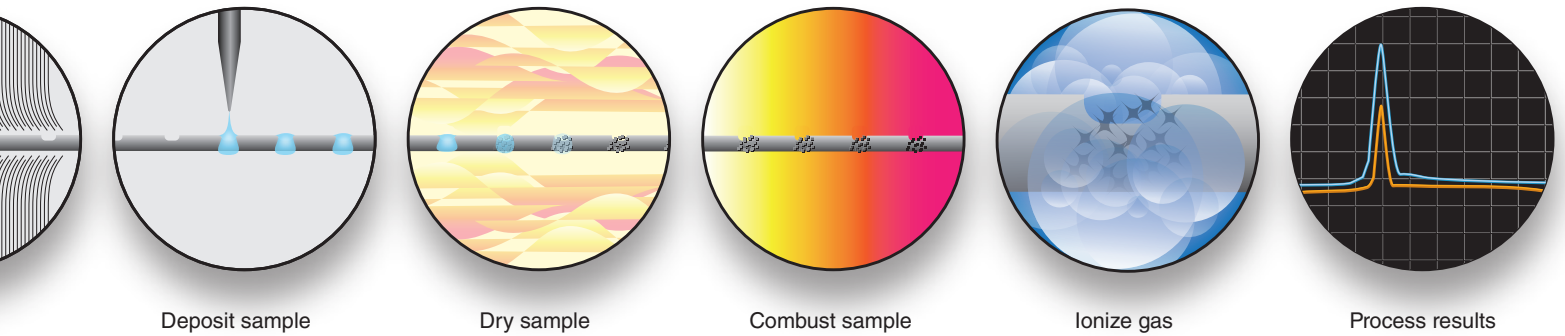
High-throughput screening techniques and combinatorial chemistry play an important role in the initial screening process for new medications. Another approach that holds great promise for drug discovery is accelerator mass spectrometry (AMS), an extremely accurate method for dating bones, tree rings, ice cores, and other carbon-bearing materials.

AMS uses an accelerator to count the carbon-14 atoms in a sample. It is sensitive enough to find one carbon-14 isotope among a quadrillion other carbon atoms. With that number, researchers

Avi Thomas works with a biological accelerator mass spectrometry (bioAMS) instrument at Livermore. The system includes a high-performance liquid chromatograph and a moving-wire interface, which allow researchers to directly study liquid samples.

can quickly and accurately deduce a sample’s age because the carbon-14 level in the sample will mirror the isotope’s level in the atmosphere at the time the material was created.

In the late 1980s, Livermore scientists were the first to apply the sensitivity of AMS to biological testing. In biological AMS (bioAMS), a substance to be studied is tagged with carbon-14 or another radioactive isotope and ingested or absorbed by a test subject. In the hours, days, or weeks that follow, the carbon-14 isotope will show up in the subject’s DNA, blood, urine, or tissue. Over the last 25 years, bioAMS research has revealed how animals



BioAMS relies on a moving-wire interface to transport liquids to the accelerator. A tiny wire is indented at regular intervals and then cleaned. When liquid droplets from the high-performance liquid chromatograph land on the wire, they are attracted to the indentations. Solvent in the sample is then removed through evaporation, and the remaining material is reduced to carbon dioxide and ionized. This gas is transferred to the accelerator, which measures the ratio of carbon-14 to either carbon-13 or carbon-12 in the material.

and humans metabolize carcinogens, vitamins, and toxins. (See the box on p. 20.)

AMS is unique in that it can measure extremely low concentrations of substances with a high level of accuracy. To study how humans respond to an existing or candidate medication, Lawrence Livermore researchers and medical collaborators developed a technique called microdosing, in which a patient takes just 1/100 of a normal therapeutic dose. The drug's "fate" in the patient's body can easily be measured and studied.

Baillie, who served as vice president of drug metabolism and pharmacokinetics at Merck Research Laboratories before moving to the University of Washington, looks forward to having AMS readily available for human trials required in the drug-discovery process. He estimates that researchers could shave at least a few months off the development timetable by using the technique.

He is not alone in his enthusiasm. "The pharmaceutical and medical research communities want to apply bioAMS in a routine, nonresearch fashion," says Ken Turteltaub, who leads the Laboratory's bioAMS program. "But the instrumentation must be a lot simpler and easier for nonexperts to use."

