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## Shock-Wave Compression of Liquid Deuterium to 0.9 Mbar\*

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Liquid-deuterium samples initially at 21°K and 1 atm were shock compressed to pressures of 200 and 900 kbar and temperatures of 4500 and 7000°K, respectively. The 900kbar points were obtained by reflecting the 200-kbar shock wave from a brass cover. A theoretical model which uses an effective intermolecular pair potential gives reasonable agreement with the data. Using this potential we obtain a lower limit for a metallic phase transition at 1.7 cm<sup>3</sup>/mole and at 4.2 and 4.8 Mbar for H<sub>2</sub> and D<sub>2</sub>, respectively.

Our interest in the equation of state of the hydrogen isotopes stems in part from their importance in determining the structure of the outer planets, Jupiter, Saturn, and Uranus. These are largely composed of hydrogen, and its presence in insulating and metallic states will have a profound influence on the observed planetary properties. It is generally believed that the outer, low-pressure regions are molecular and the interiors are metallic. This metallic transition boundary has previously been estimated<sup>1</sup> to occur at 0.52 and 0.80 times the radius in Jupiter and Saturn, respectively. Uncertainties in the equation of state of the molecular phase can significantly alter this estimate.<sup>2</sup> Recently, experimental workers<sup>3</sup> have reported a metallic transition at 2.8 Mbar, which would indicate that the above estimates are in error by about 10%.

Recent work in controlled fusion has led to the

possibility of using high-powered lasers to compress solid  $D_2$  to the high temperatures and densities required for the fusion reaction to take place.<sup>4,5</sup> An important input into the proper design of experiments will necessarily involve an accurate equation of state of  $D_2$ . Since existing technology now allows us to make measurements to nearly one million atmospheres, an improved understanding of planetary structure and controlled fusion is possible. In this report we present the results of such measurements.

Shock compression of a number of liquid-deuterium samples with projectiles from a large two-stage light-gas gun<sup>6</sup> is reported here. In this gun, a piston driven by a charge of propellant compresses a charge of hydrogen gas into a truncated conical volume at the end of a long cylindrical barrel. The gas is allowed to expand into a second stage where it accelerates a projec-



FIG. 1. Target and projectile just before impact. Aluminum target body, A; brass cover, B; sample cavity, C; annular coolant cavity, D; impact surface, E. The projectile consists of tantalum impact plate (Ta) and a polycarbonate (PC)-polyethylene (PE) sabot.

tile to the desired impact velocity.

The target and projectile are shown in Fig. 1. The sample used was deuterium, rather than the lighter isotope, since the larger inertial mass of the former facilitates compression in a shock process and allowed us to reach higher pressures. The sample is contained in the small cylindrical cavity (1 cm thick) at the center of the Duralumin target assembly. This cavity is covered by a brass plate. The whole assembly is cooled by filling the annular cavity with liquid hydrogen. Sample vapor pressures were determined and yielded a volume of  $5.90 \pm 0.015$  cm<sup>3</sup>/gm at 1 atm.<sup>7</sup>

A tantalum plate was mounted in the front face of the projectile, which was accelerated to veloccities varying between 6.996 and 6.822 km/sec for five of the six reported experiments, with one lower velocity of 6.626 km/sec. Two timedflash x-ray shadowgraphs yielded the velocity to nominally 0.1%. The plate flatness, however, was 0.007 cm (or 0.7% of the sample thickness) over its 5-cm diameter. The impact with the Duralumin target causes a plane shock to travel into it and through the deuterium sample and the brass cover plate. To determine the Hugoniot state in the Duralumin, we used the conservation conditions<sup>8</sup> of mass and momentum in the form

$$V_1/V_0 = (D_1 - u_0)/(D_1 - u_1)$$
, (1)

