

TAKING THIN DIAMONDS TO THEIR LIMIT: COUPLING STATIC-COMPRESSION AND LASER-SHOCK TECHNIQUES TO GENERATE DENSE WATER

Kanani K. M. Lee^{1a}, L. Robin Benedetti^{1b}, Andrew Mackinnon², Damien Hicks², Stephen J. Moon², Paul Loubeyre³, Florent Occelli³, Agnes Dewaele³, Gilbert W. Collins², and Raymond Jeanloz^{1a}

^{1a} Department of Earth and Planetary Science and ^{1b} Department of Physics, University of California, Berkeley, CA 94720-4767

² Lawrence Livermore National Laboratory, Livermore, CA 94550

³ Commissariat à l'énergie atomique, Bruyeres-le-Chatel, France

Abstract. Laser-driven Hugoniot experiments on precompressed samples access thermodynamic conditions unreachable by either static or single-shock compression techniques alone. Recent experiments using Rutherford Appleton Laboratory's Vulcan Laser achieved final pressures up to ~200 GPa and temperatures up to 10,000 K in water samples precompressed to ~1 GPa, thereby validating this new technique. Diamond anvils, used for sample precompression, must be thin in order to avoid rarefaction catch up; but thin diamonds fail under pressure. Anvils no more than 200 μm thick were encased in diamond cells modified to accommodate the laser-beam geometry.

INTRODUCTION

Dynamic experiments generally employ a sample initially at normal density and standard pressure, therefore providing data on the principal Hugoniot. By varying the initial density of the sample through precompression, it is possible to obtain data off the principal Hugoniot, accessing conditions unreachable by either static or single-shock compression. That is, precompression allows an investigation of the entire Pressure-Volume-Temperature (P-V-T) space between the isotherm and Hugoniot.

The method is proven here through experiments on H₂O, which has previously been studied both dynamically and statically to pressures in the multi-Mbar (~10² GPa) range. The precompression technique is shown below.

CONSTRAINTS

In laser-driven shock wave experiments, a short, high intensity pulse yields a strong shock. The shock must ideally be steady in order to obtain accurate Hugoniot data. Rutherford Appleton Laboratory's Vulcan Laser generates a 10¹³ – 10¹⁴ W/cm², 4 ns long, 400 μm diameter pulse by way of seven beams, six of which come from off-axis. This geometry results in two significant design constraints for precompressed samples. First, the intensity and short pulse length require that the anvil through which the laser shock travels be extremely thin—no more than 200 μm . In order to have a planar shock wave through the sample, the shock wave must propagate through the anvil thickness while the laser is on, thus supporting the wave propagation, else the end-of-pulse rarefaction may catch up. Side rarefaction is also an issue if the laser spot is not large enough to yield a planar front. Second, the laser spot size and beam path

require a large aperture radius (hence unsupported anvil radius), r , and aperture angle, θ , in the high-pressure cell itself (Fig. 1).

In order to satisfy the needs for a thin anvil having a large unsupported aperture, we chose the strongest material known, diamond. Use of a diamond anvil cell has numerous advantages, not the least of which are that the basic technology is well established and that the sample can be probed before and during the experiment due to the optical transparency of the anvil. Hence, the thickness, pressure and other characteristics of the pre-compressed sample can be determined at the outset of the Hugoniot experiment.

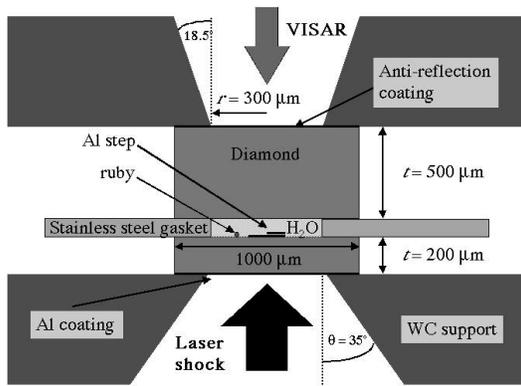


FIGURE 1. Schematic cross-section of diamond-cell configuration used for laser-driven shock experiments on precompressed samples. Wide openings (300 μm radius holes) in tungsten carbide (WC) supports allow ample shock laser entry ($\theta = 35^\circ$ opening) and VISAR access (18.5° opening). Thin diamonds are pushed together to apply pressure on a small sample of water (~30 nL) held in a hole within a stainless steel gasket 100 μm thick. An Al step is glued on the thinnest diamond and used to measure the breakout times (velocities, and ultimately shock pressure) with VISAR (1). A few ruby grains are placed in the sample chamber for precompression pressure measurements via ruby fluorescence (2). There is a 1000 Å Al flash coating on the back side of the thinnest diamond to lower the critical depth of shock ablation, and an anti-reflection coating on the thicker diamond for the VISAR measurement.

