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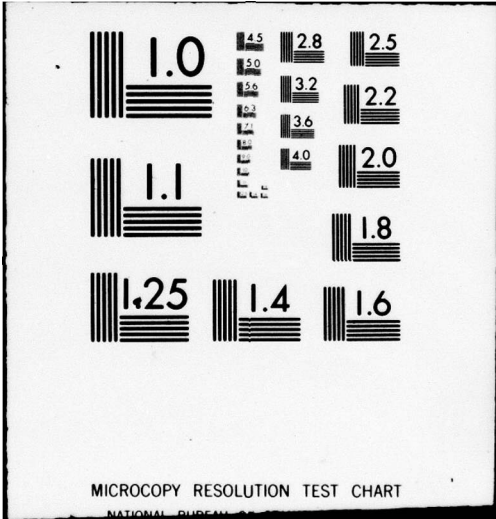
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METALLIC HYDROGEN

by

Hsu Tse-An and Chu Tse-Wan



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METALLIC HYDROGEN

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I. INTRODUCTION

In nature there is a variety of states in which matter exists. When external conditions change, the states of matter are different. For an example, we classify matter as insulator, semiconductor, or conductor according to its electrical conduction properties. The advance of modern technology, particularly modern high pressure technology, has shown that the conduction properties are not invariant. Under a sufficiently high pressure, an insulator may turn into a conductor or semiconductor. Since electrical conduction is a characteristic of metal, the phenomenon of a nonconductor turning into a conductor under high pressure is called metallization. For an example, a good insulator, polytetrafluoroethylene (Teflon) has a metallization pressure of 210-240 kilobar ($1 \text{ bar} = 10^6 \text{ dyne/cm}^2 = 0.98692 \text{ atmosphere pressure}$). The metallization pressure of phosphorus is 40-50 kilobar. Semiconductors such as germanium and silicon have metallization pressures of 100 kilobar and 125 kilobar, respectively. These phenomena proved the hypothesis of the English physicist J. B. Bernal who proposed in 1925 that any material could be turned into metal if enough pressure were applied [1].

According to solid state theory, the electric conduction of metal is caused by the overlapping of a filled band and an empty band. We know that every isolated atom has its electronic energy levels (see Fig. 1). When atoms are compressed closer together (i.e., interatomic spacing r is shortened) to form crystals, energy levels become energy bands. The bands which are filled with electrons are called filled bands. The band which has no electron is called a conduction band. Between conduction band and filled band there is a forbidden zone. At this time, the material is an insulator. When the forbidden zone is not very wide, some electrons in the filled band can be excited to the conduction band. At the same time, there are holes in the filled band. Both electrons and holes contribute to electric conduction.

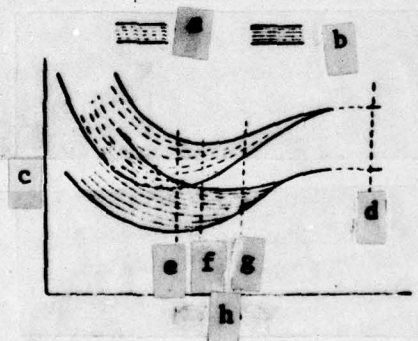


Figure 1. Relationship between interatomic spacing r and energy E .
 a - conduction band; b - filled band; c - energy; d - free atom; e - metal; f - semiconductor; g - insulator; h - interatomic spacing.

This is the characteristic of a semiconductor. The material is a semiconductor. When r decreases, the forbidden zone no longer exists and filled band and conduction band overlap each other, and the material becomes a metal. Therefore, when pressure is applied, the interatomic spacing decreases. Eventually, the material becomes metallized.

We can also explain metallization this way: In the free atom state, electrons belong to the atoms. They form an electron cloud around the atoms. When a crystalline state is formed, if the condition does not change drastically, interatomic interaction is weak, and the electron is localized around the atom. The material is in an insulating or semiconductor state. Under high pressure, the interatomic distance decreases continuously, and atomic

interaction becomes very strong. Eventually, some of the electrons leave their own atoms and are shared by other atoms. The material then becomes a metal.

Hydrogen is the first element on the periodic table. It is formed by one proton and one electron. Its atomic radius is one $1a_B$ (Bohr radius)*. It has three isotopes: H^1 (hydrogen); H^2 (deuterium) and H^3 (tritium). In nature, hydrogen is a mixture of hydrogen and deuterium.