and

$$P_1 - P_0 = (D_1 - u_0)(u_1 - u_0)V_0^{-1}, \qquad (2)$$

with the previously measured Hugoniot relations<sup>9</sup>

$$D - u_0 = A + B(u - u_0), (3)$$

for tantalum, aluminum, and leaded brass,<sup>10</sup> to determine the shock strength in the aluminum target and the brass cover. In the above equations u, V, and P are mass velocity, volume, and pressure, respectively. Subscript 1 refers to the conditions behind the shock that moves with velocity  $D_1$  in the laboratory coordinate system. The values of A and B obtained on samples for which  $T_0 = 298^{\circ}$ K and  $P_0 \simeq 0$  are, respectively, 3.414 km/sec and 1.201 for tantalum; 5.328 km/sec and 1.338 for Duralumin; 3.726 km/sec and 1.434 for brass. The room-temperature densities of these metals were 16.69 for tantalum, 2.786 for Duralumin, and 8.56 gm/cm<sup>3</sup> for brass.

Since the target was cooled down to  $21^{\circ}$ K, the Duralumin and brass Hugoniot curves had to be recalculated for initial conditions of  $T = 21^{\circ}$ K and  $P_0 \simeq 0$ . This was done using the Grüneisen equation of state,

$$P(E_1, V_1) - P(E_2, V_1) = (\gamma/V_1)(E_1 - E_2), \tag{4}$$

and the energy conservation condition along the Hugoniot

$$E_1(V_1) - E_0(V_0) = (P_0 + P_1)(V_0 - V_1)/2.$$
(5)

In (4)  $\gamma/V$  was constant and equal to 6.15 for Duralumin and 17.5 for brass. Density corrections are given in Ref. 6.

The shock pressure in the sample was found from a measurement of the shock speed and the requirement that pressure and mass velocity must be constant across the boundary between the container and the sample after the shock has passed this interface. If one applies that condition with Eq. (2), we note that the pressure in the Duralumin must decrease to reach the interface condition. The path for the release process is defined by

$$du = -\left(-\frac{\partial V}{\partial P}\right)_{s}^{1/2} dP$$

We therefore need to know the isentropic release path. This release path starting from  $P \simeq 1.75$  Mbar and  $T \simeq 6100^{\circ}$ K on the aluminum Hugoniot is entirely in the liquid phase. Therefore, the equation of state used<sup>11</sup> for this purpose included formulations for the solid, the liquid, and the mixed phase. The mass velocity and pressure behind the shock in deuterium can then be obtained from the solution of Eq. (2) (with measured values of D and  $V_0$ ) and the calculated release path.

980



FIG. 2. Solid circles, singly shocked and doubly shocked deuterium Hugoniot measurements; open circle, singly shocked hydrogen point; solid lines, deuterium and hydrogen Hugoniots; dash-dotted line,  $0^{\circ}$ K isotherm for molecular deuterium; dashed line,  $0^{\circ}$ K isotherm for metallic deuterium.

When this shock wave reaches the brass cover, a second shock is reflected back into the sample, with a mass velocity and pressure equal to that behind the transmitted shock in the brass cover. This interface condition is obtained by solving Eq. (2) for brass and hydrogen. Note that for brass  $u_0$  is 0 and for deuterium it is equal to the mass velocity behind the first shock.

The results are shown by the bars in Fig. 2. The initial volume of the liquid sample was 23.7  $\text{cm}^3/\text{mole}$  in the liquid at 1 atm. The pressure behind the first shock varied between 190 and 220 kbar for the six experiments, and reflection of this shock from the brass cover produced the states between 800 and 900 kbar. The major sources of error are the uncertainties in shock measurements (which were made with charged coaxial pins and foil switches) and the calculated release curve of Duralumin.

The two Hugoniots shown in Fig. 2 were calculated using relation (5) and a theoretical equation of state for fluid deuterium. The theoretical calculations were carried out using liquid perturbation theory<sup>12, 13</sup> and the intermolecular potential in atomic units:

$$\varphi(\mathbf{R}) = 8.2e^{-1.74R} - \left(\frac{13}{R^6} + \frac{116}{R^8}\right) \exp\left(-\frac{400}{R^6}\right). \quad (6)$$

The repulsive term in Eq. (6) was computed by

the Hartree-Fock method as previously reported<sup>14</sup> and is a weighted average of the four calculated orientations. The attractive term has been computed from an analysis of experimental oscillator strengths and theoretical calculations.<sup>15</sup> The exponential factor in this term was first suggested by Trubitsyn<sup>16</sup> and serves to damp out the attraction at small interatomic distances.

