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Monte Carlo simulation of thin-film growth on a surface with a triangular lattice

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Abstract

The thin-film growth process on a triangular lattice surface is studied by a Monte Carlo simulation (MCS). Four kinetic processes are considered: atom deposition, adatom diffusion, adatom nucleation and adatom reevaporation. We pay close attention to the substrate temperature and the interaction between the adatom and neighboring particles. The interaction energy between the adatom and the neighboring atoms around it is calculated by the Morse potential. The results show that the higher the temperature, the more compact the island. As the temperature is low, the film growth becomes fractal growth. At higher temperature, the island becomes more regular in shape that resembles the surface morphology.

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1. Introduction

Recently, the increasing number of experimental [1–8] and theoretical [9–15] works, which deal with thin-film growth and nucleation phenomena, reflect the considerable interest in the morphology of thin film and the related physical and chemical characteristics of these materials. These experimental results have shown that the film growth processes can be strongly affected by a number of fabrication conditions, such as substrate temperature, surface morphology, etc. Substrate temperature plays an important role in thin-film (or island) growth processes. Fractal-like islands resembling the diffusion-limited aggregation (DLA) islands have been often observed in systems such as Au on Ru (0 0 0 1) [1, 2], Pt on Pt (1 1 1) [3], Ag on Ni (1 0 0) [4], and Ag on graphite [5] when the temperature is lower than or equal to room temperature. With increasing temperature, the dendritic arm of the fractal island becomes wider and the island tends to compact growth [1, 5]. At very high temperature the island becomes more compact growth [3]. Some exceptions refs. [6, 7] have found compact islands at lower temperatures. Experimental results have shown that the island geometry shape is also affected by the substrate surface morphology. The fractal island has frequently been observed on a substrate with triangular or hexagonal geometry [1–3, 5, 6], also with a square lattice [8] at a lower temperature. At high temperatures, the island growth on the substrate with a triangular or hexagonal geometry resembles triangular or hexagonal geometry islands [3, 5]. These results indicate that the geometry shape of the island is mainly determined by both substrate temperature and surface morphology.

The basic reason for the dependence between the growth geometry of thin island films with the surface processes and the experimental conditions has so far not been explored. A variety of theoretical models [9–15] have been used to study the adatoms diffusion on the surface and the growth of thin films, such as the DLA [9] model and Monte Carlo simulation (MCS) [10]. However, these studies have neglected the difference in substrate morphology and is limited only to a few properties of the (island) cluster geometry. The results are independent of the experimental conditions (such as the substrate temperature and the surface morphology). Ratsch et al. [11] used MCS to examine the island growth; the coverage and the deposition rate are taken into account. Zhang [12] investigated the two-dimensional fractal shapes in metal-on-metal film growth using MCS. The model took into account the effects of the substrate temperature and the surface geometry with both a square

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and a triangular lattice. However, the interaction energy between the adatom and the clusters was very simple in these models. In Bruschi's model [14], the interaction energy between the adatom and near neighbor atoms was considered in more detail. However, the effect of the next near neighbor atoms was not taken into account. In these pioneering works, the substrate was an ideal substrate surface. Practically, there are many strains, broken, step areas and other defect areas on the substrate surface. Experimental result [2, 15] has shown that the fractal growth becomes more easily at those areas than at other areas.

In this paper, the effect of the surface morphology and the substrate temperature on the film growth process is taken into account. We consider the interaction energy between the adatom and the neighboring atoms (including nearest neighbors (NN) and next near neighbors (NNN)) by Morse potential, which depends on the distance between the adatom and the neighboring particles. The result is consistent with TEM observation of the grown thin-film.

2. Monte Carlo model

Four kinetic processes are included in the description of surface processes: (i) the deposition of atom; (ii) the diffusion of the adatom on the substrate; (iii) the nuclei of the adatom; and (iv) the reevaporation of the adatom.

