Shock Compression of Liquid Helium to 56 GPa (560 kbar)

W. J. Nellis, N. C. Holmes, A. C. Mitchell, R. J. Trainor,[a]
G. K. Governo, M. Ross, and D. A. Young
University of California, Lawrence Livermore National Laboratory, Livermore, California 94550
(Received 17 July 1984)

The first shock-compression experiments on liquid helium are reported. With a two-stage light-gas gun, liquid He at 4.3 K and 1 atm was shocked to 16 GPa and 12 000 K and double shocked to 56 GPa and 21 000 K. Liquid perturbation theory has been used to determine an effective interatomic potential from which the equation of state of He can be obtained over a wide range of densities and temperatures.

PACS numbers: 64.30.+t, 34.20.Kn

In this Letter we present shock-wave data for liquid He that has been compressed to densities up to five times greater than the normal liquid and heated to temperatures up to 21 000 K. The maximum pressure attained is 56 GPa (560 kbar). These are the first shock data ever reported for liquid He and the conditions attained are more extreme than any achieved in other experiments. The highest static pressure so far attained in He is 16 GPa on the melting curve.[1]

The equation of state and intermolecular potential for dense He and H_2 have been widely studied because they are the simplest and most abundant of the elements. Their properties are important for modeling the giant planets where they are the major constituents and are subjected to pressures up to 4.5 TPa (45 Mbar) and temperatures up to 20 000 K in Jupiter and about 1 TPa and 14 000 K in Saturn.[2] Saturn is of particular interest because studies have suggested that it has an internal energy source that is associated with unmixing and gravitational separation of the hydrogen-helium fluid at pressures below 1 TPa.[2] The existence of this phase transition depends very sensitively on the hydrogen and helium equations of state.

In a series of recent papers we have reported shock-wave data[3] and theoretical calculations[4] for liquid D_2 shock compressed to 76 GPa and 7000 K. An important result was the determination of an effective intermolecular potential from which a complete equation of state can be obtained. This paper reports the results of a similar study for He.

Because of the simplicity of the He atom the interaction between pairs of atoms has been intensively studied. Most of the *ab initio* theoretical work has been concerned with determining the potential at separations less than 2.0 Å. Here the interaction is strongly repulsive and may be tested by high-energy molecular-beam experiments.[5] Information about the two-body potential at larger separations near the potential minimum has been obtained largely from gas-phase experimental data on virial coefficients, thermal conductivity, and viscosity.[6] However, at the extreme conditions existing in shock experiments and in the giant planets, the pressures are determined mainly by the short-range interaction which, because of many-body effects, differs substantially from that of the purely two-body interaction. This has been shown to be the case for dense H_2.[4] Static measurements on the solid and fluid are at present unable to test this regime.

In our experiments, strong shock waves were generated by the impact of planar projectiles into cryogenic specimen holders. Projectiles were accelerated to velocities of 3–7 km/s by means of a two-stage light-gas gun.[7,8] Thermodynamic variables of shock pressure, density, and specific internal energy were derived from the measured impactor velocity, shock velocity, and initial liquid density by means of the Rankine-Hugoniot equations and the shock-impedance-matching method. The He specimens were initially in the liquid state at 4.3 K and slightly above atmospheric pressure.

The specimen holders[9,10] and coolant fill system[3] were similar to that used for liquid H_2. Several refinements were necessary in the cryogenic design because the heat of vaporization per unit volume is an order of magnitude smaller for He than for H_2. The specimen holder was made from pure Al for improved thermal coupling between the liquid He coolant and the specimen cavity. The sample cavity was filled by condensing He gas to liquid by flowing the gas through a heat-exchange coil located in the coolant chamber. More than 100 layers of aluminized Mylar[11] were wrapped on the complete assembly for thermal radiation shielding. The impact surface or base plate, which could not be covered, was an infrared mirror, diamond turned for smoothness and Au plated for high reflectivity. A liquid-N_2–

TABLE I. Hugoniot data for liquid He. The initial density of the Al (alloy 1100) impactors was 2.715 g/cm^3. The initial density of the Ta impactor was 16.66 g/cm^3. The initial density of the pure Al base plates was 2.733 g/cm^3, corrected for thermal contraction from room temperature. The initial helium molar volume was 32.4 cm^3/mol. u_I is the impactor velocity, T_0 is the initial temperature, and ρ_0 is the initial density.

