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SIMULATION OF FRACTAL GROWTH OF THIN FILMS AT LOW TEMPERATURE*

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Fractal growth of thin films at low temperature (50—175 K) is simulated by Monte Carlo method. It is shown that the thin film growth is quite different from the diffusion-limited aggregation (DLA) model when the coverage is larger than 0.1 ML. The average branch width of clusters increases with increasing temperature and it usually larger than the branch width (1.9 atom) in the classic DLA model. The average fractal dimension of clusters increases also with increasing coverage while the fractal dimension of DLA model remains constant. This difference comes from the weak screening effect during the late stage of thin film growth. The relationship between the saturation island number n_s and deposition interval Δt is described in a power law: $n_s \propto \Delta t^{\gamma}$, where $\gamma = -0.332$ is very close to the theoretical value $-1/3$ of rate equations from nucleation theory.

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I. INTRODUCTION

The physics of thin film growth has been a topic of much current interest. The application of improved and novel surface science techniques such as field ion microscopy (FIM)[1] and scanning tunneling microscopy (STM)[2], has indeed revealed many atomic details of the film growth. A variety of fractal structures have been observed in thin-film systems.[3—7] For example, STM images of Au on Ru(0001) grown at room temperature[4] and Pt or Ag growth on Pt(111) at low temperature[5—7] reveal fractal islands. The observation of fractal island growth has been understood by comparison with the classic model of diffusion-limited-aggregation (DLA).[8] In this model, clusters are formed as randomly diffusing particles sticking irreversibly to the perimeter of a growing aggregate and the fractal dimension keeps constant with increasing number of particles. However, in thin-film systems, the branch width and the fractal dimension of clusters increase with increasing temperature and coverage.[9]

In our previous work, some growth models have been proposed to simulate the processes of nucleation and growth of thin-films on square lattice substrates.[10,11] In this paper, a kinetic Monte Carlo simulation model is presented to simulate the thin-film growth on triangular substrates at low temperature, using realistic thin-film system-Ag on Pt(111) surface and physical parameters so as to investigate the atomic process and the morphologies of fractal growth of thin films. We pay special attention to revealing the difference of thin film growth from the classic hit-and stick DLA model. It is shown that the branch width and the fractal dimension of clusters increase with increasing temperature and coverage, which is consistent with the experimental observations, while they should remain constant in the standard DLA model.

II. SIMULATION MODEL

The simulation model is described as follows: Ag atoms are deposited randomly onto a 512×512 Pt(111) surface (triangular substrate, see Fig.1) with a deposition flux R (ML/s) and allowed to hop from site to site on the surface at a diffusion rate ν, which is given in Arrhenius form:

$$\nu = \nu_0 \exp(-E_d/k_B T)\,, \tag{1}$$

where ν_0 is the vibrational frequency of a surface atom ($\sim 10^{13}\,s^{-1}$), E_d is its energy barrier ($E_d = 0.157\,eV$[12]), T is the temperature and k_B is Boltzmann constant. The average deposition interval of successive atoms Δt it related to the deposition flux R and the surface diffusion rate ν. If we set the unit of Δt to be a diffusion step, we have $\Delta t = \nu/R$ (steps). It means that when the previous adatom diffuses Δt steps, the next atom is deposited. If $\Delta t < 1$, there are more than one atom deposited onto the substrate simultaneously. The diffusion steps of adatoms on the substrate are not limited due to the very long residence time of the atoms on the surface and the evaporation

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of atoms from the substrate is neglected at low temperature. In the temperature range from 50 to 175 K, the residence time of Ag on Pt(111) surface is in the range from 3.6×10^{13} to 2.7×10^{47} s. When the mobile atoms diffuse to their nearest-neighbor sites, they will stick with a unit probability and then a stable nucleus forms. At low temperatures, under typical growth conditions, the adatom surpersaturation on the surface is so high that the critical nucleus is just a single atom, and the stable nucleus is a dimer.[13,14] The diffusion of an adatom stops when it hits a critical nucleus or an existing island and condenses there. The effect of dissociation of islands is also negligible. This means that the adatoms stick irreversibly to the existing islands, and the dimers are stable. We focus on simulating the fractal growth at low temperatures in this paper, therefore the edge diffusion of adatoms is not considered. The periodic boundary condition is set up when adatoms diffuse to the boundaries. In this way, multiple islands can be generated on the triangular substrate, whose structure is expected to be fractal characteristic for various temperatures and coverage.

