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ABSTRACT

Using intense magnetic pressure, a method was developed to launch flyer plates to velocities in excess of 20 km/s. This technique was used to perform plate-impact, shock wave experiments on cryogenic liquid deuterium (LD_2) to examine its high-pressure equation of state (EOS). Using an impedance matching method, Hugoniot measurements were obtained in the pressure range of 30-70 GPa. The results of these experiments disagree with previously reported Hugoniot measurements of LD_2 in the pressure range above ~40 GPa, but are in good agreement with first principles, *ab-initio* models for hydrogen and its isotopes.

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The high-pressure EOS of hydrogen and its isotopes has been the subject of considerable interest, principally due to the importance of the EOS to such areas as inertial confinement fusion, planetary astrophysics, and our fundamental understanding of warm dense matter. Until recently, the most widely accepted EOS model was the Sesame model [1]. Prior to 1997, Hugoniot measurements of liquid hydrogen and deuterium had been limited to the pressure range below approximately 20 GPa [2], which is accessible by conventional gas-gun, plate-impact experiments. However, recent measurements from laser-driven experiments [3] at pressures of 20-300 GPa suggest that LD_2 is much more compressible than previously thought. The results from these laser-driven experiments suggest a maximum compression in excess of $\rho/\rho_0 = 6$, which deviates significantly from the Sesame EOS that predicts a maximum compression of approximately 4.4.

Despite several efforts to model this apparent increase in compressibility, the theoretical models based on first principles, *ab-initio* methods [4-6] are unable to describe the experimental results above ~40 GPa. Rather, these models tend to corroborate the stiffness of the Sesame EOS at high shock pressures. To the best of our knowledge, only empirical EOS models [7], in which parameters are adjusted to fit experimental data, have been able to reproduce the anomalously large maximum in compression. The inability to resolve this discrepancy has raised concern that either our understanding of the physics governing the EOS of these simple elements is lacking or there is a systematic error in the experiments.

Some of the concerns center around the small sample sizes and the method used in the laser experiments to drive the shock wave. Considering the relatively high shock velocities and the extremely high sound speeds in the shocked state, the experimental measurements were made on a few ns time scale, which limited the overall accuracy of the EOS data. Furthermore, the use of ablatively driven shock waves raises concerns regarding the duration, planarity, and constancy of the shock wave.

Given the significant discrepancy between theory and experiment, it is desirable to obtain independent EOS measurements of LD_2 with sufficiently different experimental techniques that are not subject to the limitations listed above, and that have the potential for increased accuracy. Recently, a new capability has been developed to isentropically compress materials to high pressures [8] using the intense magnetic pressure produced by the Sandia Z accelerator [9]. This new capability has been used to launch relatively large flyer plates to velocities about three times higher than that possible using conventional gas gun technology. The flyer plate technique for performing high-pressure shock wave experiments is particularly attractive for several reasons. First, the experiments are plate-impact experiments, and thus produce a well-defined shock loading of the sample, with a substantial duration of constant pressure (to 30 ns). Second, relatively large sample diameters and thicknesses are possible, thus increasing the accuracy of the EOS data. Finally, the large sample sizes allow for multiple and redundant diagnostics to be fielded which further enhance the accuracy and confidence of the data.

The experimental configuration used to obtain Hugoniot data with the magnetically driven impact technique is shown in Fig. 1. The necessary cryogenics were provided by an expendable cryocell connected to a survivable cyrostat [10]. The cavity of the cryocell was defined by a stepped aluminum (6061-T6) pusher plate and a *z*-cut sapphire window, with cavity dimensions of approximately 5 mm in diameter and 300 and 600 μm in thickness. LD_2 samples were condensed in the cryocell by filling the cavity with high purity deuterium gas at 18 psi, cooling the cryocell to its equilibrium temperature of 16-18 K, and then warming the cell to 22.0

