Under extreme pressure and strain, even everyday materials can reveal unusual behavior.
SQUEEZE a material hard enough, and its structure and properties will change, sometimes dramatically so. With enough heat and pressure, scientists can turn pencil lead (graphite), one of Earth’s softest materials, into diamond, one of its hardest. Apply even more pressure—such as might be found in explosions, detonating nuclear weapons, laser fusion experiments, meteorite impacts, or the hearts of stars and planets—and materials can take stranger forms. Deep in Jupiter’s core, for instance, where pressures likely reach 50 to 100 million times that of Earth’s atmosphere, hydrogen is predicted to be a metallic liquid rather than the familiar transparent gas.

Such conditions are difficult to imagine, let alone re-create and study in the laboratory. “States of matter reached during implosions and explosions are very extreme,” Livermore physicist Gilbert “Rip” Collins notes. “For example, to achieve fusion ignition, we’re trying to compress hydrogen from one-fifth of the density of water to 100 times the density of lead. A better understanding of the properties of matter at states relevant to ignition, weapons, planets, and stars will allow us to develop better predictive capabilities.” More-accurate and -comprehensive material models will help Livermore scientists and engineers to successfully meet formidable challenges in energy and national security missions and to advance basic science research.

One approach Livermore researchers use to shed light on the exotic phases and behavior of otherwise commonplace compounds and elements is to perform dynamic compression experiments. Unlike static compression platforms, such as diamond anvil cells, which compress materials gradually and can maintain high pressures for long durations (seconds to

(left to right) Joe Zaug, Jonathan Crowhurst, Harry Radousky, and Mike Armstrong examine Livermore’s compact ultrafast laser system, which is used for dynamic compression experiments. (Photograph by Lanie L. Rivera.)
days), dynamic compression experiments typically use lasers or gas guns to generate shock waves that rapidly raise temperature, pressure, and density for a fleeting amount of time (microseconds to nanoseconds). Different dynamic compression platforms yield different but complementary information. Lasers can access the highest pressures and temperatures. Gas guns, which achieve more moderate pressures, can provide especially accurate measurements and longer experimental timescales. Another laser platform, the ultrafast laser, yields the highest strain rates (the fastest compressions) of the three. Experimental techniques such as diamond anvil–based precompression and ramped compression further expand the realm of feasible pressure and temperature combinations, allowing researchers to reproduce the relatively cool, high-pressure conditions of planetary interiors, for instance. (See S&TR, July 2013, pp. 19–21; and March 2007, pp. 23–25.)

At Lawrence Livermore, the wide array of dynamic compression platforms and techniques available to researchers enables studies of a material across a vast range of temperatures, pressures, strain rates, and densities. The different approaches span seven orders of magnitude in strain rate, for example. Among the many dynamic compression research efforts under way at Livermore are hydrogen experiments at the National Ignition Facility (NIF), gas-gun studies on mantle minerals and aggregate materials, and explosives research using the Laboratory’s ultrafast laser system. Furthermore, multiple techniques can be used together to create a more complete picture of a material. Livermore researchers are also driving the field forward with novel experimental investigations and by marshalling new diagnostics and more powerful platforms.

**Under Pressure**

Laser-initiated dynamic compression experiments at facilities such as the Omega Laser Facility at the University of Rochester and Livermore’s NIF have attained the highest pressures and temperatures of any dynamic compression technique, enabling the study of entirely new physics regimes. (See S&TR, July/August 2012, pp. 4–11.) Collins, who leads Livermore’s Center for High Energy Density Science, says, “Right now, at NIF, we’re starting to explore the behavior of solids at terapascals of pressure with experiments that map thermodynamic variables, diffraction to determine structure, techniques to map optical properties, and soon other capabilities standard to condensed-matter laboratories.” Collins adds, “Before this work, researchers were limited to exploring maximum pressures of a few hundred gigapascals. We’re just beginning to understand how materials behave at such high pressures. In particular, we’re trying to understand how quantum mechanical laws manifest themselves at extreme conditions relevant to planetary formation and evolution.”
Our solar system’s gas giants, such as Jupiter and Saturn, and many of the extrasolar planets discovered thus far are composed largely of hydrogen and helium. Scientists have long posited that deep inside these planets, the high temperatures and pressures render hydrogen metallic and cause it to separate from helium, much like water and oil do. If so, the heavier helium might sink and release energy, which would explain, for instance, Saturn’s unexpectedly high luminosity (total energy emitted per unit time). However, high-pressure hydrogen experiments and models have produced conflicting results, particularly regarding the plasma phase transition—a sudden transformation from a molecular insulating fluid to a metallic atomic fluid at around 100 to 300 gigapascals and at a relatively cool 1,000 to 2,500 kelvin.

