Putting More Pressure on Hydrogen

Flashes of light and deafening crashes punctuated Livermore's laser experiments to transform an isotope of hydrogen into a metal. The payoff: data for revising the hydrogen equation of state, fundamental not only to the Laboratory's national security projects but also to physical science itself.

THE laser experiments team knew they had to scramble. The dismantling of Nova, the world's largest laser, was on the agenda. Lawrence Livermore's even larger laser facility, the National Ignition Facility (NIF), needed Nova's space for support facilities as NIF construction was progressing. The two-beam laser target area necessary for the experiment was scheduled to be shut down imminently, and the Nova schedule was very full. But a place in line suddenly became available. Team members knew this was their opportunity to repeat some important but difficult work using new diagnostic techniques with the Nova facility. They were going to perform another round of experiments to laser-shock and compress deuterium, an isotope of hydrogen, and turn the element most familiar in a gaseous form into a metal.

For this experiment, Nova would be used to create conditions not very different from the atmospheres of giant planets and the outer envelopes of low-mass, largely hydrogen stars. The laser would subject hydrogen to extreme and hitherto unexplored pressure regimes. It would pulverize deuterium samples, allowing experimenters to collect, analyze, and verify thermodynamic and optical information about how hydrogen goes metallic.

The results would establish a substantially improved equation of state for the element hydrogen. They would also add to our understanding of large planets and stars, make it easier to design fusion targets for NIF's 192 laser beams, and prove important for

DOE's stockpile stewardship mission by providing new high-pressure deuterium data critical to safety and reliability assurances of the nation's nuclear weapons.

Simplicity Poses Difficulties

Scientists have been attempting to metallize hydrogen for some time. The desire to do so must have materialized as soon as Eugene Wigner (later a Nobel laureate for work in quantum mechanics) theorized in 1935 that under extreme pressure, hydrogen turns into a metal. Wigner's theory concerns the high-energy-density physics of hydrogen, an area of knowledge fundamental to solving problems in astrophysics, planetary physics, nuclear explosions, and inertial fusion. However, experimental tools to test theory were not available for some 30 years. Then, in 1994, Lawrence Livermore researchers saw the first evidence of metallization during shock compression experiments with a light-gas gun (*S&TR*, September 1996, pp. 12–18). In the meantime, theorists developed models of hydrogen at extreme pressure, density, and temperature, but the models were fraught with uncertainty and disagreement.

They still are. That is because hydrogen, with its one electron and one proton, is simple only in its atomic structure. At high pressures, it is among the most difficult elements to understand. At the extreme densities of very high pressure, its various particles—atoms, molecules, ions, electrons, even strings of molecules—are free to interact strongly and nonlinearly. Hydrogen bypasses the screening mechanisms in more complexly structured elements that work to regulate particle interactions and thereby make an element's behavior easier to predict. The basic problem for theorists: What mixture of particles should constitute the hydrogen model?

Different proposed particle mixtures and interparticle forces have led to different results from hydrogen metallization models. Therefore, scientists have yet to agree on a hypothesis of how highly pressurized hydrogen transforms from a diatomic insulator into a monatomic conducting metal. A major point of contention among theorists concerns the specific mechanism causing metallization: Does it happen when hydrogen molecules separate (the theory of dissociation)? Or when they ionize? And at what pressure and temperature?

Density and temperature effects on molecular separation and ionization must be considered and evaluated for their impact on hydrogen's equation of state. (An EOS is a mathematical representation of a material's physical state as defined by its pressure, density, and either temperature or energy. It is a necessary constituent of all calculations involving material properties.) Scientists also disagree on whether metallization occurs gradually or abruptly. Models have simulated the transformation both ways. In fact, the abrupt phase transition, a controversial theory postulated in 1989 by researchers Didier Saumon of Vanderbilt University and Gilles Chabrier from the Ecole Normal Superieure in Lyon, France, intensified the pace of research into the metallization phenomenon.