At room temperature and atmosphere pressure, hydrogen is a molecular gas. The molecular bond length is $2\ell = 1.4$, and the average bonding energy for each atomic bond is $\delta_D = 0.08225$. At $20.36^\circ K$, the gaseous hydrogen is liquified. At $14.02^\circ K$, it becomes solid.

Solid hydrogen is a molecular crystal, i.e., every crystal lattice is occupied by a hydrogen molecule. Solid hydrogen forms a hexagonal, closely packed crystalline structure. At zero pressure, the crystal lattice is $a = 3.76 \text{ \AA}$, $c = 6.13 \text{ \AA}$. Under pressure, it has a face centered cubic structure with a lattice constant $a = 5.210 \text{ \AA}$. The mass density of solid hydrogen is $\rho_s = 0.089 \text{ g/cm}^3$. In 1935, E. P. Wigner and H. B. Huntington [2] showed that metallic hydrogen could exist as a metallic phase of hydrogen. With a pressure in excess of 400 kilobar, solid hydrogen can turn into metallic hydrogen. Thereafter, many authors have studied this problem and predict that the transition into the metallic phase occurs at a pressure between 0.25 and 18 mega bars.

II. METALLIC HYDROGEN TRANSITION PRESSURE

From thermodynamics, it is known that, when solid hydrogen is undergoing a transition into the metallic state, the chemical potential of solid hydrogen (subscript s) and metallic hydrogen (subscript m) are equal:

$$\mu_1 = \mu_2 \quad (1)$$

At 0°K,

$$\mu = \epsilon + pQ,$$

where ϵ is energy; p is pressure, Q is volume. Substituting into (1), we have

$$p = -(\epsilon_2 - \epsilon_1)/(Q_2 - Q_1). \quad (2)$$

Therefore, the problem of finding the pressure becomes one of finding the energies. Since hydrogen is an 1_A element in the periodic table, the energies of metallic hydrogen can be calculated by using the Wigner-Seitz cellular method which was applied to monovalent alkali metal with success. Let us assume that metallic hydrogen has the same structure as that of an alkali metal, which is body-centered cubic. Hence we can write the energy of the metallic hydrogen as

$$\epsilon_2 = \epsilon_{20} + \epsilon_{2v}.$$

ϵ_{20} and ϵ_{2v} represent the potential energy and zero point vibration energy, respectively. Then,

$$\begin{aligned} \epsilon_{20} = & \epsilon_1 Q_1^{-1/n} - \epsilon_2 Q_2^{-1/n} + \epsilon_3 - \epsilon_4 Q_1^{1/n} \\ & - \epsilon_5 Q_2^{1/n} + \epsilon_6 \ln Q_1, \end{aligned} \quad (3)$$

where the constants:

*To facilitate the discussion, unless otherwise specified, all the units used in this paper are in atomic units, and the electron mass m , charge e , and Planck's constant \hbar are all taken to be unity.

The length unit is Bohr's radius $a_B = \hbar^2/me^2 = 0.529177 \text{ \AA}$. The unit for energy is $e^2/a_B = 27.2 \text{ eV} = 2 \text{ Rydbergs}$. The unit for pressure is $C^2/a_B^2 = 294 \text{ mega bars} = 2.89 \times 10^8 \text{ atmospheres}$.

$$c_1 = 2.87137, c_2 = 2.18909, c_3 = 0.447992, \\ c_4 = 73.925 \times 10^{-6}, c_5 = 2.1856 \times 10^{-6}, c_6 = 82.5 \times 10^{-6}.$$

The calculation of the energy of solid hydrogen can be considered as follows: The interactions among hydrogen molecules are Born repulsion and Van der Waals attraction. Considering the molecules are in equilibrium, the interaction among them can be written as:

$$\varphi = a \exp(-bR) - cR^{-6}(1 + dR^{-1})e^{-eR^{-10}}, \quad (4)$$

where R is the distance to the center, $a = 3.39$, $b = 1.85$, $c = 10.9$, $d = 10.6$. The factor $e = 400R^{-6}$ is a correction term due to high pressure.

