Effect of incident kinetic energy on a-Si:H structure: A molecular dynamics simulation study

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Abstract. This paper utilized molecular dynamics simulation to investigate the influences of the incident kinetic energy on the structure of hydrogenated amorphous silicon (a-Si:H) thin films. The SiH_3 radical impinged on the Si (001) surface with substrate temperature of 500 K in the incident kinetic energy range from 0.04 to 5.81 eV. The results showed that high incident kinetic energy could smooth the surface, promote the densification of internal structure, and increase the adsorption rate of SiH_3 radical and the crystalline volume fraction. The SiH_x and dangling bonds, which are the key factors that affecting the film quality, have been further analyzed, the results reflected that the increase of the incident kinetic energy will increase the content of SiH combination pattern and decrease the dangling bonds in the films, thus improving the quality of the thin films. There is a critical value, 0.64 eV, 1.45 eV or 4.04 eV, of this variation rate. Steep variation trends is corresponding to the incident kinetic energy less than this critical value, and variation trends is gentle for that above it.

1. Introduction

Hydrogenated amorphous silicon (a-Si:H) thin films have played a crucial role as intrinsic absorber layers with doped layers in heterojunction (HIT) solar cells [1, 2]. The quality of a-Si:H thin films will determinate the conversion efficiency of HIT solar cells. The structures of thin films, including dangling bond and the proportion of SiH_x are one of the key factors which influencing the quality of a-Si:H thin films. Full understanding microstructural information is helpful to building the process-structure-property relationship, even rising high-efficieney HIT solar cells. However, the deposition conditions are interlaced each other. It is difficult to investigate the relationship between atomic structures of a-Si:H and single condition experimentally. The molecular dynamics (MD) simulation is a useful tool for investigating atomic arrangement [3] and the structural evolution [4]. Therefore, we utilizing molecular dynamics (MD) simulation to explore the microstructural information behind different incident kinetic energy. It is expected to find the correlation between the process condition and microstructure and provide theoretical basis for the experiment.

2. Computational model and methods

2.1. Computational details

The interatomic interactions are described by an extended form of the Tersoff potential, which was first proposed by Tersoff [5] and developed by Murty and Atwater [6], includes Si-Si, Si–H and H–H interactions.

A single crystal silicon of 22 atomic layers is used as the substrate. The simulation cell consisting of three regions is represented in figure 1: (i) the five bottom layers are fixed to their perfect lattice positions, (ii) the middle nine layers above the fixed are utilized Langevin method to control the temperature of 500 K, (iii) the eight top layers can move freely. Periodic boundary conditions (PBCs) are applied in both the x- and y- directions. The initial size of the model is Lx × Ly × Lz = 81.8 Å × 81.8 Å × 28.9 Å.
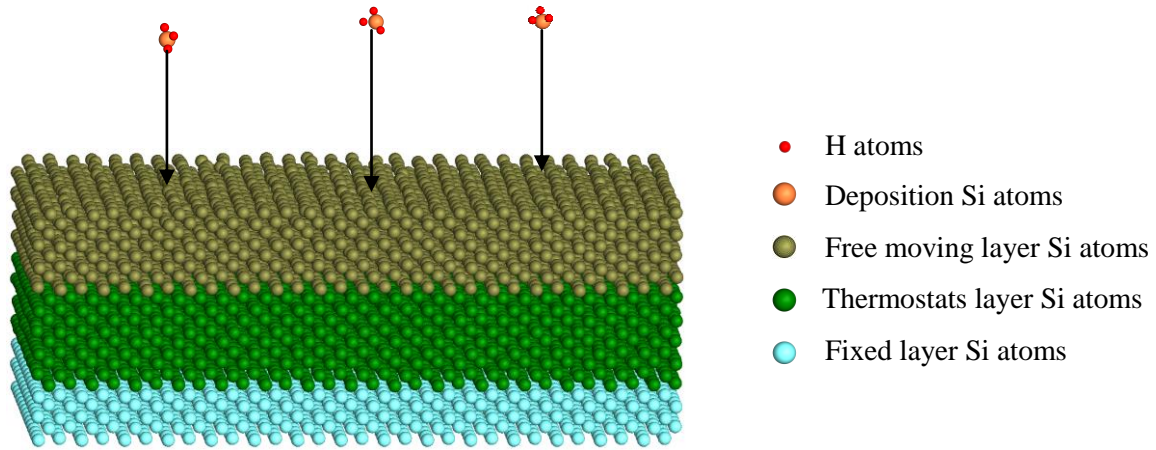