Scientists are eager to resolve this theoretical challenge so they can modify and refine the fundamentally important high-energy-density EOS for hydrogen. They realize the EOS is flawed; to improve it is to improve a necessary tool for answering important basic questions about high-energy-density matter.

Says Livermore physicist Robert Cauble in describing the goal he and his colleagues are seeking, "We're trying to fill in the box of hydrogen EOS theory. Right now, we know that the theory works in a couple of corners—we know something about plasma, condensed matter, metal, liquid metal, and insulators. If we could join those pieces together, maybe we could stretch the theory toward the other corners and produce one overriding hydrogen theory that spans all densities, temperatures, and pressures."

Experimentation to Guide Theory

Clearly, experimentation is necessary for clarifying hydrogen metallization theory. Experimentation was not possible until the 1970s, when the first tools for creating the requisite experimental conditions finally became available. At Livermore, scientists began using explosively driven systems to compress magnetic fields and, in turn, small hydrogen samples to megabar pressures. They performed hydrostatic experiments in which pistons were pressed on liquid samples inside a pressure vessel. They also used diamond anvil cells to squeeze liquid hydrogen samples. Almost 60 years after Wigner's theory, Lawrence Livermore scientists shocked deuterium, an isotope of hydrogen,* with a light-gas gun and saw evidence of metallization for the first time. The gas-gun data revealed the precise pressure at which metallization occurs at high temperature. They also demonstrated that, at high temperatures (about 4,000 kelvin), metallization occurred at pressures significantly lower than had been theorized—at 0.2 megabar instead of 3 megabars. (1 megabar is the pressure of 1 million atmospheres, 15 million pounds per square inch, or 100 pascals.)

The gas-gun data brought theory into a new realm of discovery and inspired other researchers at Livermore to extend experimentation to higher pressure regimes that are possible on the Nova laser. The laser could be used to shock liquid deuterium to a wide range of pressures above the metallic transition.

*Results apply to deuterium, hydrogen, and tritium; the experiment used deuterium for convenience.

The optical properties of the shocked state could be measured to verify that the metal–insulator boundary had been spanned and thermodynamic properties could be measured to determine the EOS.

However, the same techniques that are used in gas-gun shock experiments cannot be simply carried over to lasers. The spatial scale is about 50 times smaller, and the time scale is about 1,000 times shorter. Attempts to produce laser-driven shocks capable of yielding accurate high-pressure EOS data had been made since the mid-1970s but yielded no useful data. Unlike gas-gun shocks produced by a fast-moving but cold projectile, laser irradiation of matter produces a very hot plasma that can interfere with measurements. Before performing the hydrogen EOS experiments, the Livermore team had to overcome the challenges inherent in the technique. They did so using newly developed diagnostics and target designs.

The first set of laser shock experiments, reported in early 1997, yielded startling results.[1] When shocked to 1 megabar, the deuterium compressed to a much-higher-than-expected density. This fact raised new questions even as the viability of laser shock experiments was demonstrated, and the experimenters could not rest without attempting another round of experimentation. And so it was that a group of laser physicists found themselves working frantically to design or modify diagnostic equipment, rush fabrication, and get it all installed into the Nova chamber before the two-beam target area was dismantled. Over a long weekend, they prepared for this second round of laser shots, setting up and checking diagnostic and cryogenic target components, verifying shielding and alignment, and inspecting for leaks. They tested everything; they called in several shifts of technicians to work around the clock; then they prepared themselves and their families for the series of 16-hour days.

Tools for Shock Experiments

Although Livermore's light-gas-gun experiments marked the first time the shock compression method was used to metallize hydrogen, shock compression is common in high-energy-density physics experiments. Large amounts of energy are added suddenly to a material system, creating intense sound or pressure waves that become shock waves. Shock waves compress a material to greater pressure, changing it to a new state at higher density, temperature, and pressure.