Since  $D_2$  is diatomic, included in the total energy were the rotational and internal vibrational energy, as well as the energy due to electronic excitation. These terms were treated as independent degrees of freedom and assumed to be volume independent. They consequently do not contribute to the pressure. In addition, the energy due to dissociation was included. This theory is in excellent agreement with the points at 200 kbar and also with previous shock compression of hydrogen to 40 kbar and 10.5 cm<sup>3</sup>/mole.<sup>17</sup>

As Fig. 2 shows, we have not tried to improve the agreement at 900 kbar and 7000°K as closely as at lower pressures (200 kbar) and temperatures (4500°K). Two reasons may be given for this. First of all, the uncertainty of the data is still comparatively high. But secondly, dissociation effects are not negligible while the dissociation energy is uncertain at these densities. Calculations<sup>13</sup> of the chemical equilibrium show that at 900 kbar approximately 9% of the atoms are dissociated if a 4.7-eV dissociation energy is assumed. For this reason and because of the relatively high uncertainty of the 900-kbar points we have made no attempt to search for a better overall potential at this time.

One of the characteristics of shock-wave experiments is the high temperature generated. Because of these temperatures atoms can approach each other more closely, and as a result the maximum contributions to the thermal properties at high densities occur at collision diameters R for which  $\varphi(R) \simeq kT$ . At 7000°K (near the highest pressure point) this R (= 3 a.u.) is equivalent to compressing solid D<sub>2</sub> to a volume of 1.8 cm<sup>3</sup>/mole. Therefore the effect of the high temperatures is to allow us to study the intermolecular potential near the region of a possible metal-lic transition.

Using the potential of Eq. (6) to calculate pressure and free energy of the molecular phase and using the results of Neece, Rogers, and Hoover<sup>18</sup> for the metallic phase of hydrogen and for the difference in zero-point energy between H<sub>2</sub> and D<sub>2</sub>, we compute the insulator-metal phase-transition pressure as 4.8 Mbar at 0°K and the volumes of the metal and molecular phases as 1.4 and 1.7 cm<sup>3</sup>/mole  $D_2$ , respectively. In  $H_2$  the predicted transition is at 4.2 Mbar. If we had attempted to force fit the potential to give better agreement with the highest pressure Hugoniot points, it would have had to be made less repulsive. This would lead to a further lowering of the free energy of the molecular phase vis-à-visthe metal, leading to a higher transition pressure. Therefore the pressure of 4.2 Mbar appears to be a lower bound.

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<sup>10</sup>The brass used had a composition of Zn/Cu/Pb = 35/62.5/2.5. The material was dimensionally stable to thermal cycling between 700 and 20°K. No loss in weight was observed from possible dezincification which occurs at higher temperatures. The bulk sound speed is constant to  $\pm 2\%$  between 300 and 77°K, and no phase changes are observed on the Hugoniot. The anomalous effects observed on rod stock of higher zinc content by N. L. Coleburn and J. W. Forbes, J. Appl. Phys. <u>40</u>, 4624 (1968), should therefore not be a problem here.

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## Dispersion and Cyclotron Damping of Pure Ion Bernstein Waves

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Ion Bernstein waves are excited by a long wire in the center of a potassium Q-machine plasma column. Because the wave vector is very nearly perpendicular to the magnetic field, we observe pure ion Bernstein waves, in contrast to neutralized ion Bernstein waves which were previously observed. The frequency bands for the cyclotron damping are so narrow that we can distinguish the isotopes of potassium.

There are two different kinds of ion Bernstein waves<sup>1</sup> depending upon the departure from exact perpendicularity for propagation across the magnetic field. We will call pure ion Bernstein waves (PIBW) those which propagate almost perpendicular to the magnetic field and in which the electrons are almost stationary. As we will see, the angular region for which PIBW exist is very nar-