Use of a Wave Reverberation Technique to Infer the Density Compression of Shocked Liquid Deuterium to 75 GPa

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A novel approach was developed to probe density compression of liquid deuterium (L-D_2) along the principal Hugoniot. Relative transit times of shock waves reverberating within the sample are shown to be sensitive to the compression due to the first shock. This technique has proven to be more sensitive than the conventional method of inferring density from the shock and mass velocity, at least in this high-pressure regime. Results in the range of 22–75 GPa indicate an approximately fourfold density compression, and provide data to differentiate between proposed theories for hydrogen and its isotopes.

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Recently, there has been increased interest in the high-pressure equation of state (EOS) of hydrogen and its isotopes. The behavior of this element, which has significant implications for such areas as inertial confinement fusion and planetary astrophysics, has become the subject of several investigations, both experimental [1–4] and theoretical [5–11]. In the mid 1990s liquid deuterium (L-D_2) EOS data obtained using laser-driven shocks [1] indicated a maximum compression along the Hugoniot of approximately sixfold, a substantial increase over the approximately fourfold shock compressibility predicted by the widely used Sesame EOS [12]. Subsequent laser-driven experiments [2], in which the L-D_2 pressure was inferred upon reshock from an aluminum anvil, appeared to have confirmed this anomalously high compression. More recently, magnetically driven flyer plates were used to perform conventional plate-impact experiments on L-D_2 [3]. The results of these experiments are in better agreement with the Sesame EOS, exhibiting a stiffer response with a maximum compression of approximately fourfold. Finally, Hugoniot experiments using an independent, convergent geometry technique have recently been performed and appear to confirm this stiffer response [4]. These Hugoniot data, shown in Fig. 1, have created a controversy regarding our understanding of the high-pressure response of L-D_2.

The primary reasons for why this controversy has yet to be resolved relates to the difficulty of these experiments, and the high precision in shock and mass velocity measurements required to accurately infer compression. In typical shock wave experiments the shock velocity, U_s, and mass velocity, u_p, are measured, which is the case in all of the Hugoniot experiments shown in Fig. 1. Conservation of mass and momentum are then used (the Rankine-Hugoniot jump conditions [15]) to determine pressure, $P = \rho_0 U_s u_p$, and density compression, $\Lambda \equiv \rho_1/\rho_0 = U_s/(U_s - u_p)$. However, uncertainties in U_s and u_p are magnified when converting to the density plane; specifically, the fractional errors in U_s and u_p are multiplied by the factor $(\Lambda - 1)$. Thus, for highly compressible materials such as L-D_2, in which u_p approaches U_s, moderate uncertainties in U_s and u_p lead to significant uncertainties in Λ. Fig. 2 illustrates this point; the enhanced differences in Λ result from relatively small differences in the measured U_s-u_p response. Thus, extremely accurate measurements are needed to resolve the discrepancy using conventional measurement techniques.

In this paper we describe a novel approach to infer density compression along the principal Hugoniot that is more discriminating than the inference from measurements of U_s and u_p. This technique monitors the relative

