Absolute Equation-of-State Data in the 10–40 Mbar (1–4 TPa) Regime

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A technique has been developed that allows the measurement of equations of state of low-atomic-number materials in regimes of extremely high pressure. Using this technique we have obtained absolute multimegabar measurements on the principal Hugoniots of low-Z solids, including data on polystyrene up to 40 Mbar (60 times the previous absolutely measured pressure) and beryllium at pressures up to 15 Mbar. The polystyrene data were able to differentiate between existing theoretical models. The beryllium data confirm theoretical Hugoniots of several materials as high as 65 Mbar. [S0031-9007(98)05321-6]

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Experimental and theoretical investigations of the equations of state (EOS) of materials at high pressures and densities are of fundamental interest to astrophysics, inertial confinement fusion (ICF), and other related fields. Matter shock-compressed to pressures greater than a few Mbar (1 Mbar = 100 GPa) can be a strongly coupled, partially ionized fluid that is extremely difficult to model, putting a premium on data at high pressures. Diamond anvil cells have been used to determine low temperature isotherms at static pressures up to about 3 Mbar [1]. However, obtaining EOS data at elevated temperatures in this regime requires strong shocks. Until recently, the only driver with sufficient energy to obtain such data has been nuclear explosions [2–4].

High power lasers are capable of driving multimegabar shocks in small samples either by direct irradiation [5] or indirectly, such as mounting the target on a hohlraum into which the lasers are focused [6]. Indirect drive has some advantages over direct drive. More beams can be used in order to obtain higher pressures. The laser-matter interaction takes place on the interior hohlraum walls so that x rays reradiated from the walls are scattered uniformly in the cavity, x rays that can be used to drive very uniform shocks. In addition, the drive temperature can be much smaller than with direct drive (200 eV compared to 2 keV), reducing the potential for preheating of the sample. Hugoniot data at pressures well above 1 Mbar have been obtained for some metals with direct drive [7] and indirect drive [8]. However, these experiments derived EOS data for a sample relative to the EOS of a standard material in a method called impedance matching. The nuclear-explosion-driven data were found by the same method, nuclear impedance matching (NIM).

We have employed indirect drive to acquire absolute (not relative) EOS data for low-Z solids at pressures greater than 10 Mbar. We have obtained absolute data on the Hugoniot of beryllium between 10 and 15 Mbar and polystyrene (CH) between 10 and 40 Mbar. Both CH and Be are critically important materials for ICF; they are the primary candidates from which the shells of ICF ignition capsules will be made for the National Ignition Facility [9]. The pressure regimes accessed by ignition targets are similar to those investigated here and much higher than in previous absolute EOS measurements [10]. We were able to determine that a well-used CH EOS model is in error in this regime. The absolute Be data largely confirm present EOS models for the metal and, further, show that previous NIM data for several materials, including Mo, LiD, Al, and W, at pressures as high as 65 Mbar [3], are accurate.

A single shock drives a sample to a point on the principal Hugoniot, which is the locus of all final states of pressure, energy, and density that can be achieved behind a single shock wave passing through a material from the initial normal-density state. Conservation relations require that two independent parameters be measured to obtain absolute Hugoniot data. The shock speed U_s, particle speed U_p (the material speed after passage of the shock), final pressure P, and final density ρ are related by

$$P - P_o = \rho_o U_s U_p \tag{1}$$

and

$$\rho/\rho_o = U_s/(U_s - U_p), \tag{2}$$

where ρ_o and P_o are initial density and pressure [11]. Often one of the measured quantities is U_s; the second quantity is generally very difficult to obtain. Comparative data obtained in impedance match experiments are derived from simultaneously measuring shock speeds in a standard material and in a conjoined sample. The shock speed in the standard referenced to the standard's presumed-known EOS permits identification of a point on the Hugoniot of the sample [12]. In the experiments described here, both U_s and U_p were measured in each experiment, providing absolute Hugoniot data.