### Faster, Cheaper, and Easier

The traditional AMS technology requires that all sample material be reduced to solid graphite pellets. This step involves physicists and experts in sample preparation, precise chemistries, and days of work—all factors that make bioAMS expensive and severely limit its utility in a nonresearch setting.

Livermore scientist Ted Ognibene is working with Avi Thomas, a Lawrence scholar and physics doctoral student at the University of California (UC) at Davis, to develop a sample-preparation method that accommodates liquid samples and bypasses the graphitization process. Their technique rapidly converts the carbon content of liquid samples to carbon dioxide and transports the gas to an ion source, where it is ionized before it enters the accelerator. With the new process, samples can be much smaller—nanograms

instead of micrograms—and AMS results are available in minutes. Thomas's dissertation explains the process in detail.

A commercial high-performance liquid chromatography (HPLC) unit allows for the use of liquid samples. This separation and analysis tool is found in most biology and medical laboratories. In fact, when athletes are tested for substance abuse, HPLC units process their urine, separating each sample into individual components for analysis.

The innovation that transports liquid to an accelerator is a moving-wire interface. Moving-wire interfaces have been adapted for some applications but have not previously been used with AMS. A tiny wire, indented at regular intervals, accepts the HPLC output—a nonvolatile sample material dissolved in liquid. The surface area of the precisely made indentations attracts the liquid, ensuring that samples are exactly the same size. The wire proceeds to a drying oven to evaporate the solvent in the sample. It then moves to a combustion oven, where the sample's carbon content is converted to carbon dioxide gas. (See the movie at [str.llnl.gov/Dec12/images/bioams.wmv](http://str.llnl.gov/Dec12/images/bioams.wmv).)

AMS quantifies the carbon isotopes by separating the ions derived from a sample and identifying their nuclear charge and mass. The amount of carbon-14 is measured relative to the more abundant carbon-13 or carbon-12 isotopes in the ion beam. These separation and measurement steps follow the traditional AMS process. What has changed dramatically is the time required. With standard AMS, researchers typically had to wait days or sometimes weeks to obtain test results. In the new bioAMS process, from the moment a sample is deposited on the wire until its carbon ratio is measured, a mere 90 seconds have elapsed.

In addition, traditional AMS often required the entire sample for a single measurement. Says Thomas, "With our changes, scientists will likely have material left over after the bioAMS analysis. If the experiment needs to be tweaked or repeated for some reason, they will have something left to examine. And with the fast turnaround, the experiment can be run again almost immediately."

## Biological Accelerator Mass Spectrometry at Livermore

In 1988, Livermore scientists used accelerator mass spectrometry (AMS) to determine how low doses of a suspected carcinogen affect the DNA of mice. AMS achieved a tenfold improvement in detecting damaged DNA over the best methods then available.

In the years since, researchers at the Center for Accelerator Mass Spectrometry have expanded AMS for biomedical and pharmaceutical applications. The center has become the world leader in biological AMS research, and in 1998, the National Institutes of Health (NIH) named it the first NIH Research Resource for Biological AMS. Researchers from Livermore and institutions worldwide have applied AMS to answer questions in fundamental biology, metabolism, nutrition, toxicology, pharmacology, and more recently drug development.

The traditional AMS processes used to perform these studies have been relatively expensive and time consuming. The current work at Livermore is facilitating the use of smaller samples and reducing the time required for sample preparation and processing. These steps will save money and offer fast results to researchers, moving biological AMS one step closer to routine laboratory use.

Mass Spectrometry, physicists will help get the new system running properly, and the center's team will be on call for troubleshooting the initial experiments. The goal, however, is for biomedical researchers to use the instrument as they would any other piece of laboratory equipment, which will make the bioAMS unit different from other AMS systems.

Ralph deVere White, an M.D. and director of the UC Davis Comprehensive Cancer Center, has been a bioAMS booster for years. "Not only can AMS and microdosing be used to discover new drugs, but we are also using them right now for clinical trials with an existing chemotherapy drug for bladder cancer," says deVere White, a long-time collaborator with Turteltaub and others at Livermore. "Patients respond differently to medications, and we are trying to predict who will respond to the chemotherapy treatment and who won't. Thanks to bioAMS, we may be able to make better use of a drug we already have."