For the laser experiments, the target of the shock waves consisted of liquid deuterium loaded inside a cylinder 0.45 millimeter tall and 1.5 millimeters in diameter that had been machined into a copper block (Figure 1). One end of the cell was capped with a metal (aluminum or beryllium) disk, or pusher, that absorbed the laser energy and transmitted the shock wave into the deuterium. At the opposite end of the cell, a 0.5-millimeter-thick sapphire window allowed optical data to be taken. On both sides of the cell, thin windows of beryllium foil covered holes that were used for transverse x-ray radiography.

The metal shock pusher was coated with a polystyrene layer that cushioned it from direct laser ablation and prevented overheating. Because the laser light would shine directly through the cold polystyrene, an extremely thin (10-nanometer-thick) aluminum film was added. (After the polystyrene heated up, it would become opaque to the laser.)

Two quantities were measured by x-ray radiography during the shock compression experiments. One was the speed of the shock in the deuterium. The other was the speed to which the shocked deuterium was accelerated; this is called the particle speed. The two

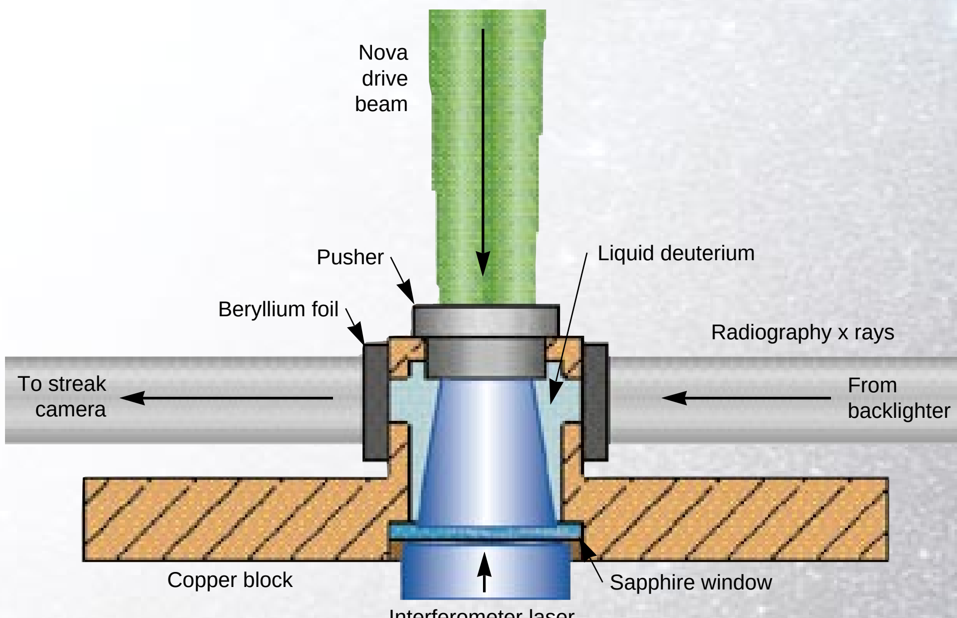

Figure 1. Schematic of the Nova laser shocking a target cell filled with liquid deuterium and machined into a copper block. One end of the cell is capped by an aluminum pusher, the other by a sapphire window used for rearview diagnostics. X-ray transmitting windows made of beryllium foil are located on each side of the cell.

quantities are used in so-called Hugoniot relations (calculations based on physical conservation laws) to determine the resulting compression and pressure of the shocked deuterium.

If a material with the same initial pressure, volume, and energy is subjected to a series of compression experiments of varying shock strengths, different pairs of initial and new compression states can be determined and plotted. The plots are the material's Hugoniot, a curve that relates the velocity of a single shock wave to the pressure, density, and total heat of the material before and after the shock wave passes. The Hugoniot is a relatively simple but well-defined curve that is unique for each material and as such is an invaluable tool for analyzing a material's EOS.