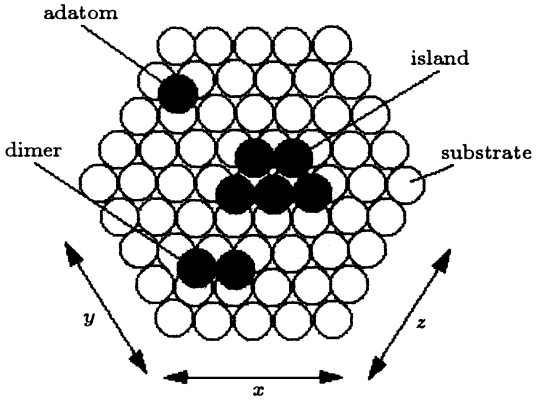


Fig.1. Model of fractal island grown on a triangular substrate.

Ⅲ. RESULTS AND DISCUSSION

Typical images of fractal aggregation simulated by this model are shown in Fig.2, which resemble the STM images closely.[6,7,9] The simulation conditions of deposition flux $R = 7 \times 10^{-5}$ ML/s, coverage $\theta = 0.1$ ML are chosen so as to compare with the experimental observation of fractal Ag aggregating on Pt(111) surface.[6,7,9] At 75 K, as seen in Fig.2(a), the Ag-adatom mobility is slow and only small clusters (2—40 atoms) grow. The branch width averaged for all clusters is $\bar{b} = 1.90$ and there is no obvious fractal property in these clusters. At 100 K (Fig.2(b)) the adatom mobility is already sufficient to grow larger clusters. The branch width $\bar{b} = 2.16$ and these clusters are shown to have self-similarity with a fractal dimension $\overline{D} = 1.67 \pm 0.06$, which is averaged for all clusters on the substrate and measured by Sandbox method (small clusters which contain less than 100 atoms are not counted for calculating the fractal dimension). This value is close to 1.6—1.7 in the classic DLA fractal growth.[8]

With increasing substrate temperature during deposition, the mobility of adatom increases, the island number decreases and larger clusters can grow. The branch width and the fractal dimension also increase with increasing substrate temperature. These results are given in Table 1. In our simulation model, we assume the substrates are smooth enough and there are no defects such as steps and kinks on the surface. In fact, these defects play an important role in the growth of thin-films at higher temperatures. A phenomenon called "step flow" will take place in this case.[6,15,16]

As seen in Table 1, the branch width in the simulation is larger than a branch width $b = 1$ atom in the classic DLA fractal growth.[14] In fact, the value of branch width $b = 1$ in DLA fractal growth is not defined. We repeat the simulation for the DLA model on a triangular substrate and get the average branch width $\bar{b} = 1.88 \pm 0.04$ atoms by using the following definition.

Table 1. The island number n, the average branch width $\bar{b}$ and the average fractal dimension $\overline{D}$ for different temperatures, averaged for four times.

T/K	50	75	100	125	150	175
Δt	5.4×10^{-6}	1	4.2×10^{2}	1.6×10^{4}	1.9×10^{5}	1.1×10^{6}
n	8671	764	120*	34*	16*	11*
$\bar{b}$		1.90	2.16	2.19	2.23	2.37
$\overline{D}$			1.67 ± 0.06	1.76 ± 0.06	1.85 ± 0.04	1.92 ± 0.03

*Saturation island number.

