[7]Nuclepore Corporation, Pleasanton, California 94566.
[8]H. Kleinert, J. Low Temp. Phys. 39, 451 (1980); D. Vollhardt, K. Maki, and N. Schopohl, J. Low Temp. Phys. 39, 79 (1980).
[9]L. H. Kjäldman, J. Kurkijärvi, and D. Rainer, J. Low Temp. Phys. 33, 577 (1978).
[10]T. Chainer, Y. Morii, and H. Kojima, Phys. Rev. B 21, 3941 (1980).
[11]J. D. Hutchins, D. S. Betts, D. F. Brewer, A. J. Dahm, and W. S. Truscott, Physics (Utrecht) 108 B+C, 1159 (1981).

Shock Compression of Liquid Xenon to 130 GPa (1.3 Mbar)

W. J. Nellis, M. van Thiel, and A. C. Mitchell
University of California, Lawrence Livermore National Laboratory, Livermore, California 94550
(Received 30 November 1981)

New data are reported for liquid xenon shock compressed to a pressure of 130 GPa (1.3 Mbar), a molar volume of 13.7 cm^3/mole, and a calculated temperature of 29 000 K. The data are consistent with the theory of Ross and McMahan, which indicates that xenon undergoes an insulator-to-metal transition at 9 cm^3/mole at about 130 GPa or greater at 0 K. The minimum molar volume achieved in these experiments corresponds to a pressure of 60 GPa on the 0-K isotherm.

PACS numbers: 62.50.+p, 71.30.+h

Xenon is the simplest material studied to understand the insulator-to-metal transition at high pressure. This material has been compressed statically to measure the 85-K pressure-volume isotherm up to 11 GPa (110 kbar) and 21 cm^3/mole,[1] and to measure electrical conduction at 32 K which indicates an insulator-to-metal transition at about 33 GPa.[2] The Hugoniot or shock-compression curve of liquid xenon has been measured previously up to a pressure of 50 GPa and a molar volume of 18 cm^3/mole.[3] Recent theoretical results are in agreement with the shock-wave data[4] but place the insulator-to-metal transition at 130 GPa or greater at 0 K.[4,5] Thus, theoretical predictions[4,5] of the transition pressure differ by a factor of 4 from the only reported experimental observation.[2]

We have measured the Hugoniot of liquid xenon to a pressure of 130 GPa and a molar volume of 13.7 cm^3/mole in order to estimate the density dependence of the narrowing of the conduction electron energy gap. Since rare-gas solids and fluids are extremely similar in their electronic structure, which is dominated by tight-binding character, these results for the fluid are expected to be representative of the solid as well. The estimate of the energy gap follows from the excellent agreement of the data with the theory of Ross and McMahan[4] which takes into account the density dependence of the electronic energy gap in xenon. The sensitivity of the data to the energy gap arises because strongly shocked xenon is heated to temperatures comparable to the gap energy; that is, xenon is a liquid semiconductor in our experiments. The heating is caused by the thermodynamically irreversible nature of the shock-compression process. A sufficient number of electrons are thermally excited so that the shock pressure is reduced by up to a factor of 3 from what it would be without electronic excitation. The reason is that the irreversible shock energy can be distributed in only two ways in a simple fluid like xenon: thermal motion and electronic excitation. If energy is absorbed internally by electronic excitation, the shock pressure will be smaller than if no excitation occurs because less energy is available for thermal pressure. Thus, the high shock temperature is a very useful probe of the electronic structure at high density and pressure.[6]

Shock waves were generated by accelerating a planar projectile to a velocity in the range 2.6–6.6 km/s with a two-stage light-gas gun[7] and impacting the projectile onto a target containing liquid xenon. The experiment is based on the Rankine-Hugoniot relations which relate measured kinematic parameters to thermodynamic variables. Diagnostic, cryogenic, and data-reduction techniques were as described earlier,[8,9] except that a cold N_2 gas system was used to cool the target assemblies and control sample temperature to 0.1 K.[10] Xenon gas was condensed until

the sample cavity was about 80% full. Thermocouples in the sample cavity were then calibrated by assuming that the measured equilibrated pressure is the saturation pressure. The sample cavity was then filled completely. The initial sample density was obtained from the initial saturation temperature[11] of the sample. The equations of state for the metal impactors and base plates (sample cavity walls) and the error analysis were published earlier.[9,12] The data are listed in Table I.