A small-radius aperture in the backing for the diamond anvil is problematic because the plasma generated by ablation of the backing plate is a source of high energy x-rays that preheat the sample (3). Also, the blow-off plasma absorbs the laser beam far from the target, thus reducing the shock intensity significantly. In order to avoid preheat, plasma blow-off and laser damage to the

DAC, a wide cone angle ($\theta = 35^\circ$) and diamond support hole (300 μm radius) were incorporated into the design of the support plate for the diamond as well as the surrounding diamond cell. This geometry, although accommodating to the laser shock, provides less than optimal support for achieving very high static pressures with thin diamond flats.

Characteristics of Thin Diamond Anvils

The diamond flat can be modeled as a uniformly-loaded circular plate, such that a simple relation exists between the maximum pressure load w and the anvil thickness t

$$w = \frac{k_1 S_m t^2}{r^2} \quad (1)$$

where S_m is the maximum stress achieved in the diamond (here, the tensile strength of diamond), r is the unsupported radius and k_1 is a constant equal to 0.833 (simply supported disc) or 1.333 (disc with fixed edges) (4). For $r = 300 \mu\text{m}$ and $t = 200 \mu\text{m}$ and using the value of tensile strength of diamond, 2.8 GPa (5), we arrive at a maximum load of between 1.0 (simply supported) and 1.7 GPa (fixed edges) (Fig. 2).

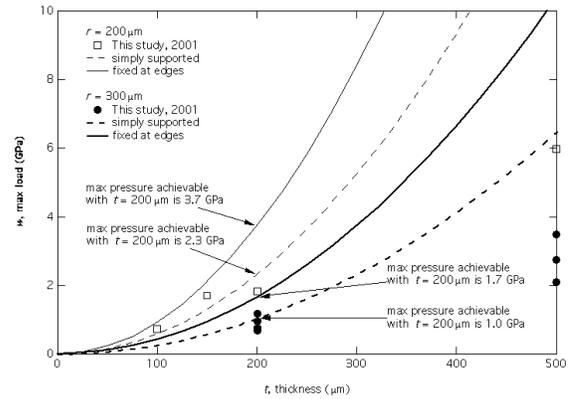


FIGURE 2. Maximum pressure load w achievable assuming a diamond disc of thickness t that is simply supported (dashed line) or fixed at edges (solid line) with an unsupported radius $r = 200 \mu\text{m}$ (thin) or 300 μm (thick). Observations of maximum pressures achieved with $r = 200 \mu\text{m}$ (open squares) and with $r = 300 \mu\text{m}$ (closed circles) are consistent with these expectations.

The thin diamond flats are fragile and flex under pressure (Figs. 3, 4), with the maximum amount of deflection y_m described by,

$$y_m = \frac{k_2 w r^4}{E t^3} \quad (2)$$

where E is the elastic modulus (1050 GPa for diamond) and $k_2 = 0.780$ (simply supported) or $k_2 = 0.180$ (fixed edges) (4). Fig. 5 shows good agreement of this simple model with our measurements.

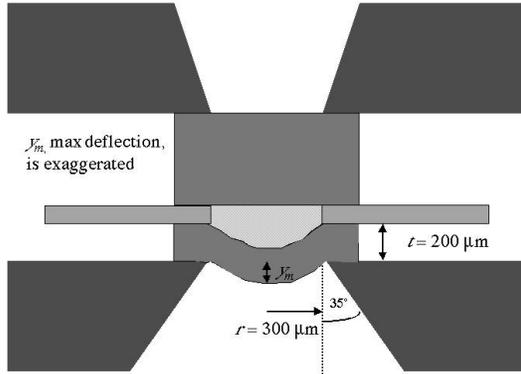


FIGURE 3. Schematic cross-section illustrating flexure of thin diamond flat (highly exaggerated).