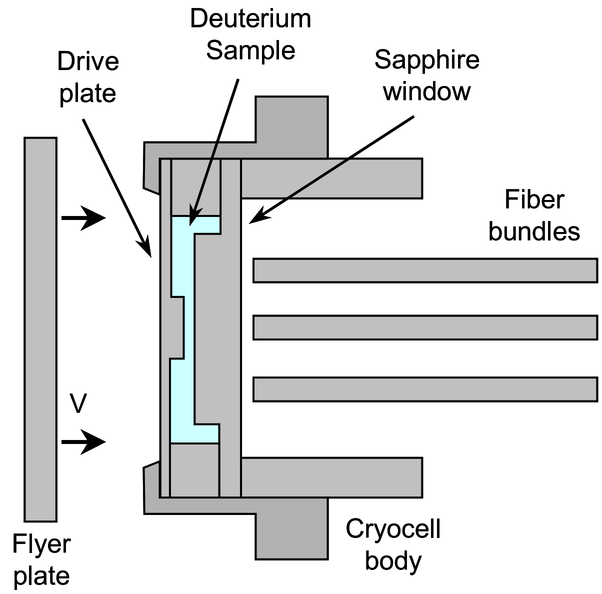

Fig. 1: Experimental configuration used to obtain Hugoniot measurements in plate-impact shock wave experiments. Note the drawing is not to scale.

± 0.1 K [10]. This produced a quiescent LD_2 sample below the boiling point of about 25 K, with nominal initial density of 0.167 g/cm^3.

Shock waves were generated by planar impact of either an aluminum (6061-T6) or a titanium (Ti-Al6V4) flyer plate onto the aluminum pusher plate at the front of the cryocell. The rectangular flyer plate, approximately 12 x 25 mm in lateral dimension and ~200-300 μm in thickness [11], was accelerated across a nominal 3 mm vacuum gap by the magnetic field. Aluminum flyer velocities as high as 21 km/s have been achieved, capable of generating shock states to ~500 GPa in the aluminum drive plate and transmitting up to ~70 GPa shock waves into LD_2. Conventional velocity interferometry [12] (VISAR) was used to directly measure the velocity history of the flyer plate from launch to impact with an accuracy of ~0.5%.

The shock response of LD_2 was diagnosed with a number of fiber-optic coupled diagnostics. Typically several optical fiber bundles of 100 and 200 μm diameter fibers were used, allowing multiple, redundant diagnostics, including (*i*) conventional VISAR, (*ii*) fiber-optic shock break out (FOSBO), and (*iii*) temporally and spectrally resolved spectroscopy. Fig. 2 shows sample data obtained from a typical LD_2 experiment. In all, 16 channels of data were obtained for each experiment, allowing up to 16 independent measurements of the shock velocity, U_s, in LD_2 and up to 4 independent measurements of U_s in the aluminum drive plate. The uncertainty in U_s was ~2-3% from the measured transit time through the cell and the initial cell dimensions. Since the uncertainties were due to random errors, statistical techniques could be used to decrease the uncertainty in U_s to approximately 1% and 2% for the LD_2 sample and the aluminum drive plate, respectively [13].

The VISAR records for the higher pressure experiments confirm the constancy of the pressure drive obtained from the flyer plate-impact, as shown in Fig. 2. In this case the VISAR velocity is indicative of U_s in the LD_2 because at shock pressures above ~30 GPa LD_2 becomes reflective [3]. From these records it was determined that the shock pressure was constant to better than 1% as the shock traversed the cryocell. It is emphasized that a correction of $1/n_0$,

