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FORMATIONS OF FRACTAL ISLANDS AGGREGATED WITH DISCS IN GAUSSIAN DISTRIBUTION ON NONLATTICE SUBSTRATES*

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The growth mechanism of fractal islands on a two-dimensional nonlattice substrate with periodic boundary conditions has been investigated by using Monte Carlo technique. Results show that the fractal dimension d_f of the final ramified islands is almost independent of the diffusion step length, mobility and rigid rotation of the islands. The characteristics of the size distribution of the discs in an island do not change the dimension d_f of the island. However, we find that d_f increases linearly with the surface coverage ρ of the system and its slope decreases with the increase of the mean diameter of the discs.

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I. INTRODUCTION

The importance of the growth mechanism of thin films lies both in establishing the experimental conditions for optimum thin film growth and in testing the theories that underpin our understanding of the nucleation, diffusion, aggregation and thin film growth process. A large number of studies have been devoted to the identification of the growth mechanism of ramified aggregates on solid substrates.[1−3] It has been found that depending on the combination of deposition and substrates, both single atoms and atomic clusters can diffuse on the solid substrates and fractal islands are formed.[4−7]

In understanding the growth mechanism of thin films, computer simulation is a very useful tool. Several simulation models, such as diffusion-limited aggregation(DLA), cluster-cluster diffusion(CCA) and deposition diffusion aggregation(DDA) models, were developed and provided important "experimental" evidences, which have explained some of the experimental results successfully.[8−10] It has been shown that the fractal islands have remarkable scaling and universality properties, and the fractal dimension of the islands depends on the surface coverage.

Recently, Ye *et al.* presented an experiment of silver atom deposition on silicone oil surfaces by vapor deposition method and they found that the compact silver atomic clusters of different sizes can diffuse quickly with various diffusion step lengths and rotate obviously with random rotation angles on the liquid surfaces.[11,12] Based on the experimental results, in our previous paper, we established a computer model to simulate the formation of islands on nonlattice substrates.[13] Results showed that the fractal dimension of the ramified islands is almost independent of the diffusion step length and rigid rotation. Furthermore, they also revealed that the fractal dimension of the ramified islands depends almost linearly on the surface coverage of the system. Unfortunately, in this model, all the discs were selected to be of identical size, which is in contradiction with the experimental evidence.[11,12]

In this paper, the Monte Carlo (MC) technique is used to investigate the fractal dimension of the fractal islands aggregated with the discs of different sizes on a nonlattice substrate. Results show that the fractal dimension of the islands is nearly independent of the diffusion step length, the mobility, and the rigid rotation angle of the islands. The fractal dimension increases linearly with the surface coverage. But the characteristics of the size distribution of the discs do not alter the fractal dimension as well as the linear relation between the fractal dimension and the surface coverage.

II. MODEL DESCRIPTION

According to Refs.[11] and [12] we suggest that there are mainly three kinds of forces acting on the silver islands on liquid surfaces. The first one comes from the collision between the liquid molecules and silver atoms, which results in the diffusion and rotation characteristics of the islands. The second one is the frictional force, which cumbers the move of islands.

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The last one is the attractive force between two discs or islands. It was observed that two compact silver clusters or islands irreversibly adhere to each other and from then on move as one unit if they touch, indicating that the attractive force among the discs really exists. Unfortunately, the physical origin of the attractive force is not very clear at the moment. We believe that the force binding the silver clusters may be resulted from the surface tension of the liquid surface.

Owing to the random collision with the liquid molecules, the diffusion step length l and rotation angle θ of islands are variable. Therefore, in our MC simulation, parameters l and θ of each island are chosen randomly at each time: (1) The diffusion step length l for each step can be randomly picked with equal probability from 1,2,3,$\cdots, l_{\rm m}$, where the maximum length $l_{\rm m}$ is fixed. (2) The rotation angle θ of an island is chosen randomly from 0 to the maximum value $\theta_{\rm m}$, also with equal probability. For the sake of simplicity, only the rigid rotation behavior about the mass centers of the islands is considered in the paper. The random choice for l and θ is constrained by a nonoverlap condition. If a diffusion step length or a rotation angle is so large that the overlap between two discs (or islands) happens, then the diffusion step length or the rotation angle will be reduced automatically until the two discs or islands just come into contact with each other. On the other hand, if two discs or islands touch, they irreversibly glue to each other and from then on move as one island. Obviously, the model described above is quite similar to the real situation of the experiment.[11,12]

It was found that the sizes (or diameters) of the compact silver clusters on silicon oil surfaces are not the same in experiments.[11,12] The size distribution of the silver compact clusters is measured for a sample at different aggregation time and the results are shown in Fig.1. Here we use a truncated Gaussian distribution $f(d) \propto {\rm e}^{-(d-\bar{d}\,)^2/2\sigma^2}, d_{\rm min} \leq d \leq d_{\rm max}$, to represent approximately the real diameter distribution, where d is the diameter of discs, $\bar{d}$ is the mean disc diameter, σ is the standard error of the distribution, $d_{\rm min}$ and $d_{\rm max}$ are the minimum and maximum of the disc diameters, respectively. Since the length scale is a relative value in MC simulatin, we select $d = 1.0$ to represent the real value $5\,\mu$m in the experiment. We find that, if we choose $\bar{d} = 1.25$ and $\sigma = 0.37$, then the truncated Gaussian distribution fits the experimental data very well (see Fig.1). Since the number of the discs with diameter $d < 0.5$ or $d > 2.0$ are very small, $d_{\rm min}$ and $d_{\rm max}$ are set to be 0.5 and 2.0 for all the simulations below. And $\bar{d}$ is also fixed as 1.25. Hereafter we use a discrete Gaussian distribution with diameter interval Δ instead of a continuous one.

