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Influence of microstructure of substrate surface on early stage of thin film growth

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Abstract

The early stage of thin film growing is studied by a Monte Carlo simulation (MCS). We focus on the effect of the microstructure of the substrate surface (lattice structure and surface defect) at different substrate temperature. The results illustrate that the surface microstructure affects strongly the film morphology with increasing substrate temperature. With increasing temperature, the growth of island film goes through fractal, dendritic and compact growth processes. At high temperature, the island geometry shape resembles the substrate lattice structure. There is a temperature region in which the island film transforms from fractal-to-compact growth, 310–400 K for triangular lattice substrate and about 250–390 K for square lattice substrate. A surface factor is introduced to represent the effect of a non-ideal surface, changing this factor, different island morphologies can be obtained at the same temperature.

1. Introduction

The study of thin films on solid surface has been carried out for many years. Many experimental results [1–8] have shown that the film growth processes can be strongly affected by fabrication conditions, such as substrate temperature, surface microstructure, etc. Fractal-like islands resembling the diffusion-limited aggregation (DLA) mode have been often observed in some experiments [1–3] at lower temperature. The experiments also showed that with increasing substrate temperature, the dendritic arm of the fractal island becomes wider and the island tends to be compact [1,3,7,8]. At very high substrate temperature, the island becomes more compact [2]. However, some authors [4,5] have investigated compact islands at lower temperature. Some experimental results have shown that the island shape is strongly affected by the lattice structure of the substrate surface. The fractal island has frequently been observed on substrate with triangular or hexagonal geometry [1–4], also with square lattice [6] at lower temperature. At high temperature, the island resembles triangular or hexagonal geometry [2,3] and square geometry [9,10]. These results illustrate that the geometric shape of islands is mainly determined by both the surface lattice structure and the substrate temperature. We also note that, in some experiments, different island morphologies can be obtained in the same temperature region. For example, the island shape of dendritic (or fractal) is reported by Hwang [1], Nishitani [3], and Taira [7] at the temperature range 300–500 K. However, the island shape is compact in Refs. [2,4] at the same temperature range. In these experiments, besides the effect of the experimental conditions, the surface microstructure (lattice structure and defects), and different interaction energies between the adatom and the surface are several important factors for islands growth.

Recently, the basic reason for the effect of the surface microstructure and the substrate temperature on the growth geometry of thin island films has so far not been explored. A variety of theoretical models [11–18] have been used to study adatom diffusion on the surface and the growth of thin films. Monte Carlo simulation (MCS) which is a simple and important method for studying random process has been used in many works [14–18]. Zhang [14] studied the fractal growth in metal-on-metal film growth process using MCS. The model took into account the effective of the substrate temperature and the surface structure with both square and triangular lattices. In Xiao's [15] model, atom attachment, detachment and surface diffusion were considered. The effect of the

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experimental conditions such as the deposition temperature and impingement rate on the film growth process were studied in detail. However, the interaction energy between the adatom and the clusters was very simple in these models. Bruschi [16] also considered the effect of the substrate temperature on the island growth process. The interaction energy between adatom and near-neighbor atoms was considered in more detail, but the effect of the next near-neighbor atoms was not taken into account. In these pioneering works, the substrate had an ideal surface and the effect of the substrate microstructure (lattice structure and defect) was not considered in detail.

In this paper, the early stage of thin film growth is studied by MCS. The model accounts for atom deposition, adatom diffusion, and adatom reevaporation. We focus on the effect of the microstructure of substrate surface (especially surface lattice structure and surface defects) at different substrate temperatures. The interaction energy between adatom and neighbor particles is calculated using a Morse potential. The interaction energy between the adatom and the neighbor atoms (including nearest-neighbors (NN) atoms and next-near-neighbors (NNN) atoms) is considered in detail. A surface factor is introduced to represent the effect of a non-ideal surface. By changing this factor, different island morphologies can be obtained at the same temperature. The results reproduce the main experimental findings.