Beryllium and polystyrene are low-Z materials, making it possible to use x-ray transmission radiography to obtain the required two quantities. Eight beams of the Nova laser [13] (λ = 350 nm) were focused into a 3-mm-long by 1.6-mm-diameter cylindrical gold hohlraum. The experimental package was mounted to the side of the hohlraum;

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x-ray ablation of the end of the package inside the cavity drove intense shocks into the package. Two beams irradiated a vanadium backlight foil; 5.4 keV x rays from the backlighter passing through the package were recorded on a streak camera in a side-on arrangement. Pinholes with 5- or 10-μm diameters were used to aperture the image. A schematic diagram of the arrangement for this experiment is shown in Fig. 1.

Each package consisted of two conjoined 0.5-mm-diameter cylinders surrounded by a 100-μm-thick Be annulus. The cylinder nearer the hohlraum was plastic doped with bromine, CH(Br), at an atomic concentration of 2% (with a density of 1.22 g cm^{-3}) or 6% (1.56 g cm^{-3}). It was 200 or 300 μm long depending on the experiment. The sample to be measured was in the farther section, either undoped CH with a density of 1.044 $\pm$ 0.007 g cm^{-3} or Be (1.85 $\pm$ 0.01 g cm^{-3}). The cylinders were glued into a gold sleeve extending from the hohlraum wall to within 50 μm of the interface between the two sections. This sleeve, as well as the annular Be tamper, served to constrain radial expansion of the package as the shock propagated through the package. This assembly was extensively modeled and tested at Nova, where it was found that the combination of sleeve, annular tamper, and reentrant hohlraum mounting produced a 500-μm-wide shock front that remained planar to better than 5% at the interface position [14]. In these experiments, only the central 250 μm of the cylinder was imaged so that planarity of the observed shock was better than 2%. Two wires were glued across the package at metrologized positions. The wires were also radiographed and served as spatial fiducials.

CH(Br) is opaque to the backlighter x rays but CH and Be are transmissive, making the interface between the two sections visible on film as the boundary between dark and light areas. As the shock crosses the interface, the interface begins to move at a speed U_p determined by the shock pressure at the interface. This motion is visible on the streak record, and U_p can be obtained by tracking the interface.

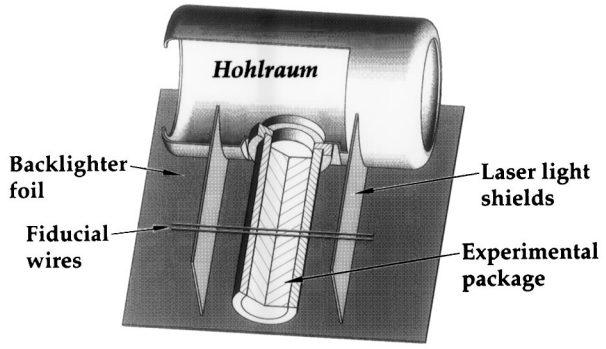

FIG. 1. Diagram of a hohlraum-driven EOS experiment for side-on radiography of a two-section package (shown as a cutaway) mounted to the hohlraum. The backlighter foil is shown in the rear. Laser beams are focused into the cylindrical hohlraum through openings in each end, producing x rays that drive a shock down the package. Backlighter x rays pass through the package and are recorded on a streak camera. Shields prevent laser light from illuminating the package. Two wires that serve as spatial fiducials on the streak film are shown.

The shock front can be similarly tracked. Higher-density shocked matter is less transmissive than material ahead of the shock. The shock front can be identified on film as the interface between these two areas of differing transmission. Following the shock front on the streak film reveals U_s.

Figure 2 displays a streak image. The interface and the shock front are noted and the two wire shadows are visible. The trajectories were fit by the least squares method to determine U_p and U_s. However, the slopes are not in general linear because the x-ray drive is off by the time the shock crosses the interface. The laser energy required to sustain a $\sim$230 eV hohlraum temperature and filling of hohlraum with plasma are the limiting factors. By the time the shock crosses the interface it has begun to decelerate. Since the shock speed is not constant, (1) and (2) must be solved at the time that the shock crosses the interface.

The shocked material density could be estimated by measuring the difference in backlighter transmission in either side of the shock front: The compression can be inferred from known cold opacities. This was done previously in Nova experiments designed to detect mixing at shocked interfaces [15]. However, it was recognized at that time that the detailed shape of the shock front was required to determine the actual x-ray path through the shocked material so that a measurement by this method would be inconclusive.