Figure 1. The schematic diagram of the deposition of a-Si:H/c-Si thin films system.

In the process of thin films deposition, the radicals deposit on the crystal Si (001) surface, and the detail parameters are as follows: Substrate temperature is 500 K, incident frequency is one radical per picosecond, the total number of the incident radicals are 8000. The incident radical is SiH_3, which is believed to be the main films precursors for high-quality films. The time step used in this simulation is 1.0 fs. The incident kinetic energies are selected as 0.04, 0.64, 1.45, 2.58, 4.04 and 5.81 eV, respectively. The final configuration obtained at different incident kinetic energy is quenched to 0.01 K or lower (eliminating thermal vibrations) before analyzing its characteristics.

2.2. Characterization methods

Surface-roughness [7] is a scalar that represents the surface-roughness of a-Si:H thin films. Surface-roughness (Rq) is defined as

$$R_q = \left(\frac{\sum_{i=1}^{N} (Z_i - \overline{Z})^2}{N} \right)^{1/2} \tag{1}$$

where: Z_i-the height of the *i*-th exposed atom on the film surface; $\overline{Z}$-the mean height of all the surface atoms; *N*-the number of the surface atoms.

Uttormark etc [8, 9] proposed a characterization index (A, equation (2)) to determine whether an atom is at crystalline state or not. The crystalline volume fraction is the ratio of the number of crystalline atoms to the total atomic number in the thin film.

$$A = \sum_{i=1}^{6} \left(\cos\theta_i + 1/3 \right)^2 \tag{2}$$

3. Results and discussion

Figure 2 (a) and (b) have shown the structures of the a-Si:H/c-Si thin films system with the incident kinetic energy of 0.04 and 5.81 eV, respectively. As can be seen, the crystal structure is at the bottom and the amorphous is at the top. For the incident kinetic energy of 0.04 eV, the atoms on the surface of the thin films emerge island structure, loosely connection and very rough appearance. For the incident kinetic energy of 5.81 eV, the surface of the film is smoother than the former.
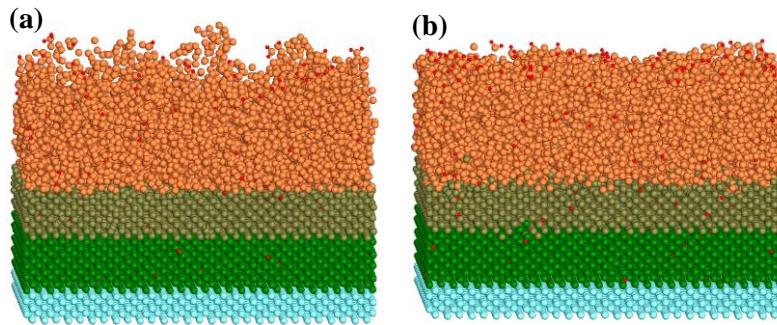

Figure 2. The structure of the a-Si:H/c-Si thin films depositing at the incident kinetic energy of 0.04 eV (a) and 5.81 eV (b), respectively.