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

The adatom diffusion before being absorbed by the clusters consists simply of the adatom moving along six directions for a triangular lattice. Only one interaction energy between the adatom and the substrate is considered in this case. The probabilities are equal (1/6), and determined by a random number R_1 (uniformly distribution in [0, 1]).

The adatom adsorbed by islands (or clusters) can migrate along the island boundary, and can also separate from the island. This process is modeled by a nearest-neighbor hopping process with the hopping probability. Six randomly migration directions are considered (nearest sites 1–6 shown in Fig. 1), the migration to next near sites (next near sites 7–18) is prohibited because this motion requires more energy. If one of the sites has been occupied, then the probability P_j is equal to zero in this direction. P_j is proportional to the adatom hopping rate

$$\upsilon_h^j = v_0 \exp(-E/k_B T) \tag{1}$$

The prefactor $v_0 = 2k_B T/h$ is the attempt rate, where T is the substrate temperature, k_B is Boltzmann's constant and h is Planck's constant. E is an energy barrier to hop, given by

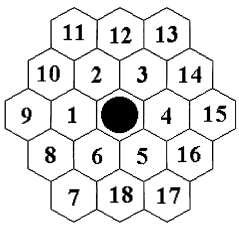

Fig. 1. Morphology of the substrate surface and local environment of an adatom for a triangular lattice.

$$E = E_S + E_D + \alpha E_B \tag{2}$$

where E_S is the interaction energy between the adatom and the substrate, E_B is the step edge (or defect) energy, as the adatom reaches a defect site, $\alpha = 1$; as there is no defect site near the adatom, $\alpha = 0$. E_D is an interaction energy between the adatom and near atoms (including NN atoms and NNN atoms). It is defined as a sum of contributions of the near atoms deposited before; the value of E_D depends on the migration direction. Voter [16] calculated the total energy of a rhodium atom for all the possible occupancies of the neighbors and determined the activation barriers for hopping. The occupancy of six adjacent sites (Fig. 1) is taken into account in his model. Each one of these sites can be either empty or occupied, creating $2^6 = 64$ barrier. Bruschi [14] considered these barriers using a recorder of the list; the recorder includes all information about the atom and atom migration. However, the interaction between the adatom and NNN atoms were not considered in his model.

In this paper, we consider the interaction energy between the adatom and the neighboring atoms (including NN and NNN atoms) by Morse potential. We use E_{ij} representating the interaction energy between an adatom at site (i, j) and the neighboring atoms around it. E_{ij} is given by $E_{ij} = \sum_{kl=1}^{m} V_{ij}^{kl}$, where V_{ij}^{kl} is the interaction energy between the adatom at site ij with the atom at site kl; m is the number of occupied neighboring sites. V_{ij}^{kl} is simply given by Mores potential

$$V_{ij}^{kl} = V_0\{\exp[-2a(r_{ij,kl} - r_0)/r_0] - 2\exp[-a(r_{ij,kl} - r_0)]\} \tag{3}$$

where $r_{ij,kl}$ is the distance between site ij and site kl, r_0 is the distance between two nearest-neighboring atoms, V_0 is interaction energy between two nearest-neighboring atoms, and a is a constant. If an adatom migration from site ij to site $i'j'$, it must overcome a barrier E_D that is given by

$$E_D = E_{i'j'} - E_{ij} + bE_l \tag{4}$$

where E_l is the saddle point energy when an adatom moves on the triangular lattice surface. For example, some atoms are put in sites 1, and 3 (Fig. 1), an adatom moves to 2, 4, or 5 must overcome saddle point energy for $b = 2$, 1, and 0, respectively.

The total hopping rate can be calculated by $\upsilon_h = \sum_{j=1}^{m_1} \upsilon_h^j$, where m_l is the number of the vacant sites near the adatom.

