22
3/22/79
24 Copies to NTIS

UCID-17977

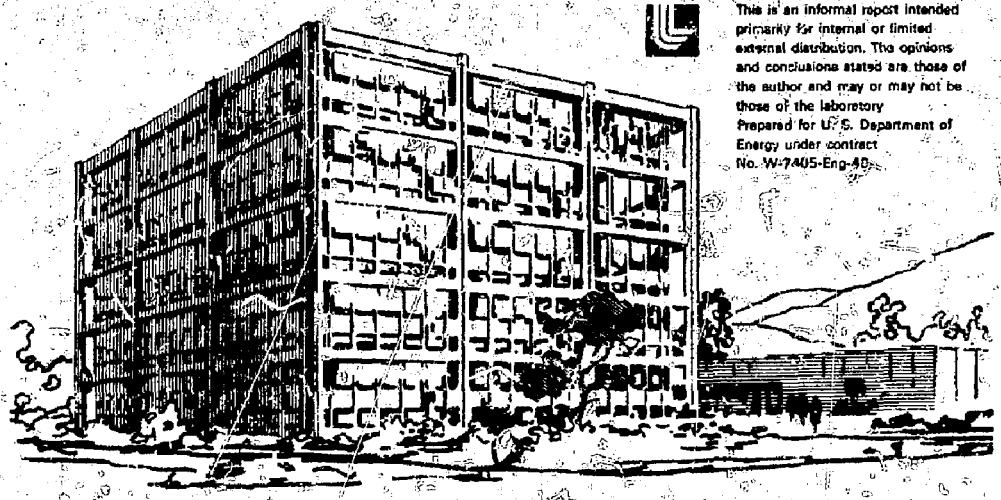
Lawrence Livermore Laboratory

METALLIC HYDROGEN RESEARCH

T. J. Burgess and R. S. Hawke

November 16, 1978

MASTER



This is an informal report intended primarily for internal or limited external distribution. The opinions and conclusions stated are those of the author and may or may not be those of the laboratory. Prepared for U. S. Department of Energy under contract No. W-7405-Eng-48.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

CONTENTS

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

METALLIC HYDROGEN RESEARCH

ABSTRACT

Theoretical studies predict that molecular hydrogen can be converted to the metallic phase at very high density and pressure, and we have achieved these conditions by subjecting liquid hydrogen to isentropic compression in a magnetic-flux compression device. We determined that hydrogen becomes electrically conducting at a density of about 1.06 g/cm^3 and a calculated pressure of about 2 Mbar. In our experimental device, a cylindrical liner, on implosion by high explosive, compresses a magnetic flux which in turn isentropically compresses a hydrogen sample; coaxial conical anvils prevent escape of the sample during compression. One anvil contains a coaxial cable that uses alumina ceramic as an insulator; this probe allows continuous measurement of the electrical conductivity of the hydrogen. A flash x-ray radiograph exposed during the experiment records the location of the sample-tube boundaries and permits calculation of the sample density. We briefly summarize the theoretical underpinnings of the metallic transition of hydrogen and describe our experimental apparatus and technique, analytical methods, and results.

INTRODUCTION

Molecular hydrogen is considered a nonmetal, but theoretical studies have indicated that extremely high pressure, on the order of millions of atmospheres, can convert hydrogen from the molecular to the metallic phase. Such conditions exist in the interiors of Jupiter and Saturn, which consist primarily of hydrogen. In the case of Jupiter, nearly all the hydrogen is thought to be in the metallic phase.[1-7]

Present interest in the properties of metallic hydrogen was stimulated by the suggestion by Ashcroft[8] that it might act as a superconductor at relatively high temperatures of 200-300 K. If the metallic phase were metastable, metallic hydrogen could prove scientifically interesting and possibly useful in commercial applications. A greater understanding of the metallic phase, the transition pressure and volume, and the electrical properties would also help describe the interior structure of the major planets and the origin of their magnetic fields. Military applications are also conceivable.

The theoretical equation of state (EOS) of metallic hydrogen has been thoroughly explored. Until recently, however, experimental techniques to produce multi-megabar pressures in a manner which would allow measurement of the EOS of "soft" and low-density materials such as hydrogen were not available. In this report, we review the experimental work performed in this area and the theoretical basis of this research. We describe in detail our application of explosive-driven magnetic flux compression techniques to the study of metallic hydrogen and include our experimental methods, the nature of the diagnostics, and our results.

THEORETICAL DESCRIPTIONS OF METALLIC HYDROGEN

Wigner and Huntington calculated nearly 40 years ago that a pressure of 250 kbar would transform molecular hydrogen to a metal.[9] At low temperatures and pressures, hydrogen exists in molecular form (H_2), but increasing pressure distorts the electromagnetic environment of the molecule until an electronic transition occurs, and the bound electrons of the diatomic insulator become the free electrons on a monatomic metal. This transition

occurs at a density at which the binding energy per molecule is approximately that of the bond dissociation energy of H_2.

The theoretical EOS of metallic hydrogen has been thoroughly explored,[1-7,9,10] mainly by the Wigner-Seitz method.[11] Incorporating recent refinements, Neece, Rogers, and Hoover[12] calculated the electronic energy of the metal using a self-consistent Wigner-Seitz procedure and determined the zero-point energy from electrostatic elastic constants. Their method works well for lithium and sodium and, therefore, has been assumed to be reliable for the simpler case of hydrogen. Schneider[13] made extensive calculations for the possible crystal stuctures and obtained a slightly denser EOS.