It is possible, however, to measure the compression ρ/ρ_o by measurement of the thickness of the shocked region, the distance between the shock front and the interface, at any given time. This is the thickness of a shocked sample layer whose original thickness was the distance between the shock front and the initial interface

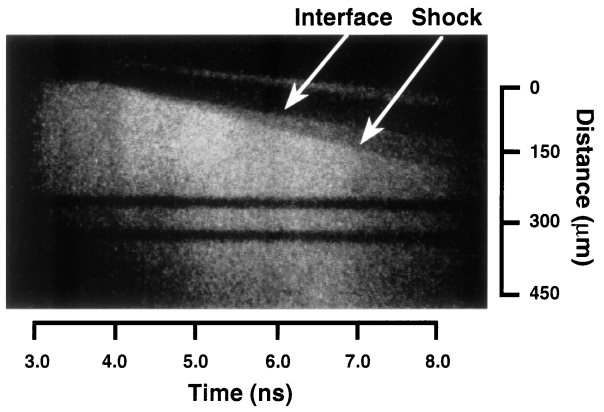

FIG. 2. A time-resolved side-on radiograph of a shocked polystyrene EOS package. Time is measured from the start of the driving laser pulse. The shock and interface trajectories are noted.

position; ρ/ρ_o is the ratio of the two values [10]. On the Hugoniot this method is strictly valid only for a constant shock speed. However, since the shock deceleration is not large in our experiments, this method is expected to be accurate up to 3 ns after the shock crosses the interface. Uncertainties in the shock density by this method are comparable to those from (2), 15%–20%, and represent a check on the U_s-U_p measurements.

Figure 3 shows the results of a series of Hugoniot experiments on polystyrene. The data vary from 10 to 40 Mbar, 60 times the pressure of previous measurements, which are shown in Fig. 3(b) [16]. The uncertainties are governed by the limitations of the diagnostic and the ability to fit the trajectories. The process of selecting the points to define the trajectories themselves produced negligible error. Since the compression is a ratio, the effects of magnification and streak camera timing do not enter in the uncertainty of the compression. The lack of distinction between the two trajectories at the time the shock crosses the interface leads to an additional uncertainty that was estimated by variation of the pusher and shock tracks. The curves in the figure are Hugoniots calculated from a Thomas-Fermi equation of state model called QEOS [17], the CH EOS table in the SESAME EOS library [18], and an extensively used EOS table [19]. The data support the less compressible SESAME and QEOS models. The other model, which is based on an extrapolation of lower pressure shock data, shows too high a compression in this pressure regime. Such a distinction between the models could not be made at lower pressures.