To interpret experimental results for xenon, Ross and McMahan used augmented-plane-wave (APW) electron-band theory to obtain the 0-K isotherm and the volume dependence of the energy gap at $T=0$ between the top of the full $5p$ valence band and the bottom of the empty $5d$ conduction band for solid xenon in the fcc crystal structure.[4] The theory shows gap closure; i.e., an insulator-to-metal transition at a molar volume of 9–11 cm^3/mole and a pressure of 130 GPa or more. Ray *et al.*[5] have performed three different calculations and obtain a 0-K isotherm for fcc xenon in agreement with the results of Ross and McMahan. Hama and Matsui[13] have performed APW calculations for both fcc and bcc xenon. They predict that at high pressure solid xenon undergoes an fcc-to-bcc phase transition prior to the insulator-to-metal transition and that bcc xenon undergoes metallization at 66 GPa and 13.4 cm^3/mole at $T=0$ K. Because of the high temperature achieved in these experiments, our data are for the fluid phase and no evidence for solid-solid phase transitions at low temperatures would be expected in these data. All these calculations are in excellent agreement with the data of Syassen and Holzapfel reduced to 0 K up to 11 GPa and show that the molar volume of 18 cm^3/mole achieved in previous shock-wave experiments corresponds to a pressure of 20 GPa on the 0-K isotherm, much smaller than the shock pressure of 50 GPa. The shock pressure is higher because of the calculated high temperature of 1.5 eV achieved in the experiment. With use of a pair potential for xenon atoms and the calculated volume dependence of the energy gap, theory[4] agrees very well with the shock-compression data up to 50 GPa. The xenon pair potential was derived from the potential for argon by using corresponding-states scaling and is in good agreement with the pair potentials derived from atomic-beam scattering experiments, and from the xenon Hugoniot data below 25 GPa where negligible electronic excitation is believed to take place.

Our data are plotted in Fig. 1 as shock pressure versus molar volume. Also plotted are the data of Keeler, van Thiel, and Alder at lower pressures.[3] Agreement between the two data sets is very good in the region of overlap. The solid lines through our data, curves A and B, are unpublished calculations of Ross which extend the earlier theory to higher pressures. The new calculations include the contribution to the thermal properties from the Madelung and polarization potentials in the partially ionized dense plasma.[14] Curve A was calculated with the same volume-dependent electron band gaps as in Ref. 4. Curve B was calculated by approximating the effect of multiple excitation by assuming that the partial derivative of the band gap with the number of electrons excited, $(\partial E/\partial N_e)$, is the same in the fluid as in the gas. The theory indicates that about 1 electron/atom is excited at a shock pressure of 130 GPa. The agreement of all the shock-wave data with theory is excellent. To illustrate the thermal effects in dense xenon, the 0-K isotherm of Ross and McMahan is also plotted in Fig. 1 together with the calculated shock-compression curve if electronic excitation is ne-

TABLE I. Hugoniot data for liquid Xe. The initial density of the Al alloy 1100 impactors was 2.714 g/cm^3, of the Ta impactors 16.67 g/cm^3, and of the Al alloy 1100 base plates 2.739 g/cm^3, higher than the impactors because of thermal contraction. u_I is the impact velocity, T_0 is the initial temperature, ρ_0 is the initial density, u_s is the shock velocity, u_p is the mass velocity, P is the shock pressure, and V is the final molar volume.

Shot	Impactor	u_I (km/s)	T_0 (K)	ρ_0 (g/cm^3)	u_s (km/s)	u_p (km/s)	P (GPa)[a]	V (cm^3/mole)
Xe6	Al	2.614	163.1	2.972	3.506 ± 0.013	1.659 ± 0.011	17.28 ± 0.17	23.28 ± 0.20
Xe3	Al	5.583	165.1	2.955	5.399 ± 0.037	3.298 ± 0.023	52.62 ± 0.65	17.29 ± 0.32
Xe4	Ta	5.368	163.8	2.965	6.905 ± 0.052	4.571 ± 0.038	93.57 ± 1.31	14.97 ± 0.40
Xe2	Ta	6.596	164.0	2.964	7.966 ± 0.048	5.502 ± 0.047	129.9 ± 1.7	13.70 ± 0.39

[a]1 GPa = 10 kbar.

