Properties of fluid deuterium under double-shock compression to several Mbar

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The compressibility of fluid deuterium up to several Mbar has been probed using laser-driven shock waves reflected from a quartz anvil. Combining high-precision (~1%) shock velocity measurements with the double-shock technique, where differences in equation of state (EOS) models are magnified, has allowed better discrimination between theoretical predictions in the second-shock regime. Double-shock results are in agreement with the stiffer EOS models—which exhibit roughly fourfold single-shock compression—for initial shocks up to 1 Mbar and above 2 Mbar, but diverge from these predictions in between. Softer EOS models—which exhibit sixfold single-shock compression at 1 Mbar—overestimate the reshock pressure for the entire range under study. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1778164]

The properties of hydrogen and its isotopes at pressures near 1 Mbar and temperatures of the order of 1 eV are fundamental to the modeling of massive planets,[1] brown dwarfs,[2] and inertial confinement fusion targets.[3] Under these conditions hydrogen is a partially degenerate, strongly correlated, conducting fluid, a complex system whose equation of state has been the subject of extensive study.[4–9]

Experimental data in this regime have been obtained using shock wave studies on a variety of different platforms. Early experiments using a two-stage light-gas gun achieved pressures in deuterium of 0.2 Mbar and 0.9 Mbar under single- and double-shock compression, respectively.[10] The development of laser-driven shock wave experiments allowed single-shock pressures up to 3.4 Mbar (Ref. 11) and double-shock pressures up to 6 Mbar to be probed.[12] More recently, single-shock pressures up to 1 Mbar (Ref. 13) were achieved using magnetically driven flyer plates and up to 1.2 Mbar using explosively driven, converging systems.[14] Although the region of phase space common to these experiments is small, a controversy has appeared to emerge. The laser-driven single- and double-shock results are consistent with a softer equation of state (EOS),[6] giving sixfold compression on the principal Hugoniot at 1 Mbar. This soft EOS was fit to reshock temperatures in earlier gas gun experiments.[15] Single-shock measurements near 1 Mbar performed on other platforms failed to observe such a high compression,[13,14] instead finding slightly over fourfold compression as predicted by the stiffer EOS models.[7–9] Tests up to 0.75 Mbar on multiple shock states also appeared to be consistent with stiffer models.[13]

To address this controversy, we have undertaken a series of highly precise, laser-driven shock experiments to study the double-shock behavior of fluid deuterium at pressures up to nearly 9 Mbar, the highest pressures yet measured in a deuterium EOS experiment. We cover a wider range of pressures and have significantly higher measurement precision than earlier double-shock experiments,[12] using an approach that determines shock velocities to 1% uncertainty and essentially eliminates any systematic errors from shock unsteadiness and nonplanarity. The double-shock technique magnifies the expected differences in the experimental observables (i.e., measured shock speeds) predicted by various EOS theories, thus providing a more sensitive platform for discriminating between models. In addition, such measurements probe a denser region of phase space—a regime more relevant to planetary interiors[1]—than can be accessed using single shocks alone, although there is no model-independent way of separating the individual contributions of first and second shocks. Results show that none of the available models accurately predict double-shock measurements over the entire pressure range under study. Stiffer EOS models agree with our data for first shocks up to ~1 Mbar and above 2 Mbar, but underestimate the reshock pressures in between; softer EOS models consistently overestimate the reshock pressure.

This experiment was performed on the OMEGA laser at the University of Rochester, a neodymium-doped phosphate glass system that operates with frequency-tripled, 0.35 μm light.[16] To generate the shock pressures explored in these experiments, laser energies of 440–3100 J were delivered using a square pulse 3.7 ns in duration. The laser focal region was smoothed using distributed phase plates, producing a uniformly irradiated spot 800 μm in diameter. Targets consisted of a z-cut, α-quartz anvil mounted on the upper step of a diamond-turned aluminum pusher which was attached to a copper cell filled with cryogenic deuterium (see Fig. 1). A plastic ablator was used to reduce hard x-ray generation.

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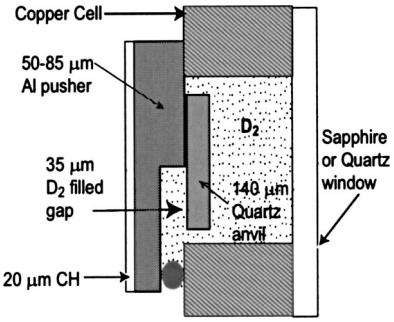

FIG. 1. Characteristic cryogenic deuterium target design. Dimensions are for one of the three types of target.

