Shock Hugoniot of diamond from 3 to 80 TPa

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The principal Hugoniot of carbon, initially diamond, was measured from 3 to 80 TPa (30 to 800 million atmospheres), the highest pressure ever achieved, using radiography of spherically-converging shocks. The shocks were generated by ablation of a plastic coating by soft x-rays in a laser-heated hohlraum at the National Ignition Facility (NIF). Experiments were performed with low and high drive powers, spanning different but overlapping pressure ranges. The radius-time history of the shock, and the profile of mass density behind, were determined by profile-matching from a time-resolved x-ray radiograph across the diameter of the sphere. Above \( \sim 50 \) TPa, the heating induced by the shock was great enough to ionize a significant fraction of K-shell electrons, reducing the opacity to the 10.2 keV probe x-rays. The opacity and mass density were deduced simultaneously using the constraint that the total mass of the sample was constant. The Hugoniot and opacity were consistent with density functional theory calculations of the electronic states and equation of state (EOS), and varied significantly from theoretical Hugoniots based on Thomas-Fermi theory. Theoretical models used to predict the compressibility of diamond ablator experiments at the NIF, producing the highest neutron yields so far from inertial confinement fusion experiments, are qualitatively consistent with our EOS measurements but appear to overpredict the compressibility slightly. These measurements help to evaluate theoretical techniques and constrain wide-range EOS models applicable to white dwarf stars, which are the ultimate evolutionary form of at least 97% of stars in the galaxy.

INTRODUCTION

Carbon (C) is widespread in nature and thought to be the fourth most common element in the universe \(^1\). Formed from the fusion of He in giant stars and at the end of the red giant phase of stars like the sun, C accumulates at high levels in the core and is a major component of white dwarf (WD) stars, where it is predicted to segregate and crystallize on cooling \(^2\), or in the crust of accreting neutron stars \(^3\). Although there is little debate about its properties at pressures high enough for the electrons to become degenerate, they are less certain when atoms are partially ionized. The ionization behavior affects the equation of state (EOS), radiative opacities, and diffusion coefficients, and is smoothed out in Thomas-Fermi (TF) models used in predicting most EOS models for WDs, which are the final state of the vast majority of stars in our galaxy \(^4\). This uncertainty limits our understanding of the convection zone in WDs and thus their cooling and evolution.

States of matter at elevated pressure and temperature are often generated in shock wave experiments \(^5\), where the high-pressure matter is confined inertially (i.e. by the finite time required for the compressed components to disassemble) and so the pressures achieved are not limited by the strength of surrounding components as is the case with static presses \(^6\). Dynamic loading experiments are ubiquitous for studies of warm dense matter with pressures in excess of 1 TPa.

Large pulsed lasers such as the National Ignition Facility (NIF) can be used to induce pressures in excess of 10 TPa, which are of interest for studies of massive exoplanets, brown dwarfs, and stars, as well as engineering problems such as inertial confinement fusion (ICF). We have previously reported the use of radiography of a spherically-converging shock to deduce a range of states along the shock Hugoniot of a sample \(^7\), up to \( \sim 40 \) TPa in polystyrene, and thus to constrain the EOS. The x-ray source was a foil, laser-heated to a plasma emitting strongly in the kilovolt regime. In the results reported previously, the technique used to analyze the radiograph used a model of the variation in mass den-

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sity behind the shock wave expressed in terms of radius and time. For shock pressures high enough to affect the K-shell electrons and hence the opacity to kilovolt-scale photons, a Lagrangian feature was used to constrain the total mass of the sample in the density model, and hence deduce the changing opacity at the shock front between instants of time [10, 11].

In the work reported here, we apply the same experimental technique to samples of carbon (initially diamond), deducing EOS data to significantly higher pressures than in polystyrene: ~80 TPa, which is the highest pressure at which material properties have been measured in the laboratory. We use a modified analysis method, parameterizing the model in terms of functions related to the EOS and reconstructing the density model indirectly, rather than the other way around. This method gives a smaller uncertainty and additional EOS data. As before, data obtained from the experiment are absolute – they are not made relative to the properties of a reference material – and the opacity was deduced simultaneously with the mass density. Absolute measurements of diamond are particularly impactful as it is used widely as a reference or benchmark in measurements of other materials.