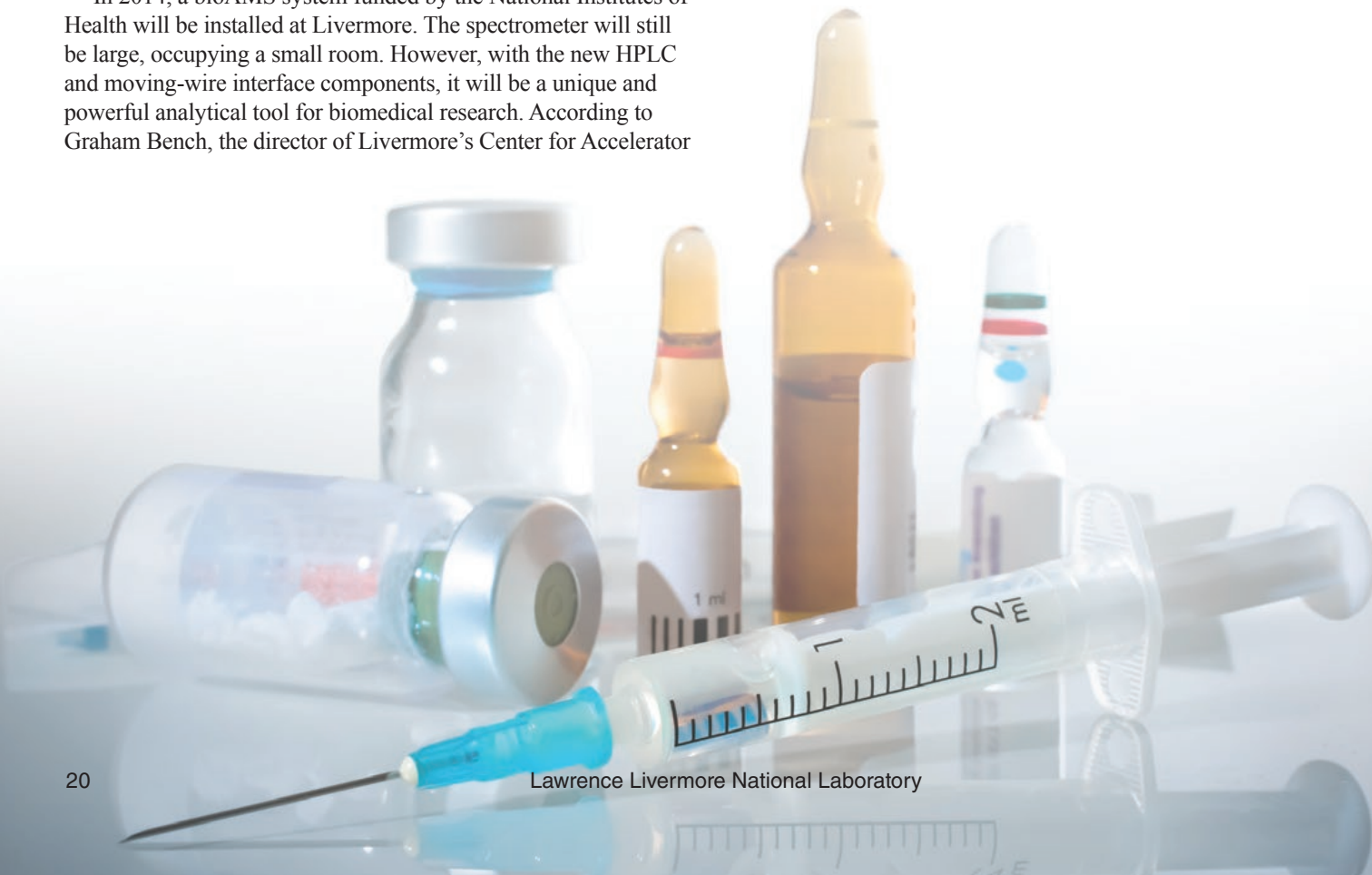
—Katie Walter

**Key Words:** biological accelerator mass spectrometry (bioAMS), Center for Accelerator Mass Spectrometry, drug discovery, high-performance liquid chromatography (HPLC), moving-wire interface, pharmaceutical research.