In order to obtain the quantitative differences, the surface-roughness and relative atomic mass density of the films with different kinetic energy are analyzed, the results are shown in figure. 3. With the incident kinetic energy increasing, the surface-roughness decreased. In the range of lower incident kinetic energy (0.04-1.45 eV), the downward trend is steep, and then, as the incident kinetic energy continues to increase, the downward trend become a little of gentle. The surface structure is smoother obtained at higher incident kinetic energy. As we can see from figure 3b, the relative atomic mass density increases with increasing the incident kinetic energy and gradually approach to 1. Its variation trend is opposite to that of surface-roughness. The above results show that: Increasing incident kinetic energy, the surface of the films is more smooth and flat, and its internal structure is more compact. The better performance of a solar cell is predictable.

We further analyzed the adsorption rate of SiH_3 radical with different incident kinetic energy. As shown in figure 4a, the adsorption rate increases as the incident kinetic energy increasing. When the incident energy is below 0.64 eV, the radical cannot be effectively adsorbed onto the surface. It is easy to combine with the surface H atoms of the film and then return to the gas phase as the SiH_4. When the incident energy is more than 0.64 eV, the adsorption rate tend to stable at about 0.97.
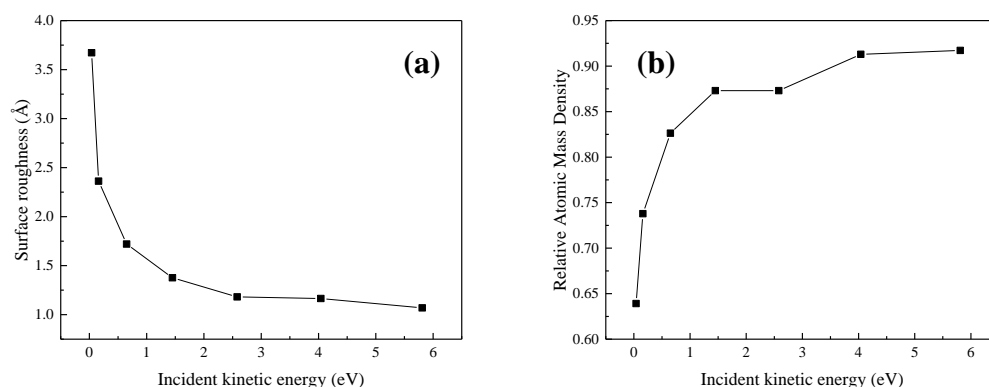

Figure 3. The surface-roughness (a) and relative atomic mass density (b) of the films depositing at different incident kinetic energy.
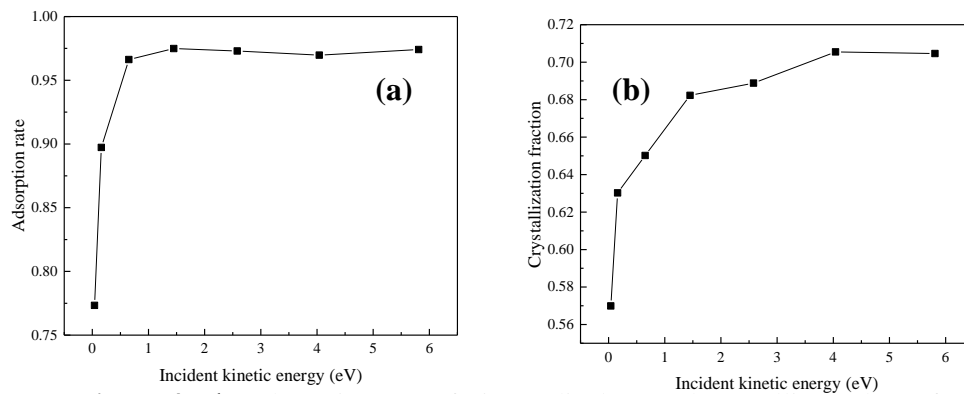
Figure 4. The adsorption rate of SiH$_3$ radical (a) and crystalline volume fraction (b) of the films depositing at different incident kinetic energy.

The crystalline volume fraction of the thin film layer at different incident kinetic energy is shown in figure 4b. The crystalline volume fraction rose gradually when the incident kinetic energy increased before 4.04 eV. After that value, the crystalline volume fraction tended to stable. It means that the incident kinetic energy can induce crystallization of atoms and improve the order degree of the atomic structure in the range of 0.04 to 4.04 eV. However, the local paracrystalline structure arrangement is not obvious varied after the energy over 4.04 eV.