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

$$\upsilon_r = \nu_{r0} \exp(-E_{ij}^r / k_B T) \tag{5}$$

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

Three processes (deposition, diffusion and reevaporation) can be selected, the probability is

$$P_i = \upsilon_i / \sum \upsilon_i, \quad i = d, h, r \tag{6}$$

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 selection process is a diffusion event, then the diffusion direction of the adatom is determined by hopping probabilities of the adatom given by $P_j = 0$ (for occupied sites) and $P_j = \upsilon_h^j / \upsilon_h$ (for vacant sites). Then the total deposition time is determined by the inverse of the sum of the rates of all the processes,

$$t = \sum \delta t = \sum (\upsilon_h + \upsilon_d + \upsilon_r)^{-1} \tag{7}$$

3. Results and discussion

The calculation is performed on a lattice surface with 240×280 for a triangular lattice. About 60 active sites (defect sites or nucleation sites) are randomly distributed on the surface. The deposition rate is $F = 0.005$ ML/s, deposition time is 30 s, then the coverage $\theta = 0.15$ ML. The calculation parameters are listed in Table 1. The substrate temperature varies from 150 to 550 K. The choice E_S, E_b, and V_0 were made in conformity with the work of Voter [16]. In this series of simulation, reevaporation is not considered.

The morphology of the islands is shown in Fig. 2 for a triangular lattice with different substrate temperatures. From the Fig. 2, we can find that the islands are strongly affected by the substrate temperature T. As T is lower ($T = 150$ K), the island morphology is very similar to the DLA fashion. At this low temperature, the fractal island growth is in conformity with that reported by Roder in Ref. [6] and Michely in Ref. [3]. It can be noted that the island morphology is not correlated to the surface lattice structure at lower temperatures. As the substrate temperature is raised ($T = 300 \sim 350$ K), the arm of the fractal island becomes thick and the average size decreases. On increasing the temperature continually ($T = 450$ K), the island becomes more compact. 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. The island shape has a hexagonal or triangular geometry at higher temperatures. This result is in good agreement with that reported by Michely in Ref. [3]. Our result is different from the calculation result reported by Bruschi in Ref. [14], because we consider the effect of the surface defect. The defect site is the center of particle nucleation; the adatom condenses in this site to form a cluster (or island). Further, the condensing of the adatom results in the cluster (or island) growing, however, the island diffusion is prohibited due to the limitation of the defect site.

Table 1
The calculation parameters

E_S (eV)	E_B (eV)	E_l (eV)	V_0 (eV)	T_0 (K)	$a = r_0$	$\upsilon_0 = \upsilon_{e0}$ (s^{-1})
0.75	0.25	0.15	0.35	200	$2.47a_0$[a]	1.0×10^{12}

[a] a_0 is atom radius.

Generally, the shape of the islands is determined both by the substrate surface morphology and by the average migration length of adatoms at the perimeter of the cluster L_d. At lower temperatures, the surface migration of adatoms is inhibited $L_d = 0$, that is, atoms attaching to a cluster stick where they hit, the dendritic island growth occurs. The samples very much resemble the shape of DLA growth. As the temperature is raised, the L_d increases, the adatom can continue edge migration until it reaches a more stable site. It can be observed that the island arms become wider with increasing substrate temperature. At elevated temperatures, 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 ramified cluster, or separate from the cluster. This process introduces more channels for island rearrangements, which lead to a more compact island shape. When the substrate temperature is 450 K, the island becomes more regularly hexagonal or triangular in shape. Observing that the transition from a dendritic island to a compact island occurs at the absolute temperature $T = 350$–450 K. This result is consistent with TEM observation of thin-film grown at a substrate temperature greater than or equal to room temperature [1, 2].