If the transition from the molecular to metallic phase is assumed to be first order and to result in a large decrease in volume, then, by analogy with the alkali metals, the crystal structure of a solid metallic phase would probably be body-centered cubic. Brovman, Kagan, and Kholas[14] analyzed the stability of a number of possible crystal structures for metallic hydrogen. Their calculations indicated that cubic structures are absolutely unstable upon relaxation of pressure, but that very anisotropic structures are stable. They thus concluded that metallic hydrogen at zero pressure would tend to crystallize into a triangular family of structures obtained from a primitive hexagonal lattice and that the resulting metastable structure would have a finite, but as yet uncalculated, lifetime.

The EOS of molecular hydrogen has always been the major uncertainty in the estimation of the insulator-metal transition pressure because the interaction of two hydrogen molecules at small intermolecular separations, such as the nearest neighbor distance corresponding to the transition density, is not well known. The inaccuracy of proposed intermolecular pair potentials is reflected primarily in the potential energy contribution to the EOS, since the zero-point energy at densities near the transition is a small quantity.

The transition pressure near zero kelvin is calculated by constructing the tangent common to both curves on a plot of the internal energy of each phase as a function of the molar volume (Fig. 1). The absolute value of the slope of the tangent line equals the transition pressure, and the points of tangency determine the transition volumes and energies. If the metallic curve is fixed on the plot and the molecular curve shifted upward (which corresponds to a more repulsive pair potential), the transition pressure decreases; a more

attractive pair potential raises the transition pressure. The sensitivity of the calculated value of the transition pressure on the "stiffness" of the molecular EOS is illustrated by the range of calculated values, e.g., from 0.7 Mbar[1] to 20 Mbar.[15] This subject is reviewed in greater detail by Ostgaard.[16]

ULTRAHIGH PRESSURE EXPERIMENTS

Static Methods

Measurement of the EOS of hydrogen at a constant temperature is limited by the current state of static press technology. Stewart,[17] and recently Anderson and Swenson,[18] have made isothermal measurements on solid hydrogen at 4.2 K up to about 25 kbar. Many groups throughout the world are pursuing megabar static press development: Ruoff at Cornell, Spain at the University of Maryland, and Kawaii in Japan. The largest press used for high pressure research was constructed by the late L. F. Vereshchagin in the Soviet Union.[19] Based on the results of our recent dynamic experiments,[20] it appears that the ultrahigh pressure required to produce the metallic transition of hydrogen should be feasible using static presses with diamond anvils.

Dynamic Methods

The pressure required to produce metallic hydrogen, even near zero kelvin, is just beyond the capability of modern static presses. To achieve pressure of many megabars, dynamic methods (i.e., shockwaves) have proven expedient in the study of high-density hydrogen. These techniques can produce a multi-megabar environment, but the temperature rise and the consequent thermal pressure generated severely limit the volumetric compression. Multiple shock techniques can reduce the thermal pressure contribution, but the precision of the overall pressure-volume measurement is progressively compromised as the number of shocks is increased.

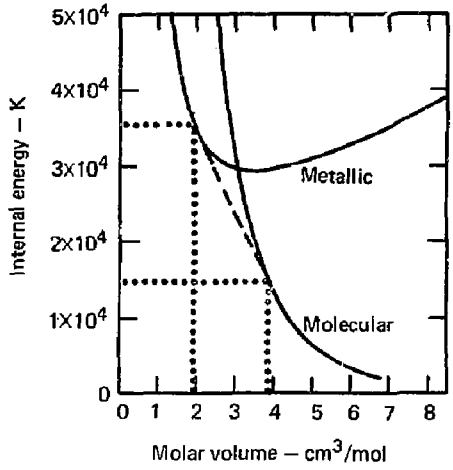


FIG. 1. Internal energy for molecular and metallic hydrogen as a function of molar volume.[16] The absolute value of the slope of the dashed tangent line equals the transition pressure, and the points of tangency determine the transition volumes and energies.

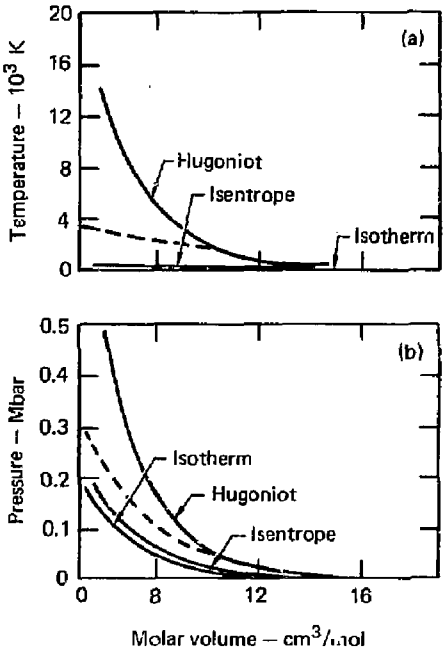


FIG. 2. Comparison of thermodynamic properties for isothermal, Hugoniot, and isentropic compressions. The dashed curves represent a reflected Hugoniot.[24]

Two methods are available to obtain high-density EOS data: shock Hugoniots and isentropic compression. A qualitative comparison of the results of these methods is shown in Figs. 2(a) and 2(b) which plot temperature and pressure as functions of molar volume.

The Hugoniot curve represents the locus of thermodynamic points that result from a given initial condition when a shock wave is passed through a material. Although shock wave methods are capable of achieving very high pressure, the process is extremely fast and hence irreversible; the resulting high temperatures and thermal pressure limit the volumetric compression (Fig. 2). Using high explosive, van Thiel and Alder,[21] shocked liquid hydrogen to 40 kbar and to an estimated temperature of 1100 K from an initial state near the normal boiling point.

