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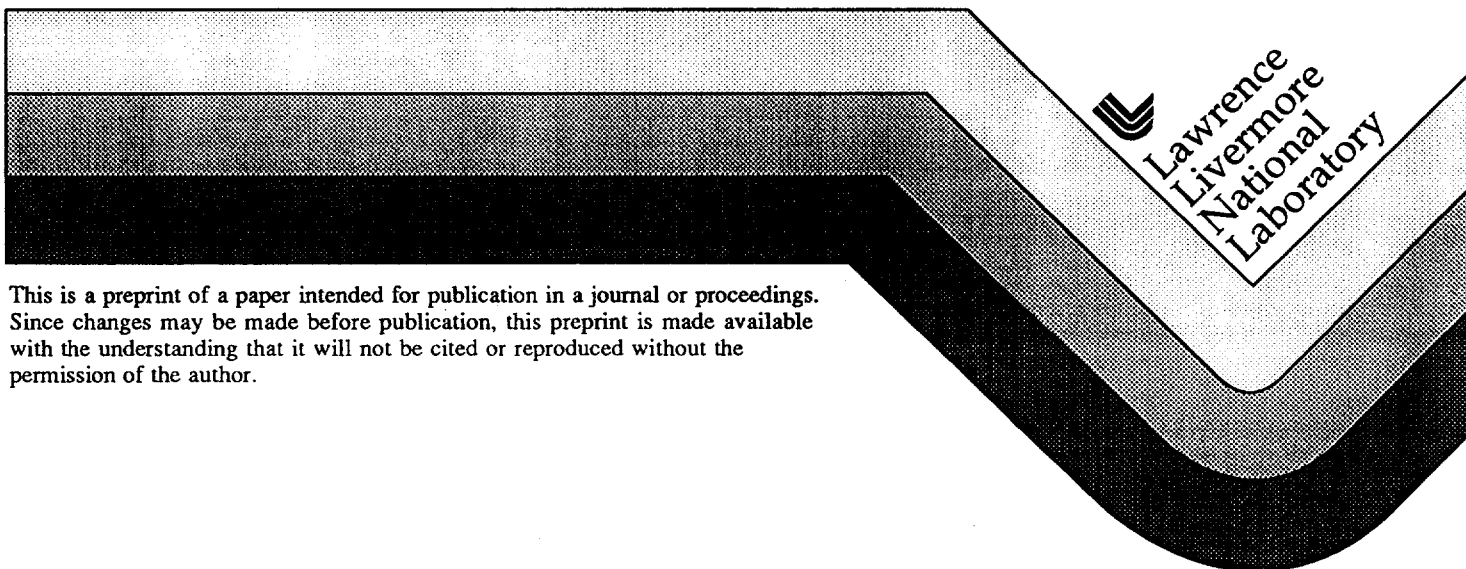
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Hydrogen at High Pressures and Temperatures: Implications for Jupiter

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Abstract

Electrical conductivities and shock temperatures were measured for shock-compressed liquid H_2 and D_2 . Conductivities were measured at pressures of 93-180 GPa (0.93-1.8 Mbar). Calculated densities and temperatures were in the range 0.28-0.36 mol/cm³ and 2000-4000 K. The resistivity data are interpreted in terms of a continuous transition from a semiconducting to metallic, primarily diatomic fluid at 140 GPa and 3000 K. Shock temperatures up to 5200 K were measured at pressures up to 83 GPa. These data are interpreted in terms of a continuous dissociative phase transition above 20 GPa. The continuous transition from a molecular to monatomic fluid means that Jupiter has no distinct core-mantle boundary. The dissociation model derived from the temperature measurements indicates a dissociation fraction of about 5% at 140 GPa and 3000 K. The isentrope of hydrogen was calculated starting from the surface temperature of Jupiter (165 K). At a metallization pressure of 140 GPa in Jupiter, the temperature is about 4000 K and about 10% of the hydrogen molecules are dissociated. The electrical conductivity was calculated along this isentrope by deriving a scaling relationship from the measured conductivities. The results indicate that hydrogen becomes metallic much closer to the surface of Jupiter than thought previously, a possible explanation of the very large magnetic field of Jupiter, but the metallic conductivity of the molecular fluid is two orders of magnitude lower than predicted for the monatomic fluid.

