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Path Integral Calculation of Shock Hugoniot Curves of Precompressed Liquid Deuterium

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Abstract. Path integral Monte Carlo simulations have been used to study deuterium at high pressure and temperature. The equation of state has been derived in the temperature and density region of $10\,000 \leq T \leq 1\,000\,000\,\mathrm{K}$ and $0.6 \leq \rho \leq 2.5\,\mathrm{g\,cm^{-3}}$. A series of shock Hugoniot curves is computed for different initial compressions in order to compare with current and future shock wave experiments using liquid deuterium samples precompressed in diamond anvil cells.

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The equation of state of dense deuterium has been the topic of intense discussions since Nova laser shock wave experiments [1] measured the Hugoniot curve up to megabar pressures in 1997 and predicted an unexpectedly high compressibility. Four years later, Knudson *et al.* measured a significantly smaller compressibility using magnetically driven flyer plates [2]. This controversy continues to initiate new efforts to determine the EOS in different regions of the high temperature phase diagram. In a new approach to reach higher densities, G.W. Collins and P. Cellier use a diamond anvil cell (DAC) to precompress samples of liquid deuterium and then launch laser shock into them. The focus of this article is to use path integral Monte Carlo (PIMC) results to determine Hugoniot curves for precompressed samples in order to make a prediction for comparison with current and future experiments.

The two series of shock wave experiments using the Nova laser [1, 3] reached pressures of up to 340 GPa and predicted a 50% higher compressibility than previously estimated. The results provided the first experimental data in a regime of extreme pressure and temperature, in which our understanding had so far been primarily based on analytical models and computer simulations. The experimental findings suggested that standard EOS models such as Sesame [4] were too *stiff*, predicting a maximum compression of about 4-fold the initial density. Instead the measured EOS was found to be closer to *softer* models like [5] that predicts a maximum compression ratio of about 6. Such a difference in the EOS of dense hydrogen and deuterium would have significant consequences for our understanding of Jovian planets, brown dwarfs, and low mass stars as well as for the design of the inertial confinement fusion strategies.

As a consequence of the publication of the Nova measurements, a number of chemical models were developed which incorporated the experimental information directly or indirectly to fit unknown model parameters and then predicted an EOS closer to Nova experiments. This is a common strategy used to improve such a complex function as the deuterium EOS, which must describe different phases and regimes and spans many orders of magnitude in temperature and pressure. Typically the EOS is pieced together from different analytically known limits, experimental data, and computer simulation results. However, the high compressions seen in the predictions of chemical models were not confirmed by first principles simulation techniques such as density functional molecular dynamics [6] and PIMC [7]. Both results are in relatively good agreement with each other, predicting a lower compressibility and a Hugoniot curve close to that of the Sesame model.

In a different series of laser shock wave experiments, Mostovych *et al.* [8] used a reshock technique to probe the EOS at higher compressions. The results were also in better agreement with the soft EOS models and differed significantly from *ab initio* simulations [9]. It should be noted that the error bars were larger than those of the Nova results.

The controversial discussion of different results changed dramatically with the publication by Knudson *et al.* [2]. Instead of a laser, the Z pulse power machine was used as magnetic drive to accelerate a flyer plate launching a shock into deuterium. The achieved pressures have not yet exceeded 100 GPa but a different trend in the Hugoniot curve can already be identified. The results clearly predict a significantly lower compressibility, which is in good agreement with *ab initio* simulations and with other stiff EOS models. Given two different experimental results, it can be expected that efforts to resolve this discrepancy will intensify. So far, no satisfactory explanation has been given despite many different attempts to interpret the data. This also included suggestions for the existence of nonequilibrium states with different temperatures for ions and electrons. Dharma-wardana *et al.* [10] suggested $T_e < T_i$ while Gygi *et al.* [11] proposed $T_e > T_i$. The later prediction resulted from a remarkable computational effort involving the first dynamic simulation of shock propagation using the Car-Parinello method.