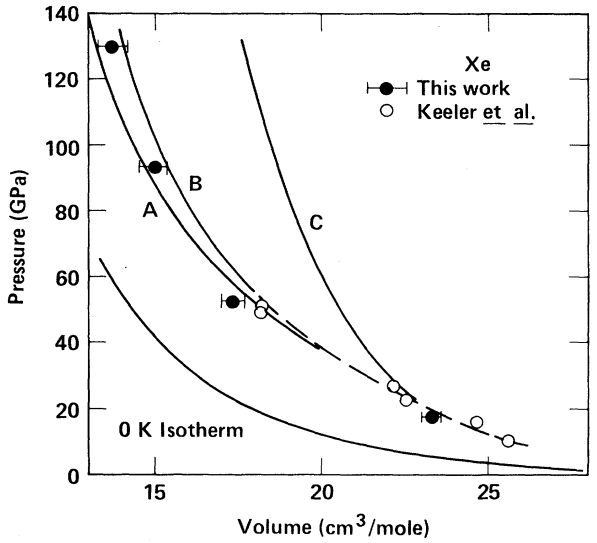

FIG. 1. Xe Hugoniot data compared to theoretical calculations. The 0-K isotherm, the low-pressure Hugoniot (dashed curve), and the high-pressure Hugoniot calculated by neglecting electron excitation (curve *C*) are after Ross and McMahan (Ref. 4). The two curves *A* and *B* were calculated by Ross (unpublished) to extend the dashed curve to higher pressures and temperatures (100 GPa = 1 Mbar).

glected in the theory (curve *C*).

In conclusion our new shock-wave data for liquid xenon in the volume range 13.7–23.3 cm^3/mole are in excellent agreement with theory[4] in which the energy gap for fcc xenon decreases from 7 to 4 eV in this volume range and goes to zero (insulator-to-metal transition) at 9 cm^3/mole and a pressure of about 130 GPa or greater at $T = 0$ K. Our measurements are sensitive to the energy gap because the calculated temperatures range between 0.6 and 2.4 eV, which are comparable to the magnitude of the gap. Our data indicate that the band gap has not closed at 13.7 cm^3/mole, which corresponds to 60 GPa on the 0-K isotherm, and thus our results provide no support for the insulator-to-metal transition to occur at 33 GPa at 32 K.[2]

We wish to thank M. Ross for use of his unpublished calculations of the high pressure Hugoniot curves of liquid xenon. We also thank A. K. McMahan and M. Ross for several helpful discussions about the interpretation of the data. We thank R. E. Neatherland for assembling the cryogenic target holders, D. E. Bakker and C. D. Wozynski for firing and maintaining the two-stage gun, H. R. Martinez for assistance with the electronic measurements, and J. Chmielewski for fabricating the projectiles. We wish to thank V. W. Morasch and D. E. Bakker for fabrication of the cooling system. We wish to thank N. Brown of Specialty Engineering Associates for providing the shock-wave detectors.

This work was performed under the auspices of the U. S. Department of Energy under Contract No. W-7405-Eng-48.

[1]K. Syassen and W. B. Holzapfel, Phys. Rev. B 18, 5826 (1978).

[2]D. A. Nelson, Jr., and A. L. Ruoff, Phys. Rev. Lett. 42, 383 (1979).

[3]R. N. Keeler, M. van Thiel, and B. J. Alder, Physica (Utrecht) 31, 1437 (1965).

[4]M. Ross and A. K. McMahan, Phys. Rev. B 21, 1658 (1980).

[5]A. K. Ray, S. B. Trickey, R. S. Weidman, and A. B. Kunz, Phys. Rev. Lett. 45, 933 (1980).

[6]M. Ross, W. Nellis, and A. Mitchell, Chem. Phys. Lett. 68, 532 (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]A. C. Mitchell and W. J. Nellis, Lawrence Livermore National Laboratory Report No. UCRL-86478, 1981 (to be published).

[11]N. B. Vargaftik, *Tables on the Thermophysical Properties of Liquids and Gases* (Hemisphere, Washington, 1975), p. 580.

[12]A. C. Mitchell and W. J. Nellis, J. Appl. Phys. 52, 3363 (1981).

[13]J. Hama and S. Matsui, Solid State Commun. 37, 889 (1981).

[14]M. Ross, unpublished.