Three different thicknesses were used for the ablator-pusher combination: 20 μm of CH on a 90–130 Al step (90 μm lower step and 130 μm upper step); 20 μm of CH on a 50–85 μm Al step, and 20 μm CH plus 80 μm of CH-Br (plastic with 2% Br by atomic weight) on a 50–85 μm Al step. The quartz anvil was glued to the upper step with a glue thickness of ~1 μm and hung over the lower step as shown in Fig. 1. The deuterium sample explored in this experiment is trapped within the 35–40 μm region between the quartz anvil and the thin Al plate. By observing the solid-liquid transition in deuterium and using the well-known properties of deuterium on the saturation line,[17] we determined that the deuterium density was 0.174 g/cm^3. At this density and at the probe laser wavelength of 532 nm, the index of refraction was calculated to be 1.1381.[17] At room temperature, the density of quartz was measured to be 2.65 g/cm^3 and the refractive index along its c axis at 532 nm was found to be 1.547. Because quartz has such a low thermal expansivity, its density changes by only 0.5% and its refractive index by 0.1% when cooled to <20 K, changes that are negligible for our purposes and can be ignored.

The shock diagnostic was a line-imaging velocity iterferometer system for any reflector[18,19] (VISAR) which measures the Doppler shift of a moving reflector. Two VISARs with different velocity sensitivities were used to resolve 2π phase shift ambiguities that occur at shock break out from the aluminum and upon transit of the shock front from deuterium into quartz. The velocity sensitivities for the two VISAR instruments were 6.069 and 14.138 μm/ns/fringe for deuterium and 4.465 and 10.400 μm/ns/fringe for quartz. Postprocessing of the VISAR images can determine the fringe position to ~5% of a fringe; since the measured shock velocities are 25–45 μm/ns in deuterium and 14–24 μm/ns in quartz, multiple fringe shifts allow the precision of the shock velocity measurement to be ~1%. The probe source was an injection-seeded, Q-switched, yttrium-aluminum garnet laser, operating at a wavelength of 532 nm with a pulse length of ~25 ns. Streak cameras with temporal windows of ~3 ns were used to detect the reflected probe signal. The time resolution of the VISAR and streak camera system was about 40–50 ps.

A sample VISAR trace is shown in Fig. 2(a) and the resulting velocity profile inferred from the fringe positions is given in Fig. 2(b). The three clear events observed in these records are marked by fringe (and hence velocity) shifts: The first shift represents the velocity jump that occurs when the shock crosses the aluminum-deuterium interface; the second shift, at time t_x, corresponds to the drop in shock velocity as the shock moves across the deuterium-quartz interface. Shock velocities immediately before and after the shock crosses the deuterium-quartz interface are the primary observables used in this work. The third shift is the jump in velocity observed in quartz when the first shock reverberating in the compressed deuterium gap catches the leading shock front in quartz.

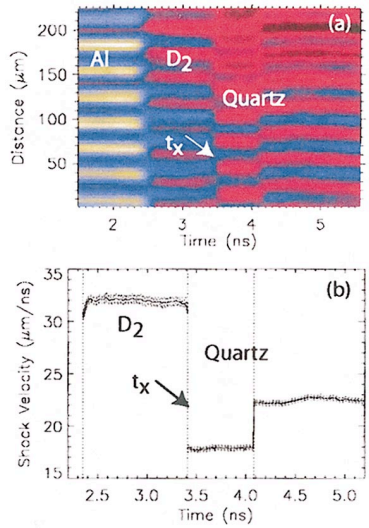

FIG. 2. (Color). (a) Sample VISAR trace showing the signal from the reflecting shock front in deuterium and quartz. (b) Resulting velocity profile extracted from the VISAR trace in (a). Dotted lines above and below the main trace indicate the error at each time step. The shock traverses the deuterium-quartz interface at time t_x.