It Took Three Laser Beams

The experiments were conducted with three simultaneous laser beams: two from Nova (Figure 2) and the third from a tabletop laser. One Nova beam was used for shocking the deuterium. The laser shot blasted the target's polystyrene layer, which heated up rapidly to drive a shock wave into the pusher and compress the deuterium. Aimed at the target cell, the laser's high energies produced a long and steady shock wave. The beam was smoothed by a phase plate to ensure a spatially planar and uniform shock front, critical for accurate measurements. If the shock had been delivered as a small, nonuniform laser spot, experimental data would have been difficult or impossible to interpret, and "edge effects" would interfere with the results as well.

Figure 2. Livermore experimenters check the setup for laser beams that will drive a shock in a tiny target cell so that transverse radiography can be performed to obtain shock-wave and reflectivity measurements.

The second Nova laser beam was used to create an x-ray source for transverse radiography by irradiating a nearby iron foil. X rays from the iron plasma illuminated the target cell from the side. The shocked deuterium absorbed and refracted the x-ray light differently because it had been changed by the propagating shock wave. The x rays transmitted through the cell were collected by a Kirkpatrick–Baez microscope, which improved data resolution, and were then focused onto a streak camera. In this way, the experimenters tracked the propagation of the shock front to find the shock speed. The pusher–deuterium interface, which moved at the particle speed, was tracked to determine that speed. Combining these speeds produced a single Hugoniot data point.

An example of a streaked radiograph of shock-compressed deuterium is shown in Figure 3. Because the pusher is opaque and the liquid deuterium is transparent, the interface between them is the boundary between the light and dark regions. When the laser-driven shock crossed the interface at 2 nanoseconds, the pusher surface accelerated to a steady speed, i.e., the particle speed. As the shock wave headed into the deuterium, a shock front (visible as a dark line because the backlight x rays refract at density differences) moved ahead of the interface. The shock and particle speeds were determined from the film. The shock propagated steadily until a second, stronger shock, caused by shock reverberation in the pusher, entered the deuterium at 6 nanoseconds.

The third laser beam was used for optical interferometric measurements. In the earlier set of laser-shock experiments, this third beam was configured as a Michelson interferometer to monitor how much the target cell heated up before the arrival of the shock wave. This "preheat" had to be accounted for, or calculations of the shocked material's initial density would be inaccurate. The experiments would determine the compression (the ratio of shocked to unshocked deuterium densities), so knowledge of the initial, unshocked density was extremely important.

The Michelson interferometer beam was directed through the sapphire window at the bottom of the target cell. Its function was to monitor the movement of the pusher surface, indicative of expansion from radiative heating. The interferometer imaged this movement by splitting its beam into two arms: a reference arm and a sample arm that bounced off the pusher surface. When the two arms were recombined, their phase differences resulted in light fringes (bands caused by diffraction) that revealed, through measurements as small as a few tens of nanometers, what motion was detected in the pusher surface. The incorporation of a polystyrene coat on top of the pusher kept its bottom surface temperature below the detection limit of 400 kelvin.

The Michelson interferometer measurements had one additional use. Its reference beam verified the planarity and uniformity of the arriving shock wave. Experimenters saw that the shock front was uniform and planar to within 2.5 micrometers over a lateral region of 350 micrometers.

With concerns over preheat and shock-wave quality out of the way, it was unnecessary to repeat the Michelson interferometer measurements for the second set of laser experiments. Instead, the third beam was set up for velocity interferometry to determine the velocity and, importantly, reflectivity of the shock front. Because of its relationship to electrical conductivity, the reflectivity measurement established the occurrence of metallization.

Then There Were Measurements

The velocity interferometer used in these experiments was a particularly accurate instrument for measuring motion—in this case, the speed of the reflecting surface of a moving shock front. Unlike a conventional interferometer that first splits a laser beam into two arms, this interferometer shot the whole beam onto the experimental sample and split the beam after it exited the sample. Then one beam was passed through a piece of glass, called an etalon, which slowed it down. Because of this induced time delay in one arm, recombining the beams generated light fringes. The fringes changed when the shock front moved, doing so in proportion to shock speed.