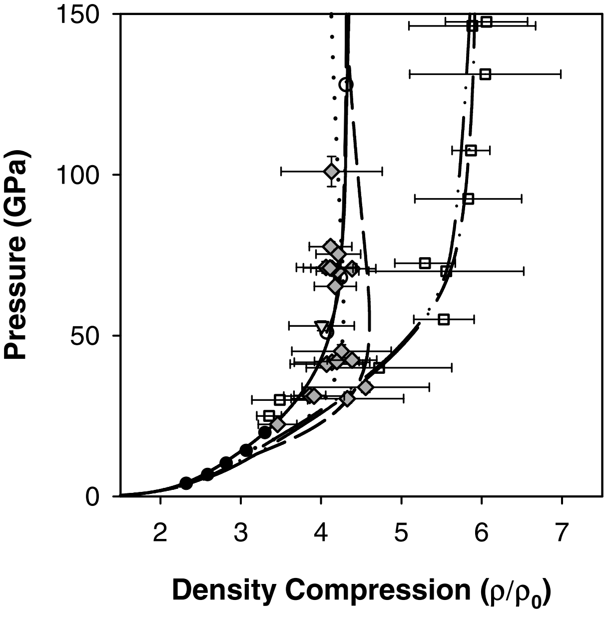

FIG. 1. L-D_2 pressure-density compression Hugoniots. Theoretical models: Sesame (solid line [12]); tight binding (dotted line [5]); GGA-MD (dashed line [6]); path integral Monte Carlo (open circles [7]); Ross (dot-dot-dashed line [8]); Young (dot-dashed line [13]). Experiments: Gas gun (filled circles [14]); laser-driven (open squares [1]); magnetically driven flyer plate (gray diamonds [3]); convergent geometry (gray triangle [4]).

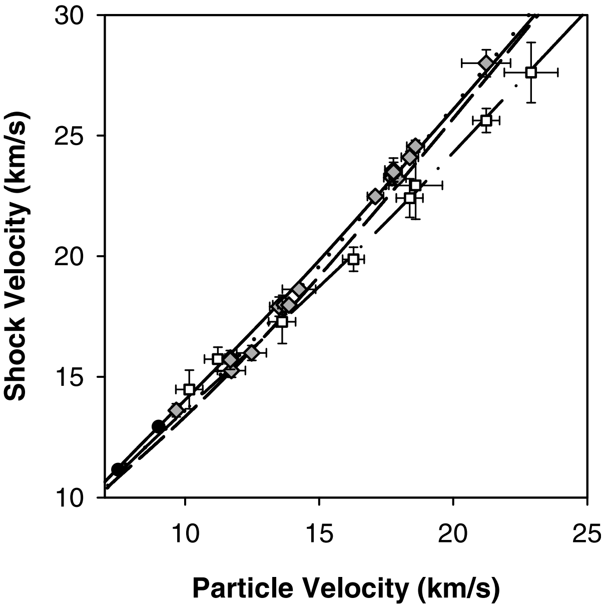

FIG. 2. L-D_2 U_s-u_p Hugoniots. Lines and symbols as in Fig. 1.

arrival time of shock waves at the interface of the L-D_2 sample and a sapphire window as the shock reverberates between the aluminum drive plate and the window. This relative timing can be used to infer the compression of L-D_2 in this higher pressure regime with better precision than the more traditional density inference from U_s and u_p. In fact, not only was it possible to confirm an approximately fourfold density compression of L-D_2 from the reverberation measurements, we were also able to distinguish between the various stiffer *ab initio* and Sesame EOS models in the pressure range studied. We point out that this experimental technique is not specific to L-D_2, but can be applied to any material. Further, since the uncertainty in such a measurement is not as sensitive to the magnitude of the density compression, it is particularly well suited for use with highly compressible materials.

The technique makes use of the necessity for containing L-D_2 in a cell composed of higher shock impedance materials (in this case aluminum and sapphire). After the shock traverses the cell, wave interactions at the L-D_2/sapphire interface result in a transmitted and a reflected shock. We exploit this by using the reflected shock to probe the location of the aluminum/L-D_2 interface. The velocities of the shock (U_s) and the aluminum/L-D_2 interface (u_p) determine the time that the reflected shock from the aluminum/L-D_2 interface reaches the L-D_2/sapphire interface. Thus, the ratio of the time between the first and second shock arrivals at the L-D_2/sapphire interface ($t_2 - t_1$) to the original transit time across the cell ($t_1 - t_0$), referred to as the reverberation ratio, is directly related to the density of the L-D_2 in the Hugoniot state.