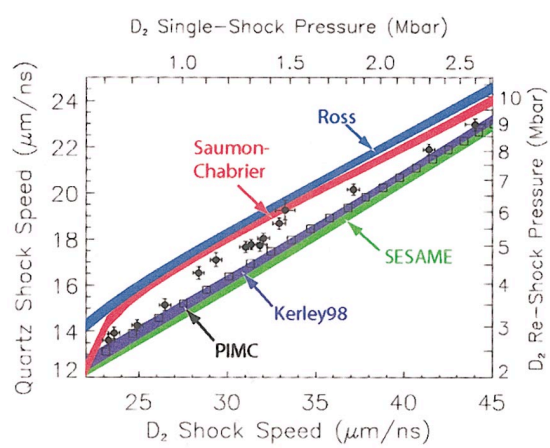

FIG. 3. (Color). Double-shock data from this study shown as filled circles with error bars. Predictions for five different EOSs are shown: SESAME (Ref. 4), Kerley98 (Ref. 7), Saumon-Chabrier (Ref. 5), Ross (Ref. 6), and PIMC (Ref. 8) (black squares) where the line thicknesses correspond to the quartz Hugoniot uncertainty. The softer models (Saumon-Chabrier, Ross) which have sixfold maximum compressibility on the principal Hugoniot, predict higher final reshock pressures than the stiffer models (Kerley98, SESAME, PIMC), which have 4–4.5-fold compressibility. The estimated D_2 single-shock and reshock pressures on the top and right axes are based on the Kerley98 model for D_2 and the measured quartz Hugoniot, respectively.

TABLE I. Measured shock velocities in deuterium $U_s(D_2)$ and quartz $U_s(Q)$ along with estimated first shock (P_1) and measured second-shock pressures (P_2). First-shock pressures are calculated using Kerley98 (first value) and Ross models (second value) for the measured $U_s(D_2)$. Superscripts on shot numbers give the target type.

Shot #	$U_s(D_2)$ (μm/ns)	$U_s(Q)$ (μm/ns)	P_1 (Mbar)	P_1 (Mbar)
27 869[a]	37.1±0.3	20.2±0.3	1.8/1.9	6.7±0.2
27 879[b]	29.4±0.3	17.117.1±0.3	1.1/1.2	4.6±0.2
27 934[a]	32.9±0.4	18.7±0.3	1.4/1.5	5.6±0.2
27 940[a]	31.8±0.4	17.7±0.3	1.3/1.5	5.0±0.2
29 021[b]	24.9±0.3	14.3±0.3	0.8/0.9	0.3±0.2
29 398[a]	44.0±0.6	23.0±0.3	2.6/2.7	8.9±0.3
29 401[a]	41.4±0.3	21.9±0.3	2.3/2.4	0.8±0.2
29 412[b]	23.3±0.3	13.6±0.3	0.7/0.8	2.7±0.2
30 122[b]	28.4±0.3	16.5±0.3	1.1/1.2	4.2±0.2
30 129[b]	33.3±0.6	19.3±0.4	1.5/1.6	6.0±0.3
30 134[b]	31.3±0.3	17.8±0.2	1.3/1.4	5.0±0.2
30 663[a]	32.0±0.3	18.0±0.2	1.4/1.5	5.2±0.2
31 359[c]	31.0±0.3	17.7±0.3	1.3/1.4	4.9±0.2
31 361[c]	26.5±0.3	15.1±0.3	0.9/1.0	3.5±0.2
31 363[c]	23.6±0.3	13.9±0.3	0.7/0.8	2.8±0.2

[a]20 μm CH+80 μm of CH–Br on 50 μm Al.
[b]20 μm of CH on 90 μm Al.
[c]20 μm of CH on 50 μm Al.

To extract the velocity profile we average the phase information at each time over a 20–30 μm region. To determine shock velocities at the deuterium-quartz interface, we take linear fits a few hundred picoseconds before and after t_x and extrapolate them to t_x. This eliminates ambiguities due to slight blurring of the measured velocity in a ±25 ps time window centered on t_x caused by the resolution of the VISAR and streak camera system. Figure 3 plots the results in terms of the primary experimental observables: the shock speed in deuterium and quartz. The data are tabulated in Table I.

