Observation of Electrical Conductivity of Isentropically Compressed Hydrogen at Megabar Pressures

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Electrical conductivities of hydrogen and neon were measured during isentropic compression in a magnetic-flux compression device. Hydrogen becomes conducting at a density of about 1.06 g/cm^3 and a calculated pressure of about 200 GPa (2 Mbar). Neon remained an insulator to an estimated pressure of more than 500 GPa.

Considerable theoretical effort has been expended on predicting the density and pressure at which molecular hydrogen will transform to the metallic phase. Shock-wave,[1,2] isentropic,[3,4] and static[5,6] compression experiments have been reported on hydrogen at pressures of about 100 GPa or more. Here, we summarize results obtained by isentropic compression in a magnetic-flux compression device (Fig. 1).[7] The cylindrical liner is imploded by TNT explosive and compresses the magnetic flux, increasing the intensity from 5 T to more than 1000 T. The increasing magnetic field compresses the silver sample tube (Fig. 2), which isentropically compresses the sample. The conical anvils prevent escape of the sample during compression. The angle between the anvils and the sample tube is small enough to avoid jetting of the hydrogen during compression.[8] One anvil is a coaxial cable that uses Al_2O_3 ceramic as an insulator.[9] The coaxial anvil is in contact with the sample and is connected to a reflectometer,[9] allowing continuous measurement of the sample conductivity. A flash x-ray radiograph is made in each experiment and used to locate the sample-tube boundaries and thus calculate the sample density at the time of the radiograph exposure.

We performed four experiments on hydrogen and one on neon (Table I). In all four hydrogen experiments, resistivity decreased to less than 1.0 Ω cm. Figure 3 shows the radii measured from the flash radiographs of the four hydrogen experiments and magnetohydrodynamic (MHD) computer-code[10] calculations of the radii versus time. The MHD calculation is based on the measured implosion velocity of the liner, initial magnetic-field intensity, initial radii, and equations of state (EOS's) for the materials,[7,11,12] which are well known, with the exception of the two samples, hydrogen and neon.

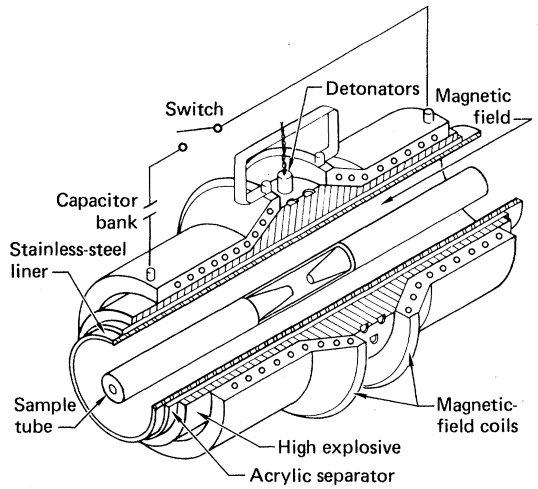

FIG. 1. Magnetic-flux compression device (Fig. 2 shows detail of the sample tube).

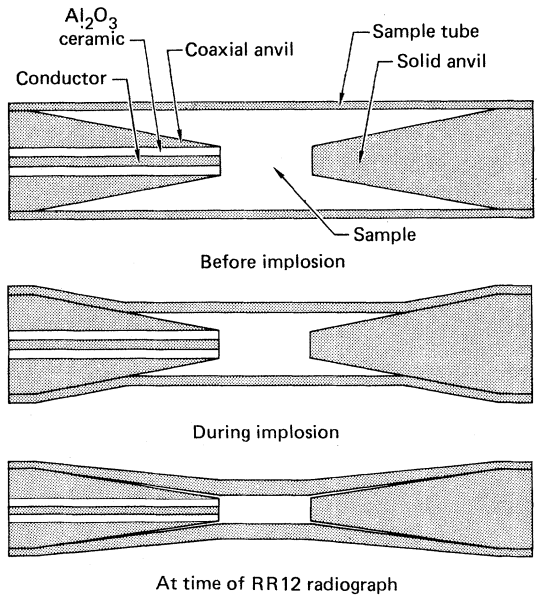

FIG. 2. Cross section of sample tube, sample, and anvils at three stages of compression.

