Thin Film Growth Behaviors on Strained fcc(111) Surface by kinetic Monte Carlo

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Abstract. We study Ag islands grown on strained Ag(111) surfaces using kinetic Monte Carlo (KMC) simulations. We employed KMC parameters of activation energy and attempt frequency estimated by nudged elastic band (NEB) method and vibration analyses. We investigate influences of surface strain and substrate temperature on film growth. As the biaxial surface strain increases, the island density increases. As temperature increases, the shape of the island changes from dendric to hexagonal and the island density increases.

1. Introduction
Thin film technology has enabled the development of applied microelectronic devices which have various functional properties, such as magnetic, thermal, chemical ones. One of the most widely used techniques which fabricate thin films is Molecular Beam Epitaxy (MBE) [1]. MBE can precisely control the thickness of films, and manufacture nanoelectronic devices. It is important to understand the growth process at the atomic level and its relationship with film characterization.

At heterointerfaces, periodic dislocation network could appear due to the lattice misfit [2-5]. The STM results show that well-ordered islands are formed upon periodic dislocation network [3-5]. The dislocation network gives rise to periodic strain on the film surface. Surface strain often governs the growth behavior. Ab initio self-consistent calculations have shown that a Co adatom on the Pt (111) surface prefers to diffuse to the largest tensile strain region [6]. The Strain-assisted self-assembly has been expected to be one of the promising methods to achieve controlled nanostructures from the thermodynamic and kinetic points of view. Thus, it is worthwhile to study effects of the surface strain on the island nucleation and the growth behavior by atomic simulations.

In this study, we simulate the homo-epitaxial growth of Ag film on strained Ag(111) surface by kinetic Monte Carlo (KMC) method. We carry out KMC growth simulations on surfaces with uniform tensile strain and investigate the surface strain dependence of the island density.
2. Kinetic Monte Carlo Simulation Model

In the KMC method, the epitaxial growth is simulated by sequentially giving rise to each elementary growth process with the event probability [7-10]:

$$\Gamma = \Gamma_0 \exp\left(-\frac{E}{k_B T}\right),$$

where $\Gamma_0$ and $E$ are the attempt frequency and the activation energy for each process, $k_B$ is the Boltzmann constant, and $T$ is the substrate temperature. The effective attempt frequency is evaluated by the vibration analyses and the activation energy is obtained from the minimum energy path (MEP) which can be reasonably determined by the nudged elastic band (NEB) method [11]. According to a set of the event probabilities, atoms diffuse thermally on the surface to form islands. In this work, EAM potential developed by Williams et al [12, 13] is utilized.

The elementary growth processes we consider here can be classified into six types of KMC events as illustrated in Fig.1:(a)adatom diffusion on terrace, (b)attachment to step, (c)descent from step, (d)diffusion along step, (e)detachment from step, and (f)addimer diffusion on terrace. In the KMC event, the elementary growth processes are distinguished in terms of the adsorption site (fcc or hcp), the initial coordination number $n_i$, and the final coordination number $n_f$ (ch. Table 1). Moreover, we consider A and B steps inherent in islands grown on the fcc(111) surface: A steps are $\{111\}$ facets, and B steps are $\{100\}$ facets, as shown in Fig. 2. For example, the $d$ type of event includes 64 elementary growth processes due to the two kinds of step edge, the two kinds of adsorption site, $n_i$ and $n_f$ ranging from 4 to 7. Consequently, total 98 elementary growth processes are considered.

| type of event                  | adsorption site | initial coordination number | final coordination number | step edge |
|-------------------------------|----------------|-----------------------------|---------------------------|-----------|
| adatom diffusion on terrace   | fcc or hcp     | 3                           | 3                         | -         |
| attachment to step            | fcc or hcp     | 3                           | 4, 5, or 6                | -         |
| descent from step             | fcc or hcp     | 3                           | 4, 5, 6, 7, 8, or 9       | -         |
| diffusion along step          | fcc or hcp     | 4, 5, 6, or 7               | 4, 5, 6, or 7             | A or B    |
| detachment from step          | fcc or hcp     | 4, 5, or 6                  | 3                         | A or B    |
| addimer diffusion on terrace  | fcc or hcp     | 4                           | 4                         | -         |

Figure 1. Six types of KMC events.  
Figure 2. Two steps on fcc(111) surface.
3. Results and Discussion

The surface morphology of epitaxial films is controlled by the substrate temperature, the deposition rate, the biaxial surface strain, and so on. In this study, we investigate influences of the substrate temperature and the surface strain on thin film growth by KMC simulations.