Introduction

Properties of hydrogen at high pressures and temperatures are needed to understand the nature of metallization and effects of electrical conductivity on the magnetic fields of giant planets, as well as to calculate the pressures, densities, and temperatures inside Jupiter and Saturn [Hubbard, 1980; Stevenson, 1982; Zharkov and Gudkova, 1992]. The interiors are at high pressures and high temperatures because of the planets' large mass and low thermal conductivity. Pressure and temperature at the center of Jupiter are about 4 TPa and 20,000 K [Zharkov and Gudkova, 1992]. Internal temperatures are well above the calculated melting curve of hydrogen [Ross, et al., 1981]. Properties of hydrogen also must be known to understand the interiors of new Jupiter-size planets now being discovered close to nearby stars [Mayor and Queloz, 1995]. These giant planets might also be composed of massive amounts of hydrogen, since hydrogen comprises 90% of all known atoms. For these reasons we have measured electrical conductivities and temperatures of fluid hydrogen and deuterium shock compressed to pressures up to 180 GPa. The conductivity experiments were performed with a reverberating shock wave to minimize the temperature and maximize pressure. The temperature measurements were performed at higher temperatures and lower pressures to investigate effects of molecular dissociation. The theoretical model for dissociation derived from the temperature measurements shows that the conductivity experiments probed primarily the molecular phase.

Experiments

High shock pressures were generated by impact of a hypervelocity impactor onto the front surface of a sample holder containing liquid hydrogen [Nellis, et al., 1983]. Al, Cu, or Ta impactors were accelerated to velocities up to 8 km/s with a two-stage light-gas gun. Both hydrogen and deuterium were used to obtain different shock-compressed densities and temperatures. In the conductivity experiments hydrogen was

contained between Al_2O_3 anvils which in turn were contained between Al disks [Weir et al., 1996a]. Hydrogen shock pressure was determined by shock impedance matching the measured impactor velocity and known Hugoniot equations of state of the metal impactor, Al disk, and Al_2O_3 anvil. The pressure in hydrogen reverberates up to the first shock pressure in the Al_2O_3 , independent of the equation of state of hydrogen [Ogilvie and Duvall, 1983]. Due to the large density mismatch between hydrogen and Al_2O_3 , the first-shock pressure in hydrogen is a factor of ~ 25 lower than the first shock in Al_2O_3 . The reverberation of the shock in hydrogen between the Al_2O_3 anvils compresses the hydrogen to the first shock pressure in the Al_2O_3 while maintaining a relatively low final hydrogen temperature. The configuration was illustrated previously [Weir et al., 1994]. The electrical resistance was measured and electrical resistivity was obtained by calibration (conductivity is the reciprocal of resistivity). Three somewhat different electrical circuits were used, depending on the expected resistance to be measured. Two or four electrode probes were used depending on whether the expected conductivity was small or large compared to $10\ (\Omega\text{-cm})^{-1}$. Thus, the conductivity was measured using two isotopes and three circuits. In the temperature experiments hydrogen was contained between an Al disk and an Al_2O_3 or LiF window [Holmes et al., 1995]. Pressure and density were obtained by shock impedance matching. The temperatures of the first shock and of this shock reflected off the window were determined by fitting the optical spectra of the emitted radiation to a greybody spectrum.

Results

Although condensed molecular hydrogen is a wide bandgap insulator at ambient pressure ($E_g=15\text{ eV}$), at sufficiently high pressure the energy gap is expected to close to zero. Previous experiments have measured the electrical conductivity of hydrogen in the fluid phase at single-shock pressures up to 20 GPa and 4600 K [Nellis et al., 1992].

