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Molecular dynamics investigation of deposition and annealing behaviors of Cu atoms onto Cu(0 0 1) substrate

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ABSTRACT

The deposition growth and annealing behaviors of Cu atoms onto Cu(0 0 1) are investigated in atomic scale by molecular dynamics (MD) simulation. The results indicate that the film grows approximately in a layer-island mode as the incident energy is from 1 to 5 eV, while surface intermixing can be significantly observed at 10 eV. The surface roughness of the film decreases with increasing the incident energy, and the film after annealing becomes smoother and more ordered. These phenomena may be attributed to the enhanced atomic mobility for higher incident energy and thermal annealing. It also indicates that atomic mixing is more significant with increasing both the incident energy and substrate temperature. In addition, the peak-to-peak distances of radial distribution function (RDF) clearly indicate that the films before and after annealing are still fcc structure except for that at the melting temperature of 1375.6 K. After annealing, the film at the melting temperature returns to fcc structure instead of amorphous. Moreover, the residual stress and Poisson ratio of the film are remarkably affected by the thermal annealing. Furthermore, the density of thin film is obviously affected by the substrate temperature and annealing process. Therefore, one can conclude that high incident energy, substrate temperature and thermal annealing could help to enhance the surface morphology and promote the microstructure of the film.

1. Introduction

As the electronic and magnetic devices [1–3] become smaller, it is very important to understand the deposition and diffusion on the surfaces or interfaces since the surface structure of these devices is strongly affected by the transport behaviors of adatoms at the early growth stage of thin films, especially during annealing process. The annealing treatment can recrystallize the films and then improve their physical properties. Therefore, the relationship between the thermal annealing and physical properties has been one of the interesting issues.

Many researches have been devoted to understand the atomic diffusion during deposition process of the film growth and investigating the effect of thermal annealing on surface morphology and microstructure of the films. In Cu–Co system [4,5], the degree of interdiffusion becomes more pronounced as the annealing temperature increases. Recently, Nakamura et al. [6] found that the elastic constant of Cu-film on Si(0 0 1) significantly increases after the low temperature annealing. The stress-induced void generation in ultrathin Cu wires during annealing process was reported by Sasajima et al. [7]. The results showed that the effects of wire width, overlayer thickness, and cooling rate on void formation are obvious. In addition, the asymmetry of surface intermixing has been observed in many metallic thin film systems, such as Co–Al [8,9], Au–Ni [10], Cu–Ta [11] and Cu–Pd [12]. In homoepitaxial growth, the influences of the incident energy and incident angle of deposited particle on the film morphology were investigated by Monte Carlo (MC) and kinetic Monte Carlo (KMC) methods in our previous studies [13,14]. Recently, we reported that transport behaviors of the deposited atom are closely related to both the local impact site and the incident energy [15]. However, it is still very difficult to analyze the microstructure evolution of the film during thermal annealing because of the limits of experimental analysis. On the other hand, molecular dynamics method is an ideal technique for investigating the microstructure evolution and growth mechanism of the films.

Until now, few studies were reported for the effect of thermal annealing on surface morphology and film microstructure in homoepitaxial growth. In this work, we take Cu atoms onto Cu(0 0 1) substrate as a prototype in order to study how the homoepitaxial film is affected by annealing process. The surface roughness, layer coverage fraction and radial distribution function of the film before and after annealing are investigated by molecular dynamics method. Finally, the explanation for these phenomena will be discussed.