Applying the above interaction field and based on the face-centered cubic structure, summing up all the hydrogen molecules, we can obtain the potential energy $\phi_{..}$. Actually, the interaction between hydrogen molecules which are far apart is small. Only those nearest neighbors have strong interaction. Because the repulsive term $a \exp(-bR)$ converges faster than the Van der Waals term, one has to sum up more molecules for the latter.

Since the atomic weight of hydrogen is small, the zero point energy ($\phi_{..}$ and $\phi_{..}$) must be included. A calculation shows that this term is about 1/10 of the total energy.

It should be pointed out in calculating energies for metallic and solid hydrogen that the reference zero point energy should be the same. Generally, an isolated hydrogen molecule is taken to be the zero point.

The energy of solid hydrogen can be calculated by some empirical formulas. We have tried to use the Morse potential and equation of states. This way one can plot the energies of metallic and solid hydrogen ϵ versus volume Q (Fig. 2). From this, one can see that

the slope of the tangent line which is common to both is the transition pressure $p = 1.35$ mega bar.

Figure 2 also shows that $Q = 20.083$, metallic hydrogen has a minimum. This corresponds to the zero pressure atomic volume of metallic hydrogen. The corresponding zero pressure density is 0.562 g/cm^3 .

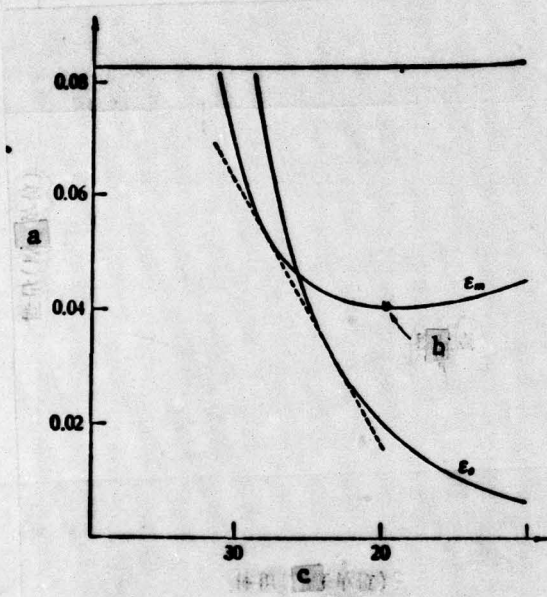


Fig. 2. ϵ - Q curves for metallic and solid hydrogen

a - energy (in a.u.);
b - equilibrium point;
c - volume (in a.u.).

Table 1. lists the calculations of different authors. It is seen that, even though the predicted transition pressures are quite different, they are all on the order of magnitude of several million atmospheres. Therefore, the formation of metallic hydrogen presents a new challenge for high pressure physics and high pressure technology.

Table 1. Calculated Values for Metallic Hydrogen Transition Pressure by Different Authors.

1	3		7	8		10	11
	4	6		9			
Wigner, Huntington	0.62	0.025				0.4	
Kronig, Koringa, De Boer	0.47		0.8	0.4		0.7	
Абрикосов			1.12	0.625		2.4	
De Marcus	0.525					1.93	
Carr		0.029					
Трубицын	0.57		1.40	1.0		4.6	
Schneider			1.39			2.52	
Neese, Roger, Hoover		0.0365	0.83	0.48		0.80	
2	0.562	0.04146	0.926	0.668		1.35	

KEY: 1 - Authors; 2 - Hsu Tse-An and Chu Tse-Wan; 3 - Metallic hydrogen; 4 - Equilibrium point; 5 - density (g/cm^3); 6 - binding energy (a.u.); 7 - transition density (g/cm^3); 8 - solid hydrogen; 9 - transition density (g/cm^3); 10 - transition pressure (megabar); 11 - not less than.

III. THE CHARACTERISTICS OF METALLIC HYDROGEN AND ITS POSSIBLE APPLICATIONS

However, man has not yet produced metallic hydrogen. Based on theoretical considerations and the practical applications of liquid hydrogen and solid hydrogen, metallic hydrogen not only has theoretical significance, but could have wide practical applications. In general, as a new material, it has high density, high energy content, high T_c (superconducting transition temperature) and good electric and thermal conducting properties.