By varying the initial conditions, Hugoniot curves that describe a broad region of thermodynamic space can be generated. The use of reflected shock waves (dashed curve, Fig. 2) constitutes the most convenient means of changing the initial conditions. In this type of experiment, the primary shock wave passes through the liquid hydrogen and is reflected from a metal anvil. The primary wave establishes the initial condition for the reflected wave. Because the reflected Hugoniot is a two-step compression, the final temperatures are lower than for the unreflected one-step Hugoniot at the same density. Recently, van Thiel *et al.*[22,23] used a two-stage gun to shock liquid deuterium to 210 kbar (primary wave) and to 950 kbar by reflection. The density increased by a factor of about 8 or 9, and the final temperature was calculated to be about 7000 K.

Although the high temperatures resulting from Hugoniot experiments limit volumetric compression, these temperatures do allow molecules to penetrate the repulsive cores of their neighbors to a greater extent than at low temperatures. This permits analysis of the data to determine the intermolecular potentials at relatively small separations. Performing such an analysis, Ross *et al.*[24] obtained satisfactory agreement with the Hugoniot data using an intermolecular potential which included many-body effects--a third body in this case. On the basis of these data, they concluded that the potential which best fitted the experimental data resulted in a metallic transition at about 3 Mbar. They also showed that a correction for the

nonsphericity of the diatomic hydrogen molecule increased this value to 3.5 Mbar.

The reflected Hugoniot is very sensitive to the density achieved by the first shock, and thus errors in volume measurement multiply with each reflection. This establishes a practical limit on the number of reflections that can be used to approach an isentropic compression, which is the limiting case of an infinitely reflected Hugoniot. Isentropic compression is defined as a process conducted in a completely reversible and adiabatic manner. These conditions can be approximated in practice by experiments in which the compression is rapid enough to prevent heat loss but slow enough to be reversible. The temperature increase along the isentrope is quite small compared to the Hugoniot and closely approaches the isotherm (Fig. 2). Large volumetric compressions and high pressures can thus be obtained with minimal temperature increase.

In an attempt to approximate isentropic compression, the Soviet group of Grigorev *et al.*[25] compressed hydrogen gas with an explosive-driven cylindrical implosion. Their only diagnostic measurement was a flash radiograph exposed at a given point in time during the implosion from which a volumetric compression was determined. Based on a discontinuous density increase at a pressure estimated to be 2.8 Mbar, they claimed experimental observation of metallic hydrogen. They considered the phase transition to be one of second order between liquid states because they calculated a temperature of 7000 K at the density discontinuity. Al'tshuler *et al.*[26] questioned the validity of these experiments because the pressure was calculated indirectly and no electrical resistivity measurements of the hydrogen were made during compression.

Working independently, two groups have successfully developed isentropic compression methods: Hawke *et al.*[27-30] in the United States and Al'tshuler *et al.*[26] in the Soviet Union. The Soviet group has suggested a compression technique in which a detonated solid explosive produces a gas that acts as the working fluid. Figure 3 shows the configuration of their proposed apparatus. A charge of high explosive assembled around a metal cylinder is uniformly ignited at its outer surface, and the shock wave detonates a layer of explosive on the interior of the shell during the first stages of implosion. The gaseous

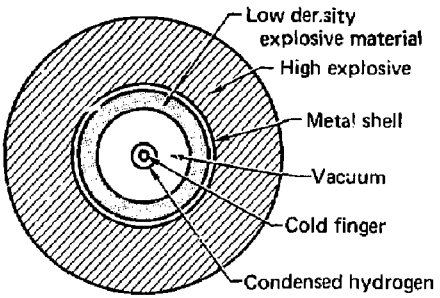


FIG. 3. Proposed configuration for isentropic compression of hydrogen by means of explosive-driven gas compression.[26] An outer ring of high explosive implodes the metal shell, and the shock wave detonates a coating of explosive on the inner side of the shell. The gaseous products expand into the evacuated region, and, as they are compressed by the imploding shell, they in turn isentropically compress liquid hydrogen condensed on the cold finger.

products expand into the vacuum, and, as they are compressed by the imploding shell, they in turn isentropically compress hydrogen condensed on a "cold finger" at the center of the system. The presumed advantage of this technique is the simplification of analog-type electrical resistivity measurements of the hydrogen during compression by avoiding time-dependent magnetic fields. Otherwise, this method appears to be computationally and technically very complex.

APPLICATION OF MAGNETIC FLUX COMPRESSION TO METALLIC HYDROGEN RESEARCH

The production of ultrahigh magnetic fields by explosive-driven flux compression was first reported by Fowler *et al.*[31] in 1960. Rather than review the subsequent evolution of the subject, which has come to be known as "megagauss physics," we refer the reader to the excellent review by Fowler.[32] In this report, we concentrate on EOS applications of intense magnetic fields.

Inspired by a suggestion in a review paper by Bitter,[33] R. S. Hawke began a magnetic compression program directed toward EOS application in 1968. By 1970, he had achieved the first successful isentropic compression in the megabar region and experimented with Lucite and cesium iodide, both medium-soft materials, up to 4 Mbar.[27,28] In 1971, he began work with liquid hydrogen and condensed noble gases, and by 1973, he reported the successful compression of hydrogen and neon above 1 Mbar.[29] Although funding for further experimental work with hydrogen was discontinued in 1973, Scudder was able to complete experiments on the compression of xenon to about 140 kbar in 1974.[34]

The principle of the isentropic magnetic compression method pioneered by Hawke is illustrated in Fig. 4. The apparatus consists of two concentric, right circular, hollow cylinders. The inner cylinder, called the "sample tube," consists of a metal of high electrical conductivity such as copper or silver and contains liquid hydrogen. The outer cylinder, called the "liner," is constructed of a metal of moderate conductivity such as stainless steel. The outer cylinder is surrounded by a ring of high explosive arranged longitudinally between a coil pair. The region between the coaxial cylinders, called the "field volume," is evacuated to prevent transmission of the shock wave to the hydrogen. Tapered "anvils" in the sample tube prevent escape of the hydrogen during compression.