Qualitatively one can see from the position-time plot in Fig. 3 (drawn to scale for an initial shock of ~45 GPa) that due to the substantial compression of L-D_2 upon first shock, the exact behavior of L-D_2 upon reshock has a relatively small influence on the reverberation ratio. The comparatively short reverberation time ($t_2 - t_1$) observed is directly related to the large density compression along the principal Hugoniot. Furthermore, since the reverberation ratio is obtained from a self-emission measurement on a single streak camera image, it can be determined to a high degree of accuracy, providing a particularly sensitive inference of the density compression of L-D_2 in the Hugoniot state.

One can also show quantitatively the strong dependence of the reverberation ratio on ρ_1. Analysis of the position-time plot reveals that the ratio of the reverberation time, t_r, to the original transit time, t_i, is given by

$$\frac{t_r}{t_i} \equiv \frac{(t_2 - t_1)}{(t_1 - t_0)} = \rho_0 U_{s_1}\left(\frac{1}{\rho_1 U_{s_2}} + \frac{1}{\rho_2 U_{s_3}}\right), \qquad (1)$$

where ρ_0 is the initial L-D_2 density; ρ_1 and ρ_2 are the L-D_2 densities due to the first and second shocks, respectively; and U_{s_1}, U_{s_2}, and U_{s_3} are the velocities of the first, second, and third shocks, respectively. Equation (1) indicates that apart from the measured quantities ρ_0 and U_{s_1}, the ratio t_r/t_i depends on ρ_1, ρ_2, U_{s_2}, and U_{s_3}. However, model predictions over the pressure range examined

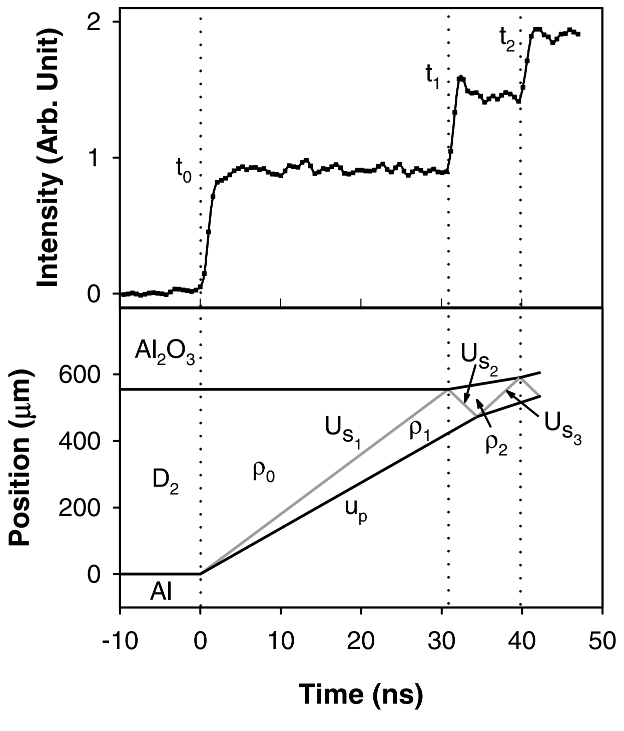

FIG. 3. Top: Typical self-emission measurement indicating shock arrival at the aluminum/L-D_2 interface (t_0), the first shock arrival at the L-D_2/sapphire interface (t_1), and the second shock arrival at the L-D_2/sapphire interface (t_2). Bottom: Position-time diagram indicating trajectories of the shock fronts and interfaces.

in this study indicate that to a very good approximation $\rho_2 \approx 1.9\rho_1$ and that $U_{s_3} \approx U_{s_2} \approx 1.1U_{s_1}$ for L-D_2; Sesame [12], tight-binding (TB) [5], molecular dynamics in the generalized gradient approximation (GGA-MD) [6], and Young [13] predictions were compared, and the variations from these relations were found to be less than 10% for each of the models. Given this very similar behavior upon reshock for the various models for L-D_2, one can show that to a good approximation $t_i/t_r \approx \Lambda/1.39 = 4.24\rho_1$, where ρ_1 is expressed in units of g/cc. Thus, the inverse of the reverberation ratio is roughly proportional to the density compression along the Hugoniot. It is to be noted, however, that when comparing experimental measurements with the various models for L-D_2 the above approximation is not needed since the models will be used to determine ρ_1, ρ_2, U_{s_2}, and U_{s_3}.