In the present conductivity experiments multiple-shock pressures of 93-180 GPa were achieved. Hydrogen is in the fluid phase because the calculated temperatures are well above the calculated melting temperatures of ~ 1000 K at 100 GPa pressures [Ross et al., 1981]. The shock-compression technique is very well suited for measuring the electrical conductivity of hydrogen because: i) the high pressure reduces the energy gap, ii) the reverberating shock maintains temperatures at a few 0.1 eV, ~ 10 times lower than the temperature which would be achieved by a single shock to the same pressure, iii) the relatively low shock temperatures activate sufficient conduction electrons to produce measurable conductivities in the condensed phase, iv) electrode dimensions and separations are of order mm's, which are straight forward to assemble, and v) thermal equilibrium is obtained in a time less than the resolution (~ 1 ns) of the measurements. In these experiments the thickness of the hydrogen layer decreases from the initial value of 500 μm down to the compressed value of ~ 60 μm . Current flow reaches its equilibrium flow pattern in ~ 1 ns for a layer 60 μm thick with a minimum electrical resistivity of 5×10^{-4} $\Omega\text{-cm}$. Supplemental experiments examining the electrical conductivity of shock-compressed Al_2O_3 [Weir et al., 1996b] show that the conductivity of hydrogen at metallization (140 GPa) is 5 orders of magnitude greater than that of Al_2O_3 . Thus, the conductivity measured is that of equilibrated hydrogen with negligible contribution from the shock-compressed Al_2O_3 holding the measurement electrodes.

The electrical resistivities decrease from about 1 $\Omega\text{-cm}$ at 93 GPa to 5×10^{-4} $\Omega\text{-cm}$ at 140 GPa and are constant at 5×10^{-4} $\Omega\text{-cm}$ at 155 and 180 GPa, as shown in Fig. 1. The change in slope is indicative of the transition to the metallic state. Since our previous data [Nellis et al., 1992] show that the electrical conductivity of hydrogen is thermally activated, we analyzed these results in the range 93-135 GPa using our previous dependence for a fluid semiconductor:

$$\sigma = \sigma_0 \exp (- E_g / 2k_B T), \quad (1)$$

where σ is electrical conductivity, σ_0 depends on density ρ , $E_g(\rho)$ is the density-dependent mobility gap in the electronic density of states of the fluid, k_B is Boltzmann's constant, and T is temperature.

The density (ρ) and temperature (T) were calculated by computationally simulating each experiment using a standard equation of state of hydrogen [Kerley, 1983]. At the conditions achieved in these experiments, this equation of state is for the molecular fluid phase. Although a computational model for hydrogen introduces systematic uncertainties in the calculated densities and temperatures, the results are useful for understanding the slope change at 140 GPa. Our least-squares fit of Eqn. 1 to the data gives $E_g(\rho) = 0.905 - (67.7)(\rho - 0.3)$, where $E_g(\rho)$ is in eV, ρ is in moles/cm³ (0.28-0.31 moles/cm³), and $\sigma_0 = 140 (\Omega\text{-cm})^{-1}$. A value of $\sigma_0 \approx 200\text{-}300 (\Omega\text{-cm})^{-1}$ is typical of liquid semiconductors [Mott, 1971].

$E_g(\rho)$ derived from this fitting procedure and $k_B T$ are equal at a temperature of 0.3 eV and a density of 0.31 mol/cm³. In this region of density and temperature the energy gap is smeared out thermally, activation of electronic carriers is complete, disorder is already saturated in the fluid, and conductivity is expected to be weakly sensitive to further increases in pressure and temperature. At 0.31 mol/cm³ the calculated pressure is 120 GPa, which is close to the 140 GPa pressure at which the slope changes in the electrical resistivity (Fig.1). At higher pressures of 155 and 180 GPa the resistivity is essentially constant at $5 \times 10^{-4} \Omega$ or a conductivity of $2000 (\Omega\text{-cm})^{-1}$, a value typical of the fluid alkali metals Cs and Rb at 2000 K [Hensel and Edwards, 1996]. Thus, fluid hydrogen becomes metallic at about 140 GPa and 3000 K via a continuous transition from a semiconducting to metallic fluid.