**EXPERIMENTAL CONFIGURATION**

The experimental configuration was as described previously [7, 8, 10], and is summarized here for convenience. Diamond spheres, 1000 µm in diameter, were obtained from Dutch Diamond Technologies B.V. A glow-discharge polymer (GDP) layer, 150 µm thick, was deposited on the spheres to act as an ablator. The coated sphere was mounted within a Au hohlraum [12], 30 µm thick, 5.75 mm diameter and 9.42 mm high, with a gas fill of 0.03 mg/cm³ He to impede filling of the hohlraum by ablated Au. Up to 176 beams of the NIF laser were used to heat the hohlraum. The resulting soft x-ray field within the hohlraum ablated the GDP, driving a shock into the bead. The overall configuration and laser pulses were based on ICF designs, to take advantage of synergies in fabrication and also the large development effort performed to give uniform drive conditions over the surface of the bead [13]. (Fig. 1.)

We consider data from two experiments, N161016-3 and N140529-2, in which the laser drive was 300 and 800 kJ respectively. The latter was based on the ‘high foot’ ICF drive [13, 14]: the former was the foot of the drive continued for 5 ns. The temperature history of soft x-rays in the hohlraum was calculated using radiation hydrodynamics and measured by the DANTE filtered diode system [15]. The peak temperature was around 205 eV for the low drive and 275 eV for the high.

The shock wave induced by ablation of the GDP strengthened as it propagated toward the center of the sample. X-ray radiography was used to measure the variation of attenuation across the diameter of the bead, from which the shock trajectory, mass distribution and opacity in the sample could be deduced as described below. The x-ray source was a Ge foil, heated by eight beams to produce a plasma that emitted strong He-like radiation at 10.2 kV, with duration ~7 ns. Slits were cut in the hohlraum wall to enable transmission of the x-rays through the sample; diamond windows were used to impede slit closure by ablated Au. The transmitted x-rays were imaged through a slit in a Ta foil onto an x-ray streak camera (Fig. 2).

The streak radiograph was used to reconstruct the radial distribution of mass density, as a function of time.
As described previously [9], the presence of undisturbed material ahead of the shock provided a strong constraint on the inference of the change in attenuation across the shock front. In order to take advantage of this constraint, the analysis was performed by adjusting a parameterized representation of the distribution of mass density until the corresponding simulated radiograph matched the measured radiograph.

In shot N140529-2, the shock became strong enough that the opacity of shocked material to the probe x-rays decreased significantly.

**DETERMINATION OF ABSOLUTE HUGONIOT AND OPACITY DATA FROM RADIOGRAPHY**

As discussed previously [9], the reconstructed radius-time distribution of mass density $\rho(r, t)$ gives an absolute measurement of the shock Hugoniot over a range of pressures, from the position of the shock $r_s(t)$ and hence its speed $u_s(t)$, and the mass density immediately behind the shock, $\rho_s(t) = \rho(r_s(t), t)$. Simultaneous knowledge of $u_s$ and $\rho_s$ gives the complete mechanical state behind the shock by solving the Rankine-Hugoniot relations [5] representing the conservation of mass, momentum, and energy across the shock. The state ahead of the shock is known, leaving five quantities to be determined: $\rho$, pressure $p$, internal energy $e$, particle speed $u_p$, and shock speed $u_s$. If any two of these quantities are measured, the Rankine-Hugoniot equations determine the rest. In particular, $p = p_0 + (v_0 - v) u_s^2/v_0^2$ where $v = 1/\rho$ and subscript ‘0’ denotes material ahead of the shock. The Hugoniot state on the Hugoniot can thus be deduced directly from the mass density distribution without reference to a standard material: an absolute measurement.

Given distributions of mass density $\rho(r, t)$ and opacity $\sigma(r, t)$ in the object, the signal along any path from the source $\hat{r}_s$ to the detector $\hat{r}_d$ at any instant of time is given by the integral of attenuation $\mu = \rho \sigma$ through the object. Given some radiographically-visible feature near the outside of the bead, defining the enclosed mass, the time-variation can be analyzed to determine changes in opacity independently from changes in density [11]. As before, with a single marker layer, we had to use a model to account for the isentropic variation of opacity with compression behind the shock.

We previously analyzed the streak radiographs by optimizing parameters in models of the distribution of mass density $\rho(r, t)$, obtaining a tabulated variation of opacity at the shock front $\sigma_s(t)$. The density model was expressed in terms of the variation between the trajectories of the shock and the marker, defined as analytic functions with parameters included in the optimization. The radiographs also include data on the EOS in the isentropic flow region behind the shock. In investigating how we might extract this off-Hugoniot data, we found that we could parameterize the radiographic model more efficiently – with fewer parameters, and leading to a lower
uncertainty – by expressing the mass density along the shock front in terms of the shock speed $u_s$ instead of time. $u_s$ is simply the derivative of the shock trajectory. The radial derivative of density at the shock front can be related to the acceleration of the shock, $\dot{u}_s$, and the isentropic sound speed $c$, by