2. Monte Carlo model

The Monte Carlo simulation was described in detail in our previous work [18]. In this section, we mainly introduce adatom diffusion on the substrate surface with square lattice, and simply describe the MCS model. It should be mentioned that the substrate surface is not an ideal surface in our model. There are some defect sites on it; these defect sites will become nucleation centers when the film deposition begins, because of the larger different interaction potential between the adatom and the defect site, so it is easy to form an island (or cluster) at a defect site.

In this paper, several kinetic processes (deposition of atoms from gas phase to the substrate, diffusion of adatom on the substrate, and reevaporation of the adatom from the substrate to the gas phase) are considered. Each process is controlled by a process parameter rate.

The deposition rate υ_d given by

$$\upsilon_d = FN. \tag{1}$$

where N is the total number of sites on the substrate and F is the deposition rate given in monolayers per second (ML/s). The coverage after deposition time t is then $\theta = Ft = n/N$, where n is the number of deposition atoms.

An atom depositing on the substrate becomes an adatom; it diffuses randomly along different directions (six directions for triangular surface and four directions for square surface). We assume that the adatom diffuses only to nearest-neighbor sites; diffusing to the next-nearest-neighbor sites are forbidden because a higher activation energy is needed. Here, we introduce the adatom diffusing on the square surface (Wei et al. [18] have described the adatom diffusing on the triangular surface). Microstructure of the substrate surface is shown in Fig. 1. For square surface, 1–4 are NN sites and 5–20 are NNN sites.

The diffusion of adatoms on the substrate consists of a series of adatom hopping from one site to another site. The hopping motion of the adatom depends on the substrate temperature and the interaction between the adatom and the neighbor atoms. There are two types of interaction energies between the adatom and the neighbor atoms: first is the interaction between the adatom and the neighbor adatoms; second is the interaction between the adatom and the substrate atoms. Before the adatom approaches an island (or there are no adatoms in NN sites and NNN sites around this diffusing atom), only the second interaction energy is considered. The adatom moves along four directions, the probability along each direction is 1/4. A random number R_1 (uniformly distributed in [0,1]) is used to determine the adatom moving direction.

If the adatom is absorbed by an island, it may stop moving and condense at the island, or migrate along the island boundary, or move away from the island. These processes are modeled by a nearest-neighbor hopping method with a hopping probability. Four random migration directions are considered (nearest sites 1–4 shown in Fig. 1b). If one of the sites has been occupied, then the probability P_j ($j = 1$–4) is equal to zero in this direction. P_j is proportional to the adatom hopping rate,

$$\upsilon_h^j = \upsilon_0 \exp(-E/k_B T). \tag{2}$$

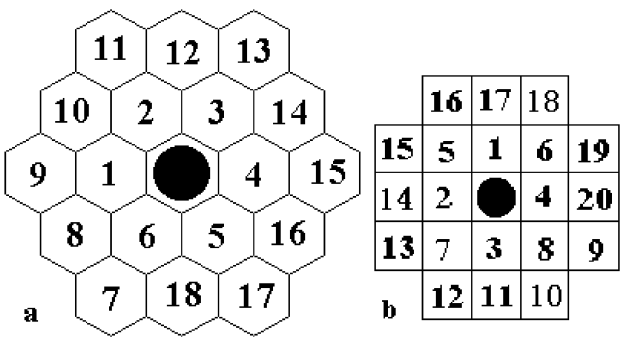

Fig. 1. Morphology of the substrate surface (a for triangular lattice surface, b for square lattice surface.)

The prefactor v_0 is the attempt rate and is taken to be the vibration frequency of a surface adatom, $v_0 = 2k_B T/h$, where T is the substrate temperature, k_B is Boltzmann's constant and h is Planck's constant. The E is an energy barrier to hop [18], given by $E = E_S + E_D + \alpha E_B$. E_D is interaction energy between adatom and nearby atoms (including NN atoms and NNN atoms). It is defined as a sum of contributions of the nearby atoms deposited earlier; the value of E_D depends on the migration direction. E_{ij} represents the interaction energy between an adatom at site (i, j) and all the neighbor adatoms around it. E_{ij} is given by $E_{ij} = \sum_{kl}^{m} V_{ij}^{kl}$, where V_{ij}^{kl} is the interaction energy between an adatom at site ij with the adatom at site kl; m is the number of occupied neighbor sites. V_{ij}^{kl} is simply given by the Morse potential [18]. If an adatom migrates from site ij to site $i'j'$, it must overcome a barrier E_D that is given by $E_D = E_{ij} - E_{i'j'}$.

