Growth mode variations of thin films on nano-faceted substrates

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We have investigated the growth mode of thin films on substrates with a faceted surface structure of a nanometer scale by means of computer simulations. The simulations have been performed for various combinations of the growth parameters such as the substrate temperature, the lattice mismatch between the film and the substrate, the interaction between the film and substrate atoms. In order to perform realistic growth simulations we have developed the molecular dynamics (MD) aided kinetic Monte Carlo (KMC) method. In the method MD deals with non-equilibrium structural relaxation of surface clusters and KMC deals with thermally activated adatom migrations. The film growth is classified into three types. The growth is promoted in the valley region or in the ridge region, or it has no site preference, depending on the growth parameters. The relation between the film growth on faceted substrates and that on flat substrates is discussed.

Keywords: faceted substrate, growth mode, molecular dynamics, kinetic Monte Carlo

1. Introduction

Fabrication of nanostructures is one of the promising research areas in the next decades. There are two trends in the methods of fabricating nanostructures like dots and wires. One is the artificial method which utilizes electron beam lithography and/or atom manipulation with a scanning tunneling microscope (STM).

The other is to utilize self-organization phenomena of materials themselves. The advantage of the latter is that the structures are formed in a wide area simultaneously though structural fluctuations in sizes and in positions inevitably exist as a disadvantage of the method. Faceted surfaces are useful candidates for templates of nanostructures.

They consist of unidirectional periodic grooves like a ridge-and-valley grating. Sugawara et al. have produced Fe dots and wires aligning along the ridges of the faceted NaCl(110) substrates [1]. The regular ridge-and-valley structure of the substrates whose period is as small as 10 nm is also formed spontaneously to reduce the total surface energy on annealing. The faceted surface structures with even smaller period have been reported for NaCl type transition-metal carbides, e.g., TaC [2,3], HfC [4], and NbC [5].

Although the faceted substrates have already been applied to producing some artificial structures in meso- and nano-scales, the thin film growth process on the faceted substrates has never been studied so far. It is important to know the growth process in order to insight the resulting film structures and to develop a method to fabricate well-defined fine structures.

We have performed computer simulations in order to classify the growth mode on the faceted substrates and to reveal its dependence on the substrate temperature, the interatomic interaction between the film and substrate atoms, and the lattice mismatch between the film and the substrate.

The simulation results are compared with the growth mode map for the growth on flat substrates [6]. The stochastic or kinetic Monte Carlo (KMC) method has been developed as a powerful tool for thin film growth simulations in the last few decades [7]. KMC can deal with a system with a large number of atoms and can follow realistic time evolution of the system.

But using the so-called solid-on-solid model where atom positions are restricted to the lattice points is not adequate for our purpose. It is easily predicted that the lattice of the film becomes incoherent and disordered near the valley and the ridge of the faceted substrate.

The crystallographic orientation could also be varied depending on the characters of the film and substrate materials. On the other hand molecular dynamics (MD) is a useful method to simulate the film structure. No MD simulation of film growth, however, can be realistic in time scale, since the time required to form a monolayer in real experiments is much longer than the longest time scale simulated by MD.

We have proposed a new simulation method, MD aided KMC where non-equilibrium structural relaxation of the surface clusters are dealt with by MD and thermally activated adatom migrations are dealt with by KMC [8].

2. Simulation method

Here we briefly summarize the simulation method. More details have been given elsewhere [8]. The basic simulation procedure follows that of the KMC method. MD is performed when a new atom is deposited or the number of nearest neighbors of a moved atom is increased by the KMC procedure.Activation energies of surface events have been calculated beforehand by making potential maps around the sites where the events take place.

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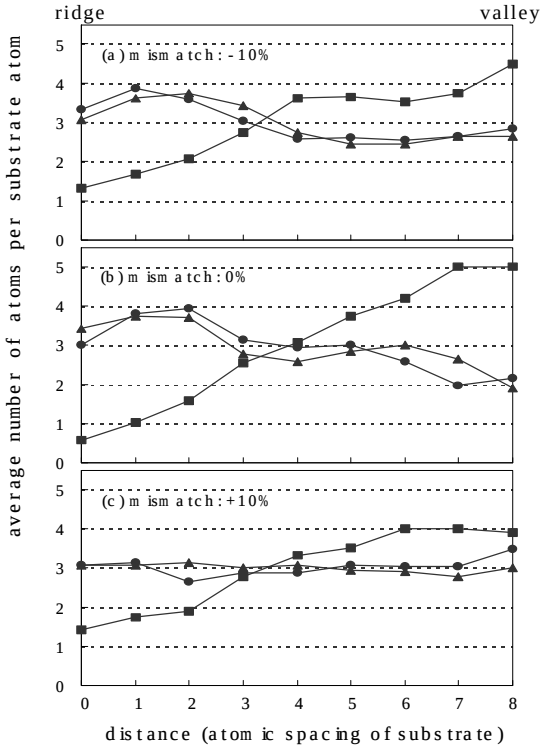