Figure 4 shows a streak velocity interferogram. For times before $t = 0$, the fringes are reflections of the stationary pusher surface. For $t > 0$, the reflection is from the shock front in the deuterium. The amount of the fringe shift at $t = 0$ is proportional to the speed of the shock front.

The difference in reflected light intensity originating from the motionless pusher surface and that from the shock front moving in the deuterium reveals the reflectivity of the shock. The pressure at which a change in reflectivity occurs can be determined because particle and shock speeds can be measured. The measured reflectivity at low shock pressure (0.2 megabar) is only a few percent. Above 0.55 megabar, however, the measured reflectivity is about 60 percent—characteristic of a poorly reflecting metal. This measurement proves that the deuterium changed from an insulating state to a conducting one. The data also show that the transition occurs simultaneously with the earlier observed high compression. These effects are linked: the high compression is a result of the transition.

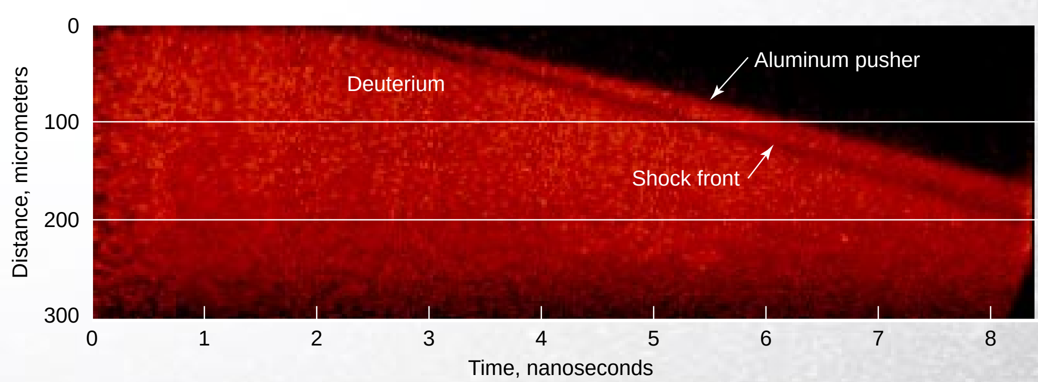

Figure 3. The image of the deuterium is moved across the film over time, producing a streak radiograph. In the figure, the pusher is above the deuterium, so the shock travels from top to bottom.