The island growth for different coverages at a high temperature ($T = 550$ K) is also studied in this paper. The results are shown in Fig. 3. The island growth at 450 K with a coverage of 0.15 ML (Fig. 3a) exhibits uniform island size distribution. However, as the temperature is raised (550 K), the island size distribution is strongly affected by the coverage. As the coverage is

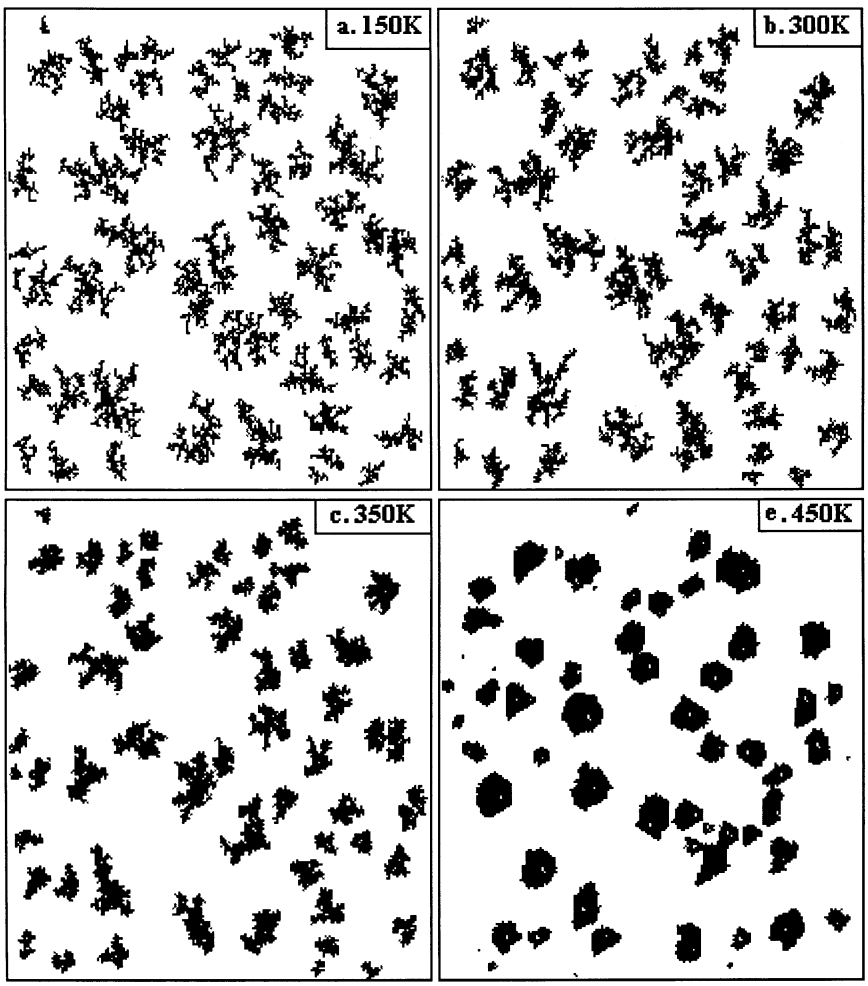

Fig. 2. Results of simulated depositions performed at different substrate temperatures.

smaller, the size distribution of the islands is uniform (Fig. 3b). With the increasing of the coverage, some islands grow fast and become large. Contrary, other islands grow slower (Fig. 3c). It can be noted that the larger island is the consequence of the incorporation of the near small clusters (show in the circle in Fig. 3b). As the coverage rises continually, the larger islands keep growing fast and incorporate with near larger islands (Fig. 3d). These results can be explained by the fact that the adatom diffusion distance becomes very long at high temperatures. The adatom can move to a more stable site near an island. In other words, the interaction energy between the adatom and the island is the largest at this site, so the adatom can condense more easily with a larger island.

To quantify these observations we have performed fractal analysis of the island morphology using the box-counting algorithm $N \propto L^{-D}$. The box dimension D for different substrate temperatures is shown in Fig. 4 ($T_0 = 200$ K). The dimension is found to be typicallyhigher than the DLA dimension $D \approx 1.72$. It is shown that the fractal dimension remains constant ($D = 1.890$) on increasing the temperature from 130 to 280 K. As the temperature increases from 280 to 400 K, the fractal dimension decreases fast. The decreasing of the dimension represents the island shape transition from fractal fashion to a compact shape. This result can be explained by the fact that as islands become more compact each occupied box tends to contain more atoms. Since the number of the surface adatoms is maintained (in this paper), the number of occupied boxes must decrease.

