Theoretical study of density condition of solid metallic hydrogen for fuel pellet

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Abstract. Hydrogen fuel pellet is often used in the laser fusion. In many cases, the hydrogen state as a fuel pellet is kept in low temperature in order to achieve high density solid or liquid states. Such states are very delicate and not easy to handle. On the other hand, a solid metallic state is said to possess similar high density. Additionally, it could be metastable in ambient temperature and utilized as a maintenance free fuel pellet. We have focused on the solid atomic hydrogen and studied theoretically the density condition of the metallization. In this study, we have picked up seven crystalline structures and compared the transition density for each structure. It is found that the density becomes higher as its structural filling factor becomes larger.

1. Introduction

Hydrogen target is often used as a fuel pellet for the laser driven inertial fusion research. As a target, deuterium and tritium are mixed and kept in solid or liquid state to obtain high initial density. Typically, the density is about 0.2 g/cc. Such a high density state is maintained in low temperature because it is not kept in ambient temperature and pressure. On the other hand, as a state of high density hydrogen, a solid metallic state is expected to exist. The solid metallic hydrogen is supposed to have similar or higher density compared with the hydrogen state as a fuel pellet. If there is a metastable solid metallic state in ambient temperature and pressure, it can be used as a new fuel pellet [1] which is very easy to handle. In this paper, we focus on the solid atomic hydrogen and investigate the metal-insulator transition (MIT) density for several structures. The solid metallic hydrogen was first predicted over 70 years ago [2] and challenges to make it are still continued. The solid metallic state is thought to exist in the low temperature (up to 1000K) and high pressure (more than 350GPa) region. In the limit of high pressure (and about room temperature), monatomic solid metallic hydrogen is said to exist. At the present time, the highest pressure achieved in room temperature is about 320GPa [3]. However, there was no evidence for the metallization and the dissociation of a hydrogen molecule. The extrapolation of this experiment and other recent studies [4] indicate that about 400GPa is necessary to make the solid metallic hydrogen. It is uncertain which of the metallization and the dissociation of hydrogen molecule is takes place at lower pressure. This is because to execute experimental research on ultra-high pressure region (more than about 300GPa) in room temperature is quite difficult[5]. Unfortunately, numerical simulations in such density and temperature region are also difficult. At the moment, in many cases, solid molecular...
hydrogen is research subject of the numerical studies on the metal-insulator transition (MIT) [6]. Few calculations of the MIT of solid atomic hydrogen were done in body centered cubic structure [7]. In this study, we consider the MIT of solid atomic hydrogen. We have assumed several crystal structures and figured out the transition densities for each structure. We have discussed the relationship between the densities and the structures.

2. Solid atomic hydrogen
In this study, we consider a crystal of solid atomic hydrogen and discuss metal-insulator transition of the material. All the electrons are bounded in 1s state. We consider inter-electronic interaction only in the case that two electrons are in the same site. Such two electrons are both in 1s state and possesses opposite spin. We ignore the interaction if two electrons are in different sites and also, we ignore the overlap integral for different sites. The model Hamiltonian is Hubbard Hamiltonian and it is given by

\[ \hat{H} = -E_{1s} \sum_i \hat{n}_{i\sigma} - t \sum_{\langle i,j \rangle} \hat{C}^\dagger_{i\sigma} \hat{C}_{j\sigma} + U \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}. \]  

Here, \( \hat{C}^\dagger_{i\sigma} \) is creation and \( \hat{C}_{i\sigma} \) is annihilation operator for an electron in \( i \) site and possesses spin \( \sigma \). \( \hat{n}_{i\sigma} = \hat{C}^\dagger_{i\sigma} \hat{C}_{i\sigma} \) is number operator. \( E_{1s} \) is bound energy of 1s electron. The first term is negligible because this term always brings the eigenvalue \( -E_{1s}N \) (\( N \) is particle number). \( t \) is hopping energy which represents an energy that an electron moves toward a neighbor atom by the Coulomb attractive potential of the atom in the neighbor site.

\[ t = \frac{\hbar^2}{4\pi \varepsilon_0} \int \frac{\phi_{1s}(\vec{r} - \vec{R}_j) \hat{\phi}_{1s}(\vec{r} - \vec{R}_i)}{|\vec{r} - \vec{R}_j|} \phi_{1s}(\vec{r} - \vec{R}_i) d\vec{r} = \frac{\hbar^2}{4\pi \varepsilon_0 a_B} (\alpha + 1) e^{-\alpha}. \]  

Here, \( \phi_{1s}(\vec{r} - \vec{R}_i) \) is 1s wave function bounded in site \( i \). \( a \) is distance of first nearest neighbors and \( \alpha = a/a_B \). \( a_B \) is Bohr radius. \( U \) is so called Hubbard U which represents the energy of the electronic interaction. This value is given by a mean value,

\[ U = \langle \psi_U | \hat{V}_{int} | \psi_U \rangle = \frac{5}{8} \frac{\hbar^2}{4\pi \varepsilon_0 a_B} = 17.0 (eV). \]  

Here, \( \psi_U = \phi_{1s}(\vec{r}_1) \phi_{1s}(\vec{r}_2) \) and \( \hat{V}_{int} = \frac{4\pi}{\sqrt{4\pi \varepsilon_0 a_B} |\vec{r}_1 - \vec{r}_2|} \). Comparing \( t \) and \( U \), we obtain \( t/U = 1.6(\alpha + 1)e^{-\alpha} \). The density region we are concerned is about \( \alpha > 3.3 \) (see, fig. 2(a)) and then, \( t/U < 0.254 \). It indicates the third term of the Hamiltonian is dominant for the MIT. It means, the MIT is Mott transition. By using this Hubbard model, we consider below seven structures to compare each other. The seven structures are simple cubic (sc), body centered cubic (bcc), face centered cubic (fcc), hexagonal close-packed (hcp), diamond structure, wurtzite structure and graphite structure.