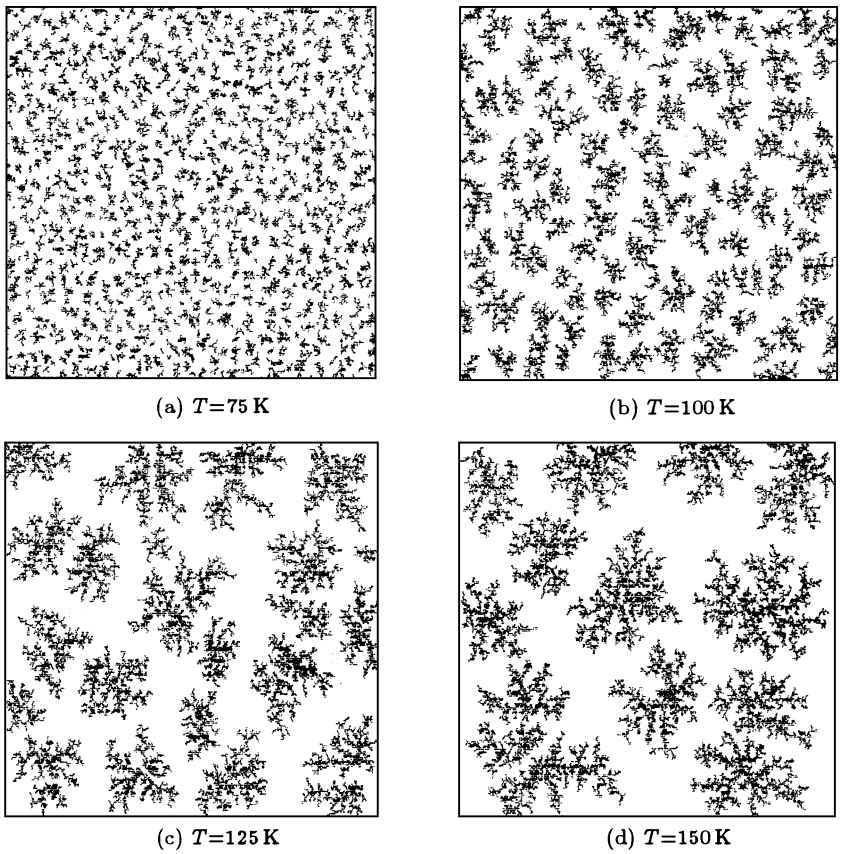


Fig.2. Images showing fractal Ag aggregates grown on Pt(111) at different temperatures.

The branch width of a single cluster is defined as the average number of continuous atoms from any atoms in the cluster along three directions of the triangular substrate, which can be described as:

$$b = \frac{1}{3}(b_x + b_y + b_z)\,, \tag{2}$$

where b_x, b_y and b_z are the average numbers of continuous atoms along x, y and z directions respectively (as seen in Fig.1). So the average branch width of all clusters on the substrate is:

$$\bar{b} = \frac{1}{n}\sum_{i=1}^{n} b_i\,, \tag{3}$$

where n is the island number on the substrate and b_i is the average branch width of the ith cluster.

In can be seen clearly from Table 1 that the average fractal dimension increases obviously with increasing temperature. The average branch width increases also with increasing temperature. These results are distinctly different from those obtained within the DLA model. Owing to the fact that in thin film growth, the diffusing atoms are not limited to be launched far from the growth centers, the screening effect is weaker than that in the classic DLA model. This is the main difference between our simulation and the DLA model. The dependence of the average branch width $\bar{b}$ and the average fractal dimension $\overline{D}$ on substrate temperature T is shown in Fig.3. The clusters growth at 50 K are too small to obtain $\bar{b}$.

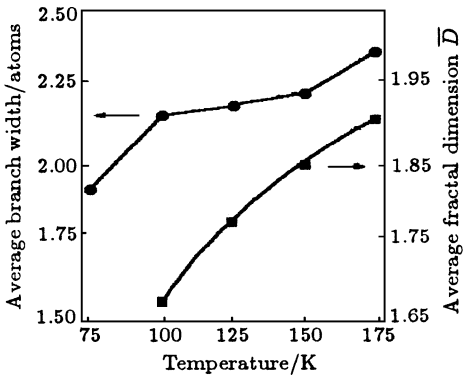


Fig.3. Temperature dependence of the average branch width and the average fractal dimension.