Chemical models have been constructed to predict Hugoniot curves ranging from 4 to 6-fold compression while different types of first principles simulations, with the exception of wave packet molecular dynamics [12], consistently predict a stiffer Hugoniot curve. However, it is generally assumed that primarily experimental work is now needed to resolve this controversy. New efforts are on the way and novel techniques are being developed. While the principle Hugoniot represents only one line in the phase diagram, other methods can probe different densities. Double and multiple shock reverberation measurements can access higher density regimes. Also, a pulse shaping technique is being tested by Knudson *et al.* to generate compression states on an isentrope rather than Hugoniot states. One new way to access higher densities is to use a DAC to precompress deuterium before launching a single shock through the diamond into the sample. This method allows one to choose a different initial state characterized by

($\rho_0 = m/V_0, P_0, E_0$) leading to shock states ($\rho_1 = m/V_1, P_1, E_1$) given by [13],

$$P_1 - P_0 = \rho_0 \, u_{\rm s} \, u_{\rm p} \ , \tag{1}$$

$$\frac{\rho_1}{\rho_0} = \frac{u_{\rm s}}{u_{\rm s} - u_{\rm p}} \ , \tag{2}$$

$$0 = (E_1 - E_0) + \frac{1}{2} \, (V_1 - V_0) \, (P_1 + P_0) \ . \tag{3}$$

For a given EOS, shock velocity $u_{\rm s}$ and particle velocity $u_{\rm p}$ can be derived from, $u_{\rm p}^2 = \xi/\eta$ and $u_{\rm s}^2 = \xi\eta$ with $\xi = (P_1 - P_0)/\rho_0$ and $\eta = 1 - \rho_0/\rho_1$.

In our calculations, we use $E_0 = -15.886\,\text{eV}$ per atom and $P_0 = 0$ as used in [7]. We expect that there will be small corrections to the initial state based on the initial pressure and temperature, which can be applied to our results when the initial state of a specific experiment is known. Different initial conditions like δE_0, caused primarily by a higher initial temperature, and δP_0, from the precompression, shift a particular Hugoniot curve characterized by a fixed ρ_0 from density ρ_1 to $\rho_1 + \delta\rho_1$. The density correction $\delta\rho_1$ to first order is given by,

$$\delta\rho_1 = \frac{\rho_1^2}{m\,(P_1 + P_0)} \ [\, 2\,\delta E_0 + (V_1 - V_0)\,\delta P_0 \,] \ . \tag{4}$$

We expect these corrections to be relatively small compared to the difference between the Nova and Z-pinch results, which correspond approximately to a difference of 3 eV per atom in the internal energy or to an EOS change of $-2\,\text{eV}$ in PV per atom (see [9]). Furthermore, to convert the following results for deuterium to hydrogen, one can simply divide the densities ρ_0 and ρ_1 by 2 because the isotope effect in the EOS is smaller than the error bars for the considered final shock temperatures. (ρ_0, P_0, E_0) must then match the initial hydrogen state.

The purpose of this article is to make a prediction based on PIMC simulations for the Hugoniot curves in current and future experiments using samples of precompressed liquid deuterium. Due to the length restriction for this article, we cannot describe any details of the PIMC method, which can be found in [14, 15, 16, 17]. Results from consecutive EOS efforts have been reported in [18, 19, 20]. For this particular calculation, we used EOS data published in [7, 9, 17] and results from a number of additional simulations. We simulated 32 pairs of electrons and deuterons in periodic boundary conditions. The imaginary time path integral was discretized into M slices with a time step of $\tau = 1/k_{\rm B}TM$ that was varied with density. For the evaluation of the pair action, we raised τ^{-1} from 2 to $8 \times 10^6\,\text{K}$ as the density increased from 0.6 to $2.5\,\text{g}\,\text{cm}^{-3}$. For the fermionic contributions, we used even smaller time steps ranging from 8 to $16 \times 10^6\,\text{K}$. This required simulations with up to 1000 time slices.

The precompression in a DAC allows one to probe regions of higher density as shown in Figs. 1 and 2. The different curves show predictions based on PIMC simulations for different initial compressions. The densities $\rho_0 = 0.25$ and $0.30\,\text{g}\,\text{cm}^{-3}$ were chosen to approximately represent conditions in a first set of experiments using laser driven shocks in samples precompressed in a DAC [21]. The range of initial pressures and densities