$$\dot{u}_s = \frac{\partial \rho}{\partial r} \frac{\partial p}{\partial \rho_s} \left[ c(u_s) + u_p(u_s) - u_s \right] \frac{\partial u_s}{\partial p_s} \tag{1}$$

where $\partial p/\partial \rho_s = c^2$, and so we used an optimizable function $c(u_s)$ to capture the radial density gradient. We refer below to constructing $\rho(r, t)$ indirectly via $\rho(u_s)$ and $c(u_s)$ as Hugoniot functions. We also used an optimizable function to describe the opacity in the shocked state; informed by comparisons with theory that were made when investigating the results for polystyrene [10], we chose a Fermi-like function of shock pressure, the pressure being obtained from the Hugoniot relations using the shock speed and mass density. Low-order polynomials were found adequate for the other fitting functions over the range of the data. More details and comparisons of alternative analysis methods are described elsewhere [10].

Constructed in this way, the functions over which parameters are optimized are properties of the sample material and do not depend on the specific experiment, in contrast to parameterizing $\rho(r, t)$ explicitly. It is then possible to obtain the solution that best fits multiple experiments simultaneously.

**ANALYSIS OF EXPERIMENTAL DATA FOR DIAMOND**

The streak radiograph was analyzed to deduce the radius-time distribution of mass density, represented using smooth functions as described above. The analysis was performed in several different ways to explore the sensitivity [16]. The locus of the shock was represented well by the function $r_s(t) = \alpha(t_c - t)^\beta$ with parameters $\alpha$, $\beta$, and $t_c$.

Given a set of fitting parameters, the Hugoniot was obtained directly from the function $\rho_s(u_s)$, the sound speed along the Hugoniot similarly from $c(u_s)$, and the opacity from the function $\sigma(p)$. The goodness-of-fit of the simulated radiograph to the data was used to assign a probability to the model. By perturbing the fitting parameters about the best fit, properties were deduced with corresponding probability, and loci were accumulated as probability amplitudes. The nominal best fit for each property was taken to be the locus of maximum likelihood, very similar to the corresponding from the best-fitting parameters. Statistical fitting uncertainties were obtained as contours from the probability distribution. Systematic uncertainties from the uncertainty in instantaneous sweep rate of the streak camera and magnification affect the absolute values of each locus, but perturbed the shape to a much smaller degree, as found previously [9, 11].

We were able to obtain data along the principal Hugoniot between 3 and 80 TPa. The statistical uncertainty was substantially higher for the low-drive shot, consistent with the lower signal level in the radiograph. Fitting both shots together gave a significant reduction in the uncertainty at low pressures. The shape was in significantly better agreement with the theoretical EOS constructed to account for shell effects [26] than one based on TF predictions [27] (Fig. 5). For the variation of sound speed with shock speed, we were not able to detect any significant variation from a straight line, consistent with both EOS models, though at high pressures the data favored the shell structure EOS. No significant variation in opacity was found in the low-drive shot. In the high-drive shot, the opacity was deduced to drop by ~20% between 10 and 80 TPa. The most rigorous prediction available to us was the ATOMIC model [21], which accounts for detailed configurations of excited electrons. Plasma opacity theory has difficulty at low temperatures, and values below 10 eV are interpolations between the ambient opacity and the first calculated value. Combined with the shock Hugoniot states from the shell structure EOS, the opacity was predicted to drop consistently with the variation deduced from the experiment. (Figs 6 and 7.)
CONCLUSIONS

These experiments have again pushed the limits of pressures accessible in laboratory measurements of the equation of state, now approaching 100 TPa. Compared with our previously-reported experiments on polystyrene, these measurements on higher-density diamond were affected less by the drop in x-ray opacity induced by shock heating, even though the peak pressure was over a factor of two higher and the peak density 3.5 times higher, because of the greater starting density and stiffness of diamond. Together with improvements in the method of analysis, the resulting data were more consistent with theoretical predictions accounting for electronic shell structure, rather than TF theory. As before, the measurements are absolute rather than with respect to a standard material of assumed equation of state, in contrast to previous measurements in this regime which employed nuclear detonations as an energy source, and were more susceptible to preheating from radiation in a planar configuration. The modified method of analysis also made it possible to deduce the sound speed along the Hugoniot, providing a direct connection to the Grüneisen parameter and off-Hugoniot states. Together with the previous measurements on polystyrene, these data directly probe states occurring in the envelope of white dwarf stars, and show the importance of electronic shell structure effects.

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The data that support the findings of this study are available from the corresponding author upon reasonable request.

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