Nucleation and growth are competing processes, because an arriving adatom can either form a new island with another free adatom or diffuse into an existing island. With the increase of temperature, both the diffusion rate v and the deposition interval Δt increase, and the number of adatoms diffusing simul-

taneously on the substrate decreases. So the island number n decreases with increasing temperature. The large diffusion rate means a high probability for an arriving adatom to find an existing island before another adatom is deposited to provide a change for nucleation, yielding a lower number density of stable islands. As the substrate temperature increases to 100 K, saturation sets in and all of the adatoms will attach to existing islands before they find a second mobile atom to create a new nucleus. The relationship between the saturation island number n_s and deposition interval Δt can be described in a power-law:

$$n_s \propto \Delta t^{\gamma} . \tag{4}$$

A plot of $\lg n_s$ vs $\lg(\Delta t)$ is shown in Fig.4. The slope of this curve is $\gamma = -0.332$, which is very close to the theoretical value $-1/3$ of rate equations from nucleation theory.[12] It is seen that this relationship is not related directly to the substrate geometry because the similar result was also obtained in a square lattice substrate.[10]

To get insight into the fractal growth on triangular substrates and show the difference between our simulation and the DLA model, the island number n, the average branch width $\bar{b}$ and the average fractal dimension $\overline{D}$ as well as those obtained within the DLA model for various coverage are obtained. These results are shown in Table 2, where the substrate temperatures are set to be $T = 100$, 125, 175 K, respectively, and the deposition flux $R = 1.1 \times 10^{-3}$ ML/s.

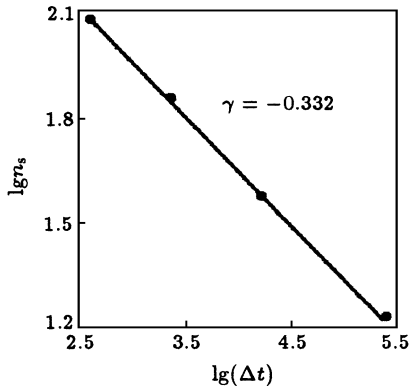


Fig.4. Relationship between saturation island number and deposition interval.

Table 2. The island number n, the average branch width $\bar{b}$ and the average fractal dimension $\overline{D}$ for various coverages at three different temperatures and those for the DLA model.

θ/ML		0.005	0.01	0.02	0.05	0.10	0.20	0.30	0.40
n	100 K	61	85	100	114	120	120	120	120
	125 K	22	26	29	33	34	34	34	34
	175 K	10	11	11	11	11	11	11	11
$\bar{b}$/atoms	100 K				1.87	2.16	2.73	3.99	7.61
	125 K			1.88	1.99	2.19	2.82	4.07	8.35
	175 K		1.93	2.14	2.26	2.37	3.41	6.23	11.25
	DLA	1.88	1.89	1.88	1.89	1.89	1.90	1.88	1.89
$\overline{D}$	100 K				1.65 ± 0.03	1.74 ± 0.03	1.80 ± 0.01	1.89 ± 0.04	1.93 ± 0.03
	125 K			1.63 ± 0.03	1.68 ± 0.02	1.76 ± 0.03	1.87 ± 0.01	1.94 ± 0.02	1.97 ± 0.03
	175 K		1.67 ± 0.03	1.71 ± 0.03	1.76 ± 0.02	1.82 ± 0.03	1.91 ± 0.01	1.96 ± 0.02	1.98 ± 0.02
	DLA	1.65 ± 0.03	1.64 ± 0.02	1.64 ± 0.03	1.66 ± 0.03	1.65 ± 0.01	1.66 ± 0.02	1.66 ± 0.02	1.67 ± 0.01

At 100 K, the island number n increases with increasing coverage for coverage below 0.10 ML, and then remains a constant. This means that saturation sets in and island growth predominantly occurs at higher coverage. At this stage, existing islands are so dense that each adatom has sufficient mobility to reach them with a higher probability than to hit a second adatom and form a new nucleus. At 175 K, saturation occurs even when the coverage is very low ($\theta = 0.01$ ML) because of the larger diffusion rate. obviously, the threshold value of coverage for saturation occurrence θ_s decreases with increasing temperature. These threshold values for various temperatures are shown in Table 3.