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### AMS in a Biology Lab

In 2014, a bioAMS system funded by the National Institutes of Health will be installed at Livermore. The spectrometer will still be large, occupying a small room. However, with the new HPLC and moving-wire interface components, it will be a unique and powerful analytical tool for biomedical research. According to Graham Bench, the director of Livermore's Center for Accelerator



*In this section, we list recent patents issued to and 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

### **Solution-Grown Crystals for Neutron Radiation Detectors, and Methods of Solution Growth**

**Natalia P. Zaitseva, Giulia Hull, Nerine J. Cherepy, Stephen A. Payne, Wolfgang Stoeffl**

U.S. Patent 8,207,507 B2

June 26, 2012

In one version of this method, an organic crystal is grown from solution so that it exhibits a signal response signature for neutrons from a radioactive source. When the crystal, which has the physical characteristics of formation from solution, exhibits the signal response for neutrons emitted by a radioactive source, a photodetector detects the signature. In another setup, the organic crystal grown from solution is large enough to exhibit a detectable signal response signature for neutrons from a radioactive source. This organic crystal has a length of greater than about 1 millimeter in one dimension.

### **Model-Based Tomographic Reconstruction**

**David H. Chambers, Sean K. Lehman, Dennis M. Goodman**

U.S. Patent 8,207,886 B2

June 26, 2012

A model-based approach to estimating wall positions for a building is developed and tested using simulated data. This approach combines two techniques from geophysical inversion problems, layer stripping and stacking, with a model-based estimation algorithm that minimizes the mean-square error between the predicted signal and the data. The technique is designed to process multiple looks from an ultrawideband radar array. The processed signal is time-gated, so that each section will detect the presence of a wall and estimate its position, thickness, and material parameters. A building's floor plan is determined by moving the array around the building exterior. The steps for combining the stacking and layer-stripping algorithms are described, and results from a simple numerical example with three parallel walls are shown.

### **Laser Diode Package with Enhanced Cooling**

**Robert J. Deri, Jack Kotovsky, Christopher M. Spadaccini**

U.S. Patents 8,208,508 B2 and 8,208,509 B2

June 26, 2012

A laser diode package assembly includes a reservoir filled with a fusible metal in close proximity to a laser diode. When the laser is operated, the fusible metal absorbs heat from the laser diode and undergoes a phase change from solid to liquid. The metal absorbs heat during the phase transition. Once the laser diode is turned off, the liquid metal cools and resolidifies. The reservoir is designed so that the metal remains in the reservoir even when in a liquid state. The laser diode assembly also includes a lid with one or more fin structures that extend into the reservoir and are in contact with the metal in the reservoir.

### **Preparation of Membranes Using Solvent-Less Vapor Deposition Followed by In-Situ Polymerization**

**Kevin C. O'Brien, Stephan A. Letts, Christopher M. Spadaccini, Jeffrey D. Morse, Steven R. Buckley, Larry E. Fischer, Keith B. Wilson**

U.S. Patent 8,211,499 B2

July 3, 2012

This system for fabricating a composite membrane from a membrane substrate uses solventless vapor deposition followed by in situ

polymerization. Two monomers are directed into the mixing chamber inside a deposition chamber to provide a mixture of the two. The mixed monomers are then deposited via solventless vapor deposition onto the membrane substrate in the deposition chamber. The membrane substrate and the mixed monomers are heated to induce in situ polymerization, which produces the composite membrane.

### **Hazardous Particle Binder, Coagulant and Re-Aerosolization Inhibitor**

**Paula Krauter, David Zalk, D. Mark Hoffman**

U.S. Patent 8,216,965 B2

July 10, 2012

This copolymer and water-ethanol solvent solution binds with airborne contaminants or potential airborne contaminants, such as biological weapon agents or toxic particulates. The solution coagulates as the solvent evaporates, adhering the contaminants to a surface and inhibiting their resuspension. The solution uses a water or ethanol-water mixture for the solvent and a copolymer with one of several functional group sets so that its physical and chemical characteristics include high adhesion, low viscosity, low surface tension, negative electrostatic charge, substantially neutral pH, and a low dissociation constant (pKa). Use of the copolymer solution prevents reaerosolization and transport of unwanted, reactive species, thus increasing health and safety for personnel charged with decontaminating buildings and areas.