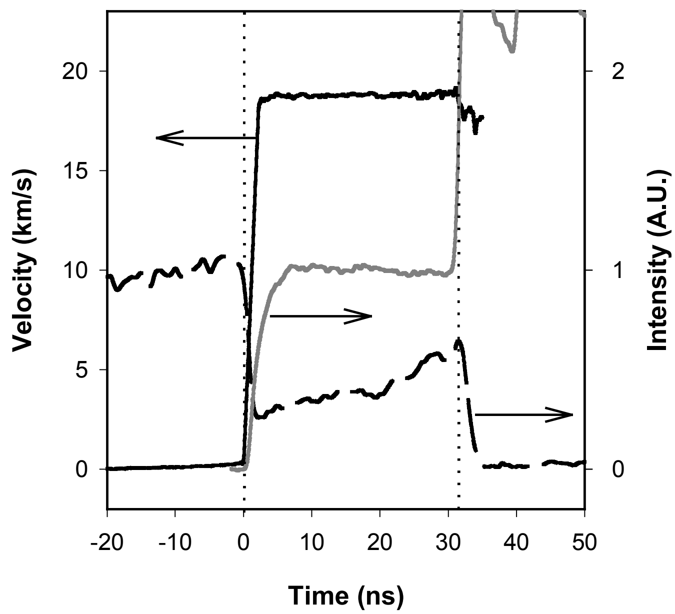

Fig. 2: Typical data obtained in LD_2 experiments; (*i*) VISAR record of the shock front (solid line), (*ii*) FOSBO record (dashed line), and (iii) self-emission record (gray line). Vertical dotted lines indicated break out of the shock from the aluminum/LD_2 interface and the arrival of the shock at the LD_2/sapphire interface.

where n_0 is the refractive index of the ambient LD_2, was applied to the usual velocity per fringe (VPF) constant for the interferometer to account for the diminishing thickness of LD_2 through which the laser light propagated as the shock front traversed the cryocell. We found that this index correction, amounting to 11.5%, was necessary to obtain consistency between U_s directly measured by the VISAR and U_s inferred from the transit time measurements.

For the lower pressure experiments, the shock front was not sufficiently reflective to obtain VISAR measurements. However, U_s was obtained for all experiments using the FOSBO and self-emission data. As seen in Fig. 2, both of these measurements provided a clear signature of shock arrival at the aluminum/LD_2 and the LD_2/sapphire interfaces. Also, in all experiments, high quality spectra were obtained over the continuous wavelength region between 250 and 700 nm. The detailed analysis of the spectral dependence of the self-emission, which provides a measure of the temperature of the shocked LD_2, will be discussed in a future publication. We emphasize that the constancy of the emission signal during the traversal of the shock through the cryocell further verifies the constancy of the pressure states achieved with the flyer plate-impact, as the intensity of emission is proportional to the pressure of the LD_2 to the ~1.75 power [14].

An impedance matching method, utilizing the Hugoniot jump conditions [15], was used to obtain Hugoniot points for the shocked LD_2. As shown in Fig. 3, the initial shocked state of the aluminum drive plate is described in the pressure-particle velocity (P-u_p) plane by the point labeled A, and the shocked state of LD_2 is constrained to lie on a straight line, with the slope of the line given by $\rho_0 U_s$, where ρ_0 is the initial density of the LD_2 sample. An EOS model for aluminum [16] was used to calculate the release isentrope from state A in the aluminum drive plate. The intersection of the calculated release isentrope and the line defined by the LD_2 shock velocity determines u_p of the shocked deuterium sample. The uncertainty in u_p for LD_2, typically

2-3%, was determined from the uncertainty in the shocked state of the aluminum drive plate, and thus from the uncertainty in U_s for the aluminum drive plate. The density compression was then determined from the jump conditions using the expression $\rho/\rho_0 = U_s/(U_s - u_p)$.

The accuracy in the impedance matching technique depends upon two factors: the accuracy in the measurement of U_s in the LD_2 and the accuracy of the calculated release isentrope for aluminum. The quality of the data shown in Fig. 2, and the multiplicity of the U_s measurement for each experiment indicates that U_s in the LD_2 is determined quite accurately. To determine the accuracy of the calculated release isentrope, release experiments in aluminum using a low density (200 mg/cm^3) silica aerogel were performed. This technique is similar to that used by Holmes, et al., to measure the aluminum release from ~80 GPa [17]. Direct impact experiments were performed to generate Hugoniot data for the aerogel between 30 and 75 GPa. Experiments were then performed in which a shock was transmitted from the aluminum drive plate into the silica aerogel, which simulates unloading to the LD_2 state. The measured U_s for the aerogel in the release experiment, along with the measured aerogel Hugoniot, determines a point in P-u_p space that the aluminum release isentrope must pass through. A typical result is shown in Fig. 3, in which a release point in aluminum was measured from an initial shock state of ~500 GPa (the point labeled B in Fig. 3). These measurements confirm the validity of the release calculations in aluminum over the pressure range of interest, and make a strong case for the procedure indicated in Fig. 3 to obtain the LD_2 Hugoniot results reported in the present work.

