

Resistivity behavior of hydrogen and liquid silane at high shock compression

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ABSTRACT

To study the electrical properties of hydrogen rich compounds under extreme conditions, the electrical resistivity of density hydrogen and silane fluid was measured, respectively. The hydrogen sample was prepared by compressing pure hydrogen gas to 10 MPa in a coolant target system at the temperature of 77 K. The silane sample can be obtained with the same method. High-pressure and high-temperature experiments were performed using a two-stage light-gas gun. The electrical resistivity of the sample decreased with increasing pressure and temperature as expected. A minimum electrical resistivity value of $0.3 \times 10^{-3} \Omega \text{ cm}$ at 138 GPa and 4100 K was obtained for silane. The minimum resistivity of hydrogen in the state of 102 GPa and 4300 K was $0.35 \Omega \text{ cm}$. It showed that the measured electrical resistivity of the shock-compressed hydrogen was an order of magnitude higher than fluid silane at 50–90 GPa. However, beyond 100 GPa, the resistivity difference between silane and hydrogen was very minor. The carriers in the sample were hydrogen, and the concentration of hydrogen atoms in these two substances was close to each other. These results supported the theoretical prediction that silane was interpreted simply in terms of chemical decomposition into silicon nanoparticles and fluid hydrogen, and electrical conduction flows predominately dominated by the fluid hydrogen. In addition, the results also supported the theory of “chemical precompression”, the existence of Si–H bond helped to reduce the pressure of hydrogen metallization. These findings could lead the way for further metallic phases of hydrogen-rich materials and experimental studies.

1. Introduction

As the lightest and putatively simplest of the elements [1], hydrogen is one of the most abundant elements in the planets Jupiter and Saturn. The interiors of these giant planets are fluids, at high pressures and temperatures, because of their large masses and low thermal conductivities. The magnetic fields of giant planets are produced by the convective motion of electrically conducting hydrogen by dynamo action. Researchers have wondered what happened to hydrogen in this state.

In 1935, Wigner and Huntington [1] predicted that solid molecular hydrogen would dissociate and form the atomic state, with metallic phase under high pressures. The transformation from molecular hydrogen to a metal has generated lively debate in experimental [2–9] and theoretical [10,11] work. In 2017, the lively debate has been resolved to the cross over to metallic fluid hydrogen as discussed in “Ultracondensed Matter by Dynamic Compression” by W. J. Nellis [12]. In the same year, Dias and Silvera (DS) reported on production of

metallic hydrogen in a diamond anvil cell at 495 GPa at 5.5 and 83 K [13]. These studies showed that the conditions for the preparation of metallic hydrogen were too harsh. To circumvent this problem, it was proposed to compress group IV hydrides, in which the electron density on the hydrogen atoms is equivalent to pure hydrogen, to megabar pressures, where they are expected to metallize at these much lower pressures [14]. Silane has been the subject of most theoretical and experimental research so far [15–18].

However, little information is available about the high-pressure and high-temperature behavior of silane and hydrogen, and experimental data from these materials, under high pressure and temperature, are scarce. The interesting question is whether or not silane could undergo a transition to an eventual metallic state under lower pressure. Measuring the electrical resistivity of samples is a good basis on which to potential metallization, and will provide additional insight into this possibility. In this letter the electrical resistivity of silane and hydrogen under multiple shock compression is obtained. A principal purpose of this paper is to report experimental results on silane and hydrogen

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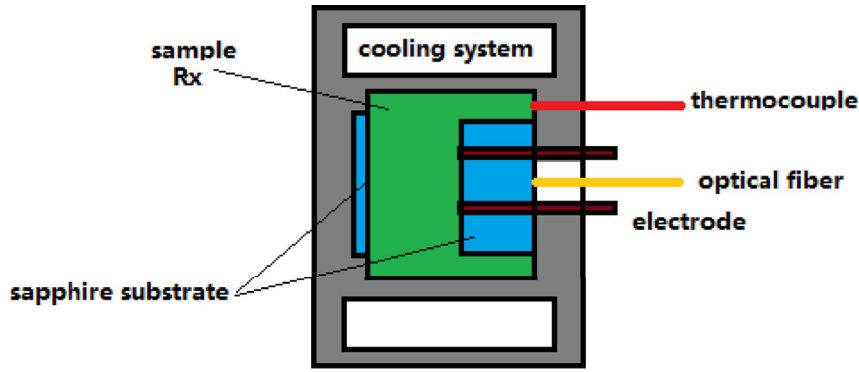


Fig. 1. The schematic diagram of low temperature target, including sample pool, cooling circulation channel and liquid-nitrogen storage chamber.

under shock compressed conditions.