3. Way of calculations
In order to treat the Mott MIT in the solid atomic hydrogen, we use Coherent Potential Approximation (CPA)[8]. In this approximation, single electron Green function \( G(\omega) \) and its self energy \( \Sigma(\omega) \) for an electron of energy \( \omega \) is decided through two equations,

\[ \Sigma(\omega) = \frac{nU}{1-(U-\Sigma(\omega))G(\omega)}, G(\omega) = \int \frac{\rho_0(E)}{\omega - E - \Sigma(\omega) + i\delta}. \]  

Here, \( \rho_0(E) \) is the density of state (DOS) without interaction between electrons. The DOS is obtained by the eigenvalues of first and second term of the Hamiltonian (1). \( n = 1/2 \).
represents paramagnetic state and \( \delta \) is infinitesimal. After finishing the calculation, we consider \( \delta \to +0 \) and we can obtain the density of state with interaction between electrons as \( \rho(\omega) = -(1/\pi) \lim_{\delta \to +0} \text{Im} G(\omega) \). By definition, \( \rho_0(E) \) varies with structures. This is because possible direction and number of the hopping is different from the structures and the eigenvalues are changed. To simplify our discussion, we normalize the DOS so that the normalized DOS \( \rho_0(x) \) satisfies \( \int \rho_0(x)dx = 1 \) and the energy is also normalized and restricted \(-1 \leq x \leq 1\). In fig. 1, we show a rough sketch of \( \rho_0(x) \) for the case of \( N \to \infty \). In this limit, \( \rho_0(x) \) between fcc-hcp and diamond-wurtzite are almost the same (of course, these \( \rho_0(x) \) are different in the case of finite \( N \)).

![Graphs](image)

**Figure 1.** The density of state without interaction for each structure. Particle number \( N \to \infty \) is shown. In this limit, the DOSs of fcc-hcp and diamond-wurtzite are corresponding each other.

The relationship between the band width \( W \) and the hopping energy \( t \) also varies with the structures and it is given by \( W = 12t \) for sc, \( W = 16t \) for bcc, fcc and hcp, \( W = 8t \) for diamond and wurtzite, and \( W = 2(\sqrt{10} + 1)t \) for graphite structure. The order of the proportional coefficient is like this: (a) diamond=wurtzite<graphite<sc<bcc=fcc=hcp.

4. **Calculation result**

We calculate and obtain the electronic states and the energy gap. Figure 2(a) represents the relationship between \( \alpha = a/a_B \) and the energy gap for every structure we treat. Recall the
sequence (a), we can say that large band width contributes to the metallization at larger distance of the neighboring atoms. Because the density $n$ is proportional to $\alpha^{-3}$, it can be expect that the transition density becomes larger as opposite to the sequence (a). However, this is not obvious (and as we show soon, this is incorrect) because the proportional coefficient between $n$ and $\alpha^{-3}$ is also different from each structure. The coefficient (we denote it as $c$) is related to the structural filling factor and is given by $n = c \times \frac{m_p}{(a_B \alpha)^3}$ ($m_p$ is mass of a proton). It is calculated as $3\sqrt{3}/8$ for diamond and wurtzite, $4\sqrt{3}/9$ for graphite, 1 for sc, $3\sqrt{3}/4$ for bcc, and $\sqrt{2}$ for fcc and hcp. The order is like this: (b) diamond=wurtzite<graphite<sc<bcc<fcc=hcp. The sequence (b) is almost the same as the sequence (a). However, the sequence (b) conflicts with the sequence (a) because the density for same $\alpha$ becomes larger as the sequence (b). In fig. 2(b), we show the density dependence of the energy gap. The density for the energy gap disappears is same as the sequence (b). It indicates the contribution of the filling factor (sequence (b)) is more effective than that of the broadening of the band width (sequence (a)). The value of the MIT of each structure is $n=0.20$ for diamond and wurtzite, $n=0.24$ for graphite, $n=0.25$ for sc, $n=0.28$ for bcc, and $n=0.30$ for fcc and hcp (unit is g/cc).

Figure 2. (a) The relationship between the energy gap and $\alpha = a/a_B$. As is obvious from the curves, energy gap decreases as the density becomes higher (smaller $\alpha$). (b) The density dependence of the energy gap for each structure.

5. Conclusion
In this study, we have investigated the structure dependence of the density when the MIT takes place in solid atomic hydrogen. We also calculated the transition density for each structure. As a result, the transition density varies with the structure and the density becomes higher for the structure of larger filling factor. This is because the filling factor is the dominant factor to decide the transition density. The transition density we calculated in this study is similar to the transition density region obtained for bcc structure[9]. This means the assumptions and parameters we used here are reliable at least qualitatively. If we consider deuterium or tritium, the expected MIT density becomes twice or three times larger.

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