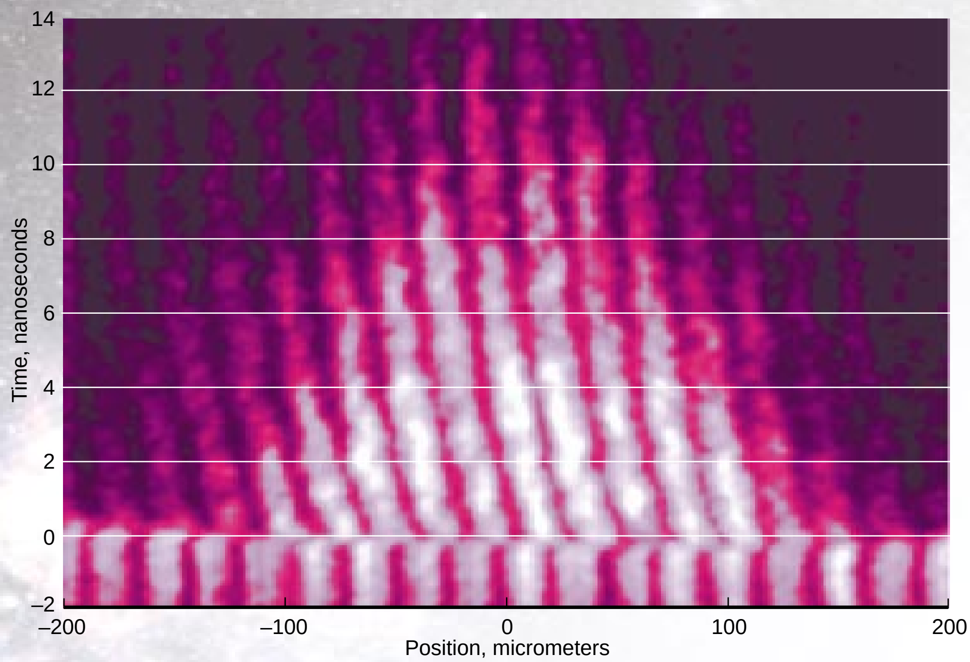

Figure 4. This velocity interferogram shows the deuterium in the Livermore Nova experiment had changed to a conducting state.

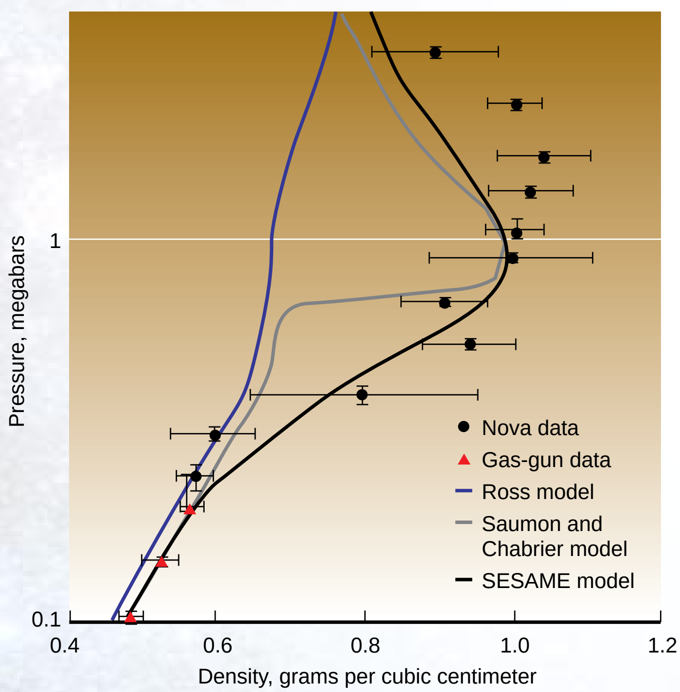

Figure 5. Livermore Nova data are significant because they show much higher compressibility than the SESAME EOS model, and they are similar to the gas-gun data and the Ross model of strong dissociation.

In addition to these measurements, the temperature of the shock was determined by recording the light emission of the shock front in several wavelength bands. An optical pyrometer viewed the shock through the sapphire window in the cell. Temperature is a fundamental component of the EOS, but it cannot be derived from the Hugoniot relations. It must be found separately. Because the form of the wavelength-dependent light intensity is a known function of temperature, fitting the emission data into that formula allowed the experimenters to find the value of temperature.

Implications for Hydrogen's EOS

The experimental team was tired but elated with their results. The data obtained from the latest round of effort would once again recharge their work on hydrogen theory and, furthermore, bring experimentation to another level. Their work had provided the first direct evidence on the Hugoniot to support the hypothesis that liquid deuterium transforms from a molecular fluid into a monatomic metallic fluid at lower pressures than postulated by earlier theoretical models.

Figure 5 shows the measured Hugoniot as pressure versus density. The figure compares Hugoniot curves for the laser data with a linear mixing model proposed by Livermore scientist Marvin Ross, an earlier model in the SESAME EOS library, the prediction of Saumon and Chabrier, and the Livermore light-gas-gun data. At the lowest compression, the laser data are in agreement with the gas-gun results, while at higher compressions, the data significantly deviate from the SESAME prediction. The data at 0.25 megabar are significant because they overlap the gas-gun data, providing confidence in current results. The current data show an enhanced compressibility similar to that of the Ross linear mixing model in the region where strong molecular dissociation is predicted. Although the shocked density at 1 megabar is close to that of Saumon and Chabrier, the data do not show the abrupt transition predicted by their model. The conclusion is that molecular dissociation and ionization are significant factors in hydrogen isotopes compressed to megabar pressures. Reflectivity measurements, using the measured Hugoniot to find the pressure, are shown in Figure 6.