In September at NIF, Marius Millot, Peter Celliers, and several other Livermore researchers looked for signs of this transition along with colleagues from the Carnegie Institution for Science, the French Alternative Energies and Atomic Energy Commission, the University of Edinburgh, and the University of California at Berkeley. The giant NIF laser launched a series of small shocks of increasing strength at a cryogenic hydrogen liquid sample. Each shock was allowed to bounce forward and back through the sample before the next hit, keeping the sample temperature lower than would hitting it with one large shock, better reproducing planetary conditions. “Part of what makes these experiments possible is NIF’s pulse shaping—the ability to tune and vary the laser power during a single pulse, rather than delivering a consistent level of power,” says physicist Millot, who led the campaign, which was supported by Livermore’s Laboratory Directed Research and Development (LDRD) Program. “Omega is limited to 4-nanosecond pulse shapes. However, on NIF we can use a highly accurate 35-nanosecond pulse shape, which gives us more flexibility.” Preliminary results show dramatic changes in the optical properties of dense hydrogen that are suggestive of a phase transition, although precise determination of the pressure and temperature at which this transition occurs will require a deeper dive into the data. The team intends to compare the new data with experimental results from diamond anvil cells, the Omega laser, and the Z Pulsed Power Facility at Sandia National Laboratories, as well as with existing models, to determine the nature of the unusual behavior of hydrogen around this metallic transition. (See S&TR, September 2015, pp. 4–11.) Ultimately, the new data will contribute to researchers’ understanding of planetary evolution and the ultradense hydrogen phase diagram.

Meanwhile, a “microphysics” experimental campaign in support of
fusion ignition will continue the study of high-pressure hydrogen. Livermore researchers will be measuring the microscopic properties of ignition-relevant materials on NIF. “It is amazing that NIF can explore compression paths leading toward ignition and the fundamental properties associated with both the molecular-to-atomic and insulator-to-metal transitions in hydrogen,” observes Collins.

**Impactful Research**

Gas gun–launched impactors are the most uniform method of compression, delivering a shock velocity tunable to within a margin of error of less than half a percent. This high level of accuracy makes gas guns, particularly those at the Joint Actinide Shock Physics Experimental Research Facility in Nevada, the platform of choice for measuring plutonium and many other materials relevant to weapons. (See *S&T*, April/May 2013, pp. 20–23.) In addition, gas guns can accommodate relatively large (tens of millimeters in diameter) samples and relatively long (microseconds in duration) experiments and are therefore especially useful for studying material strength and other mechanical properties.

Livermore’s High Explosives Applications Facility is home to a two-stage gas gun that can propel objects at up to 8 kilometers per second and produce pressures of up to 500 gigapascals. Now, researchers can also probe a material’s microstructure at the facility, thanks to an x-ray diffraction (XRD) capability installed there in late 2014. One of the first experiments to use the new diagnostic involved compression of forsterite, a magnesium-rich form of the mineral olivine, which is found in abundance in Earth’s Mantle and in meteorites and comets. Re-creating its transformation under lower-mantle pressure and temperature conditions could contribute to researchers’ understanding of mantle structure and will also provide a good test of the new XRD diagnostic.

“Forsterite is known to undergo a distinct high-pressure phase change,” says physicist Minta Akin. “Using our standard diagnostics, we can see that a transition happens from the volume change, but we didn’t know what type of transition it was. We suspected chemical decomposition, but to see the structural changes, we needed x-ray diffraction.” A comparison of diffraction patterns captured before and after applying 52 gigapascals of pressure showed a clear shift in crystal structure and orientation, as predicted.

X-ray diagnostics also play a pivotal role in a new project to study the dynamic response of granular materials at multiple strain rates, temperatures, and pressures. Granular materials—a broad category that encompasses additively manufactured
materials, foams, aggregates (such as concrete), and woven materials, such as Kevlar—do not necessarily behave the same way that a homogeneous mass of the same substance would. Dynamic compression experiments performed by other scientists on wet and dry sand, for instance, have shown that small differences in water content and sand morphology can produce dramatic and unexpected changes in shock velocity. Physicist Ricky Chau says, “A fundamental assumption regarding shock physics is that materials will have a uniform response. These experiments on heterogeneous materials suggest we need to change our assumptions.”