Shot	Impactor	u_I (km/s)	T_0 (K)	ρ_0 (g/cm^3)	u_p (km/s)	u_s (km/s)	P (GPa) [a]	V (cm^3/mol)
LHe4	Al	2.539	4.31	0.1233	2.47 ± 0.02	4.05 ± 0.06	1.23 ± 0.02	12.7 ± 0.4
LHe5	Al	6.243	4.31	0.1233	5.97 ± 0.03	8.82 ± 0.06	6.50 ± 0.07	10.5 ± 0.2
LHe2	Ta	6.658	4.30	0.1235	9.39 ± 0.05	13.5 ± 0.1	15.6 ± 0.2	9.78 ± 0.30

[a] 1 GPa = 10 kbar.

cooled radiation shield was positioned in front of the impact surface to shield thermal radiation from all directoins except the axis of the launch tube of the gun. Aluminum foil was contoured to the surrounding black inner wall of the gun target chamber. The specimen temperature was measured by a calibrated Ge resistance thermometer. The initial specimen density was liquid saturation density[12] at the measured temperature.

The Hugoniot or single-shock equation-of-state data are listed in Table I. The shock velocity (u_s)–mass velocity (u_p) relation is very accurately linear, $u_s = C + Su_p$, where a least-squares fit gives $C = 0.712$ km/s and $S = 1.36$. This fit was used to obtain by shock impedance matching the first-shock state in the double-shock experiment. Table II lists the equation-of-state data for the double-shock experiment.

A theoretical analysis should combine a statistical-mechanical theory of liquids with an effective pair potential to yield accurate predictions of the experimental data. The liquid theory used here is a modified hard-sphere variational perturbation theory.[13] It has been thoroughly tested against Monte Carlo simulations for a variety of classical fluids, and good agreement is obtained at temperatures high compared to the depth of the attractive well.

We have used the experimental data to test the adequacy of various potentials and finally to construct one which fits the data optimally. Because He, unlike H_2, has no internal degrees of freedom, we expect that this procedure will yield very accurate results. At $T = 21\,000$ K, the thermal energy is an order of magnitude smaller than the energy of the first excited state. Three useful potentials for comparison with data are (1) molecular beam,[5] (2) Aziz *et al.*,[6] and (3) linear muffin-tin orbitals (LMTO).[14] The molecular-beam potential is derived from the scattering of high-energy He beams. The potential of Aziz *et al.* is semiempirical, derived from experimental gas data and theoretical Hartree-Fock calculations. The LMTO potential was derived from a pair-potential fit to LMTO band-structure calculations on solid He up to 25 TPa. The first two potentials are strictly two-body, but the third, having been obtained from solid-state theory, is an effective potential which incorporates many-body effects.

These potentials are compared in Fig. 1. The LMTO and beam potentials are purely repulsive exponential functions and are therefore unrealistic in the region where the true potential has an attractive well. However, because of the very high temperatures along the Hugoniot, short-range repulsive contributions dominate the thermodynamic proper-

TABLE II. First-shock and second-shock states for double-shock experiment, LHe7. The first-shock state was obtained by shock impedance matching. The initial He density and molar volume were 0.1245 g/cm^3 and 32.1 cm^3/mol, respectively, at 4.25 K. The initial density of the Ta impactor was 16.68 g/cm^3 and the impactor velocity was 6.203 km/s. The initial density of the pure Al base plate and anvil was 2.733 g/cm^3. The Ta impactor was 1.5 mm thick, the Al base plate was 2.0 mm thick, the liquid He was 2.5 mm thick, and the Al anvil was 0.911 mm thick. The measured shock velocity in the anvil is $u_{s\mathrm{An}}$.

u_{p1} (km/s)	P_1 (GPa)	V_1 (cm^3/mol)	$E_1 - E_0$ (kJ/mol)	$u_{s\mathrm{An}}$ (km/s)	u_{p2} (km/s)	P_2 (GPa)	V_2 (cm^3/mol)	$E_2 - E_0$ (kJ/mol)
8.79 ± 0.06	13.8 ± 0.2	9.79 ± 0.06	155 ± 2	8.59 ± 0.17	2.38 ± 0.15	55.8 ± 4.7	5.87 ± 0.71	291 ± 17

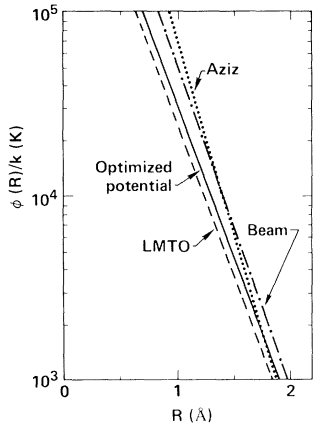

FIG. 1. Pair potentials for helium.