2. Experimental principles and methods

2.1. Sample preparation and shock loading

The high pressures and high temperatures were obtained with a two-stage light gas gun, which accelerates impactor velocities in the range 3.05–5.34 km per second. A strong shock wave was generated on impact with a sandwiched target, which was composed of aluminum/sapphire/samples/sapphire as illustrated in Fig. 1 [19,20]. The fluid silane was condensed from pure gas in a cavity cooled to 88 K by an outer cooling jacket filled with liquid N₂. The advantage of the approach can avoid the leaking of silane, which may cause a fire or possibly explosion in air. The same method was used for the preparation of high density hydrogen samples. This special arrangement will generate wave reverberation in samples place between two stiff sapphire anvils. The samples will be repeatedly shocked to much higher pressure and temperature by the reverberation effects [19,21]. A reverberating shock wave was used to measure the electrical resistivity of samples. An optical fiber was used to collect light signals. It is worth mentioning that the recorded light signal was synchronized with the measured conduction voltage of the compressed sample. By combining the light signal with the electrical signal and results of the one-dimensional hydrodynamic simulation, several points on the electrical resistivity versus pressure curve in a single shock experiment were recorded. The shocked states in the samples were found using impedance matching [19,21]. For the first shocked state, we can determined the pressure, density, and temperature, by using the impactor velocity and initial densities of the sample, impactor, and baseplate. In a similar fashion, the reshocked states were found using the known first shocked state.

2.2. Test approach

The electrical resistance of a sample was measured by inserting electrodes through the sapphire anvil, the two electrodes were separated from each other by 3 mm. Trigger pins were used to turn on the recording system, Tektronix TDS 684C digital oscilloscopes with ~2 ns time resolution at the appropriate time and ~15.6 mV voltage resolution. The measured electrical resistivity of shock-compressed sapphire is five orders of magnitude larger than that of hydrogen at metallization, it is necessary to show that the electrical resistivity of shocked sapphire anvils is large, compared to that of a sample, so that the current is not shunted through the shocked sapphire rather than conducted through the sample [22]. In order to accurately measure the potential difference across the two electrodes, two modes of operation of the measurement circuit were used, the two circuits are constant voltage source and constant current source, as illustrated in Fig. 2. Ref. [20] gives us the principle and structure of the on-line test circuit. To

derive electrical resistivity from the measured resistances, the cell constant must be identified. The cell constant is a geometry-dependent parameter, calculated using the models of Nellis, the method is described elsewhere in more detail [19], so that $R = C/\sigma$. σ is conductivity of sample.

In our experiments, the initial state of liquid silane was at 88.5 K, and its initial density was 0.711 g/cm³ [20]. We also measured the resistivity of high density hydrogen by using the same experimental method. The initial state of density hydrogen was at 77 K, and its initial pressure was 10 MPa. The hydrogen equation of state was that of Ref. [23]. All thermodynamic calculations were complete by a computation program. Table 1 shows the experimental conditions.

3. Thermodynamic state

The materials involved in the impact loading experiments include copper, LY12 aluminum and sapphire. Their shock Hugoniot parameters [24,25] are shown in Table 2. The equation of state (EOS) of these materials, and the impact of the Hugoniot equation, are calculated with the Gruneisen model, and simulated with an approximate value using $\rho\gamma = \rho_0\gamma_0$. As noted above, we need to use a theoretical EOS for the sample to perform impedance matching calculations for shocks which reflect from the window. The EOS of the dense sample is calculated using perturbation theory [26]. The intermolecular potential of the exp-6 formula parameters are determined by the critical temperature and density of the fluid samples [20,26–28] based on the corresponding states principle [29]. The parameters (α , ϵ , r^*) of the molecular interaction potential in exp-6 formula are shown in Table 3.

The exp-6 formula is given by

$$\varphi(r) = \epsilon \left[\frac{6}{\alpha - 6} e^{\alpha \left(1 - \frac{r}{r^*}\right)} - \frac{\alpha}{\alpha - 6} \left(\frac{r^*}{r}\right)^6 \right] \quad (1)$$

and the Helmholtz free energy is given by the free energy of the ideal gas and interaction term. We write the Helmholtz free energy for a system of molecules as

$$F(V, T) = F_{id}(V, T) + A_p(V, T) \quad (2)$$

The free energy of an ideal gas comes from the molecular translational motion and internal degrees of freedom, including internal vibration and rotation

$$F_{id}(V, T) = F_{trans}(V, T) + F_{int}(T) \quad (3)$$

where $F_{int}(T)$ is the contribution from the rotational and vibrational degrees of freedom. The various terms are given by:

$$F_{trans}(V, T) = \sum_i NkT \left[\ln \left(\frac{N\lambda^3}{V} \right) - 1 \right] \quad (4)$$

$$F_{int}(T) = - \sum_i NkT \ln(j(T)) \quad (5)$$

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