Molecular dynamics simulation of cluster beam Al deposition on Si (1 0 0) substrate

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Abstract

A molecular dynamics simulation to study thin film properties and formation of Al cluster beam on a Si (1 0 0) substrate is presented. In this simulation, a new modified Lennard-Jones potential is used to describe the interaction between aluminum atoms. The migration of Al-atom clusters and the structure of grown film have been studied as functions of substrate temperature and cluster energy. The migration distance of the Al cluster reaches maximum when the substrate temperature is 473 K. The crystalline of Al on the Si (1 0 0) substrate was obtained under room temperature depositing and subsequent 673 K annealing. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Thin film deposition assisted by ions beam has gained increasing interest because it enables tailoring of specific material properties in a wide range and well-defined manner. Ionized cluster beams (ICB) techniques for thin film deposition were first developed at Kyoto University in 1972. Subsequently several applications have been used in industry [1]. Recently, various thin-film materials have been prepared by this method. These include semiconductors, metals, optical coatings, optoelectronic devices, magnetic materials, dielectrics for electronic devices, thermoelectric and organic materials. Since high quality thin film can be obtained at relative low temperature and deposition parameters such as the kinetic energy of the ion cluster beam and the percentage of the ionized cluster can be controlled easily, it has become a promising film growth technique.

The use of single-crystal or epitaxial metal films has been proposed in order to reduce the integration size threshold of VLSI circuits, and a lot of experimental and computational work of metal films on Si substrate has been done [2–6]. The epitaxial growth of Al on Si (1 1 1) and Si (1 0 0) by ICB was investigated by Yamada et al. [7], molecular dynamics simulation of cluster beam deposition of thin film was studied by Müller [8], and...
another molecular dynamics study on epitaxial growth of SiGe on Si (1 0 0) was also performed by Ethier and Lewis [9]. In this paper, Al ionized cluster beam depositing on Si (1 0 0) substrate is investigated by molecular dynamics simulation. The cluster migration distance under different conditions and Al thin film structure on Si (1 0 0) substrate have been paid more attention.

2. The simulation model

2.1. Interactions potentials

Many empirical potentials have been developed to describe the structural properties of silicon. Among them, the one proposed by Stillinger and Weber [10] has certainly been the most extensively used, so we chose this three body potential for the interaction between silicon atoms.

In order to simplify the simulation procedure and reduce CPU time, we have employed two body Lennard-Jones potentials for Al–Si and Al–Al. The parameters for the mixed interactions of Al–Si were determined by the standard combination rules, i.e., arithmetic mean for lattice parameter, and geometric mean for energy well depth.

In order to simulate the Al film structure more accurately, having referred to the work of Ercolessi and Adams [11] we proposed a modified empirical Lennard-Jones potential for Al–Al, which can be expressed as follows:

\[
U(R) = 4 \times 0.368 \left[ \left( \frac{2.5479711}{R} \right)^{12} - \left( \frac{2.5479711}{R} \right)^{6} \right], \quad R < 3.4,
\]

\[
U(R) = 4 \times 0.368 \left[ \left( \frac{3.608}{R} \right)^{12} - \left( \frac{3.608}{R} \right)^{6} \right], \quad 3.8 < R < 5.6,
\]

\[
U(R) = aR^3 + bR^2 + cR + d, \quad 3.4 \leq R \leq 3.8,
\]

\[
a = -0.7897537, \quad b = 7.1675137, \quad c = -21.053315, \quad d = 19.550741.
\]

The final potential, shown in Fig. 1, was built by two traditional Lennard-Jones potentials that have potential minimax at 0.286 nm and 0.405 nm, respectively, and with a polynomial function as transition. All the parameters in Al–Al potential were calculated from thermodynamic data and the connection polynomial function was fitted by cubic spline.

2.2. The simulation system

The molecular dynamics simulation in this work is fully dynamical three-dimensional calculations. Our simulation system includes a Si (1 0 0) substrate with two immobile layers and four movable layers. The former models the underlying crystal at the bottom; the latter consists of two upper unscaled layers and two lower rescaled layers. In order to prevent the substrate temperature from rising too high during the metal atoms depositing, the velocities of the two lower layers are rescaled for every 10 time steps. A cube cell with dimensions 3.07 nm and consisting of 8 \times 8 Si atoms per layer is used. The system is open along the positive z-axis and periodic boundary conditions are applied in the x–y plane.

In our simulation, the Verlet algorithm was used to integrate Newton’s equations of motion. The time step for a collection of substrate and deposited Al atoms is 5 fs which is smaller than the silicon-optical-phonon period of 0.0628 ps and is
also proper for the maximum cluster velocity. 
(Al)_{20} clusters have a simple cube structure and 
their velocity has a range of 0.2–3 eV per atom and 
the substrate temperature varies from 300 to 673 
K. In all cases, clusters were deposited every other 
4 ps.