About 5% of the hydrogen molecules are dissociated at metallization in the fluid at 3000 K (discussed below). The sizes of the hydrogen molecule and atom and their

initial electronic energy gaps are similar, which suggests that in the mixed-phase fluid at high temperatures the molecule and atom are mutually soluble and form a common mobility gap in their electronic density of states. Thus, the energy gap measured is that of the mixture. Additional experiments varying temperature and, thus, dissociation fraction are needed to determine the sensitivity of the band gap to the relative composition of molecules and atoms.

The metallization pressure of 140 GPa in the warm fluid is substantially lower than 300 GPa, the typical theoretical value for the crystalline solid at 0 K [Ceperley and Alder, 1987; Ashcroft, 1990]. The lower metallization pressure in the fluid is probably caused by the fact that hydrogen molecules approach closer to one another in the warm disordered fluid than in the cold crystal. Also, the electronic energy gap is known to be very structure dependant [Chacham and Louie, 1991; Kaxiras et al., 1991] and the disorder allows the fluid to effectively sample many crystalline structures with lower band gaps. In addition several interactions occur in a lattice occupied by diatomic hydrogen molecules which have a high zero-point vibrational energy, orient their molecular axes relative to the crystal structure, and undergo charge transfer. These interactions in the crystal inhibit band-gap closure [Ashcroft, 1995] and are eliminated by melting, thus reducing the metallization pressure relative to that of the solid.

We have also measured temperatures of liquid deuterium and hydrogen shocked at pressures up to 83 GPa and 5200 K [Holmes et al., 1995; Nellis et al., 1995]. Temperatures were measured by fitting the optical radiation emitted from shock-compressed hydrogen to a greybody spectrum. The first-shock temperatures up to 20 GPa are in excellent agreement with predictions based on molecular hydrogen [Ross et al., 1983]. The second-shock temperatures up to 83 GPa, obtained by reflection of the first shock off a window, are lower than predicted for the molecular phase. The lower measured temperatures are caused by a continuous dissociative phase transition above 20 GPa. This partial dissociation from the molecular to the monatomic phase absorbs

energy, which causes lower temperatures than expected if hydrogen were to remain molecular. The theoretical model for molecular dissociation derived from the temperature data was used to calculate the dissociation fraction in the conductivity experiments. At 140 GPa and 3000 K the dissociation fraction is calculated to be about 5%. Since only about 5% of the hydrogen molecules are dissociated at metallization, electronic conduction is probably caused by electrons delocalized from H_2^+ ions [Ashcroft, 1968]. This is a different phase than the monatomic one predicted initially by Wigner and Huntington [1935].

Application to Jupiter

The shock temperature data were used to derive a model for molecular dissociation at high pressures and temperatures. Because hydrogen is in equilibrium in these experiments, this model was used to calculate the isentrope of hydrogen starting from the surface conditions of Jupiter, 165 K and 1 bar pressure [Nellis et al., 1995]. Since Jupiter is ~90 % hydrogen by number, this simple approach gives important general features of Jupiter. That is, the presence of He is neglected because the equation of state and electrical conductivity of Jupiter are dominated by hydrogen. This approach is generally sufficient, except for the situation of convection in the Jovian mantle, as discussed below.

When plotted as temperature versus pressure, the isentrope of hydrogen increases modestly and monotonically up to about 40 GPa, at which point the temperature reaches a broad plateau; the temperature of hydrogen might even decrease ~2% from 40 to 180 GPa, as shown in Fig. 2. This weak temperature dependence is caused by the continual increase in molecular dissociation with increasing pressure. Thus, these calculations indicate that the interior of Jupiter is cooler and has much less temperature variation than believed previously. Of course, Jupiter contains about 10% He by number. Monatomic He is much hotter at a given density and pressure than is