The reevaporation process is determined by the reevaporation rate υ_r given by

$$\upsilon_r = v_{r0} \exp(-E_{ij}^r/k_B T), \tag{3}$$

where E_{ij}^r is the activation energy for reevaporation, and v_{r0} is a constant.

Three processes (deposition, diffusion and reevaporation) can be selected by using rates υ_d, υ_h and υ_r given by (1), (2) and (4). If the deposition process is selected, then a new atom is placed on the surface randomly. If the reevaporation event is selected, then a surface adatom is removed from the surface randomly. If the diffusion process is selected, then the diffusion direction of the adatom is determined by a hopping probability. It is given by $P_j = 0$ for occupied sites, $Pj = \upsilon_h^j/\upsilon_h$ for vacant sites, where $\upsilon_h = \sum_{j=l}^{m_l} \upsilon_h^j$; m_l is the number of vacant neighbor site.

The total deposition time is determined by the inverse of the sum rate

$$t = \sum(\upsilon_d + \upsilon_h + \upsilon_r)^{-1}. \tag{4}$$

Generally, formulas (1)–(4) can describe the film growth on ideal substrate surface. However, Some experimental results have shown different island morphologies in the same temperature region [1–4]. Besides the effect of the substrate temperature, we believe that this is because of the different surface conditions. For a non-ideal surface, many factors affect the film growth process, such as surface defects, steps, terraces, etc. Another important reason is the different interaction energy between adatom and the surface. In our work, the effect of these factors is considered by introducing a dimensionless parameter β — called surface factor characterizing the effective contribution to E_S from those factors mentioned above. Then the energy barrier E_S' becomes

$$E_S' = (1 + \beta) E_S. \tag{5}$$

As $\beta = 0$, $E_S' = E_S$ represents the interaction energy between adatom and ideal substrate surface. With increasing β, the surface becomes more rough, and E_S' increases.

3. Results and discussion

The calculation is performed on a lattice surface with 240×280 sites for a triangular lattice and 240×240 sites for a square lattice. About 60 active sites (nucleation sites) are randomly distributed on the surface. The substrate temperature varies from 150 to 450 K. The E_S, E_B, and V_0 are made in conformity with the work of Voter [19]. In this series of simulation, reevaporation is also not considered. The energy parameters are $E_S = 0.75$ eV, $E_B = 0.25$ eV, and $V_0 = 0.35$ eV, $r_0 = 2.47\, a_0$ (a_0 is the atom radius). $\upsilon_0 = 1.0 \times 10^{12}\ \mathrm{s}^{-1}$.

The morphology of the island film is shown in Fig. 2(a)–(c) for a triangular lattice surface and Fig. 2(d)–(f) for a square lattice with different substrate temperature (150, 350, 450 K). From Fig. 2, it is obvious that the island film is strongly affected by the substrate microstructure and the substrate temperature. Comparing Fig. 2(a)–(c) with (d)–(f), the morphology of island film growth on a triangular surface and that on a square surface are different at the same substrate temperature except at low temperature (150 K). At lower temperature ($T = 150$ K, Fig. 2(a) and (d)), the island morphology is very similar to the DLA mode. The fractal island growth is in conformity with some experimental results reported by Roder [6] and Michely [3]. It can be noted that the island morphology is slightly correlated to the surface lattice structure, at lower temperature the fractal arm of the island growing on the square surface is a bit wider than that on the triangular surface. As the substrate temperature is increased (300 K, Fig. 2(b) and (e)), the island growth on the triangular surface maintains fractal growth, only the arm of the fractal island becomes thick. However, the island growth on the square lattice surface begins to transform to compact growth at this temperature. By increasing the temperature continually (450 K, Fig. 2(c) and (f)), both of the islands growing on the triangular and the square surface become more compact, and the shapes of the island resemble the surface crystal structure. Comparing the island morphology growth at low temperature with that at high temperature, it is obvious that the higher the temperature, the stronger the effect of the substrate lattice structure on the island morphology. At higher temperature, the island growth on hexagonal lattice surface has hexagonal or triangular geometry shape; the islands growing on square lattice surface have regular square geometry shape. This result is in good agreement with that reported by Michely [2]. Our result is different from the calculated result reported by Bruschi [16], since here we consider the effective of the surface defects. The defect site is an adatom nucleation centre, the adatom condenses at this site and forms