Fig. 1. Variations of the average number of atoms per substrate atom as a function of the distance along the slope from the ridge for the various combinations of the lattice mismatch: (a) -10, (b) 0, (c) +10% and D_{fs}: 0.125 (square), 0.250 (circle), 0.375 eV (triangle) after the deposition of 3 ML at 300 K. 0 and 8 in the lateral axis correspond to the positions of the ridge and the valley, respectively.

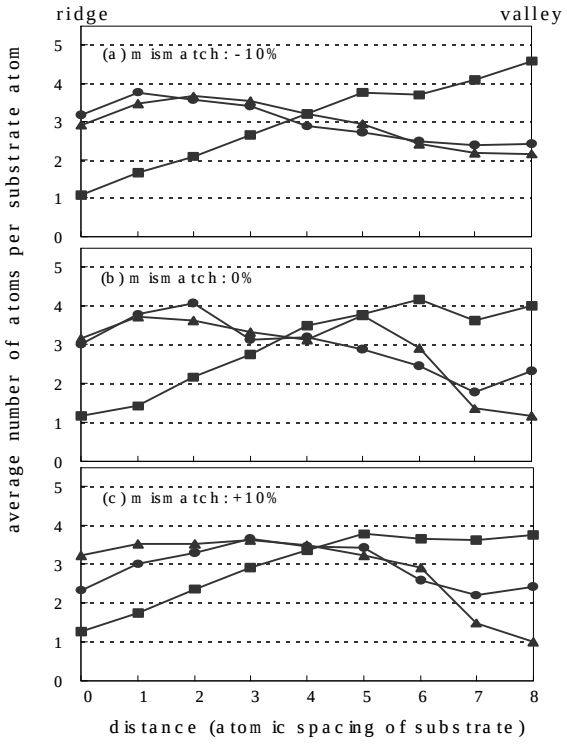

Fig. 2. Variations of the average number of atoms per substrate atom as a function of the distance along the slope from the ridge for the various combinations of the lattice mismatch: (a) -10, (b) 0, (c) +10% and D_{fs}: 0.125 (square), 0.250 (circle), 0.375 eV (triangle) after the deposition of 3 ML at 200 K. 0 and 8 in the lateral axis correspond to the positions of the ridge and the valley, respectively.

In the present study the atoms with more than 5 nearest neighbors are assumed to be immovable. The events which decreases the number of the nearest neighbors of the selected atom are forbidden, for example, dissociation of a dimer, detaching from a step to the terrace, detaching from a kink to the step, ascending a step, etc.Since the purpose of this study is to reveal the growth mode dependence on the growth parameters, we have performed the simulations for the film and the substrate of model elements using the Morse potential. We adopt the elliptic screening function proposed by Baskes [9] and adjust the screening parameters to include the influence of the second nearest neighbors partially, which reproduces the Ehrlich-Schwoebel barriers at step edges and at the ridge of the substrate.

We assume that substrates have the NaCl type structure and that the (110) plane parallel to the macroscopic surface. The atomic potentials between a film atom and the two kinds of substrate atom are assumed to be the same; the substrates have the simple cubic structure essentially. The substrates have 16 atoms in the [1-10] or periodic direction and 12 atoms in the [001] direction parallel to the ridge (or the valley).

The periodic boundary conditions are applied in both directions. The substrate atoms are kept fixed during the KMC and MD procedures.

The strength of interatomic interaction is tuned through the Morse parameter, D which is related to the binding energy. We fix D_{ff} = 0.250 eV for the film atom pair, and change D_{fs} for the film atom and the substrate atom pair as 0.375, 0.250, and 0.125 eV. We set the Morse parameter corresponding to the first nearest neighbor distance of the film so as that the lattice mismatch between the film and the substrate becomes -10, 0, and +10%. The substrate temperature is set at 200 K and at 300 K. The deposition rate is chosen as 1 ML/s.