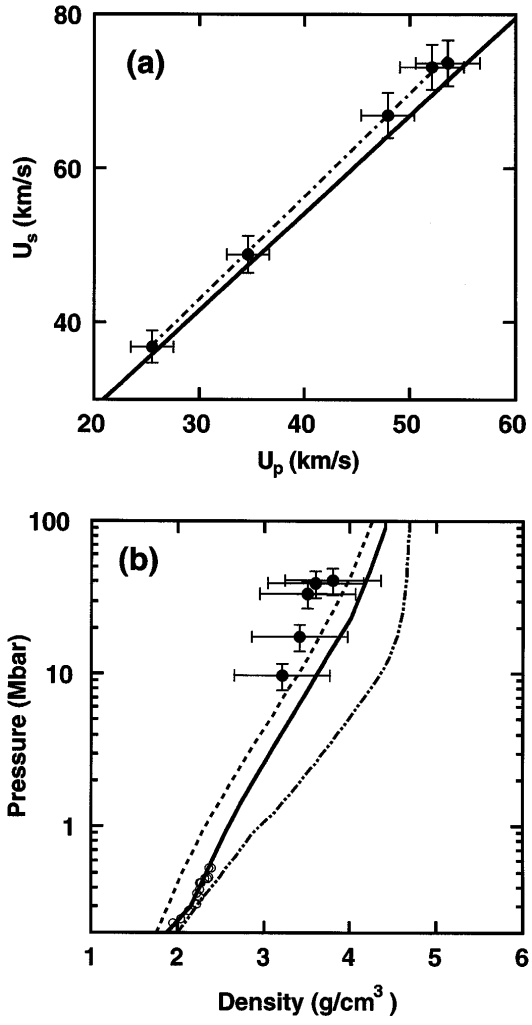

FIG. 3. Polystyrene Hugoniot results are shown as shock speed vs particle speed (a) and pressure vs density (b). The solid curve in each case is the SESAME Hugoniot [18]. The dot-dashed curve in (a) is a straight line fit to the data. In (b), Hugoniots from the Thomas-Fermi model QEOS [17] (dashed) and an extensively used EOS table [19] (dot-dashed) are shown. Previous shock data are shown as open circles below 0.6 Mbar [16].

Although the data suggest that CH may be even slightly less compressible than QEOS and SESAME predict in this regime, it is possible that the data could be systematically shifted to a lower density by unobserved preheating of the CH sample. Simulations show that at a preheat level of 0.05 eV, ~0.5% of the estimated shock temperature is possible in the sample. However, a preheat level of 0.5 eV is required to shift the QEOS Hugoniot to match the data. Experiments designed to estimate the actual preheat level of the sample are in good agreement with simulations. An unrealistic amount of preheat (several eV) would be necessary for the data to match the more compressible tabular Hugoniot.

Figure 4 shows results of two experiments for Be compared with NIM data [3,4] and the SESAME Hugoniot [18]. While the laser data have larger uncertainties than the NIM date, they are absolute measurements, whereas the NIM data are relative to a calculated EOS of another "standard" metal.

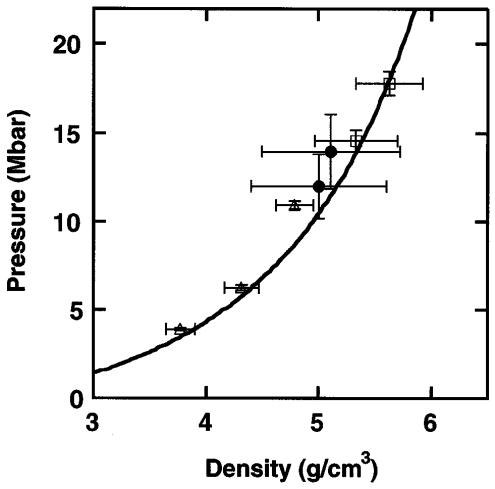

FIG. 4. Laser-produced beryllium Hugoniot data are shown as solid circles compared with Hugoniots derived from (solid line) SESAME [18]. Nuclear impedance match data are plotted as open squares [3] and triangles [4]. Shock (particle) speeds in km/s for the shots were 32 (20.2) and 34.4 (22).

The Be NIM data from Ref. [3] (open squares, Fig. 4) are part of a set of data of different materials all tested on the same nuclear experiment where molybdenum was used as the EOS standard. A Mo plate was shocked to 49 Mbar and derived pressures for the samples ranged from 9.5 Mbar for LiD to 65 Mbar for W. All of the data in the set were made consistent with the Mo EOS, although the Mo EOS was calculated, not measured. The absolute data presented here tend to confirm the derived NIM Hugoniot for Be. This also implies that the Mo EOS at 49 Mbar is accurate, as well as the remainder of the data presented in [3]. However, to more fully assess the accuracy of the NIM data, uncertainties in the absolute data need to be reduced.

In conclusion, we have presented a technique for measuring absolute Hugoniot data for low-Z materials in the multimegabar regime using a high energy laser in an indirect drive configuration. Measurements of the Hugoniot of polystyrene from 10 to 40 Mbar and of beryllium near 15 Mbar were obtained using this technique. The polystyrene data are sufficiently accurate to differentiate between theoretical EOS models. The absolute Be data are in the regime of comparative nuclear-driven EOS measurements and tend to confirm those results, indicating that the theoretical EOSs for a number of materials are accurate from 10 to as high as 65 Mbar. These are the first noncomparative EOS measurements made in this pressure regime.

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