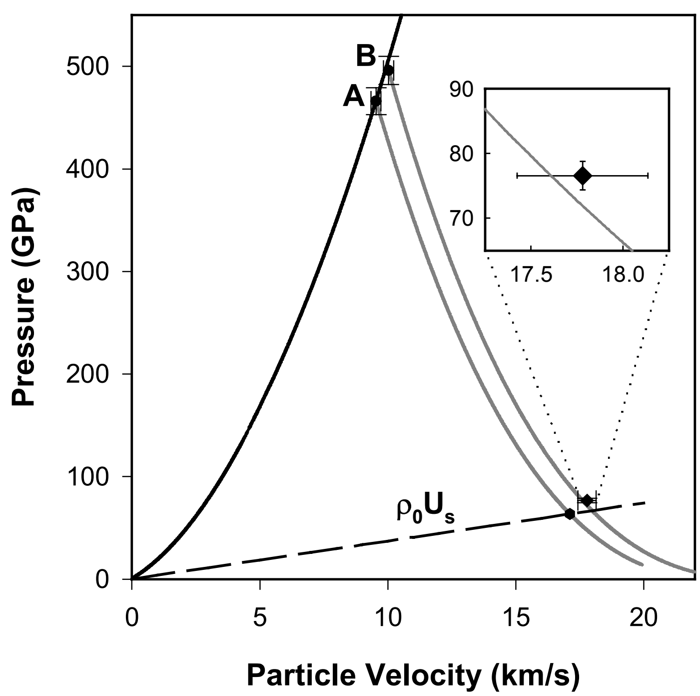

Fig. 3: Impedance matching method and aluminum release measurement. Solid gray lines are calculated release isentropes from two separate experiments: state A indicates the initial shocked state of the aluminum drive plate in a LD_2 experiment; state B indicates the initial shocked state of an aluminum sample in a silica aerogel release experiment.

The pressure-density compression states determined in this way for a total of eight experiments are displayed in Fig. 4. The lowest pressure experiment was found to be in good agreement with the results reported from the earlier gas gun experiments and the lower pressure laser experiments. However, at higher pressures, particularly the data centered around 70 GPa, there is a distinct deviation between the present results and those reported from the laser-driven experiments. Further, the data obtained from our study are in quite good agreement with the predictions from the *ab-initio* models throughout the entire range of pressures investigated.