molecular hydrogen because He has no internal degrees of freedom to absorb energy in this regime. Thus, the warmer He probably causes the temperature of Jupiter to always increase monotonically with increasing pressure. In this case the volume coefficient of thermal expansion is also positive and a positive volume coefficient drives convection. Jupiter is known to be convective because it has a large external magnetic field which varies from 14 G at the north magnetic pole to 11 G at the south magnetic pole [Smith et al., 1976], versus 0.5 G on Earth. The magnetic field is produced by the convective motion of electrically conducting hydrogen by dynamo action [Stevenson, 1983]. In addition, convective heat transfer to the surface is substantial and the reason why Jupiter radiates more internal energy than it receives from the sun [Hubbard and Lampe, 1969; Hubbard, 1968]. It is possible, however, that Jupiter is convectively quiescent over a radially thin region. A possible maximum in temperature versus pressure might induce an additional layer in the molecular region, as has been predicted to occur at 42 GPa by Zharkov and Gudkova [1992]. That is, a quiescent boundary layer over a long period of time might facilitate settling of ice and rock from hydrogen and cause an abrupt density change [Nellis et al., 1995].

The continuous dissociative phase transition from the molecular to the monatomic phase means that there probably is no sharp boundary between the molecular mantle and the monatomic metallic-hydrogen core of Jupiter. Most recent models of Jupiter assume a sharp boundary between a molecular mantle and monatomic core at an internal pressure of 300 GPa.

A major issue about Jupiter is the region in which the magnetic field is produced. On the basis of calculations of electrical conductivities of dense fluid hydrogen [Stevenson and Ashcroft, 1974; Stevenson and Salpeter, 1977; Hubbard and Lampe, 1969; Hubbard, 1968; Kirk and Stevenson, 1987], the magnetic field has been thought to be produced primarily in the monatomic metallic core and in the molecular mantle, as well [Kirk and Stevenson, 1987; Smoluchowski, 1975; Hide and Malin, 1979]. However,

the relative contributions of each region have been uncertain. Here we derive a scaling relationship for electrical conductivity and evaluate it as a function of pressure, density, and temperature along the isentrope of hydrogen starting from the surface conditions of Jupiter. To demonstrate that electrical conductivities were measured at pressures and temperatures representative of those in Jupiter, both our hydrogen isentrope and the temperatures and pressures achieved in the electrical conductivity measurements are plotted in Fig. 2.

In a semiconducting fluid electrical conductivities fit the relation in Eqn. (1) [Nellis et al., 1996]. Energy gap E_g versus density was derived from the recently measured conductivities in two different density ranges [Nellis et al., 1992; Weir et al., 1996a]. The fit to these two data sets is:

$$E_g = 20.3 - 64.7\rho, \quad (2)$$

in the range $0.13 < \rho < 0.3 \text{ mol/cm}^3$ with E_g in eV. The prefactors σ_0 determined in the two sets of experiments differ by 10^4 at densities of 0.13 and 0.30 mol/cm^3 (the corresponding pressures differ by a factor of 10). We obtain values of σ_0 at intermediate densities by exponential interpolation of these data:

$$\sigma_0(\rho) = 3.4 \times 10^8 \exp(-44\rho). \quad (3)$$

Eq. (3) fits the value of σ_0 determined at 0.13 mol/cm^3 ($1.1 \times 10^6 \text{ } (\Omega\text{-cm})^{-1}$), is close to the value obtained near 0.3 mol/cm^3 ($1.4 \times 10^2 \text{ } (\Omega\text{-cm})^{-1}$), where hydrogen is undergoing a continuous transition from a semiconducting to metallic fluid, and σ extrapolates to the metallic value of 2000 $(\Omega\text{-cm})^{-1}$ (resistivity = $5 \times 10^{-4} \text{ } \Omega\text{-cm}$) at 140 GPa.