### **Ponderomotive Phase Plate for Transmission Electron Microscopes**

**Bryan W. Reed**

U.S. Patent 8,217,352 B2

July 10, 2012

This ponderomotive phase-plate system produces highly tunable phase-contrast transfer functions in a transmission electron microscope for high-resolution and biological-phase contrast imaging. The system includes a laser source and a beam transport system to produce a focused laser crossover as a phase plate. The ponderomotive potential of this crossover induces a scattering-angle-dependent phase shift in the electrons of the post-sample electron beam, which corresponds to a desired phase-contrast transfer function.

### **Filter Casting Nanoscale Porous Materials**

**Joel Ryan Hayes, Gregory Walker Nyce, Joshua David Kuntz**

U.S. Patent 8,226,861 B2

July 24, 2012

A method for producing nanoporous material involves combining a liquid with nanoparticles to produce a slurry. Removing the liquid from the slurry produces a monolith.

## Awards

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**Carolyn Hall**, a microbiologist in Livermore's Biodefense Knowledge Center, was selected as a **2012–2013 Fellow** of the **Emerging Leaders in Biosecurity Initiative**. A competitive program sponsored by the Center for Biosecurity at the University of Pittsburgh Medical Center, the initiative is designed to create and sustain a multidisciplinary and intergenerational biosecurity community of young professionals and current leaders. Hall joined the Laboratory in 2009, after earning a Ph.D. in microbiology and immunology from Stanford University.

Postdoctoral researcher **Dina Weilhammer** in the Physical and Life Sciences Directorate is one of ten **Public Policy Fellows** chosen by the **American Association of Immunologists (AAI)** for 2012–2013. AAI is dedicated to advancing the knowledge of immunology and related disciplines as well as addressing the potential integration of immunologic principles into clinical practice. Weilhammer, who joined the Laboratory in 2011, received a Ph.D. in molecular cell biology from the University of California at Berkeley. She is part of a team developing a treatment that would activate a person's immune system to protect against bacterial and viral pathogens.

**Stephen Klein**, a climate scientist working in the Program for Climate Model Diagnosis and Intercomparison, received the **Ascent Award** from the **Atmospheric Sciences Section** of the **American Geophysical Union (AGU)**. The award recognizes Klein's research "elucidating the role of clouds in climate change and the fidelity with which climate models simulate clouds." AGU created the Ascent Award this year to acknowledge outstanding midcareer scientists in atmospheric and climate sciences.

The **American Nuclear Society** honored **Susana Reyes** with the **2012 Mary Jane Oestmann Professional Women's Achievement Award**. A nuclear engineer working at the National Ignition Facility (NIF), Reyes was recognized for her "leadership in developing detailed hazard and safety analyses for both inertial and magnetic fusion facilities, including NIF and ITER, and future power reactors." The award is given annually for outstanding personal dedication and technical achievement by a woman in the fields of nuclear science, engineering, research, or education.

The **Federal Laboratory Consortium's Far West Region** honored three teams of Livermore researchers and their collaborators with technology transfer awards.

An **Outstanding Partnership Award** recognized IntelliProbe, an optical breast cancer diagnostic system that can provide immediate cancer diagnoses and may eliminate or substantially reduce the need for biopsies. The IntelliProbe project included

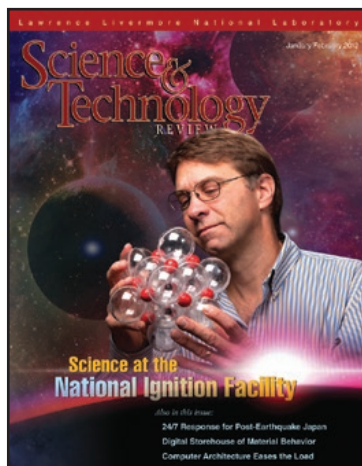
scientists from the Russian Federal Nuclear Center in Sarov and the Livermore-based company BioTelligent, Inc., as well as Laboratory physicist **Alexander Rubenchik**, program manager **Paris Althouse**, and business development executive **Annemarie Meike**. The project was managed by Global Initiatives for Proliferation Prevention, a National Nuclear Security Administration program that teams weapons scientists, technicians, and engineers from former Soviet states with U.S. national laboratories and industrial partners to work on high-technology commercial research and development projects.