The experimental configuration is identical to that described in Ref. [3]. Shock waves were generated by planar impact of magnetically driven aluminum (6061-T6) or titanium (Ti-Al6V4) flyer plates [16] onto the aluminum drive plate at the front of the cryocell. Previous experiments have shown that the pressure applied to the L-D_2 sample is constant to within 1%–2% and that the motion is one-dimensional for the duration of the experiment [3]. The shocked state of the L-D_2 was diagnosed using velocity interferometry (VISAR [17]), fiber optic shock break out (FOSBO), and temporally and spectrally resolved spectroscopy. U_{s_1} was directly measured with VISAR and was inferred through transit time measurements with FOSBO and spectroscopy as described in Ref. [3]. The reverberation time was obtained from the time-resolved spectroscopy measurement; a typical spectroscopy measurement is shown in Fig. 3. The self-emission from the L-D_2 sample provides a clear indication of shock arrival at the aluminum/L-D_2 interface(at time t_0) and the first and second shock arrivals at the L-D_2/sapphire interface(at times t_1 and t_2, respectively) [18].

We emphasize that the experimental measurements reported here were possible due to the large sample sizes and long pressure drive times achievable with the flyer plate impact. The position-time plot shown in Fig. 3 demonstrates the need for constant pressures at the L-D_2/sapphire interface for times on order of 30–40 ns in these experiments. The constancy of the emission signal during the initial transit time and the reverberation time indicates that the pressure remained constant through the time duration of the experiment, as the intensity of emission is proportional to the pressure of the L-D_2 to the ~1.75 power [3].

Results of reverberation measurements at several initial pressure states are plotted in Fig. 4, along with predictions of various L-D_2 EOS models. We chose to plot the initial shock speed in the L-D_2, U_{s_1} (the increase in U_{s_1} correlates to an increase in P), as a function of the inverse of the reverberation ratio, t_i/t_r (the increase in t_i/t_r correlates to an increase in ρ_1 and Λ), to allow for a clearer comparison with the P-ρ principal Hugoniot shown in Fig. 1. As shown in Fig. 4, the measured reverberation ratios are in much better agreement with the various stiffer *ab initio* and Sesame EOS models. In particular, the data at the highest shock velocities, corresponding to ~75 GPa, are in excellent agreement with the Sesame and TB models. This agreement corroborates the principal Hugoniot results obtained through magnetically driven flyer plate experiments [3]. If the density compression was approximately sixfold along the Hugoniot, as inferred from the laser-driven experiments [1], t_i/t_r would continue to increase with increasing pressure. The fact that t_i/t_r is observed to decrease slightly from ~45 to ~75 GPa is a strong indication that approximately fourfold compression is not exceeded along the Hugoniot and that the Hugoniot begins to stiffen at pressures above ~45 GPa. Furthermore, the maximum in t_i/t_r observed at ~45 GPa implies that a maximum in the density compression along the Hugoniot occurs at ~45 GPa, which is also consistent with the flyer plate results [3].