The electrical conductivity of monatomic metallic hydrogen was predicted theoretically to be higher, $1\text{-}2 \times 10^5 \text{ } (\Omega\text{-cm})^{-1}$ [Stevenson and Salpeter, 1977; Hubbard and

Lampe, 1969], than measured for the molecular phase, $2 \times 10^3 (\Omega\text{-cm})^{-1}$, by Weir et al [1996a] at the density and temperature at which metallization is observed (0.31 mol/cm³ and 3000 K). The metallic hydrogen conductivity predicted theoretically is expected to be accurate at higher pressures for densities above 0.5 mol/cm³, where the molecules are expected to be fully dissociated into a monatomic metallic fluid.

The electrical conductivity of hydrogen along the isentrope in Fig. 2 was calculated with Eqns (1) - (3). In Fig. 3 the resulting conductivity is plotted versus pressure along this isentrope. The conductivity was calculated up to 120 GPa, where the electronic energy gap approaches the temperature, and was then extrapolated up to the metallic value of $2000 (\Omega\text{-cm})^{-1}$ at 140 GPa.

For comparison, we also calculated electrical conductivities in the molecular envelop of Jupiter [Kirk and Stevenson, 1987] using Eqn. (1) with $\sigma_0 = 300 (\Omega\text{-cm})^{-1}$, a value typical of a liquid semiconductor [Mott, 1971], and different relations of $E_g(\rho)$ at 0 K for molecular hydrogen calculated by Freidli and Ashcroft (FA) [1977] and by Min et al (M) [1986], along an isentrope of hydrogen calculated by Saumon et al. [1995]. These two curves, FA and M in Fig. 3, can only be calculated up to 200 GPa where the temperature on the isentrope is equal to the bandgap of the molecular phase. As shown in Fig. 3, for pressures below 20 GPa all three models approach a common value. For pressures above 40 GPa our calculated conductivities are 1 to 2 orders of magnitude greater than those calculated with FA and M. There is no physically meaningful way at present to interpolate from FA and M at 200 GPa in the molecular phase up to the calculated value of S for the monatomic phase at 300 GPa. Further theoretical results [Stevenson and Salpeter, 1977; Hubbard and Lampe, 1969] imply that the conductivity of monatomic metallic hydrogen at what was thought to be the core-mantle boundary, 300 GPa, 0.65 mol/cm³, and 10,000 K, should be $10^5 (\Omega\text{-cm})^{-1}$ (S in Fig. 3).

The metallization pressure of 140 GPa in the fluid observed by Weir et al [1996a] is substantially lower than 300 GPa, the typical theoretical value for the solid at 0 K

[Ceperley and Alder, 1987; Ashcroft, 1990]. This lower pressure implies that hydrogen in Jupiter becomes metallic at about 0.9 of the radius, as predicted by Smoluchowski [1975], rather than about 0.8 of the radius for 300 GPa. Because of the large volume of Jupiter this corresponds to about 50 more Earth masses of metallic hydrogen than thought previously. Also, an electrical conductivity as low as $20 (\Omega\text{-cm})^{-1}$ might contribute to the magnetic field of Jupiter because a conductivity of this value is thought to be responsible for producing the magnetic fields of Uranus and Neptune [Nellis et al., 1988]. As shown in Fig. 3 a conductivity of $20 (\Omega\text{-cm})^{-1}$ is achieved in Jupiter at 80 GPa. For these reasons, the external magnetic field of Jupiter would be produced in the molecular envelop substantially closer to the surface with a metallic conductivity about two orders of magnitude smaller than thought previously.

The above results imply that the magnetic field of Jupiter is produced in the region in which fluid molecular hydrogen undergoes a continuous dissociative phase transition. Using the theoretical model for molecular dissociation derived from shock temperature experiments, about 10% of the hydrogen molecules are dissociated at metallization in Jupiter (140 GPa and 4000 K). Once hydrogen metallizes, additional changes in electrical conductivity caused by the continuous molecular transition to complete dissociation at higher pressures are expected to be relatively small for an additional 2-3 fold increase in pressure. That is, in a fluid the increase in the electrical conductivity is dominated by the increase in the number of conduction electrons. Once they are all excited to form a metal, further changes in conductivity are expected to be relatively small ($\sim$ factor of 2) as density and temperature increase somewhat with increase in pressure and depth. So that in the regime up to 2-3 times the observed metallization pressure, the electrical conductivity is about $2000 (\Omega\text{-cm})^{-1}$. At higher pressures, temperatures, and depths at which molecular dissociation becomes complete, the pure monatomic phase is expected to form a metallic plasma and the higher predicted conductivity [Stevenson and Salpeter, 1977; Hubbard and Lampe, 1969] is

expected to be the case. However, the external magnetic field is caused primarily by a conductivity of about $2000 (\Omega\text{-cm})^{-1}$ in the outermost portion of Jupiter.