Fig. 5 illustrates the average island size with different temperatures. The result shows that the higher the substrate temperature, the smaller the island average size. It is clear that the result of the reduction of the island average size is due to the island becoming more compact. We can also note that the island size has a sharp fall as $T = 250$–400 K, this corresponds to the transition of the island shape from dendritic to compact. Comparing the Fig. 4 with Fig. 5, we can find that two results about the transition temperature are in good agreement with each other.

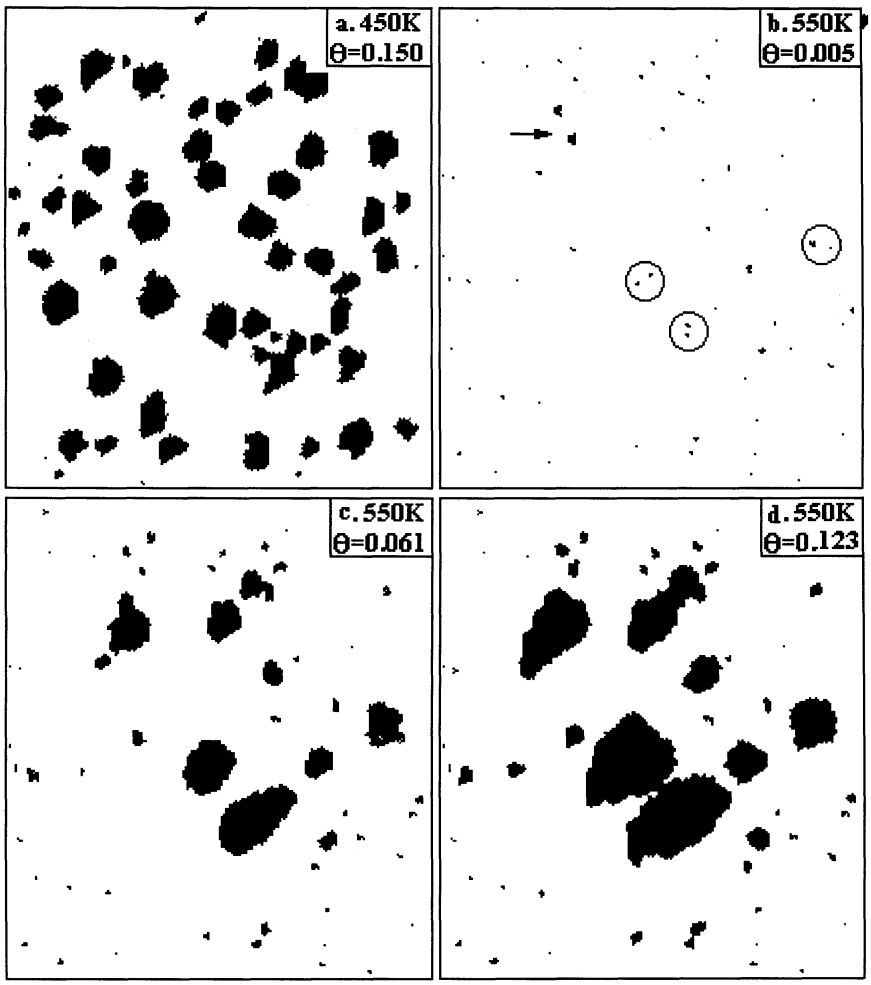

Fig. 3. Results of simulated depositions performed at different substrate coverages with a temperature $T = 550$ K.