1. High Density, High Energy Content Material

At present, liquid hydrogen and solid hydrogen are widely used in rocket technology. Because metallic hydrogen has a density of 0.562 g/cm^3 compared to 0.089 g/cm^3 for solid hydrogen, it is about

6.3 times higher. It is about 7.9 times higher compared to liquid hydrogen (0.071 g/cm^3). Therefore, when used as rocket fuel, it is far superior than solid or liquid hydrogen. It is possible to reduce the volume and weight of rockets, and advance the technology of rockets and missiles.

Calculations show that the binding energy of metallic hydrogen is about 414.6×10^{-4} Kcal/mole, which is 24.7 times that of TNT. Therefore, if metallic hydrogen is stable, it can be used as a high explosive material. From the ρ - E relationship of metallic hydrogen, the bulk modulus is 1.232 megabar. Assuming the Poisson ratio $\sigma = 0.35$, and the Young's modulus $E = 1.109$ megabar, it is very close to that of copper. From the Lindermann equation, it can be calculated that the fusion point of metallic hydrogen is $T_m = 408^\circ\text{K} = 135^\circ\text{C}$. Since the binding energy of metallic hydrogen is close to sodium (25.9 Kcal/mole), its boiling point should also be very close. One concludes then that it is not likely that metallic hydrogen will lose its stability through surface evaporation.

Since hydrogen can be burned, metallic hydrogen can be a non-polluting energy source. It could be applied to aviation, transportation and batteries. Besides, deuterium is the main fuel for nuclear fusion. Since the density of metallic deuterium is high, the fusion temperature can be lowered. The existence of metallic deuterium will accelerate the implementation of controlled nuclear fusion. It will be a revolutionary advance for energy sources. At the same time, it will make an important contribution to modernization of national defense.

2. High T_c Material

As we know, due to the advance of low temperature technology, superconductivity is becoming more useful. At present, the material which has the highest transition temperature is Nb_3Ge . Its T_c is 23.2°K . Consequently, all superconductive devices have to be used with liquid helium. Ashcroft [3] pointed out that metallic hydrogen

could be a room temperature superconducting material, i.e., metallic hydrogen can raise the superconducting temperature from more than two hundred degrees below zero to room temperature. Hence, metallic hydrogen could be applied to transporting electric energy without loss, superconducting magnets, energy storage coils, high velocity magnetic suspension trains, supersensitive superconduction gyros and superconducting antennas without depending on low temperature equipment. This will greatly enlarge the scope and utility of the applications of superconductivity. Based on Bernal's theory with Ashcroft's correction, we derived the equation for calculating the transition temperature T_c for metallic hydrogen. The calculation shows that T_c is about 80°K , which is much lower than the one predicted by Ashcroft, but still about three times higher than Nb_3Ge . We believe that the superconducting property of metallic hydrogen can be realized around the temperature of liquid nitrogen (77°K). This certainly will produce revolutionary advances of superconducting technology.

3. Good Thermal and Electric Conducting Material

From the Zeeman equation, one can calculate the electric resistivity of metallic hydrogen under zero pressure to be 0.638×10^{-6} ohms-cm which is smaller than copper (1.692×10^{-6}) which is a good electric conductor, we expect it to be a good thermal conductor also. Calculations show that the thermal conductivity of metallic hydrogen is $10.5 \text{ watt}/^\circ\text{C-cm}$, which is twice that of copper.

As we know, there are many so-called "hydrogen stars" in the universe. Jupiter and Saturn, which belong to the same solar system as the Earth, are these kinds of stars. Their temperature is very low. The internal pressure is high, which provides the condition for the existence of metallic hydrogen. They also have a very strong magnetic field which shows that superconducting metallic hydrogen exists. C. DeMarcus [4] thinks that the contents of metallic hydrogen are 78% and 63% for Jupiter and Saturn, respectively.

Since metallic hydrogen is a good thermal conductor, in the interior of these stars no large nonuniformity of temperature should exist.

IV. PRESENT RESEARCH STATUS OF METALLIC HYDROGEN

In 1935 when Wigner calculated metallic hydrogen, it was considered that the necessary pressure for metallic hydrogen to exist is about 400,000 atmospheric pressures and was considered not achievable. However, technical advances have been made in both dynamic and static high pressure methods.