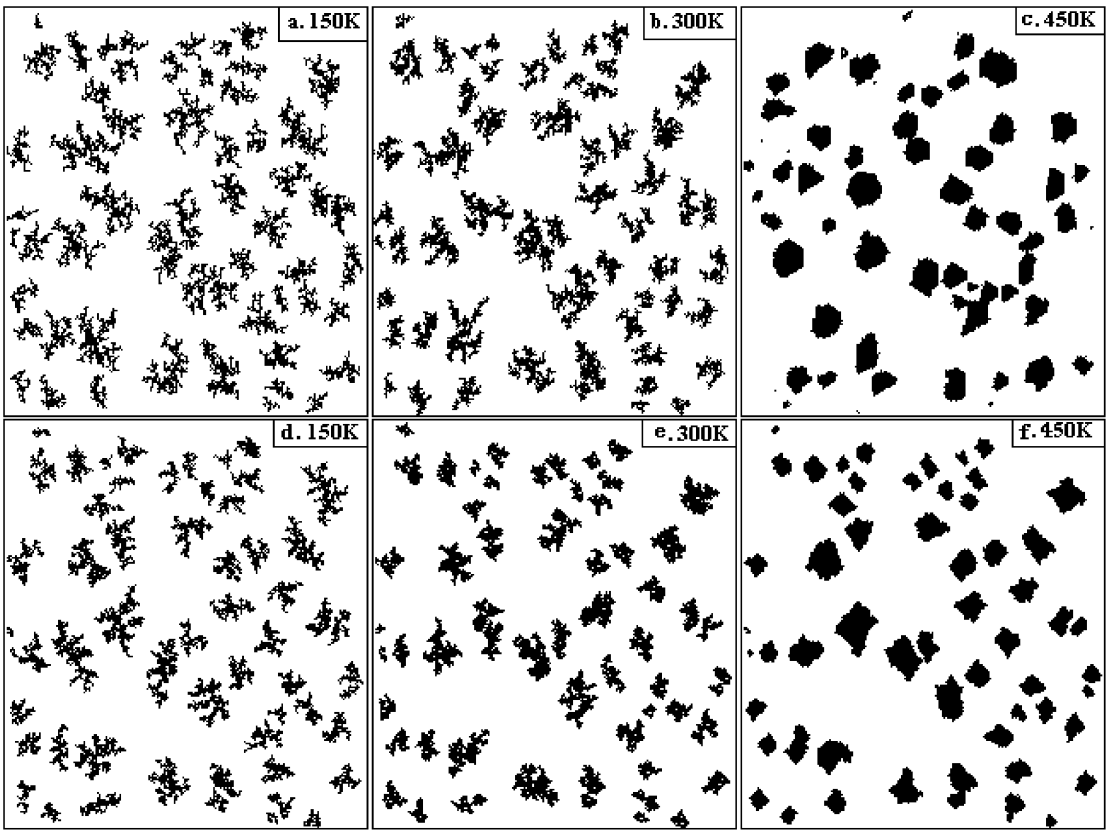

Fig. 2. The morphology of island film growing on triangular lattice surface (a,b,c) and square lattice surface (d,e,f) at different substrate temperature ($\theta = 0.15$ ML, $\beta = 0.0$).

clusters (or islands). Further, the cluster (or island) grows by absorbing other adatoms.