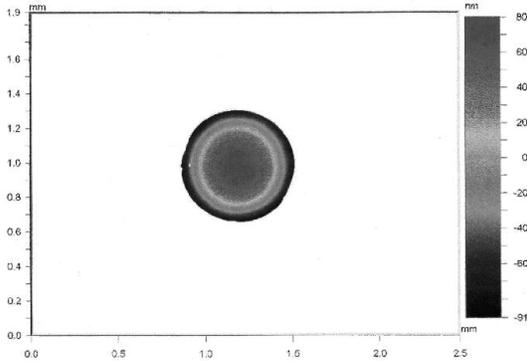


FIGURE 4. Typical white-light interferometric measurement showing the symmetric deflection of a diamond flat under compression: total deflection is $0.170 \mu\text{m}$ for $t = 200 \mu\text{m}$, $r = 300 \mu\text{m}$ at a pressure of 0.4 GPa .

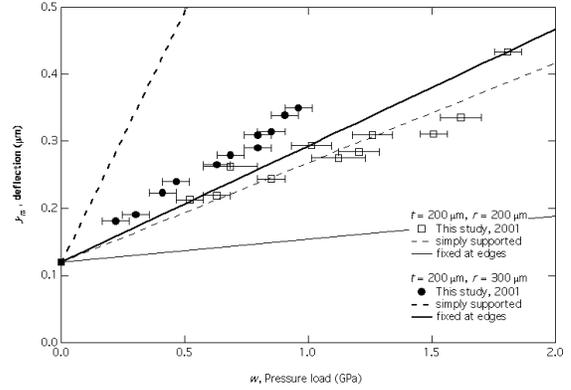


FIGURE 5. Pressure load vs. maximum deflection for $t = 200 \mu\text{m}$, $r = 200 \mu\text{m}$ (open squares, thin lines) and $t = 200 \mu\text{m}$, $r = 300 \mu\text{m}$ (closed circles, thick lines). Maximum deflection assuming simply-supported (dashed line) and fixed-at-edges (solid line). For the smaller unsupported radius, the measurements agree with the simply-supported theory, whereas for the larger unsupported radius the measurements are nearer to the fixed-at-edges condition.

SAMPLE CHARACTERIZATION

Determining the initial pressure–density–internal energy conditions (P_0 , ρ_0 , E_0) in the precompressed sample is key for Hugoniot measurements. The precompression pressure P_0 was measured in the water via ruby fluorescence (2). Using the equation of state of water by Saul and Wagner (6), the initial density ρ_0 and energy E_0 were determined.

White-light interferometry was used to determine the product of the index of refraction and distance nd (7). The height of the Al step (known *a priori*) placed into the sample to aid in the VISAR measurements was used as d ; determining the index of refraction for the compressed water sample was therefore straightforward (Fig. 6). As the index of refraction of water increases with pressure, it is necessary to obtain an accurate starting value for VISAR measurements, which look through the precompressed water.

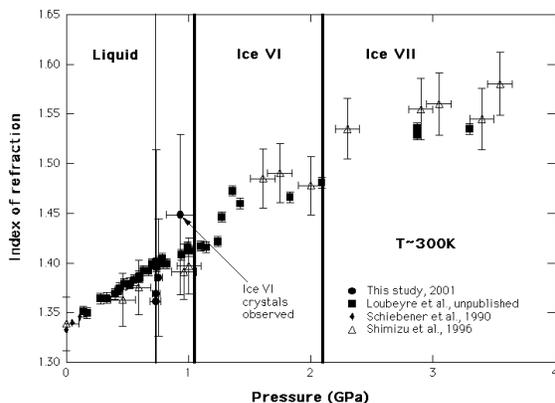


FIGURE 6. Index of refraction as a function of pressure for H₂O water, ice VI and ice VII at room temperature. The values obtained in the present study (closed circles) compare well with previous results of Loubeyre et al., (closed squares) (8); Schiebener et al., (closed diamonds) (9); and Shimizu et al., (open triangles) (10). In one run, ice VI crystals were observed at a pressure less than the nominal freezing pressure, indicating a possibly metastable condition (11).

CONCLUSION

Despite the fragility of the diamond flats, it was possible to precompress water to ~1 GPa ($\rho_0 \sim 1.2 \text{ g/cm}^3$), as predicted by the above models. These precompressed samples were laser-shocked up to pressures of ~200 GPa and temperatures to ~10,000 K (see 12, 13). In order to achieve higher initial pressures (and therefore densities), the laser intensity and duration must be increased to allow for thicker diamonds flats. For instance, the maximum load that a 500 μm thick diamond flat could withstand is predicted to lie between 6.5 (simply-supported) and 12.5 GPa (fixed-at-edges).

These results also pave the way for laser-Hugoniot measurements on mixtures, including H₂ and He, at high densities and variable temperatures.

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