It is unlikely that small amounts of He, the ices, and rock contribute significantly to the electrical conductivity in Jupiter. The electrical conductivities of these materials are small compared to that of hydrogen. In addition, their concentrations are too small to affect the conductivity if they are in solution and would produce at most a tiny filamentary percolation path if they phase separate. These materials would be a small, electrically-inert volume fraction within the hydrogen.

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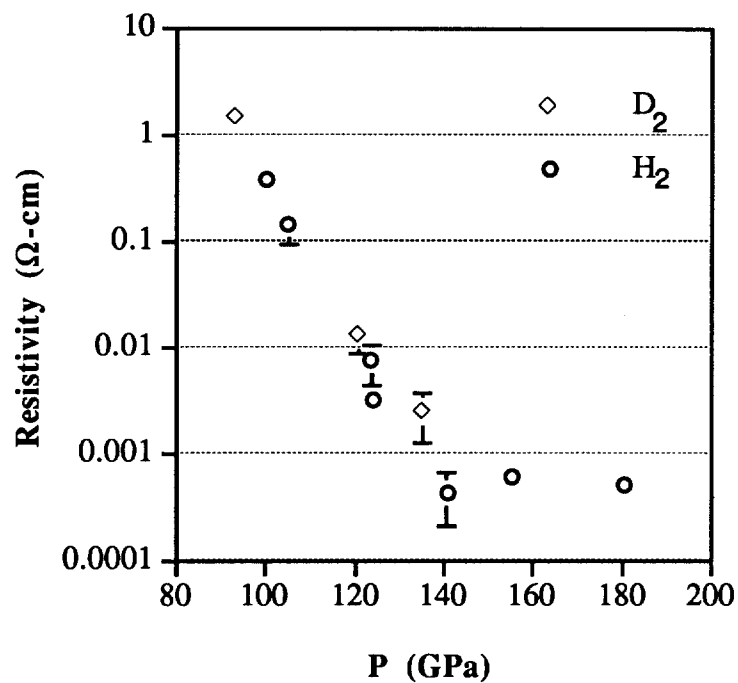


Fig. 1. Electrical resistivity versus pressure for fluid hydrogen and deuterium. The saturation resistivity of $500 \mu\Omega\text{-cm}$ above 140 GPa is that of the metallic fluid.