During an experiment, discharge of a capacitor bank into the coil pair produces a magnetic field which readily penetrates the liner, because of its

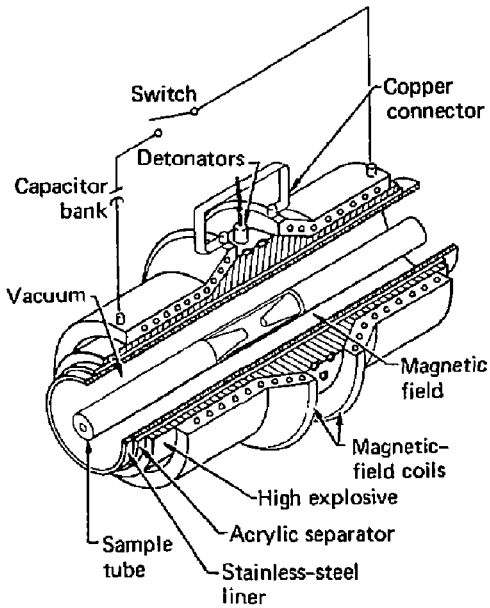


FIG 4. Experimental apparatus for isentropic compression of hydrogen using explosive-driven magnetic flux compression. An initial magnetic field generated by a pair of coils and a capacitor bank diffuses through a stainless steel liner surrounded by a charge of high explosives. When the diffused magnetic field approaches the peak value, the explosive is ignited to implode the liner cylindrically. The implosion compresses the magnetic flux, increasing the magnetic flux field density. During the flux compression, eddy currents generated in the sample tube and liner interact with the magnetic field and exert an outward pressure on the liner and an inward pressure on the sample tube.

moderate conductivity, to fill the field volume with flux (Fig. 5). When this field reaches a specified peak value (typically a flux density of 60 kG), the explosive is ignited, and the liner uniformly implodes. The time scale of this implosion is very short compared to the characteristic time for escape of the magnetic field; flux is approximately conserved, and because the cross-sectional area of the field volume decreases, the magnetic flux density present therein greatly increases to about 12 MG. Lorentz forces on the sample tube compress the hydrogen to very high densities, but, by careful design of the experiments and specification of the initial conditions with magneto-hydrodynamic (MHD) computer codes, the compression rate can be limited to values sufficiently low to prevent shock waves.

STATUS OF MAGNETOHYDRODYNAMIC (MHD) COMPUTER CODES

Various groups at the Lawrence Livermore Laboratory have devoted considerable effort to the development of MHD computer codes. Solutions are obtained by the method of finite differences using the Lagrangian formulation. The first to be written was a one-dimensional code called MAGPIE[35]; a more recent development was a two-dimensional code called HEMP.[36-38]

Two options exist in MAGPIE: one may calculate geometries with θ-component magnetic field or with z-component fields. This latter option was applicable to the geometry of our experiments. The one-dimensionality of MAGPIE requires that the magnetic field be a function only of radius and, of course, time. Using this code was thus equivalent to modeling the experiment only in one plane orthogonal to the longitudinal axis of the system and neglecting all end effects.

MAGPIE can model relatively complicated systems of many concentric shells of different materials or regions subject to specified initial conditions of temperature, density, magnetic field, and radial velocity. The latter condition is appropriate for a magnetic field oriented along the longitudinal axis. The calculations take into account the hydrodynamics of the constituent materials, Maxwell's equations for moving media, and nonlinear diffusion of a magnetic field into materials subject to the input equation of state and magnetic diffusion models.[39] Figures 6 and 7 show the variety of outputs generated by this code. MAGPIE is a useful code because it is unique,

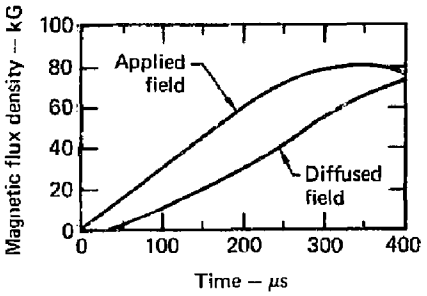


FIG. 5. Magnetic flux densities as a function of time. Both the externally applied field and the effective internal field existing in the field volume are plotted.

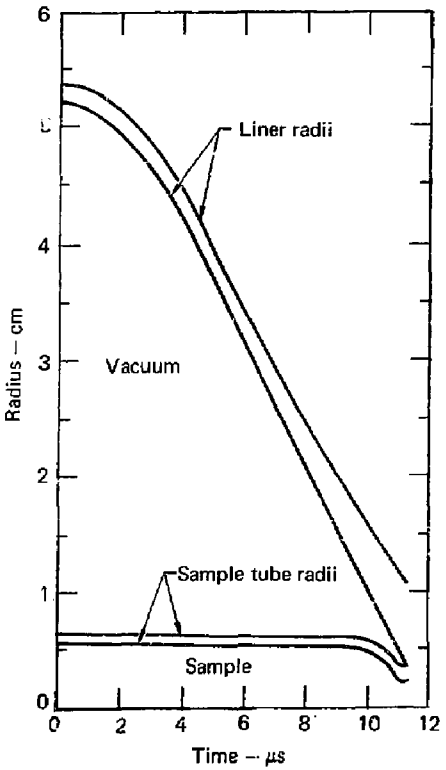


FIG. 6. Liner and sample tube radii during implosion.