With LDRD Program funding, Akin, Chau, and colleagues will be performing gas-gun experiments on granular materials at Argonne National Laboratory. A successful proof-of-concept experiment compressed a set of micrometer-scale glass spheres, while a series of phase-contrast x-ray images recorded their response, including the fracturing and explosion of the spheres (see the box on p. 11).

“We’re trying to understand pressure and density effects on uniformly sized spheres before we move to irregular sizes and shapes, such as real sand or other complex aggregates,” notes Akin. Although still in its early stages, the project could eventually benefit research areas as diverse as seismology, explosives development, and additive manufacturing.

**Explosive Results**

Few data exist on the chemical reactions that occur in shocked materials. To fill this gap, Laboratory scientists developed a compact laser system to both generate shocks and quickly determine their speed and the resulting strain rates. (See *S&TR*, April/May 2012, pp. 17–19.) This capability enables them to monitor how a reaction occurs in real time, not simply observe the final results. One of the technique’s greatest strengths is that its short compression times—tens of picoseconds rather than the nanoseconds needed by most laser drives—allow compressed states to be reached with far less energy than in longer experiments.

Physicist Jonathan Crowhurst observes, “Because we can examine the dynamic response of materials on extremely short timescales, we can investigate phenomena that may occur too quickly to observe with other techniques.”

The reduced timescale also means that researchers can use very small material samples, which is helpful when working with explosives, a particular focus for Crowhurst and his colleagues. Physicist Mike Armstrong, who has led the group’s efforts, explains, “We’re particularly interested in initiation—what causes explosives to begin detonating. For instance, how will an explosive behave in a new situation? This information is useful for safety, modeling, and engineering purposes.”

A paucity of experimental data exists on shock-induced chemical ignition and the hydrodynamic and other processes leading to detonation. Moreover, the accuracy of relevant Laboratory molecular dynamics (MD) simulation codes cannot be effectively validated without measurements obtained on the same scale.
ultrafast timescales as MD. To directly address this gap, physical chemist Joe Zaug proposed a fast, tabletop-laser-based compression platform with ultrafast velocity measurement diagnostics. (See S&T, April/May 2012, pp 17–19). On the success of these diagnostics, Zaug states, “With our coordinated group efforts, we have been paying dividends to our national defense programs and to the wider scientific community.”

In a detonating explosive, the increase in pressure caused by the shock wave causes chemical reactions to occur and gas to be released at high speed, producing a self-sustaining reaction. That reaction can last for tens of nanoseconds to microseconds, but Livermore researchers have shown through an LDRD-funded effort that they can capture its key beginning stages with the ultrafast laser system.

Livermore researchers explored how hydrogen peroxide, a model reactive system, responds to a high-impact shock. “Hydrogen peroxide, composed of one oxygen–oxygen bond and two oxygen–hydrogen bonds in a hydrogen-bonding network, afforded us the opportunity to study a very complex process using a relatively simple molecular liquid,” says materials scientist Sorin Bastea, who led the hydrogen-peroxide research effort. The team used the ultrafast laser to blast a 1-micrometer-thick aluminum film in contact with a peroxide sample. Fifty picoseconds after the peroxide was shocked, it began to tear apart, and chemical bonds were completely broken by 100 picoseconds. During the experiment, pressure topped 20 gigapascals. “At the initiation threshold, we were able to directly observe a significant jump in the shock velocity, indicating that we had mechanically initiated chemical reactions in the sample,” Armstrong notes.

Another special feature of the ultrafast experiments is that the time and length...
scales involved are similar to those of MD simulations. Thus experimental data can be incorporated into existing models relatively easily, and experimental and simulation results can be readily compared. For this study, team members performed first-principles MD simulations of the chemical initiation and detonation of hydrogen peroxide and compared them to the experimental findings. They matched well. Furthermore, having both experimental data and theoretical predictions available enabled the team to calculate the amount of chemical reaction observed in the experiments—approximately 50 percent. The two-pronged approach provided the team with a more comprehensive understanding of reactivity on ultrafast timescales. As they study more-complex energetic materials, these types of insights will aid scientists in tailoring reactivity and other properties.