The current data provide an important benchmark for a revised EOS model of hydrogen and its isotopes in a regime relevant to high-energy-density physics applications. Additionally, the experiments demonstrate that laser-driven shock waves can effectively be used for EOS studies at pressures beyond those attainable by traditional techniques. The new hydrogen EOS will change the way planets such as Jupiter are modeled, especially the size of its metallic core. For fusion occurring on Earth, the higher compressibility of deuterium will make the goal of laboratory thermonuclear fusion easier to achieve than previously thought.

Prize-Winning Basic Science

The work in developing the techniques to perform laser-driven EOS experiments and in getting surprising data on an important material earned Robert Cauble, Peter Celliers, Gilbert Collins, and Luiz Da Silva—the principal members of the research team—the 1998 American Physical Society Award for Excellence in Plasma Physics Research. The "exquisite series of experiments" cited by the award were a fitting follow-up to the earlier Lawrence Livermore gas-gun shock compression experiments, which also pushed hydrogen EOS theory up another rung. The principal investigators for that work, William Nellis and Arthur Mitchell, received the 1997 American Physical Society Award for Shock Compression Science. It may be that Laboratory researchers, in furthering the fundamental science so important to Laboratory missions, are also setting themselves new standards for scientific execution.

— Gloria Wilt

Key Words: equation of state (EOS), gas gun, high-energy density, Hugoniot, metallized hydrogen, Nova laser.

Reference

1. L. B. Da Silva et al., *Physical Review Letters*, Vol. 78, No. 3 (January 1997), pp. 483–486.

For further information contact Robert C. Cauble (925) 422-4724 (cauble1@llnl.gov).

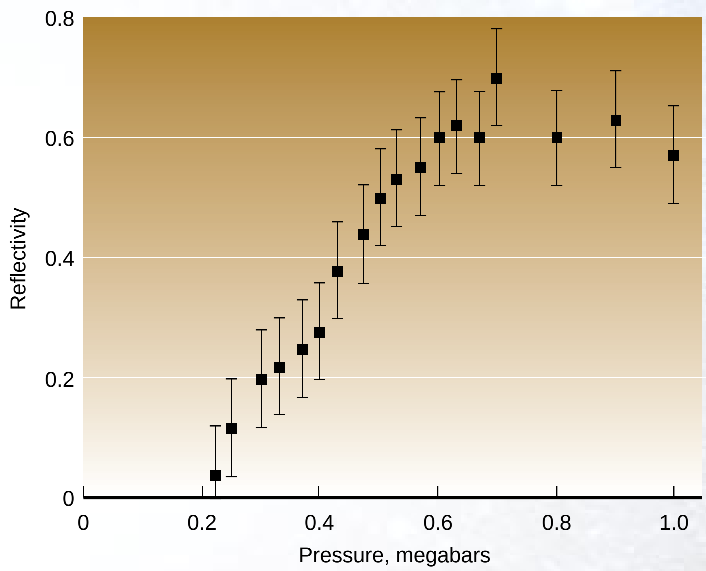

Figure 6. The steep curve between pressures of 0.4 and 0.6 megabar shows higher compressibility for deuterium than previously thought.

About the Scientist

ROBERT C. CAUBLE received his B.S. in physics in 1974 from the University of Arizona and his Ph.D. in nuclear engineering from the University of Michigan in 1980. In 1980, Cauble worked at the Naval Research Laboratory on theoretical predictions of the effects of high-density plasma on atomic transitions and particle transport. He came to Lawrence Livermore in 1985 to work on projects to produce atomic models for plasma simulations and to design and analyze experiments to use laboratory x-ray lasers as high-density-plasma probes and interferometers. More recently, he has worked on large-scale simulations and designs of experiments to elicit material properties, mainly equation-of-state data at extreme pressures using intense, laser-driven shocks.