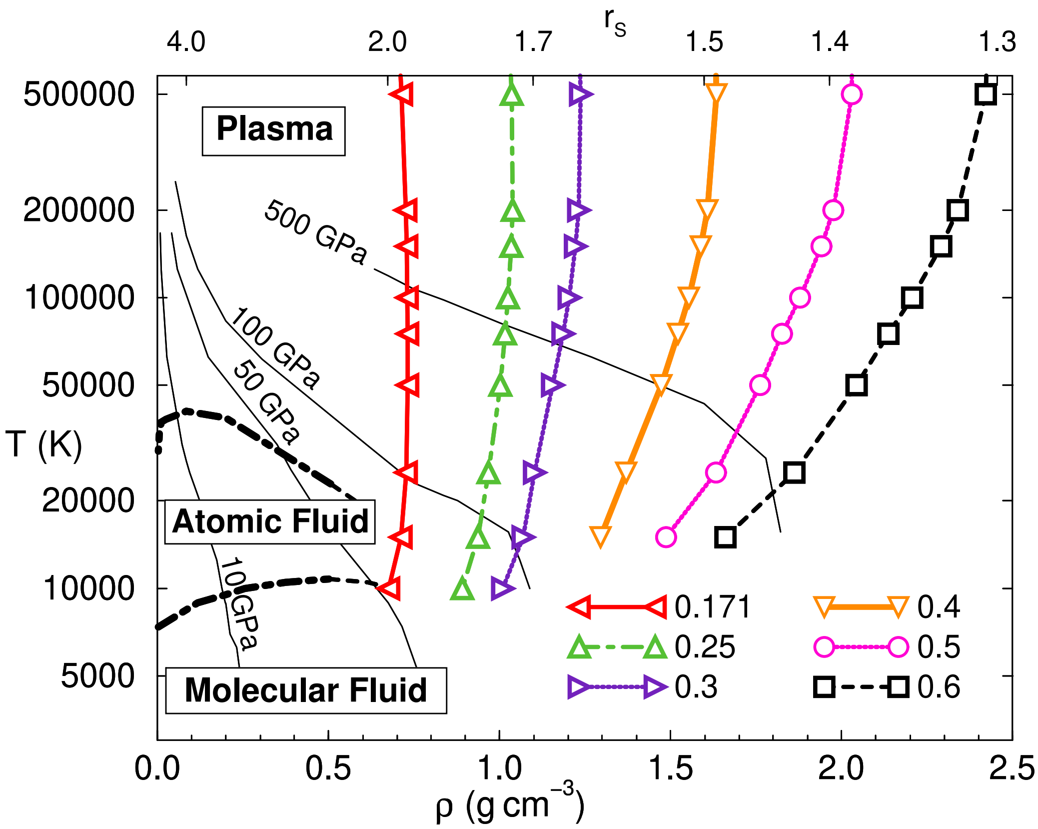

Figure 1. Deuterium temperature density phase diagram with principle Hugoniot curves calculated from PIMC simulations for different initial densities ρ_0 given in the legend in units of $\mathrm{g\,cm^{-3}}$. The thin solid lines represent isobars, dash-dotted lines indicate the approximate boundaries of the molecular, the atomic, and the plasma regime.

is limited by the currently available laser power, which restricts the thickness of the diamond anvils and therefore also the maximum achievable precompression.

The equation of state from our PIMC simulations has been used to derive the different Hugoniot curves in Fig. 2. Even a relatively small initial compression to $\rho_0 = 0.25\,\mathrm{g\,cm^{-3}}$ yields a maximum final density of $1\,\mathrm{g\,cm^{-3}}$, which is about 6 times the uncompressed initial density of $0.171\,\mathrm{g\,cm^{-3}}$ and also close to the densities predicted by the Nova experiments. However, it is expected that the maximum in compression will be at much higher pressures of about 1500 GPa, which is one order of magnitude higher than the maximum in the uncompressed case, which is estimated to be at 100 to 200 GPa.

Fig. 1 shows different Hugoniot curves for initial density up to $\rho_0 = 0.6\,\mathrm{g\,cm^{-3}}$ predicting final shock densities of up to $2.4\,\mathrm{g\,cm^{-3}}$. The maximum compression was always close to 4-fold the initial density. An unexpected increase in compression similar to the predictions of the Nova experiments has not been observed for any initial compression. In fact, the maximum compression ratio actually decreased slightly from 4.3-fold in the uncompressed case to about 4.1-fold for $\rho_0 = 0.6\,\mathrm{g\,cm^{-3}}$.