TABLE I. Experimental parameters.

Parameters Common to All Experiments

Initial liner implosion velocity, 0.3945 ± 0.0081 cm/μs

Liner – initially 5.25-cm i.r., 5.38-cm o.r. – stainless steel 304 with 0.0029-cm copper plated on inside

Sample tube – initially 0.546-cm i.r., 0.635-cm o.r. – silver.

	Individual Experimental Parameters				
	Experiment				
	RR11	RR17	RR13	RR12	RR16
Sample material	H_2	H_2	H_2	H_2	Ne
Initial temperature, K	15.5	15.8	16.1	15.8	28.2
Initial pressure, kPa	281	310	319	310	248
Initial density, g/cm^3	0.0757	0.0755	0.0752	0.0755	1.213
Initial magentic-field intensity, T	4.45	6.80	5.98	4.55	6.42
Radiograph data:					
Time of exposure, μs	$11.002^{+0.047}_{-0.041}$	$11.523^{+0.050}_{-0.043}$	$11.542^{+0.037}_{-0.035}$	11.948±0.010	11.598±0.050
Sample-tube i.r., cm		0.330±0.05		0.1585±0.0070	0.328±0.03
Sample-tube o.r., cm	0.550±0.03	0.462±0.045	0.466±0.04	0.3268±0.0055	

The flash radiograph of experiment *RR*12 was taken 0.030 μs before the time T_c at which we observed a sudden increase in electrical conductivity. Because the radius of the sample changes little during 0.030 μs, the sample radius at T_c can be accurately calculated with the MHD code if the sample radius at the time of the x ray is accurately known. We put considerable effort into image enhancement and data reduction of the *RR*12 radiograph, resulting in the small errors shown for these data in Fig. 3.

To test the sensitivity of the MHD calculations to the EOS of hydrogen, we increased the stiffness (i.e., the intermolecular repulsive potential, hence pressure versus density) by a factor of 2, which changed the extrapolated sample-tube inside radius (i.r.) from 0.144 to 0.145 cm. The difference, 0.001 cm, is small compared to the standard deviation (0.007 cm) of the radiograph boundary measurement. The uncertainty in the time of the radiograph exposure leads to an 0.010-μs uncertainty in the time T_c, which lends an additional ± 0.005-cm uncertainty to the extrapolated i.r. The root mean square of all three

TABLE II. Nominal value and variation of extrapolated sample-tube i.r., and the resulting density, pressure, and temperature of hydrogen at the time of observed conduction.

	Sample-tube i.r.[a] (cm)	Density[b] (g/cm^3)	0-K pressure[c] (GPa) Metallic	Molecular
Minimum[d]	0.135	1.24	315	680
Nominal	0.144	1.06	200	420
Maximum[d]	0.153	0.96	142	305

[a]Measured from radiograph and extrapolated with MHD code (Ref. 10).
[b]Calculated from sample-tube i.r.
[c]From Ref. 13.
[d]Based on standard deviation of extrapolated i.r.

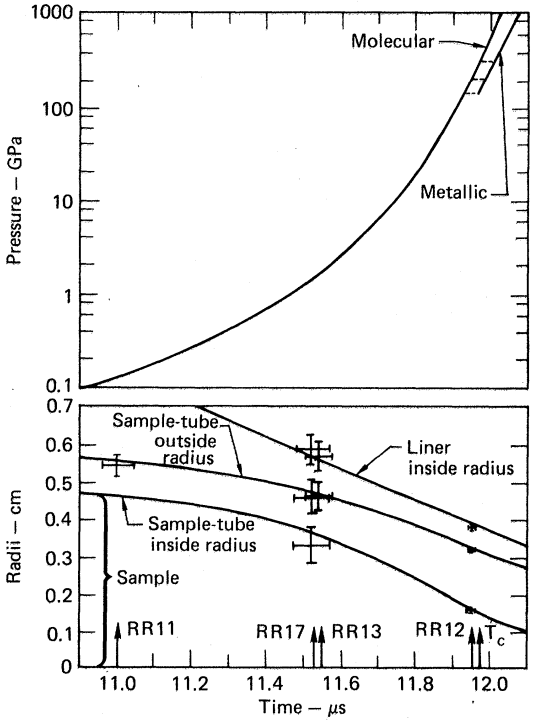

FIG. 3. MHD-code calculation of hydrogen-sample pressure and apparatus radii vs time, with experimental boundaries from radiographs.

possible errors is 0.009 cm.