The incident kinetic energy affects not only the structure of the silicon thin films, but also the combination pattern of hydrogen and silicon atoms. Besides the Si-Si bond and Silicon dangling bond, there are many SiHx radical, include SiH, SiH$_2$ and SiH$_3$. All of them will influence the conversion efficiency of a-Si:H/c-Si thin films heterojunction solar cells [10]. However, it has been reported [11] that only the SiH can reduce the density of defect states in the band gap and improve properties of the thin films.

The evolutions of dangling bond, SiH, SiH$_2$ and SiH$_3$ in the thin films with different incident kinetic energy are shown in figure 5. No matter what the incident kinetic energies are, the SiH combination pattern is the dominated, the contents of SiH$_2$ and SiH$_3$ are very less. Moreover, the content of SiH combination pattern increases with the increase of the incident kinetic energy, the contents of SiH$_2$ and SiH$_3$ decrease. This phenomenon can be understood from two aspect. One is that the initial SiH$_3$ radical changed easily to SiH due to more H atoms escaping with high kinetic energy. The other is that the higher incident kinetic energy will induce the more crystalline volume. The content of SiH$_2$ and SiH$_3$ will decrease because the more Si atoms have replaced the sites of H atoms during the process of crystalline. The content of dangling bond decreases with increasing the incident kinetic energy (figure 5a). The above two results show that the increase of the incident kinetic energy will increase the content of SiH combination pattern and decrease the dangling bonds in the films, thus improving the quality of the thin films.
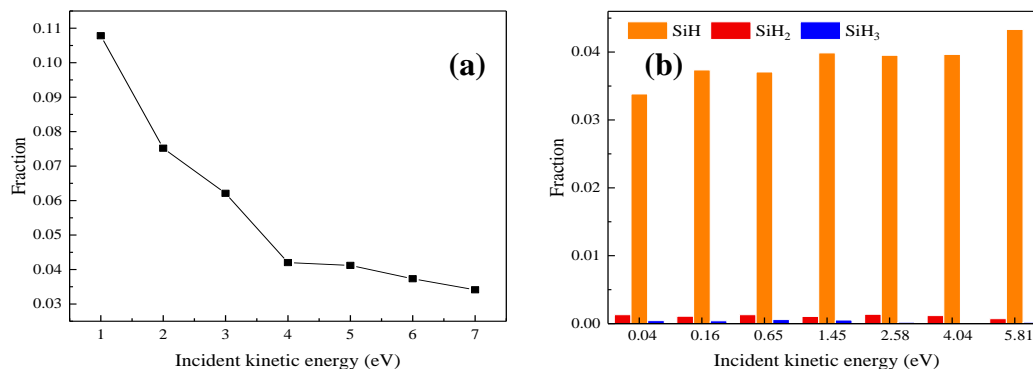

Figure 5. The evolutions of dangling bond (a), SiH, SiH$_2$ and SiH$_3$ (b) in the thin films with different incident kinetic energy.

4. Summary

In the present work, the influence of the incident kinetic energy on the structure of hydrogenated amorphous silicon thin film is investigated on atomic scale using molecular dynamics simulation. The results indicated that the surface-roughness decreases, internal structure is more compact and the probability of appearing the cavity structure decreases as the incident kinetic energy is from 0.04 to 5.81 eV. The content of SiH combination pattern increases and the dangling bonds in the films decreases with increasing the incident kinetic energy, and consequently improving the quality of the thin films. There is a critical value of these variation rate. Steep variation trends is corresponding to the incident kinetic energy less than this threshold value, and gentle variation trends for that above it. For the adsorption rate, this critical value is 0.64 eV; For surface-roughness and relative atomic mass density, the value is 1.45 eV; For crystalline volume fraction is 4.04 eV.

5. References

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