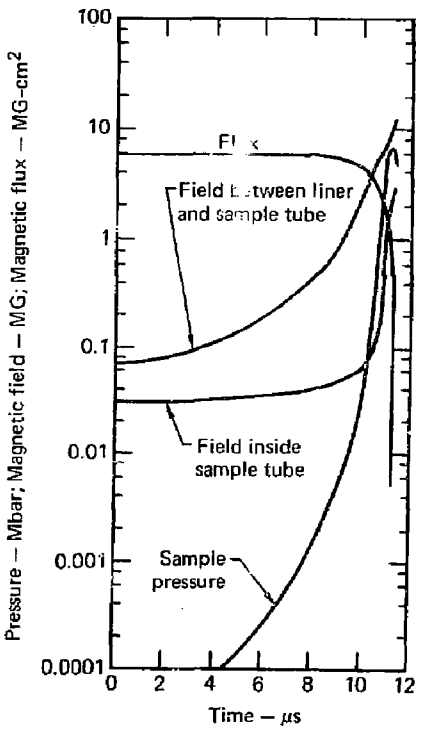


FIG. 7. Pressure, magnetic field strength, and magnetic flux in evacuated region during implosion.

well-proven, and requires comparatively little computer time to calculate a typical problem in this application. All of our magnetic compression experiments were designed and analyzed using MAGPIE.

The HEMP (Hydrodynamic, Elastic, Magneto, and Plastic) code, however, represents a very great advance. This code can solve the equations of electromagnetics, fields-materials interactions, and the conservation equation of two-dimensional elastic-plastic flow in either r-z coordinates or θ-z coordinates with cylindrical symmetry about the z-axis.

Provisions for calculation of fixed boundaries, free surfaces, pistons, and boundary slide planes have been included as well as many other special conditions. Unlike MAGPIE, the HEMP code calculates the burn of high explosive so that initial velocities are not ordinarily required. References 36-38 contain a complete description of the use of the HEMP code. Recently, a three-dimensional version of HEMP has become available.[40,41]

EXPERIMENTAL APPARATUS AND DIAGNOSTIC METHODS

Cylindrical flux compression devices to compress longitudinal magnetic fields with high explosive are not unique. Fowler's group[31] pioneered this technique and applied it to the production of intense magnetic fields, to magneto-optic studies of solids, to the production of high voltage pulses, and to the compression of plasmas.

Hawke and coworkers designed our experimental apparatus explicitly for the isentropic compression of materials for EOS studies. We describe here its design and performance, and we discuss the diagnostic techniques developed for this application.

An ideal liner would be of infinitely high resistivity to allow complete diffusion of the initial magnetic field into the sample cavity before implosion and of infinitely high conductivity after implosion to trap the magnetic field. In practice, an all-copper liner, though highly conductive, cannot be used because the initial magnetic field would be unable to diffuse through the liner in the time available, and buildup of high magnetic pressures outside the liner would destroy the coils and distort the liner. A cylindrical liner of stainless steel 1.5 mm thick with 0.025 to 0.050 mm of copper plating on the inside proved satisfactory. This resistive stainless steel liner retains the magnetic flux during the implosion almost as efficiently as an all-copper liner.

The initial magnetic field is generated by discharging a 1-MJ capacitor bank through a pair of coils. The bank consists of 120 capacitors; each has a capacitance of 14.7 μF and is charged to 35 kV. All capacitors are fused individually to prevent the accidental delivery of the bank energy to a shorted capacitor. The bank is connected to the coils by ten 40-m RG/8 cables and is switched to the coils by detonators that punch through several layers of Mylar insulation.

The coils are constructed from copper bar stock 3 × 6 mm; each contains 11 bifilar turns wrapped and wound in fiberglass and encapsulated in epoxy resin. The coils produce a magnetic flux density of approximately 0.5 G/A, and the total inductance is about 34 μH. The capacitor bank incorporates a series inductance of 15 μH to prevent a short circuit in the field coils from opening the capacitor fuses. Typically, a peak current of about 140 kA flows through the coils and generates about 60 to 65 kG of magnetic flux density in the central region.

The sample tube is copper or silver with 1.27-cm outer diameter and 0.089-cm wall thickness. Silver or copper anvils are inserted to prevent axial escape of the liquid hydrogen from the high pressure region during compression. The anvils are tapered as shown in Fig. 4 to match the volumetric compression with the axial pressure distribution along the sample tube. The liquid hydrogen typically has an initial temperature of about 16 K at pressure of about 3 bar.

The liner is imploded by a cylinder of TNT high explosive. This explosive assembly is detonated simultaneously at 168 points on a 5-cm-wide band around the exterior of the cylinder. Lucite, which separates the liner from the high explosive, is used to attenuate the von Neumann spike and to reduce the possibility of spalling on the interior surface of the liner. The liner accelerates to a velocity of 5.1 mm/μs in 2 to 3 μs.

Figure 6 plots the radii of the liner and the sample tube against time. Figure 7 shows the magnetic flux densities (B) inside the sample tube and between it and the liner, and the pressure flux (Φ) between the sample tube and the liner, and the pressure (p) on the sample tube--all as functions of time. These parameters are computed with a magnetohydrodynamic (MHD) code.[35,39] As the liner begins to implode, virtually all of the flux is trapped except a small amount which escapes because of the finite resistivity of the liner. As eddy currents heat the liner, resistance increases and flux escapes more readily as the implosion progresses. Although the trapped flux diminishes with time, the magnetic intensity and pressure increase until the velocity difference between the liner and sample tube equals the diffusion velocity of the escaping magnetic field. The sample pressure reaches a value that is slightly higher than the magnetic pressure ($p = B^2/2\mu_o$ in SI units) because the sample tube gains kinetic energy that is then transferred to the sample.