Table II summarizes the extrapolated minimum, nominal, and maximum sample-tube i.r. and the

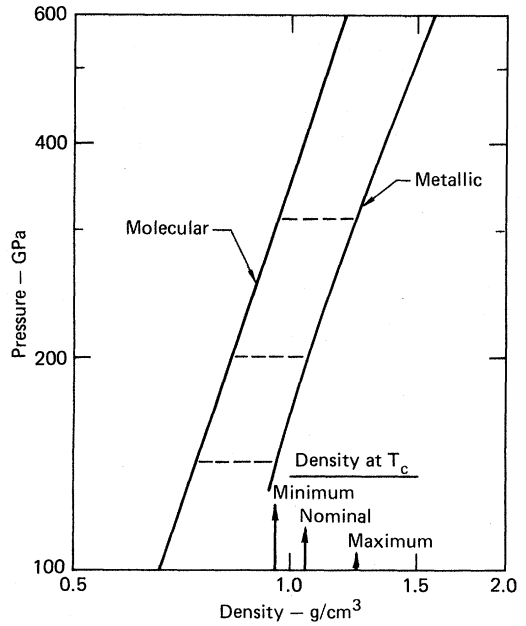

FIG. 4. Pressure-density relationships for the molecular and metallic phases of hydrogen.

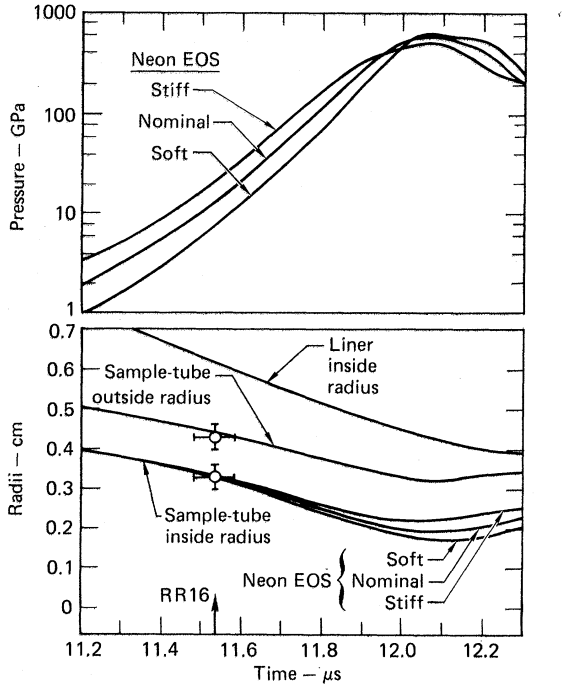

FIG. 5. MHD-code calculations of neon pressure and apparatus radii for three different neon EOS's.

resulting hydrogen density and pressure. Figure 4 is a plot of the 0-K isotherms[13] of molecular and metallic hydrogen at densities near that of the observed conduction. The isentrope temperature and its contribution to pressure above the 0-K isotherm pressure (200 GPa) are estimated to be about 400 K and 2 GPa, respectively.

In our experiment with a liquid neon sample, the electrical resistivity remained greater than 100 Ω cm throughout the compression. Because the EOS of neon is the major unknown influencing the calculation of the maximum density and pressure reached in the experiment, we made MHD calculations using the EOS of Ross[14] and two extreme EOS's having half and twice the nominal stiffness. Figure 5 and Table III show the results of these three MHD calculations. The EOS used does not significantly change the maximum pressure but does influence the maximum density. We concluded that neon remains an insulator up to a pressure of at least 500 GPa and up to a density of about 9 g/cm^3. This agrees with the prediction, based on the Herzfeld theory of metallization,[15,16] that neon will remain an insulator up to about 2000 GPa or more.