3. Results and discussions

Variations of the average film height measured in the number of atoms per substrate atom as a function of the distance from the ridge along the slope after the deposition of 3 ML are depicted in figures 1 and 2 for the growth at 300 K and 200 K, respectively. The averaging is taken over the equivalent two rows on both slopes of the substrate except for the rows at the ridge and the valley.

The left end and the right end of the graphs correspond to the positions of the ridge and the valley, respectively. (a), (b), and (c) in the figures correspond to the lattice mismatch of -10%, 0%, and +10%, respectively. The dependence on the interatomic interaction between the film and substrate atoms is seen as a difference among the three plot lines (circle, square, triangle). We classify the simulation results into three types.

(I) The film tends to fill the valley for D_{fs} = 0.125 eV regardless of the substrate temperature and the lattice mismatch. (II) The film growth is advanced near the ridge for the cases of D_{fs} = 0.250, 0.375 eV and the lattice mismatch = 0, -10% at both temperatures. The rest is categorized as (III). When D_{fs} = 0.250, 0.375 eV and the lattice mismatch = +10%, the film is highest in the middle of the terrace at 200 K, and it looks to trace the shape of the substrate perfectly on average at 300 K (fig. 1(c), circle and triangle) though the height fluctuation in the film is not very small.

Ozawa, et al. have reported a map of the thin film growth mode on flat substrates as a function of the lattice mismatch and the ratio of the interatomic interactions between film atoms and between film and substrate atoms that is obtained by Monte Carlo simulations [6]. It is interesting to compare our simulation results with their growth mode map. The film grows faster near the valley than near the ridge when the Volmer-Weber (VW) mode is expected, while the film growth is promoted near the ridge when the Frank-Van der Merwe (FM) mode is expected on flat substrates. The growth type (III) and the Stranski-Krastanov (SK) growth occur under similar conditions.

This is understood as follows. The first nucleation always occurs at the valley when the surface diffusion on the substrate is active, which is seen by the simulations. When the VW mode is favored, adatoms aggregate to the island nucleated at the valley and it grows three dimensionally. Actually part of the substrate surface stays bare even after the deposition of 3 ML. When the FM mode is favored, the film of a monolayer height grows from the valley laterally. The film forms a coherent square lattice except for the atomic row at the bottom of the valley and those next to it.

The (100) oriented crystals near the valley on both slopes conflict so much after the growth of a few monolayers that the atomic density above the valley is reduced. On the other hand, island nucleation can occur near the ridge because adatom density is higher near the ridge than in the middle of the terrace due to the Ehrlich-Schwoebel barrier at the ridge. Once an atomic layer begins to grow from the ridge side, its shadowing effect prevents deposition atoms from reaching around the valley. Thus the film grows faster near the ridge than near the valley after the deposition of a few monolayers. In the cases of large positive lattice mismatch and small D_{ff}/D_{fs}, where the SK mode occurs on flat substrates, the triangle and distorted square lattices cover the substrate surface. Adatom mobility is larger on the triangle lattice than on the square lattice. The close packed atomic planes on both slopes conflict less than the (100) oriented ones above the valley. Those make the averaged atomic distribution along the slope. The large height fluctuation of the film originates in the nature of the SK growth.

The film morphologies at 200 K for D_{fs} = 0.250 and 0.375 eV are considered to be the consequence of the diffusion limited growth rather than the results of the growth mechanisms described above, though they are similar to those at 300 K.

4. Conclusions

The thin film growth on the nano-faceted substrate is simulated with changing the substrate temperature, the lattice mismatch between the film and the substrate, and the interaction between the film and substrate atoms. We have developed the MD aided KMC method in order to perform realistic growth simulations where non-equilibrium structural relaxation of surface clusters are dealt with by MD and thermally activated adatom migrations are dealt with by KMC. Three types of film growth are recognized under the conditions considered in this study: the valley preferred growth, the ridge preferred growth, the averaged growth.

The film fills the valley when the VW mode is favored, while the film grows faster near the ridge when the FM mode is expected on flat substrates. No site preference of the film growth is observed but the film is rough for large positive lattice mismatch and small D_{ff}/D_{fs} where the SK mode takes place on flat substrates. The crystal structure of the film particularly at domain boundaries under equilibrium growth conditions, i.e. at a higher temperature will be analyzed and discussed in the coming paper.

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