A rotating-mirror framing camera was used to record the liner shape and dimensions as a function of time. High-intensity illumination, produced by an explosive-driven shock traveling through argon gas, was used to backlight the imploding cylinder. The time between frames, 168 ns, provides sufficient resolution to give a good record of the liner velocity and an indication of any departure from the original cylindrical shape.

The initial magnetic field was measured with an inductive pickup loop made of five turns of Teflon-coated No. 30 wire wrapped around a fiberglass rod

1.5 mm in diameter. The signal was recorded both directly on an oscilloscope (to obtain dB/dt vs time) and through operational amplifier plug-in units used as integrators (B vs time). Because the probes were destroyed during compression, only the data for the initial field and first few microseconds of the implosion are reliable.

A radiograph was obtained using a pulsed 35-MeV linear accelerator to bombard a 0.5-mm-thick copper foil target with a burst of electrons for approximately 0.2 μs. The target emitted an x-ray pulse of 170 ns duration FWHM which passed through the experimental assembly perpendicular to the longitudinal axis. The x rays impinging on a treated metal screen produced light that exposed the film. The film and screen were contained in an aluminum cassette that protected them from the explosive charge and allowed recovery after the experiment.

The radiograph was scanned with an optical densitometer to generate a plot of x-ray absorption vs radius. The boundaries of the anvils, sample tube, and liner were determined from this plot, and the specific volume of the hydrogen was calculated.

Pressure was deduced by indirect computations 1) by measuring the volumetric compression of metal parts of the assembly (i.e., sample tube and liner for which the EOS are considered well known), and 2) by calculating the pressure using an MHD computer code to model the experiment with an assumed EOS for hydrogen.

The electrical resistivity of the hydrogen was measured during the compression by a radio frequency reflection coefficient apparatus (Fig. 8). An electrical probe was constructed in the hydrogen volume by introducing a Kovar wire through one of the anvils. The Kovar wire was insulated from the anvil, and hence from the sample tube, by an alumina sleeve. The inner and outer diameters of the alumina insulator were 1.52 mm and 3.18 mm, respectively. An extensive development program was necessary to discover an insulator material which would remain an insulator at the very high pressure developed in these experiments.[46] Polymers such as Lucite and Teflon proved unsatisfactory.

The anvil in contact with the sample tube, insulating sleeve, and Kovar center conductor represented a coaxial transmission line (Fig. 8). When in contact with liquid molecular hydrogen, which is an insulator, the circuit was open, and a radio frequency pulse train delivered to this structure through an

impedance-matched coaxial cable was therefore reflected without inversion of the waveform (Fig. 9). When the hydrogen became electrically conductive at the metallic transition, the circuit was closed, and the pulse train was reflected with inversion of waveform. In actual practice, a high-pass filter was used to exclude electrical noise pulses caused by rapid variations of the magnetic fields due to the flux compression; the inversion was thus manifested as a phase change.

The pulse generator produced a train of 20-ns pulses at a repetition rate of 16.45 MHz. The system impedance was 50 Ω. A pair of 6-db directional couplers was used to monitor both the incident and reflected pulse trains.

Although this system was originally intended to provide a gross indication of a change in the electrical resistivity of the hydrogen at the transition point, an estimate of the actual resistivity change could be obtained. This was accomplished by computer-aided field mapping of the experimental geometry; the hydrogen was assumed to be homogeneous and isotropic. The accuracy of any estimate of the actual change of the reflection coefficient was limited by the width of an oscilloscope trace. The minimum resolvable hydrogen resistivity using this 50-Ω system was 0.035 Ω•cm.

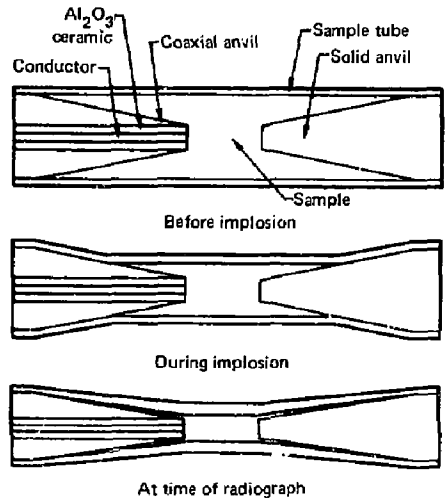


FIG. 8. Cross section of sample tube, electrical resistivity probe, and anvils before and during implosion. Material boundary locations after implosion are obtained experimentally by plotting a set of axially displaced densitometer scans across a radiograph exposed during implosion. The center conductor and insulator of the resistivity probe are not resolved.

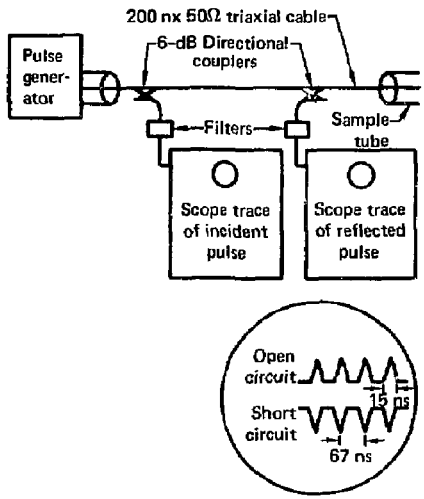


FIG. 9. Measurement of electrical resistivity of hydrogen by means of radio frequency reflection coefficient. A pulse generator provides a 15-MHz pulse train reflected with unchanged polarity by an open circuit and an inverted polarity by a short circuit. Inversion of the waveform thus indicates that hydrogen has become a conductor. The directional couplers sample the incident and reflected pulse train, and high-pass filters reject the spurious voltage induced by the rapidly changing magnetic field. The pulse trains are recorded with oscilloscopes. Fiducial time marks are used to establish the time of pulse waveform attenuation or inversion, if any. This latter time can then be cross-correlated with the MHD code calculations and with volumetric compression measurements to determine the pressure and volume at the time of increased conductivity.