Recent theory has proposed [17-19] that hydrogen might become conducting in the molecular phase because of band-gap closure before the monatomic metallic phase is formed. Estimates of the

TABLE III. Variation of calculated maximum density and pressure obtained in neon.

Stiffness of neon EOS	Max density (g/cm^3)	Max pressure (GPa)
Half nominal	11.81	600
Nominal	9.32	560
Twice nominal	7.34	510

possible density of band-gap closure lie between 0.40 and 0.81 g/cm^3, whereas our results indicate that the lowest possible density for the onset of conduction is 0.925 g/cm^3. Our experimental results do not rule out the possibility of a conducting molecular phase but do suggest that the density at which conductivity occurs is close to the predicted[1] transition density of the monatomic metallic phase.

In conclusion, we find that hydrogen becomes electrically conducting at a density of about 1.06 g/cm^3, which corresponds to a calculated pressure of 200 GPa in the metallic phase. We find that neon remains an insulator to an estimated pressure greater than 500 GPa and a density of about 9 g/cm^3.

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[1]M. Van Thiel, M. Ross, B. L. Hord, A. C. Mitchel, W. A. Gust, M. J. D'Addario, R. N. Keeler, and K. Boutwell, Phys. Rev. Lett. 31, 979 (1972).

[2]C. G. M. van Kessel and R. Siegel, Phys. Rev. Lett. 33, 1020 (1974).

[3]F. V. Grigor'yev, S. B. Kormer, O. L. Mikhailova, A. P. Tolochko, and V. D. Urlin, Zh. Eksp. Teor. Fiz. 69, 743 (1975) [Sov. Phys. JETP 42, 378 (1975)].

[4]R. S. Hawke, D. E. Duerre, J. G. Huebel, R. N. Keeler, and H. Klapper, Phys. Earth Planet. Inter. 6, 44 (1972).

[5]L. F. Vershchagin, Ye. N. Yakovlev, and Yu. A. Timofeyev, Pis'ma Zh. Eksp. Teor. Fiz. 21, 190 (1975) [JETP Lett. 21, 85 (1975)].

[6]A. L. Ruoff, in *Proceedings of the International Conference on High Pressure, Low Temperature Physics, Cleveland, Ohio, 1977,* edited by C. W. Chu (Plenum, New York, 1978).

[7]R. S. Hawke, D. E. Duerre, J. G. Huebel, H. Klapper, D. J. Steinberg, and R. N. Keeler, J. Appl. Phys. 43, 2734 (1972).

[8]J. M. Walsh, R. G. Shreffler, and F. J. Willig, J. Appl. Phys. 24, 349 (1953).

[9]R. S. Hawke, D. E. Duerre, J. G. Huebel, R. N. Keeler, and W. C. Wallace, J. Appl. Phys. 49, 3298 (1978).

[10]R. E. Kidder, in *Proceedings of the Conference on Megagauss Magnetic Field Generation by Explosive and Related Experiments, Frascati, Italy, 1965* (EURATOM, Brussels, 1966), p. 37.

[11]R. Gover, in *Proceedings of the Seventh Symposium on Thermophysical Properties, Washington, D. C., 1977* (American Society of Mechanical Engineers, New York, 1978), p. 67.

[12]J. M. Walsh, M. H. Rice, R. G. McQueen, and F. L. Yarger, Phys. Rev. 108, 169 (1957).

[13]M. Ross, J. Chem. Phys. 60, 3634 (1974).

[14]M. Ross, private communication.

[15]K. F. Herzfeld, Phys. Rev. 29, 701 (1927).

[16]D. Brust, Phys. Lett. 40A, 255 (1972).

[17]A. K. McMahan, Ref. 6, p. 21.

[18]D. E. Ramaker, L. Kumar, and F. E. Harris, Phys. Rev. Lett. 34, 812 (1975).

[19]C. Friedli, Ph.D. thesis, Materials Science Center, Cornell University (unpublished).