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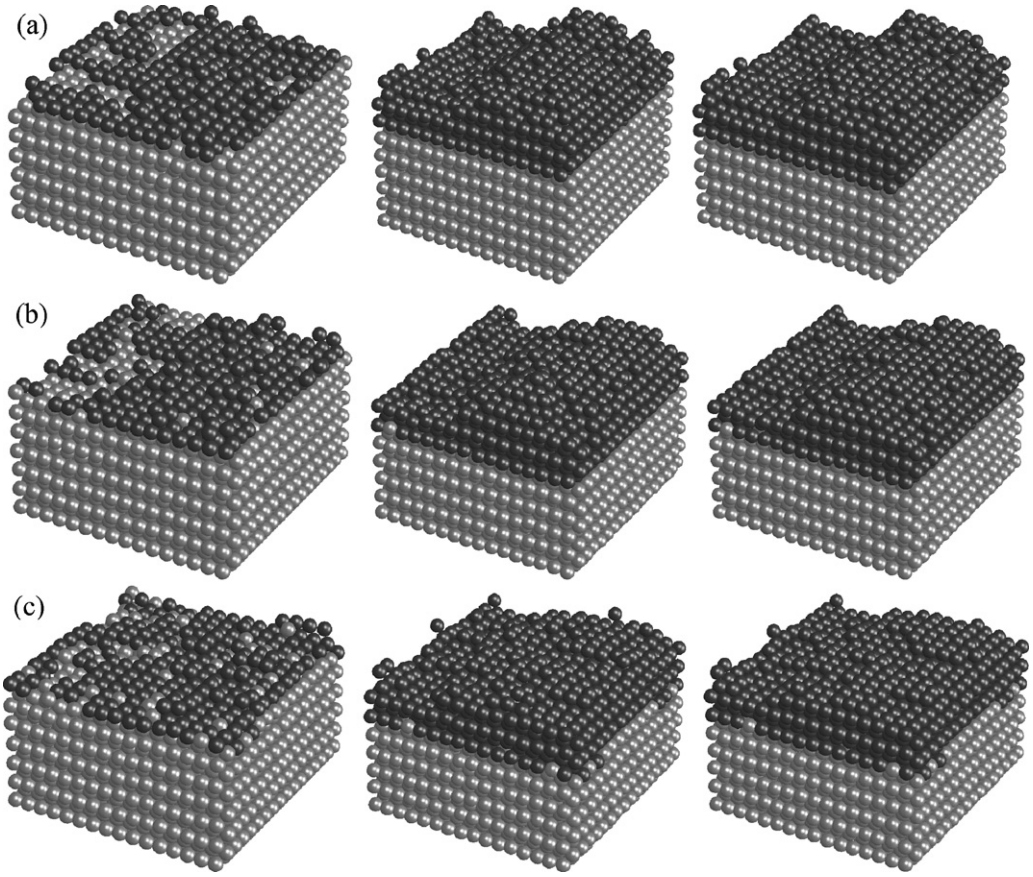

Fig. 1. The surface morphology of the film before and after annealing. The substrate temperature is 300 K and annealing temperature is 600 K. The incident energies are: (a) 1 eV, (b) 5 eV, and (c) 10 eV. From left to right in each row: after 1 ML, 5 ML (unannealed) and 5 ML (after annealing). Here, the bottom two fixed atomic layers of the substrate are not shown.

2. Computational details

In our molecular dynamics (MD) simulation, the size of the Cu(0 0 1) substrate is prepared to 40.9 Å × 40.9 Å × 23.5 Å with the surface perpendicular to the z axis. The periodic boundary conditions are applied in both the x and y directions. The bottom two atomic layers in the simulation cell are frozen to their perfect lattice to mimic the bulk. The three atomic layers above the fixed region are constrained at a specified temperature. The velocities of the atoms in the thermostat layers are rescaled at each time step (1 fs) in the simulation. All the other atoms are unconstrained with the initial temperature. In this simulation, a standard velocity–Verlet algorithm [16] is applied to integrate numerically the motion equation of the atoms. Before starting the depositions, the sample was fully relaxed about 5 ps to eliminate the internal stress in the substrate. The incident energy of deposited Cu atoms is from 1 to 10 eV, and the substrate temperature is set from 300 to 500 K. The incident atom is deposited at a distance of 16.23 Å from the substrate surface, which is farther than the cutoff distance (5.5 Å) of the adopted potential. The initial positions of the deposited atoms are randomly selected in the x–y plane, and the incident angles are normal to the surface. The time interval between two successive depositions is 0.5 ps. Totally, 1280 atoms are deposited on the substrate, in which each monolayer corresponded to 256 atoms.

After deposition, the thermal annealing process is then implemented on the sample. In this simulation, a complete annealing process consists of three stages: heating to the specified annealing temperature of 600 K, annealing for a period of time (about 20 ps) at annealing temperature and cooling to the room temperature of 300 K. In addition, the heating and cooling rates are 50 K/ps and 20 K/ps, respectively. When the substrate temperature is higher than 600 K, the annealing process only contains the cooling process.

The embedded-atom method (EAM) potential [17–19] can accurately describe the atomic many-body interactions in metallic systems. In this work, the EAM potential proposed by Mishin et al. [19] is adopted to describe the Cu–Cu interaction. Generally, the total energy of one system can be expressed as

$$E_t = \frac{1}{2}\sum_{i,j}\phi(r_{ij}) + \sum_i F(\rho_i), \tag{1}$$

$$\rho_i = \sum_{j\neq i} f(r_{ij}), \tag{2}$$

where $\phi(r_{ij})$ is the pair potential as a function of distance r_{ij} between atoms i and j, $F(\rho_i)$ is the embedding energy as a function of the host electron density ρ_i at the site of atom i by all other atoms, and $f(r_{ij})$ is the electron density at the site of atom i arising from atom j.The pair potential and embedding functions could be expressed as follows:

$$\Phi(r) = [E_1 M(r, r_0{}^{(1)}, \alpha_1) + E_2 M(r, r_0{}^{(2)}, \alpha_2) + \delta] \times \psi\left(\frac{r - r_c}{h}\right) - \sum_{n=1}^{3} H(r_s{}^{(n)} - r) S_n (r_s{}^{(n)} - r)^4, \tag{3}$$