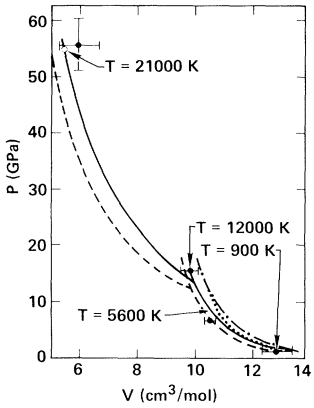

FIG. 2. Comparison of experimental (points with error bars) and theoretical (curves) helium Hugoniots. The curves correspond to potentials as in Fig. 1. Calculations with four different pair potentials are compared with the single-shock data, but only the best two potentials are compared with the double-shock point. The open circle on the theoretical curve at 55 GPa is the double-shock point predicted by the theory for the experimental conditions (10 GPa = 100 kbar).

ties, and the error due to omitting the attractive potential is negligible except at the lowest-pressure experimental point.

We calculate the theoretical Hugoniot by satisfying the condition $E-E_0=\frac{1}{2}(P+P_0)(V_0-V)$, where V, P, and E are the volume, pressure, and energy, respectively, and the subscript refers to the initial state. We use the values $E_0=100$ J/mol, $P_0=0.1$ MPa, and we use the measured value of V_0 for each of the potentials examined. Since the Hugoniot condition relates the initial and final states only; the intermediate range of states for which the attractive part of the interatomic potential may be important does not enter into the calculation. The resulting pressure-volume curve is the theoretical principal Hugoniot for the specified potential and initial state. For the double-shock curve, the calculation is repeated with an initial state now on the theoretical principal Hugoniot. The theoretical curves are compared with experiment in Fig. 2.

It is clear from Fig. 2 that the beam and Aziz potentials are too stiff, while the LMTO potential is somewhat too soft. These tendencies are strongly amplified in the calculation of the double-shock curves. This shows that many-body contributions to the effective potential have the effect of softening the two-body repulsion, and are very important in dense He. Because the LMTO and beam potentials are nearly parallel in Fig. 1, we have chosen an optimum potential which is parallel to the LMTO, but which has a larger preexponential coefficient. We find this potential to be $\phi(r)=1.1\times10^5\epsilon\times\exp(-11r/r_m)$, where $\epsilon/k=10.8$ K and $r_m=2.9673$ Å. For the highest experimental point on the principal Hugoniot, the liquid model predicts a temperature of approximately 12 000 K, and for the double-shock point, the prediction is approximately 21 000 K.

In conclusion, the high densities and temperatures reached in the shock-wave experiments have determined the effective He pair potential in a previously unexplored region and confirm the importance of many-body effects. This potential can now be used to generate a He equation of state for the study of planetary interiors.

We thank D. E. Bakker and C. D. Wozynski for setting up the specimen holders in the target chamber and firing the two-stage gun, and J. Chmielewski for machining portions of the specimen holder and fabricating projectiles. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48, with partial support from the U.S. National Aeronautics and Space Administration under Contract No. 83-033.

[a]Present address: Los Alamos National Laboratory, Los Alamos, N. Mex. 87545.

[1]P. Loubeyre, J. M. Besson, J. P. Pinceaux, and J.-P. Hansen, Phys. Rev. Lett. **49**, 1172 (1982).

[2]D. J. Stevenson, Annu. Rev. Earth Planet. Sci. **10**, 257 (1982).

[3]W. J. Nellis, A. C. Mitchell, M. van Thiel, G. J. De-

vine, R. J. Trainor, and N. Brown, J. Chem. Phys. **79**, 1480 (1983)

[4]M. Ross, F. H. Ree, and D. A. Young, J. Chem. Phys. **79**, 1487 (1983).

[5]P. B. Foreman, P. K. Rol, and K. P. Coffin, J. Chem. Phys. **61**, 1658 (1974).

[6]R. A. Aziz, V. P. S. Nain, J. S. Carley, W. L. Taylor, and G. T. McConville, J. Chem. Phys. **70**, 4330 (1979).

[7]A. H. Jones, W. M. Isbell, and C. J. Maiden, J. Appl. Phys. **37**, 3493 (1966).

[8]A. C. Mitchell and W. J. Nellis, Rev. Sci. Instrum. **52**, 347 (1981).

[9]W. J. Nellis and A. C. Mitchell, J. Chem. Phys. **73**, 6137 (1980).

[10]W. J. Nellis, F. H. Ree, M. van Thiel, and A. C. Mitchell, J. Chem. Phys. **75**, 3055 (1981).

[11]Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Department of Energy to the exclusion of others that may be suitable.

[12]R. D. McCarty, Nat. Bur. Stand. (U.S.) Tech. Note No. 631 (unpublished).

[13]M. Ross, J. Chem. Phys. **71**, 1567 (1979).

[14]D. A. Young, A. K. McMahan, and M. Ross, Phys. Rev. B **24**, 5119 (1981).