The island shape is ordinarily determined both by the substrate surface morphology (or lattice structure) and by the average diffusion length of adatoms on the substrate surface L_a and the average migration length along the perimeter of the cluster L_d. From Eq. (2), L_a and L_d depend on the substrate temperature and the surface morphology. At lower temperature, for triangular lattice the surface migration of adatoms is almost inhibited ($L_d = 0$), that is, atoms attaching to a cluster stick where they hit, the dendritic island growth occurs. The morphology of the island resembles the shape of DLA growth. However, the diffusion motion of the adatom on the square lattice is different, because there is no saddle point energy, and the interaction between the adatom and the NN atoms and NNN atoms is smaller than that on triangular lattice. From Eq. (2), the energy E is lower, so the adatom hopping rate υ_h is high, then L_a and L_d are also high. The adatom can move a long distance to find a more stable site; therefore, the arm of the fractal island growing on square surface is thicker than that on the triangular surface at the same temperature.

By increasing the substrate temperature, L_a and L_d increase, so the adatom remains on edge, migrates along an edge until it reaches a more stable site. At intermediate temperature, L_a and L_d are not very large; therefore, the fractal growth still remains on triangular surface. Because the interaction energy E is lower on square surface, it is obvious that the shape becomes compact growth. At elevated temperature, atoms adsorbed on the edge of the cluster have a larger migration length $L_d \gg 0$, the atom can move to the root of the ramify cluster, or separate from the cluster. This process introduces more channels for island rearrangement, which leads to a more compact island shape. In this case, the shape of the island depends on the surface lattice. This result is consistent with STM observation of thin film grown at substrate temperature greater than or equal to room temperature [1,2].

The surface factor β represents the substrate surface roughness. In this section, we investigate the island growth on a hexagonal lattice surface with different β at a same temperature. The results as shown in Fig. 3 and it is clear that the island is a more regular compact shape as $\beta \leqslant 0.3$ (Fig. 2(c) and 3(a)). The result shows that the lower the value of β, the more compact the island. The fractal growth occurs at $\beta \geqslant 0.7$. These results illustrate that the island growth transforms from compact growth to fractal growth by increasing the value of β at a same

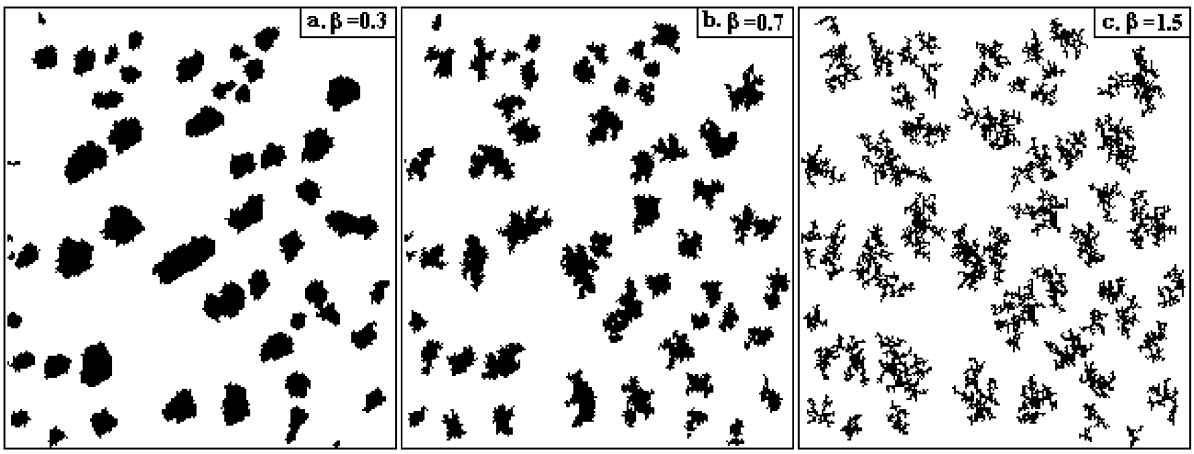

Fig. 3. Results of simulated depositions performed at different surface factors β with a temperature $T = 450$ K and coverage $\theta = 0.15$ ML.

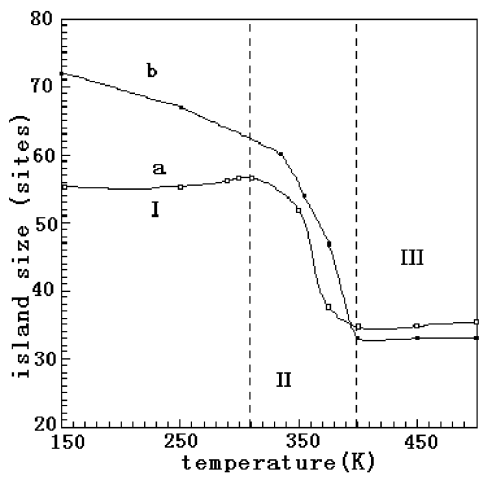

Fig. 4. The variation of the average size of the island with different temperatures.