To compare these observations with EOS models it is necessary to know the high-pressure U_s-U_p relation for quartz.[20] To determine this we performed extensive laser-driven shock measurements on quartz, complementing earlier data reported by Russian workers obtained using nuclear explosives,[21] and found that $U_s=3.798+1.312U_p$.[22] Taking into account the errors in the fit coefficients, this is in good agreement with the relation found in the early Russian work[23] ($U_s=4.200+1.280U_p$) over the range of pressures in our study.[24]

Using this fit and the impedance-matching conditions at the deuterium-quartz interface we calculate the reflected shock curves for the different EOS models. These are shown in Fig. 3, where the thickness of the lines represents the uncertainty in the linear fit to the quartz Hugoniot [for clarity, the Path Integral Monte Carlo (PIMC) results, being so close to the Kerley98 predictions, are shown as squares]. Plotting the data in terms of the experimental observables thus allows uncertainties in the quartz Hugoniot (a systematic error in *all* the data) to be separated from measurement errors in the deuterium reshock data (which are given by the error bars on the data points). Results indicate that the double-shock compressibility of deuterium at first-shock pressures between 0.7 and 1 Mbar and above 2 Mbar are consistent with predictions based on stiffer EOS models[7,8] which exhibit 4–4.5-fold compression on the principal Hugoniot (see Fig. 3). Between 1 and 2 Mbar the reshock pressure is slightly higher than predicted by these models though lower than the predictions based on softer EOS models.[5,6]

It is particularly important to recognize that these results are independent of any calculated EOS. The quartz Hugoniot is measured relative to the experimentally determined aluminum Hugoniot which is a fit to data that includes several absolutely measured points.[25]

We have considered a number of potential systematic effects that could compromise our data and address each of them below. The steadiness of shock wave velocities in our experiments varied from shot to shot, depending on the quality of the laser drive, ranging from fractions of a percent to several percent over a few nanoseconds. Our new technique of determining shock velocities at essentially a single point in time using continuous measurements is not affected by such variations, unlike the transit time measurements which were used in the earlier reshock experiments.[12] To establish this we performed extensive hydrodynamic simulations of our experimental arrangement using shock waves with a wide range of unsteadiness, rising and decaying. We saw no deviations from the steady shock case if the velocities were extrapolated to time t_x. This is confirmed experimentally, where we observe no difference between the shots which were essentially steady and those which had several percent unsteadiness.

Shock nonplanarity is also a potential problem, especially for an experiment that requires measurement of a break-out event at spatially separated positions.[12] However, since our measurement is performed at a localized point in space, we are not subject to such errors. Nonplanarity could affect our measurements if the wave is incident on the deuterium-quartz interface at a large enough angle to undergo significant refraction. Based on our measurement of the small curvature observed at the deuterium-quartz interface, we infer that the largest incident angles present in our experiments are 3° to target normal. The resulting change in the projected shock speed is less than 0.1% and can be neglected.

X-ray preheating of our target system is a process which would tend to make our deuterium sample less compressible. Using an etalon sensitive to motions as low as 0.1 μm/ns, we did not observe any expansion of the aluminum pusher prior to shock break out. In addition, for targets shot at similar laser energies, we did not see any difference in the results whether we used a 50 μm or 90 μm thick aluminum pusher. Since the attenuation length for a 1.55 keV x ray (just below the K edge of aluminum) is 10 μm, the extra 40 μm of Al would be expected to reduce the x-ray fluence by a factor of 50. The absence of any difference between results from these targets indicates that x-ray preheat is negligible for these experiments. (See Table I.)

Using laser-driven shock waves, we have probed the be-

havior of fluid deuterium up to reshock pressures near 9 Mbar. For single-shock pressures up to ~1 Mbar (~ 3.5 Mbar under double shock) we find for the first time that laser-driven, magnetically driven flyer plate,[13] and high-explosive driven[14] shock experiments all give results consistent with a stiffer EOS. Our results are also consistent with stiffer models above single-shock pressures of ~2 Mbar; however, between 1 and 2 Mbar, measured reshock pressures are greater than predicted by these models though lower than predicted by softer models. Since reshock pressures are the combined result of first- and second-shock compressions our results between 1 and 2 Mbar indicate either that the first shock generates compressions higher than ~4.3–4.4, or that the second shock, which achieves states around eightfold to ninefold compression, is less compressible than predicted by the stiffer models. Given the initial controversy surrounding the measurements below 1 Mbar it is critical that these higher pressure results be reproduced on other experimental platforms.

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