Table 3. Threshold values θ_s and θ_c for various temperatures.

T/K	75	100	125	150	175
θ_s/ML	0.35	0.10	0.06	0.02	0.01
θ_c/ML	0.10	0.06	0.02	0.015	0.007

The average branch width of clusters increases with increasing coverage in our simulation, which is consistent with the experimental observation of thin film growth. But it is independent of the coverage and almost keeps a constant 1.9 for the DLA model. Obviously, this increase of average branch width in our simulation is nonlinear, e.g., at 175 K, when the coverage is 0.01 ML, the average branch width $\bar{b} = 1.93$; when the coverage is increased to $\theta = 0.4$ ML, i.e., larger than the previous coverage by a factor of 40, the average branch width is still found to be $\bar{b} = 11.25$, i.e., it has increased only by a factor of 6. This behavior can be related to the screening effect of fractal growth. The dependence of the average branch width on coverage at three different temperatures is shown in Fig.5.

The average fractal dimension increases also with increasing coverage at the same temperature. However, the fractal dimension for the DLA model is still independent of the coverage and almost keeps a constant 1.66. The relationship between the average fractal dimension and coverage at three different substrate temperatures is shown in Fig.6.

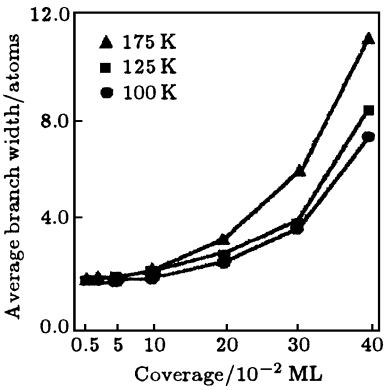


Fig.5. The average branch width vs coverage for three temperatures.

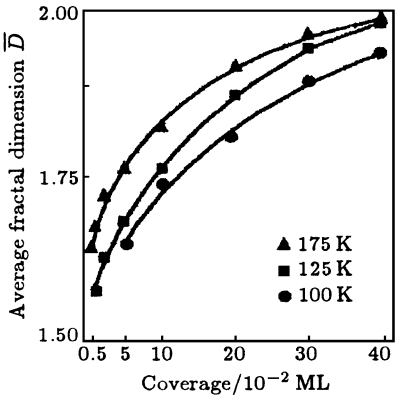


Fig.6. The average fractal dimension vs coverage for three temperatures.

In can also be seen, from comparing our simulation with the DLA model, that at low coverage, the branch width and the fractal dimension of DLA model are close to our simulation and consistent with the experiments of thin film growth. For example, at 125 K, the branch width and the fractal dimension of our simulation are very close to those formed within the DLA model when the coverage is 0.02 ML. For $\theta > 0.02$ ML, the difference between them becomes greater and greater. This means that above this coverage, the DLA model is not valid any more for describing the growth of thin films. Similar results are obtained for various temperatures. These threshold values of coverage θ_c are also shown in Table 3.

IV. SUMMARY

In summary, by using computer simulation, the fractal growth of thin films at low temperatures is investigated. The simulation results well illustrate the fractal growth processes under various conditions and are consistent with the experimental observation of thin film growth. The average branch width and the average fractal dimension of clusters are dependent on the substrate temperature and coverage, which is different from the classic DLA model. It is shown that the DLA model is only valid for describing thin film growth at low coverage.

Of course, at higher substrate temperature, the adatoms can diffuse not only on the substrate but also around the perimeter of a cluster, i.e., the edge diffusion cannot be neglected. The growth morphology will transit from fractal to dendrite and compact. On the other hand, the adatoms may evaporate from the substrate when the substrate temperature is high enough and the resident time of adatoms on the substrate is not long, and the diffusion steps of adatoms should also be limited. Furthermore, small clusters such as dimers and trimers can also diffuse on the substrate and the disociation of islands will set in at higher temperature, resulting in the formation of larger clusters by coalescence. Further results will be described in our forthcoming paper.

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