3. Results and discussion

3.1. Migration distance

Adatom migration on the substrate is one of the 
most important mechanisms for the control of film 
growth. The migration effect helps the adatoms to 
interact with each other and to form stable nuclei. 
The energy of the cluster and the temperature of 
the substrate are the two main factors affecting the 
migration distance. Diffusion distance index, a 
mean distance from the initial position for every 
atom, is defined as

\[ d(t) = \sqrt{\frac{\sum_{i=1}^{N} (x_i(t) - x_i(0))^2 + (y_i(t) - y_i(0))^2}{N}} \]

where \( x_i(t) \) and \( y_i(t) \) are the coordinates of the \( i \)th 
atoms in the cluster at time \( t \), and \( x_i(0) \) and \( y_i(0) \) 
are the initial position of the \( i \)th atoms.

The diffusion distance index under different 
levels of cluster energy is shown in Fig. 2. The 
diffusion distance increases with increasing of 
cluster energy and the adatoms reach equilibrium 
after 1 ps. The simulation shows that in order to 
obtain high quality thin film, high cluster energy 
is necessary, and that the migration distance 
remarkably increases with increasing of cluster 
energy.

Substrate temperature is another important 
factor that can dramatically affect adatoms mi-
gration and the quality of thin film. In this paper, 
we investigated the diffusion distance index at 
different substrate temperatures. According to the 
work of Levenson et al. [12], we choose five kinds 
of substrate temperature: 300, 373, 473, 573 and 
673 K. Three clusters were used at each tempera-
ture and the mean migration distance was ob-
tained after Al adatoms reached equilibrium, 
which means the diffusion distance index was cal-
culated after 2 ps. The result is shown in Fig. 3.

Obviously, the migration distance increases 
with increasing of substrate temperature. How-
ever when the temperature reaches 473 K, the 
migration distance does not increase any longer. 
In fact, it decreases dramatically and reaches the 
minimum when the temperature is 673 K. In the 
study of anisotropic surface mobility of alumi-
num on Si (1 1 1) by Levenson et al. [12], it was 
found that the diffusion distance is near maxi-
mum at a surface temperature of 200°C for both 
0 and 3 kV acceleration. A similar phenomenon 
was found in our simulation. We think two fac-
tors have contributed to this result. First and 
foremost, the high substrate temperature makes
re-evaporation a high possibility. The migration distance increases with increasing substrate temperature until the higher kinetic energy of adatoms causes larger number of atoms to desorb. That means around 473 K there may be a balance point for absorbing and desorbing. Second, sole adatoms tend to re-evaporate more than those adatoms that interact with each other. So the farther the atoms migrate, the bigger the possibility of the re-evaporation.

The result mentioned above indicates that high cluster energy and proper substrate temperature are beneficial for the high quality thin film. But extremely high cluster energy can make the substrate defective and degrade the thin film properties.

3.2. Thin film structure

According to the experiment of Yamada et al. [13], we have performed a deposition simulation of 15 Al clusters on a small system – Si (100) 5 x 5 x 6 substrate at 300 K with 2 eV/atom. The time interval between two deposited clusters is 4 ps, and the whole system is annealed at 673 K after depositing. Before the deposition, we first let the substrate attain thermodynamic equilibrium by performing an undisturbed molecular dynamics relaxation.

The deposited thin film is shown in Fig. 4 and part of it after annealing is shown in Fig. 5. The graph clearly indicates that compact Al thin film can be obtained by ICBD at the room temperature, and that subsequent to 673 K annealing, part of Al thin film can form Al (110) crystal on Si (100) substrate. The crystalline orientation of Al on Si (100) is shown in Fig. 6.

![Fig. 4. The thin film structure of 15 Al clusters depositing on Si (100) by ICBD under the following conditions: substrate temperature is 300 K and cluster energy is 2 eV/atom.](image)

![Fig. 5. Part of thin film structure in Fig. 4 after annealing at 673 K.](image)

![Fig. 6. The micrograph of crystalline orientation of an Al film on Si (100) surface.](image)
4. Conclusions

We have used the molecular dynamics simulation to investigate the Al cluster properties on Si (100) substrate, especially the migration distance with different cluster energy and at different substrate temperatures. Our simulation shows high cluster energy is beneficial to form thin film and proves Levenson’s experimental result that migration distance can reach the maximum at 473 K. The decrease of surface mobility above 473 K is mainly caused by the increased evaporation rate of Al. Also Al thin film formed by ICBD at room temperature and subsequent annealing at 673 K proves the experiment of Yamada that Al (110) crystal can be obtained on Si (100) substrate. Meanwhile, the simulation result has testified that the modified Lennard-Jones potential we proposed is feasible in this molecular dynamics simulation.

References