RESULTS AND DISCUSSION

We have evidence that we transformed hydrogen from the molecular to the metallic phase during our exploratory experiments on isentropic compression of hydrogen. A measurement of the radio frequency reflection coefficient during a magnetic compression experiment indicated the discontinuous decrease of the electrical resistivity of hydrogen from that of a dielectric insulator to that of a conductor or semiconductor. We interpreted this observation as an indication of a metallic transition and calculated the transition pressure to be about 2 Mbar using an assumed EOS and a volumetric compression measurement of the hydrogen.[20] The latter was determined from a flash radiograph taken when the resistivity dropped. Our observations agree with the predictions of Ross *et al.*[24] and the Soviet workers.[25]

The diagnostic technique we used could not resolve a resistivity less than 0.035 Ω-cm, but we concluded that the hydrogen resistivity decreased to at least this value. We precluded the possibility of thermal contributions to this conductivity observation on the basis of theoretical calculations[42] of the hydrogen temperature rise from a liquid state along an isentrope during these experiments (1500 K). An identical experiment using liquid neon in place of liquid hydrogen produced no decrease in resistivity. Neon is expected to have a much higher transition pressure than the experimental apparatus could develop.

We can conceive of three possible alternate explanations for the observed resistivity decrease during a hydrogen compression experiment: the shorting out of the alumina insulator due to shock loading, the collapse of the sample tube on the anvil which injects ionized material into the resistivity probe region, or band-gap closure.[43-45] The results of previous studies of alumina insulators[46] and our neon experiment preclude failure of the insulator.

Figure 8 shows the locations of material at a late stage of an explosion-driven magnetic compression experiment, based on a flash radiograph. The sample tube has collapsed about halfway down the surface of the anvil, pushing hydrogen in front of it. Under certain conditions, the oblique collision of two metal surfaces results in the formation of a high-speed "jet" of molten metal,[47,48] which, if created at the sample tube-anvil interface, could

inject ionized material into the electrical probe, causing a spurious resistivity decrease. According to a jet analysis,[49] however, the angle between the sample tube and the anvil is too small for jet formation. Calculations using the HEMP code could be used to resolve this question finally, but because a jet, if it exists, must interact with the magnetic field that is trapped and subsequently compressed with the hydrogen in the sample tube, the calculation would be more complex than conventional jet analyses. Our results do not exclude the possibility of a conducting molecular phase due to band-gap closure, but do suggest that the density at which conductivity occurs is close to the predicted transition density[22] of the molecular phase.

Examination of Fig. 8 also suggests that the anvil taper is not an optimum shape for maximum compression because the collapse of the sample tube upon the anvil is only partial, resulting in the possible entrapment of hydrogen at large radii and hence low compression. Shape optimization, based on extensive HEMP calculations, should lead to more efficient volumetric compression of the hydrogen.

The transition pressure was extrapolated from a flash radiograph taken at a time as close as possible to that of the resistivity decrease. This time was selected on the basis of previous experiments. Volumetric comparison data was obtained from this radiograph for both the hydrogen and the containing assembly (sample tube and anvils). Because of the complex shape and changing configuration of the hydrogen-containing region time, it was difficult to obtain unambiguous data from a single experiment. Cross-correlation of three experiments, as described in Ref. 20, yielded a pressure of 2 Mbar at a density of 1.06 g/cm^3 at the time of the conductivity change. If a method could be developed to measure the hydrogen pressure at the same time that the radiographic measurement of volumetric compression is made, the indirect calculation of the pressure by a computer code using an assumed hydrogen EOS could be eliminated.

CONCLUSIONS

We have provided evidence that isentropic compression of hydrogen at a pressure of about 2 Mbar changes the electrical conductivity of hydrogen from that of an insulator to that of a conductor. This suggests the possibility of the occurrence of the metallic transition. The device that we developed to produce these very high pressures can be used for isentropic compression of a variety of materials, ranging from the softest, hydrogen, to one of the stiffest, sapphire. Our work also confirmed the efficacy of a 5-Mbar electrical insulator. We believe that our investigation demonstrates that discovering the high pressure EOS and metallization parameters of hydrogen are well within the capabilities of present technology.

We conclude that further research in this area should pursue the following objectives:

- Development of EOS data for molecular hydrogen up to volumetric compressions of at least 15 to 1 or the metallic transition, whichever occurs first.
- Demonstration of the occurrence (or nonoccurrence) of the metallic transition by experiments that "bracket" the resistivity change.
- Measurement of the pressure and density at the metallic transition.

ACKNOWLEDGMENTS

The authors thank D. E. Duerre, R. Grover, W. H. Gust, A. T. Harris, L. S. Hord, W. G. Hoover, J. G. Huebel, R. N. Keeler, H. Klapper, J. R. Long, R. F. McLain, A. C. Mitchell, J. Mosher, M. Ross, J. K. Scudder, J. W. Shaner, E. Teller, M. van Thiel, W. C. Wallace, W. C. Wilson, and the bunker crews of W. M. Halladay and H. A. Massey. In addition, special acknowledgment is due Max Fowler, Fritz Herlach, and Jim Shearer.