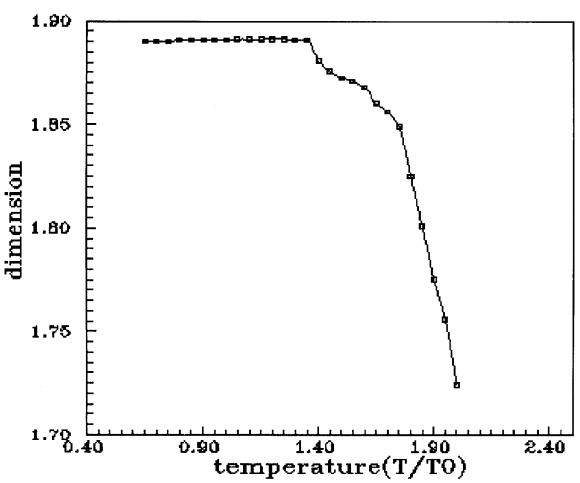

Fig. 4. The box-counting dimension with different temperatures.

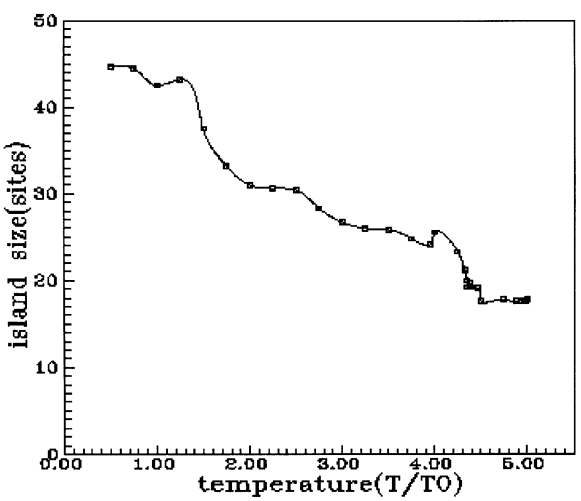

Fig. 5. The average island size with different temperatures.

4. Conclusions

The island film growth on a substrate surface with a triangular lattice is studied by a Monte Carlo simulation. The effect of several experimental conditions such as the substrate temperature, the substrate morphology, surface defection and the non-ideal surface condition are considered. The difference between our work with the early models is that the interaction energy between the adatom and the neighboring atoms around it is calculated by Morse potential. In order to describe the effect of the non-ideal conditions, a number of defect sites is introduced to describe the adatom nucleation process.

First, the effect of the substrate temperature on the island growth process is observed. The results illustrate that the higher the substrate temperature, the more compact the island. The fractal island growth occurs as the temperature is lower, and the island shape resemble the DLA fashion.

As the substrate temperature is raised, the branch of the fractal island becomes wider and the size of the island decreases. At higher temperatures, the island becomes more regular in shape that is, it resembles the surface morphology. These results are consistent with TEM observation of a grown thin-film. The island morphology growth at low temperatures is not correlated with the surface structure. However, with the increasing of the temperature, it is obvious that the higher the temperature, the stronger the effect of the substrate lattice structure on the island morphology.

Second, the effects of the surface coverage at high temperature ($T = 550$ K) are studied. It has been shown that the growth rates for different islands have great difference with the increasing of the coverage. The result is obvious that the larger the island, the faster the island growth. This is because the adatom diffusion distance increases at high temperatures. The adatom can move to a more stable site near an island where the interaction energy between the adatom and the island is the largest, so the adatom can condense more easily with a larger island.

Finally, the fractal dimension and the island average size are studied. It is shown that the fractal dimension remains constant ($D = 1.890$) in a temperature range (from 130 to 280 K). As the temperature increases from 280 to 400 K, the fractal dimension decreases fast. The decreasing of the dimension represents the island shape transition from fractal fashion to a compact shape. It should be noted that the island size falls fast as the temperature changes from 250 to 400 K, this corresponds to the transition of the island shape from dendritic to compact.

Acknowledgements

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