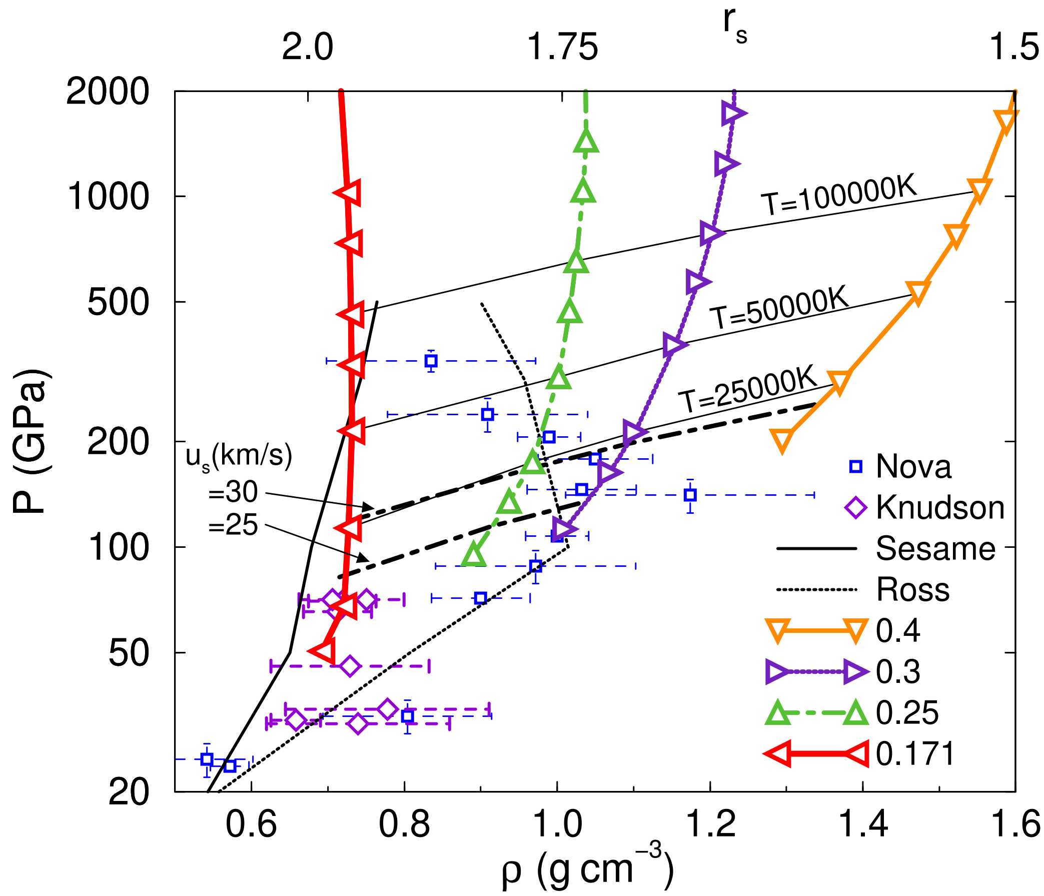

Figure 2. Pressure density diagram showing Hugoniot measurements [1, 2, 3] and results from chemical model calculations [4, 5] starting from the uncompressed liquid deuterium state ($\rho_0 = 0.171\,\mathrm{g\,cm^{-3}}$). The triangles indicate Hugoniot curves from PIMC simulations for different initial densities ρ_0 given in the legend in units of $\mathrm{g\,cm^{-3}}$. The thin solid lines are isotherms, the dash-dotted lines represent final shock states generated with the same shock velocity ($u_s = 25$ and 30 km/s) starting from different initial compressions.

It remains to be seen which of the characterized conditions of dense deuterium will be realized in future experiments. It depends on the experimental limitation such as the power of the shock drive and the pressures that can be reached in the DAC. For example, if the sustainable shock velocity is limited to 30 km/s, only pressures up to about 200 GPa can be reached if $\rho_0 \leq 0.4\,\mathrm{g\,cm^{-3}}$. The pressure-density points obtainable at constant shock velocity are shown in Fig. 2. The locus of those points is very similar to the isotherm of $T = 25\,000$ K.

Much higher temperatures and pressures can be reached in a shock reverberation measurement using the same initial shock velocity. Comparing the precompressed single shock experiment with the double shock experiment using an aluminum back plate, one finds that an initial compression to $\rho_0 \approx 0.37\,\mathrm{g\,cm^{-3}}$ leads to a primary Hugoniot curve very similar to the secondary Hugoniot curve in the double shock experiment (compare

Fig. 1 and [9]). However, for a fixed primary shock velocity, the states reached in the double shock experiment are at about twice the temperature and 1.8 times the pressure compared to the compressed single shock experiment with $\rho_0 \approx 0.37\,\mathrm{g\,cm^{-3}}$ using the same primary shock velocity. A more detailed discussion of these curves is expected to follow when the first experimental data become available.

In conclusion, the EOS from PIMC simulations of dense deuterium has been used to compute shock Hugoniot curves for different initial precompressions. The study makes precise predictions solely based first principles calculations to compare with shock experiments using deuterium samples, which have been precompressed in DACs. The main purpose of this work is to provide these curves as a reference for the interpretation of future experiments as the precompression technique is further advanced.

Acknowledgments

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