Dynamic high pressure is created by explosions or high velocity pulses. This induces a shock wave inside the sample and creates a very high instantaneous pressure. This method can create pressure up to several million atmospheres of pressure. The shortcoming of this method is that the duration is too short. Los Alamos Laboratory [5] of the U. S. uses the magnetic compression method. They used explosives to compress the magnetic field which creates an instantaneous magnetic field of 10 million Gauss and compresses the liquid hydrogen stored within the sample. The pressure reaches about 1-4 megabar. Because of the use of magnetic compression, the temperature of the sample rises rather moderately when compared with using explosives. The duration also increases from 10's of ns for explosion to about 10 μ s. Data taken during compression show that liquid hydrogen becomes conducting metallic hydrogen under high pressure. Gregor'yev and his associates in the USSR used explosives to create shock waves to compress liquid hydrogen directly. They discovered that the p (pressure) - ρ (density) curve of the hydrogen has a turning point at $p = 2.8$ megabar. They believe this corresponds to the phase transition point for metallic hydrogen [6].

To prepare metallic hydrogen by the static compression method, the pressure has to reach about a megabar. Consequently, many countries are manufacturing large oil hydraulic pressure machines to create this kind of high pressure. The USSR Institute of High Pressure Physics and Chemistry designed a hydraulic pressure machine

of fifty thousand tons which stands fifty meters in height. They claim that, after this machine goes into operation, metallization of hydrogen can be realized.

However, the first hydrogen metallization is achieved by using a small diamond with a sharp point. In 1975, Vereshchagin, et al. [7] published an experimental report which applied a force of no more than 30 kg to solid hydrogen.

Their experiment set-up is shown in Fig. 3. At a low temperature (4.2°K), container 1 has a pair of diamonds. One of them denoted as 3 has a flat surface and is fixed at the bottom. Another diamond 2 has a sharp point and is allowed to move up and down. Hydrogen gas is blown into 1. It forms a layer of solid hydrogen "frost". Now a weight is placed on top of 2. The point of contact between 2 and 3 has a very high pressure. This is due to the fact that the point of 2 is very sharp and has a very small area. High pressure is created in this way. No more than 30 kg is needed to metallize hydrogen.

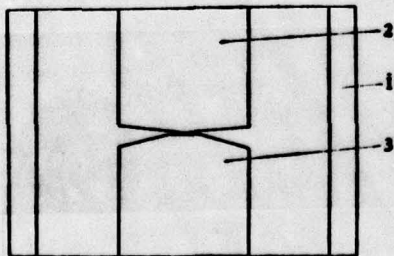


Fig. 3. Static pressure experimental set up for metallic hydrogen.

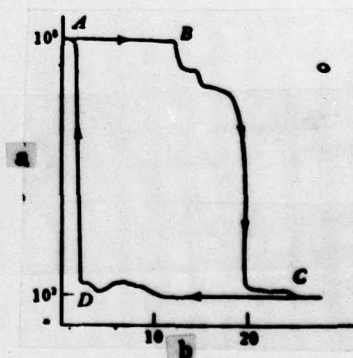


Fig. 4. Experimental curve for metallic hydrogen transition

a - resistance (Ohm);
b - total force (kilogram).

Figures 4, 5, and 6 show their experimental results. Figure 4 shows that when pressure begins to increase (point A), because solid hydrogen is an insulator, the resistance between 2-3 is very large - about 10^8 ohms. When the weight reaches 20 kg at point B, solid hydrogen starts to transform into metallic hydrogen. The electric resistance starts to decrease. At point C, the process is completed. When we reduce the pressure, metallic hydrogen retains its property until it reaches point D, and back to solid hydrogen.

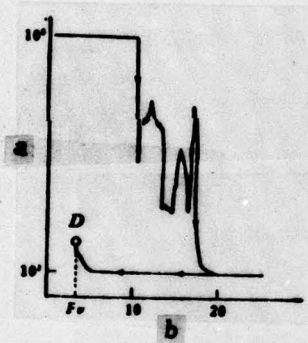


Fig. 5. Maintain metallic hydrogen phase under pressure F_v

a - resistance (ohm);
b - total force (kg).

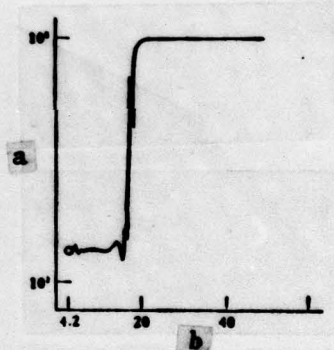


Fig. 6. Metallic hydrogen returns to solid hydrogen phase as temperature increases.

a - resistance (ohm);
b - temperature ($^{\circ}K$).