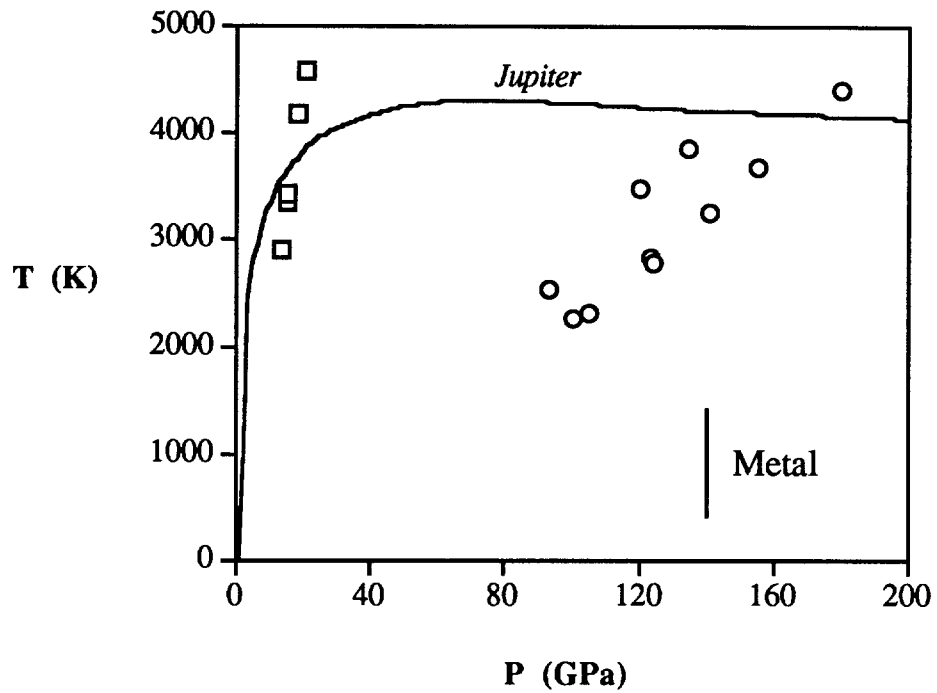


Fig. 2. The solid curve is the isentrope of hydrogen calculated from surface conditions of Jupiter (165 K and 1 bar) plotted as temperature versus pressure. Open circles and squares are temperature and pressure at which electrical conductivities were measured. Metallization of hydrogen in Jupiter occurs at 140 GPa.

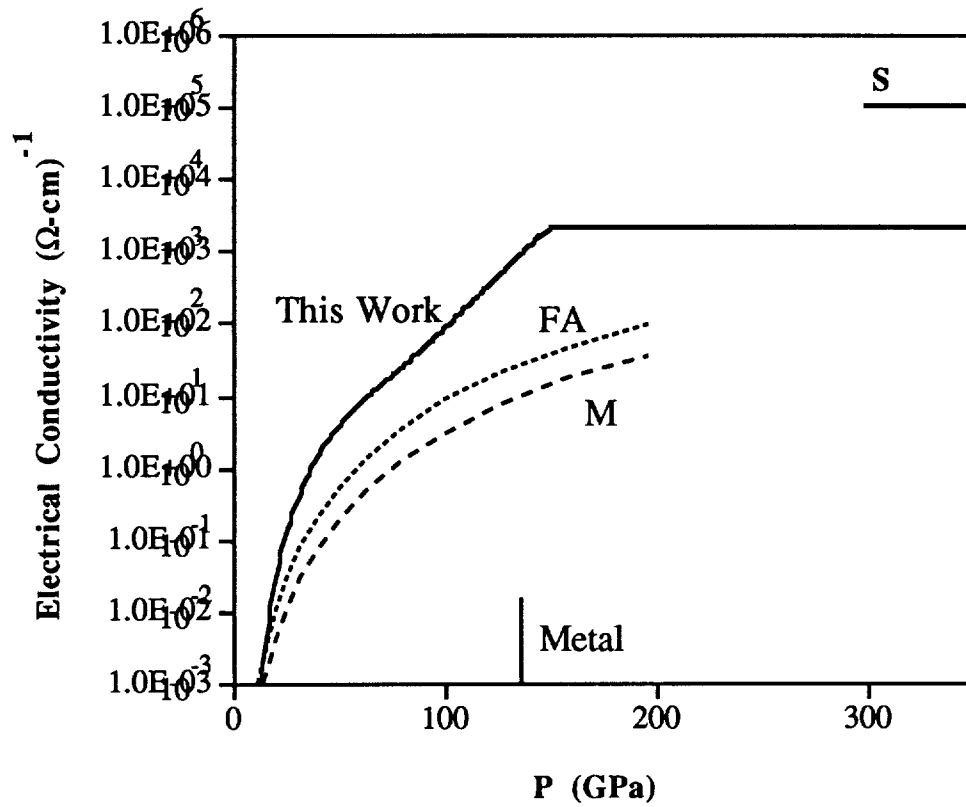


Fig. 3. Electrical conductivities of hydrogen in Jupiter plotted versus pressure along isentropes of hydrogen. FA, M, and S are defined in text.

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