A Case for Diversity

On its own, each shock physics platform has helped to advance dynamic compression research, but together they pack even more of a punch. The lengthy quest to understand iron’s phases illustrates how data from experiments performed on different, and evolving, experimental platforms can be used to build a more complete picture of a material’s behavior across a broad range of conditions. The sixth most abundant element in the universe, iron is a key constituent of rocky planets, including Earth. Melting is likely the most important process in the evolution of a planet’s interior, determining whether a rocky planet will form a distinct crust, mantle, and core and whether that core will generate radiation-shielding magnetic fields, thought to be a necessary criterion for supporting life.

In the early 2000s, Livermore gas guns provided the first measurements of the melting temperature of iron at Earth’s core pressures (360 gigapascals), which helped establish the core’s size. Melting points are determined by measuring the sound speed in the material—because when the material melts, the sound velocity suddenly drops.

This pressure regime was revisited in 2005 using x-ray diffraction at the Omega Laser Facility to probe the start and finish of melting along the same thermodynamic path. The work included the first direct observation of one of the best-known phase transitions in shock physics, that of iron at approximately 13 gigapascals. In 2014, Crowhurst and colleagues used the ultrafast laser to make the highest ever time-resolved measurements of that phase change. These experiments demonstrated that at ultrahigh strain, the transition occurs at double the pressure of
A scanning electron microscopy image shows the free surface of an iron sample after five shots from an ultrafast laser. The craters are the result of compression waves generated by the intense laser energy applied to the reverse surface.

experiments using lower strain rates, such as those performed on a diamond anvil cell or even at the Omega laser.

Livermore researchers have explored more extreme regimes, as well. In 2013, using Omega, they made record-setting pressure measurements in solid iron at up to 560 gigapascals, further constraining iron’s melting conditions and demonstrating that high-pressure iron has unexpected strength. Experiments at NIF have since captured the pressure versus density of iron at 800 gigapascals—more than double the pressure at Earth’s core. Notes Collins, “We’re presently working to explore solid and liquid iron to several terapascals, and these data will be crucial to understanding the evolution of extrasolar terrestrial planets.” Gas-gun experimentalists are also making the first measurements of iron’s emissivity—how efficiently its surface radiates energy—to calculate an even more precise melting point.

Livermore researchers also verified the shock-induced formation of diamond by an insensitive high explosive. Many international shock wave programs had failed to experimentally verify this long-predicted product because of the high-temperature conditions of long-timescale shock measurements. However, using the Laboratory’s ultrafast-timescale compression-and-release platform, Zaug and his colleagues conducted an experiment that produced recoverable diamond as an intermediate product. This effort thus provided needed validation data for Laboratory predictions.

Dynamic compression researchers have often built on data from other platforms, as in the case of iron. Up until now, however, most researchers have pursued their investigations separately, and multiproject collaborations have been rare. That situation has begun to change, as Livermore researchers are increasingly finding that the problems with which they are grappling are best solved using multiple methods. Chau explains, “If we squeeze a material, it will eventually change phase, but how does the rate at which we squeeze affect the material’s transformation? Are there any intermediate phases? These are some of the questions we are starting to ask, and the solutions require studying materials at multiple timescales, using multiple platforms.”

By investing in platforms for dynamic compression, the Laboratory has helped to advance many missions at once, from fundamental Earth and planetary science to fusion energy, to national security work and beyond. Just as important as the tools, however, is assembling the right research teams—something at which Livermore excels. Collins notes, “Because our ultrahigh-pressure experiments unveil so many new and unpredicted phenomena, we have built an outstanding and diverse team of scientists to understand these phenomena and to build the foundations for this new scientific frontier.”

—Rose Hansen

Key Words: Advanced Photon Source (APS), dynamic compression sector (DCS), explosives, forsterite, gas gun, granular material, High Explosives Applications Facility (HEAF), hydrogen peroxide, ignition, iron, Joint Actinide Shock Physics Experimental Research Facility, Jupiter, Linac Coherent Light Source (LCLS), Laboratory Directed Research and Development (LDRD) Program, luminosity, magnetic field, molecular dynamics (MD) simulation, National Ignition Facility (NIF), Omega Laser Facility, optical pyrometer, phase transition, Saturn, shock physics, shock wave, strain rate, ultrafast laser system, velocity interferometer, x-ray diffraction, x-ray imaging.

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