Figure 5 shows that, after the transition to a metallic state is completed and the pressure is reduced to point D and does not reduce further, the metallic state could be stable. Therefore, to maintain hydrogen in a metallic state, a constant pressure F_v is required. At constant pressure F_v but increasing temperature, Fig. 6 shows that, after the temperature reaches $20^{\circ}K$, a metallic state can still be maintained. At a higher temperature, metallic hydrogen reverts back to solid hydrogen. This experiment not only shows that metallic hydrogen could be an unstable phase of room temperature, it also shows that the conduction phenomenon in the

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experiment is not due to a short-circuit, but it is due to the phase transition into a metallic state.

V. CONCLUSION

Vereshchagin's experiment was the first successful phase transition from solid hydrogen to metallic hydrogen. It was an important advance in the technology of metallic hydrogen. However, since the structure of a high pressure container can produce only very small amounts of metallic hydrogen, physical properties of metallic hydrogen cannot be tested. Therefore, we are still far from obtaining metallic hydrogen. In order to obtain enough metallic hydrogen, first, a sufficient high pressure has to be produced. This is the direction of current high pressure technology research. Besides, since the compression ratio of solid hydrogen is very large, some present day high pressure technology cannot be directly applied. Therefore, research on metallic hydrogen will stimulate the advances of high pressure technology.

Even though Vereshchagin's experiment showed that at room temperature metallic hydrogen is unstable, but a critical temperature to maintain the metallic state at a high pressure could exist, the value of it is obviously very important. Also, metallic hydrogen created in the experiment is very small in quantity, is that quantity smaller than the critical crystal lattice which could exist at constant temperature and pressure? All these questions have to be answered by further research. It is interesting to note that, in the previous discussion, to realize metallic hydrogen, the starting point is always solid hydrogen. In reality, since in all the present phases of hydrogen, the energy of solid hydrogen is the highest and is most close to the energy of metallic hydrogen, using solid hydrogen as a beginning state will facilitate the phase transition into the metallic state.

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However, are there any other ways one can begin with? We know that the problem did not begin with Wigner's calculation. As early as 1869, Graham studied the barium-hydrogen property [8], and found that, as the amount of hydrogen increased, the mechanical properties and electric resistivity of barium-hydrogen also changed. It maintained its metallic appearance and the property of being stretched. He considered barium-hydrogen was actually the combination of the metallic phases of hydrogen was called "metallic hydrogen". In recent years, studies on Ba-H show that, after hydrogen enters Ba, it donates part of its electrons. At the same time, even though Ba is not a superconductor, injecting hydrogen into Ba will cause superconductivity to occur. Ba-H has a T_c above 1°K . Ba-H_{0.9} has T_c in excess of 5°K . Foil of the alloy of Ba-Cu can be made to be superconducting through the ion implantation method. The T_c is 16.6°K . Some people think that this is due to the high T_c of metallic hydrogen.

Baranowski [9] determined the crystal lattice constant changes as the amount of hydrogen increases. Based on his experiment, we have calculated the volume of the hydrogen atom in barium. Calculations show that metallic hydrogen has a volume of 20.2 (a.u., the unit of volume is a_B^3), which is very close to the atomic volume of metallic hydrogen, which is 20.083. It is very different from that of the negative ion volume of 60-100 for hydrogen. This shows that the hydrogen in Ba exists in the form of metallic hydrogen.

Actually, to convert carbon (graphite) to diamond under high pressure, some metal which will react with carbon to form a compound is used as a catalyzer to reduce the required pressure considerably. We assume that, if hydrogen compounds are used as catalyzers, the pressure required for the metallic hydrogen transition to occur may not be that high. At the same time, it might also stabilize metallic hydrogen at constant pressure. However, initial estimates show that metallic hydrogen cannot be obtained from Ba-H under pressure.

Research in foreign countries also has been along the same line. Gilman [10] argued for using LiH and HF to create a new compound. - LiH_2F and the possibility of forming a stable metallic hydrogen lattice in the LiF lattice, even though using a different initial state of hydrogen to form metallic hydrogen has not been very successful so far. This is perhaps one of the more promising ways to obtain metallic hydrogen in quantity. This remains a difficult task.

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