The dynamic transmission electron microscope (DTEM), a Livermore technology licensed to Integrated Dynamic Electron Solutions (IDES) in Belmont, California, won an **Outstanding Commercialization Award**. DTEM captures images at 15-nanosecond (15-billionths-of-a-second) intervals to reveal micrometer- and nanometer-scale details of material processes. Livermore physicists **Thomas LaGrange** and **Bryan Reed** led the DTEM development. **Ida Shum**, a business development executive in the Industrial Partnerships Office, coordinated the commercialization effort. In 2011, after working with the Laboratory team to develop a prototype machine, IDES sold and installed a DTEM at École Polytechnique Fédérale de Lausanne in Switzerland.

The Innovation Hub Initiative established by i-GATE (Innovation for Green Advanced Transportation Excellence) was honored with an **Outstanding Partnership Award**. I-GATE is a regional public-private partnership that supports small businesses and bolsters green transportation and clean-energy efforts. Award recipients include **Buck Koonce**, Lawrence Livermore's director of economic development; i-GATE program leader **Bruce Balfour** of Sandia National Laboratories; **Rob White**, economic development director for the city of Livermore; and **Louis Stewart** of the California Governor's Office of Business and Economic Development.

The **Focused Advanced Persistent Threat (FAPT) Group**, a multilaboratory collaboration led by Livermore scientist **Matt Myrick**, received a **2012 National Cybersecurity Innovation Award** from the **SANS Institute**. The group was recognized for developing the Master Block List (MBL), a service and data-aggregation tool that creates filters to block cyber attacks. MBL allows Department of Energy laboratories and plants to share information in real time on domain names that are known or suspected to be untrustworthy. In addition to Lawrence Livermore, FAPT member organizations include Sandia, Los Alamos, Pacific Northwest, Argonne, and Idaho national laboratories; the Kansas City and Pantex plants; National Renewable Energy Laboratory; and the MOX Fuel Fabrication Facility.





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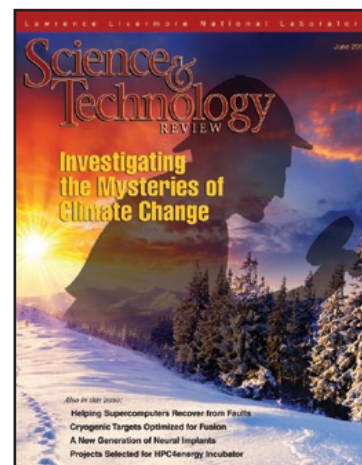


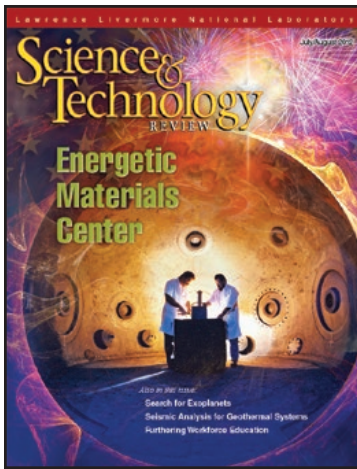
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## Positively Scintillating Neutral Particles Brighten Scientific Prospects

In collaboration with academic and research institutions, Laboratory physicists are building ultrasensitive detectors that can identify rare neutral particle interactions. In one effort, the detectors precisely track the number and flux of antineutrinos emitted by nuclear reactors. A water-based detector shows great promise for long-range monitoring, potentially up to hundreds of kilometers from a reactor core. As part of the Double Chooz experiment in France, Livermore researchers simulated reactor outputs to confirm neutrino oscillation parameters. Laboratory physicists also helped develop the Large Underground Xenon (LUX) detector, built to detect dark-matter particles for the first time. The LUX technology serves as the basis for a compact argon-based detector designed to measure coherent neutrino nucleus scattering, a yet unconfirmed interaction predicted by the Standard Model of particle physics.

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# The Data Science Challenge



Innovative algorithms and computational methods developed at the Laboratory help researchers deal with data overload in fields as diverse as bioinformatics, cyber-surveillance, and climate science.

### *Also in January/February*

- *First experiments at the Linac Coherent Light Source have revealed unexpectedly fast effects in graphite.*
- *A new flow-through electrode for capacitive desalination produces freshwater faster than conventional methods and uses very little energy.*
- *Diffraction optics experts at Livermore are partnering with industry to create flexible, efficient lenses for a space telescope prototype.*

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