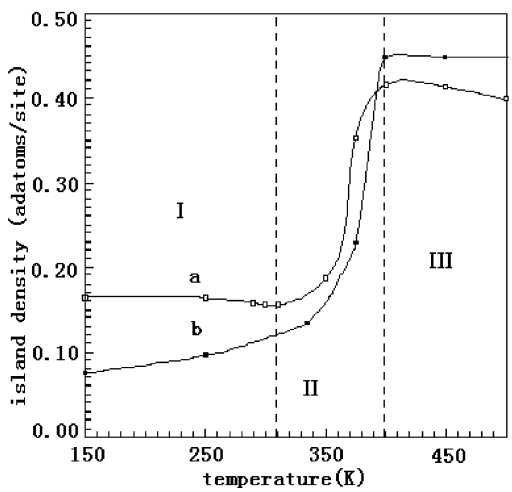

Fig. 5. The variation of the average density of the island with different temperatures.

temperature. Some experimental reports have shown that the island shapes are different in the same temperature range in different experiments. For example, the island shape is dendritic (or fractal) as reported by Hwang [1] and Nishitani in [3] at a temperature of about 300–500 K. However, the island shape is compact in [2,4] in the same temperature range. In these experiments, besides the effect of the experimental conditions, the non-ideal surface and different adatom–surface interaction energy are two important factors for islands growth. Using different surface factors, different islands can be formed from dendritic shape to compact shape at the same temperature. These results are in conformity with some experimental results.

Figs. 4 and 5 illustrate the average island size and the density of the island (which represents that the average numbers of the adatom per site in an island) with different substrate temperatures. At lower substrate temperature (<300 K) and higher temperature (>400 K), the island size (Fig. 4(a)) and the island density (Fig. 5(a)) for deposition on the triangular substrate do not vary. The reason for this is that the shape of the island is almost fractal at lower temperature and compact at high temperature. There is a little difference for the square substrate, at lower temperature the island size (Fig. 4(b)) falls slowly and the island density (Fig. 5(b)) rises slowly with increasing temperature. This is because the interaction energy between adatom and the neighbor adatoms is lower on the square surface than that on the triangular surface, the island growth is not completely fractal. We also note that the island size has a sharp fall as $T = 310$–400 K for triangular surface and 250–400 K for square surface. In this temperature region the density of the island has a sharp rise. It is clear that in this temperature region the growth of the island changes from fractal shape to compact shape.

4. Conclusions

The island film growth on substrate surfaces with hexagonal and square lattice is studied by Monte Carlo simulation. The effect of the substrate temperature and the substrate morphology is considered. The difference between our work and in earlier models is that the interaction energy between the adatom and the neighbor atoms is calculated by the Morse potential. In order to describe the effect of the non-ideal conditions, a surface factor is introduced to describe the surface roughness. The effect of the substrate lattice structure on the island growth process is firstly observed at different temperatures. The results illustrate that the higher the substrate temperature, the more compact the island. The fractal island growth occurs at lower temperatures, and the island shape resembles the DLA mode. At higher temperature, the island becomes more regular in shape and resembles the surface morphology. These results are consistent with STM observations of thin film growth [1,2]. The island morphology at low temperature is not correlated with the surface structure. However, with increasing temperature, it is obvious that the higher the temperature, the stronger the effect of the substrate lattice structures on the island morphology. Then, the effects of the surface factor at high temperature ($T = 450$ K) are studied. The result is obviously that the smaller the surface factor β, the more compact the island. With increasing β from 0.3 to 1.5, the island geometry shape changes from a compact shape to a fractal shape. That is to say that different islands can be formed from fractal shape to compact shape at the same temperature by changing the surface factor β. These results are in conformity with some experimental results.

Acknowledgements

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