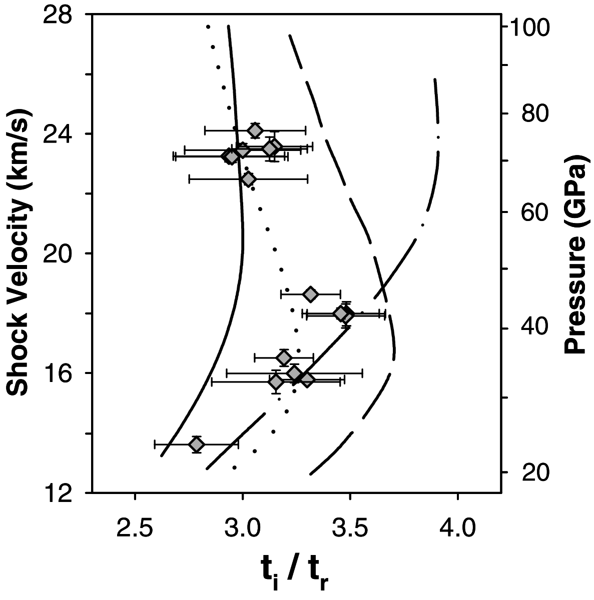

FIG. 4. Measured reverberation timing compared with theoretical predictions. Lines and symbols as in Fig. 1.

These measurements also discriminate between the stiffer *ab initio* and Sesame EOS models in the pressure range of 25–50 GPa. The Hugoniot measurements over this pressure range are unable to distinguish between the various EOS models, all of which fall within the scatter and uncertainty of the measurements. However, as illustrated in Fig. 4, the differences in the model predictions for the reverberation ratio in this pressure range are significantly larger and exceed the measurement scatter and uncertainty. In particular, the reported data exhibit the best agreement with the TB model [5,19]. This result is likely due to the fact that the TB model is a semiempirical model in which the potentials used in the *ab initio* method were fit to match molecular and solid-state properties of hydrogen and deuterium [5].

While the present results may not completely resolve the discrepancy in the high-pressure response of L-D_2, these results along with the flyer plate Hugoniot data previously reported [3] and the recent Hugoniot measurements using convergent geometry [4] make a strong case for an approximately fourfold limited shock compression. The consistency of these measurements lends confidence to each set of results, and suggests that systematic errors are likely not present. In particular, the agreement between results obtained using two completely independent loading techniques strengthens this argument.

There still remains a discrepancy with the laser-driven Hugoniot and reshock experiments [1,2]. A possible explanation for this difference is the pressure drive produced in the laser-driven experiments. As mentioned earlier, extremely precise experiments are needed to address this issue, which places stringent requirements on experimental parameters such as the constancy and planarity of the pressure drive. Given the strong sensitivity of the inferred density to uncertainties in the U_s-u_p measurements, a rather modest deviation in the constancy and/or planarity of the pressure drive could result in large enough systematic errors to explain the observed differences.

It has also been proposed that the laser-driven experiments may not produce an equilibrium state [11], and thus the laser results are not representative of the Hugoniot response of L-D_2. However, given that the collision time is several orders of magnitude smaller than the time scales of these experiments [20], both sets of experiments are expected to reach thermodynamic equilibrium states. Therefore, it is unlikely that the discrepancy can be ascribed to nonadiabatic effects in the laser experiments, as suggested in Ref. [11].

For completeness, it should be noted that an additional source of the discrepancy could be a perturbed or preconditioned sample in one or both sets of experiments (laser-driven and flyer plate). The consistency between the present reverberation data and the flyer plate Hugoniot data [3] does not completely rule out this possibility in the flyer plate experiments. However, as discussed in Ref. [3], we have gone to great pains to ensure that the samples were not perturbed prior to shock compression. Furthermore, the agreement of the present results and the recent Hugoniot measurements using a completely independent, conventional loading technique [4] corroborates this assertion.

In conclusion, we have exploited a novel reverberation technique to probe the density compression of L-D_2 along the principal Hugoniot. The reverberation measurements confirm the stiffer Hugoniot response at high pressures inferred from recent magnetically driven flyer plate experiments [3] and predicted by the various *ab initio* and Sesame EOS models [5–7,12]. The results are consistent with a maximum density compression along the Hugoniot of ~4.2. Furthermore, the reverberation technique was found to be sufficiently sensitive to discriminate between the stiffer EOS models in the pressure range investigated. Best agreement with the measured reverberation ratios was found with the TB model [5,19].

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