Fig. 3 shows the simulation results of the Ag homo-epitaxial growth on a uniform tensile strained Ag(111) surface. The number of deposited atoms is 54000, which corresponds to 0.06[ML]. The substrate temperature, T, is 100 [K], and the deposition rate, R, is 0.01 [ML/s]. The biaxial surface strains \((\varepsilon = \varepsilon_{11} = \varepsilon_{22})\) are (a) 0, (b) 0.01, and (c) 0.02, respectively. The island shape doesn’t depend on the surface strain so much. Fig. 4 shows the island density as a function of the deposition amount in the simulations. For evaluating the island density, we count islands consisting of more than two atoms and neglect adatoms and addimers. As the biaxial surface strain increases, the island density gradually becomes higher as shown in Fig. 4. Thus, the lattice region with the highest tensile strain should be most favorable for nucleation. At the tensile strained region, the lattice spacing is large, and the adatoms are easy to be accommodated.

Next, we investigate influences of the substrate temperature on thin film growth. Fig. 5 shows the simulation results of the Ag homo-epitaxial growth at (a) T=150, (b) T=200, and (c) T=250, respectively. The number of deposited atoms is 54000, \(R=0.01\) [ML/s], and \(\varepsilon =0.02\). At T=150 [K], the islands have a dendritic shape with a triangular envelope as shown in Fig. 5(a). At T=200 [K] and T=250 [K], the islands have round and hexagonal shapes, respectively, as shown in Figs. 5(b) and 5(c). At high temperature, the probability of adatom diffusion along step edge is so enhanced that an adatom at the step edge easily attaches to the kink site, and the island has a few kink sites and the shape of the island becomes compact. At low temperature, the probability of adatom diffusion along step edge is so reduced that an adatom at the step edge is associated with another adatom at the step. Then, the new kink site is generated, and the shape of the island becomes dendric. As T increases, the kinetic anisotropy of adatom diffusion along two kinds of step edges is suppressed, so the shape of the islands is more isotropic. On the other hand, the island density is higher at lower temperature, as shown in Fig.6. At high temperature, an adatom can move to the existing island and absorbs at step edge within the deposition interval, and so the formation of new nuclei by isolated adatoms hardly occur.

![Figure 3](image)
Figure 4. Island density as a function of the deposition amount at $T = 100$ K and $R = 0.01$ ML/s.

Figure 5. Simulated surface images of self-organized Ag nanoislands on a uniform tensile strained Ag(111) surface at (a) $T=150$, (b) $T=200$, (c) $T=250$ K. All simulations are carried out by the deposition amount of 0.06ML (54000 atoms), at $R = 0.01$ML/s and $\varepsilon = 0.02$. The size of the images is $740 \times 1230$ lattice sites.

Figure 6. Island density as a function of the deposition amount, at $R = 0.01$ ML/s and $\varepsilon = 0.02$. 
4. Conclusions
We investigated Ag islands grown on uniform strained Ag(111) layer using Kinetic Monte Carlo simulations. We found that the island density depend on the biaxial surface strain and the substrate temperature. As the biaxial surface strain increases, the island density increases. On the other hand, the shape of the island hardly depends on the surface strain. We have also shown that while at low temperature the shape of the island is dendric and the island density is higher, the shape changes to hexagonal and the island density decreases as temperature increases. Thus, the growth behavior on thin film growth, such as the density and shape of island, is tunable by controlling the substrate temperature and the surface strain.

References

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