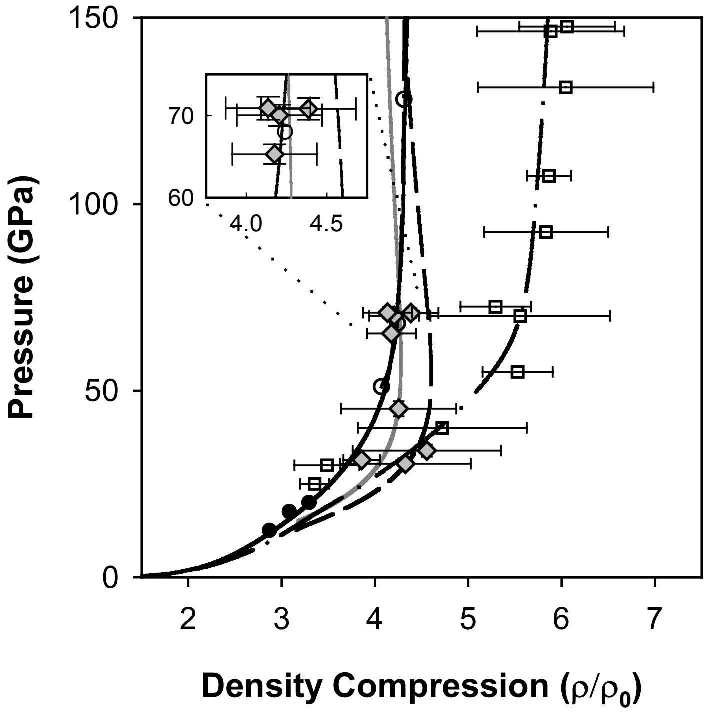

Fig. 4: Deuterium Hugoniots. Theoretical models: Sesame (solid line [1]); Tight Binding (gray line [4]); GGA-MD (dashed line [5]); PIMC (open circles [6]); Ross (dot-dash line [7]). Experiments: Gas gun (filled circles [2]); Laser-driven (open squares [3]); This work (gray diamonds).

It should be noted that both the laser technique and the magnetically driven flyer technique are new and not entirely proven, and are therefore subject to potential systematic errors. We took great pains to identify, address, and minimize the potential systematic errors in the present work. In particular we assessed the constancy of the pressure drive obtained with the magnetically driven flyer through the VISAR and spectroscopy measurements, and the accuracy in the impedance matching technique through the silica aerogel experiments. Further we validated the magnetically driven flyer plate technique through aluminum symmetric impact experiments to ~500 GPa. The results of these experiments, which will be described in a future publication, were in very good agreement with published data on aluminum at high-pressure. Finally, we have also examined potential density and pressure gradients in the flyer through improved MHD simulations [18]. The results of all of these studies indicate that the experimental technique and the conclusions drawn from the measurements are internally consistent.

There are also sources of potential systematic error in the laser-driven work that center around the diagnostic used to determine the density compression in the shocked state. The use of transverse radiography to determine the location of the shock front and the interface is a non-traditional shock diagnostic, which has not been validated on a known material. A few potential problems associated with this type of measurement warrant discussion. First, any deviation from a constant pressure shock will result in an incorrect inference of density when using the Hugoniot jump conditions. In particular, hydrocode simulations that emulate the radiography measurements indicate that errors of up to 10-15% in the determination of density can result from modest deviations from constant pressure shocks, depending upon the pressure history. Second, it is difficult to infer velocity from a trajectory measurement to a high degree of accuracy. In particular, it was reported in the laser work that good agreement was found between a VISAR measurement of the shock front and the radiography results. However, the index correction to the VPF was not used in the analysis of the VISAR result in that study, which leads to concern regarding the accuracy of the radiography result. Finally, the pressure at which LD_2 appears to undergo a large increase in compression in the laser results corresponds to drive pressures at which one expects the aluminum driver to melt under compression [19]. This suggests the possibility of an ill-defined interface between the aluminum drive plate and LD_2 sample.

As a final point, it should be noted that recent laser-driven, double shock experiments have been reported as an independent confirmation to the laser-driven Hugoniot measurements [20]. Recent theoretical work [21], in which these double shock experiments are compared with *ab-initio* models, shows that these experiments are not able to distinguish between the theoretical models for initial shock pressures below ~100 GPa. Thus, there is no disagreement between the double shock experiments and the present work over the range of pressures studied. Furthermore, these are integrated experiments that depend not only on the principal Hugoniot, but also on the LD_2 properties upon re-shock. Thus, conclusions regarding the principal Hugoniot cannot be unambiguously determined and should be viewed with caution.

In conclusion, we have performed high-velocity plate-impact, shock wave experiments to investigate the high-pressure EOS of LD_2. The results of these experiments are in agreement with theoretical models based upon first principles, *ab-initio* methods, and corroborate the stiff shock response at pressures up to ~70 GPa predicted by the Sesame EOS. Further, the present results disagree with earlier results reported from laser-driven experiments at pressures above ~40 GPa. Clearly there is a need for further theoretical and experimental work to resolve this discrepancy, and to address whether there are systematic errors in either experimental technique, or whether there is a physical phenomenon responsible for the different response at the two differing time scales of these experiments. However, in light of the fact that both experimental techniques are new and not entirely proven, it is critical that they both be subjected to intense scrutiny.

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