REFERENCES

1. R. Kronig, J. DeBoer, and J. Korringa, Physica, 12, 245 (1946).
2. W. H. Ramsey, Monthly Notices, Roy. Astron. Soc., 110, 325 (1950).
3. A. A. Abrikosov, Astron. Zh., 31, 112 (1954).
4. W. C. DeMarcus, Astron. J., 63, 2 (1958).
5. P. J. E. Peebles, Astrophys. J., 140, 328 (1964).
6. R. Smoluchowski, Nature, 215, 691 (1967).
7. W. B. Hubbard. Astrophys. J., 152, 745 (1968).
8. N. W. Ashcroft, Phys. Rev. Lett., 21, 1748 (1968).
9. E. P. Wigner and H. B. Huntington, J. Chem. Phys., 3, 746 (1937).
10. W. J. Carr, Phys. Rev., 128, 129 (1962).
11. E. P. Wigner and F. Seitz, Phys. Rev., 43, 804 (1933).
12. G. A. Neece, F. J. Rogers, and W. G. Hoover, J. Comput. Phys., 7, 621 (1971).
13. T. Schneider, Helv. Phys. Acta, 42, 957 (1969).
14. E. G. Brovman, Yu. Kagan, and A. Kholas, Sov. Phys.-JETP, 34, 1300 (1972).
15. B. J. Alder and R. H. Christian, Phys. Rev. Lett., 4, 450 (1960).
16. B. Ostgaard, Phys. Lett., 45A, 371 (1973).
17. J. W. Stewart, J. Phys. Chem. Solids, 1, 145 (1956).
18. M. S. Anderson and C. A. Swenson, Phys. Rev., B10, 5184 (1974).
19. L. F. Vereshchagin, Private Communication.
20. R. S. Hawke, T. J. Burgess, D. E. Duerre, J. G. Huebel, R. N. Keeler, H. Klapper, and W. C. Wallace, Phys. Rev. Lett., 41, 994 (1978).
21. M. van Thiel and B. J. Alder, Mol. Phys., 10, 427 (1966).
22. M. van Thiel, M. Ross, B. L. Hord, A. C. Mitchell, W. H. Gust, M. J. D'Addario, and R. N. Keeler, Phys. Rev. Lett., 31, 979 (1973).
23. M. van Thiel, *et al.*, Phys. Earth Planet. Interiors, 9, 57 (1974).
24. M. Ross, R. H. Ree, and R. N. Keeler, Proc. 4th Int. Conf. on High Pressure, Kyoto, Japan, Nov. 25-29, (1974).
25. F. V. Grigorev, S. B. Kormer, O. L. Mikhailova, A. P. Tolochko, and V. D. Urlin, JETP Lett., 16, 201 (1972).
26. L. V. Al'tshuler, E. A. Dynin, and V. A. Svidinskii, JETP Lett., 17, 13 (1973).
27. R. S. Hawke, D. E. Duerre, J. G. Huebel, R. N. Keeler, and H. Klapper, Nature, 233, 79 (1971).

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

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

30. G. B. Lupkin, Phys. Today, 26, 17 (1973).

31. C. M. Fowler, W. B. Garn, and R. S. Caird, J. Appl. Phys., 31, 588 (1960).

32. C. M. Fowler, Science, 180, 261 (1973).

33. F. Bitter, Sci. Am., 213, 65 (July 1965).

34. J. K. Scudder, Diagnostics Development for Magnetic Flux Compression Experiments, Lawrence Livermore Laboratory, Livermore, Calif., UCRL-51767 (1975).*

35. D. Steinberg, A One-dimensional Magnetohydrodynamics Code, Lawrence Livermore Laboratory, Livermore, Calif., UCRL-14931 (unpublished).

36. M. L. Wilkins, Calculation of the Elastic-plastic Flow, Lawrence Livermore Laboratory, Livermore, Calif., UCRL-7322, Rev. 1 (1969).*

37. E. D. Giroux, HEMP User's Manual, Lawrence Livermore Laboratory, Livermore, Calif., UCRL-51079, Rev. 1 (1973).*

38. M. L. Wilkins, Magnetohydrodynamics of HEMP, Lawrence Livermore Laboratory, Livermore, Calif., UCRL-51715, (1973).*

39. R. E. Kidder, Proc. Conf. on Megagauss Magnetic Field Generation by Explosive and Related Experiments, Frascati, Italy, September 21-23 (1965) Euratom, Brussels, (1966), p. 37.

40. M. L. Wilkins, R. E. Blum, E. Cronshagen, and P. Grantham, A Method for Computer Simulation of Problems in Solid Mechanics and Gas Dynamics in Three Dimensions and Time, Lawrence Livermore Laboratory, Livermore, Calif., UCRL-51574, Rev. 1 (1975).*

41. S. J. French, Jr., User's Manual for Fortran IV HEMP 3D, Lawrence Livermore Laboratory, Livermore, Calif., UCRL-52359 (1977).*

42. M. Ross, Private Communication.

43. A. K. McMahan, in Proc. Int. Conf. High Pressure, Low Temperature Physics, Cleveland, Ohio, 1977 (Plenum Press, New York, 1978), edited by C. W. Chu.

*Available from Nat. Tech. Info. Service, U. S. Dept. of Commerce, 5285 Port Royal Rd., Springfield, Virginia 22151.

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

45. C. Friedli. "Band Structure of Highly Compressed Hydrogen," PhD Thesis, Materials Science Center, Cornell University.

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

47. G. Birkhoff, D. P. MacDougall, and E. M. Pugh, J. Appl. Phys., 19, 563 (1948).

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

49. F. H. Harlow and W. E. Pracht, Phys. of Fluids, 9, 1951 (1966).

KMG:dlh