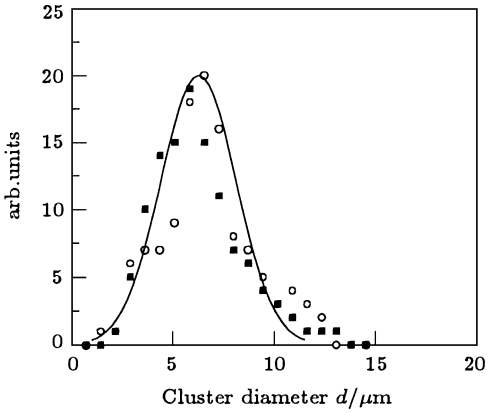


Fig.1. The distributions of the disc diameters. Circles and squares are the experimental data, measured at time $t = 18$ and 168 h, respectively; the solid line is the Gaussian distribution of disc size.

Generally, the diffusion mobility of an island decreases with the increase of the island size or area. The probability p that the island actually move, which is proportional to its mobility, can be described as:[10]

$$p \propto s^{-\gamma}, \tag{1}$$

where s is the area of the island and the exponent γ characterizes the dependence of the probability p on the island size s. A small γ denotes that even large islands can diffuse easily while a very large γ indicates that only the diffusion of very small islands can be observed.

Therefore, the simulations presented here correspond to the main characteristics found in the experiment of the silver islands diffusing and aggregating on liquid surfaces. Initially, n discs with a Gaussian distributions in their sizes are randomly deposited (avoiding overlap) on a nonlattice square of area $L \times L$ with periodic boundary conditions. Each time, only one island is randomly picked to move with a random manner (parallel diffusion in one of four directions: $\pm x$ and $\pm y$, or rigid rotation). The fractal dimensions of the final ramified islands are calculated by the box method.[14] We draw a grid consisting of N^2 equal square cells on the island and then determine the number of effective cells $C(N)$ that are needed to cover the island. The fractal dimension $d_{\rm f}$ is the slope of the linear fit from a log-log of $C(N)$ vs. N. All the values of $d_{\rm f}$ are averaged over ten simulation

runs. All the simulations in this paper are carried out on a square of size $L = 200$.

Ⅲ. RESULTS AND DISCUSSION

A. Influence of the diffusion step length, mobility and rigid rotation on the fractal dimension

The simulations are carried out for the system with the total disc number $n = 2000$, $\Delta = 0.05$ is used in these simulations. Figure 2 shows that in the system consisting of the discs with Gaussian distribution in size, the fractal dimension d_f of the islands is nearly independent of both the maximum step length l_m and the parameter γ. Similar results are also found for the systems with $n = 1000, 3000$ and 4000. We also find that the rotation behavior of the islands has nearly no effect on the fractal dimension of the islands. These results are in good agreement with the conclusion obtained from the system with single kind of discs and strongly suggest that the characteristics of the Gaussian distribution in disc size does not alter the fractal dimension d_f and the relation between d_f and the other diffusion parameters, i.e., l_m, θ_m and γ.

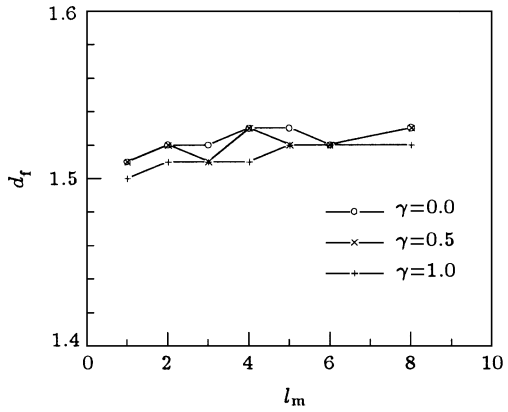


Fig.2. Dependence of fractal dimension d_f on the maximum step length l_m and the parameter γ.

We propose that the phenomena mentioned above result mainly from the random nature of diffusion. Our computer model describes a homogeneous system in which all the discs are deposited randomly on a nonlattice substrate. Thus the system possesses random diffusion behavior statistically. The motion of the discs and ramified islands with random diffusion step length and rotation angle would not change this homogeneous property. Thus the growth mechanism as well as the fractal dimension of the aggregates could not be obviously influenced by the random motion. In other words, the fractal dimension is not a good parameter for distinguishing the details of the aggregates. However, to make a final conclusion for the physical origin of the phenomena, further experimental and theoretical evidences are still needed.

B. Surface coverage

Several MC simulation results indicate that the fractal dimension d_f of the final islands depends on the surface coverage ρ, which is defined as the ratio of the occupied area by the discs to the area of the square.[10,13] In this section, the dependence of d_f on ρ will be studied. For the sake of simplicity, in the following study, we select $\gamma = 0$, $l_m = 5$, and $\theta_m = 0$, which is reasonable since, according to the discussion above, d_f is independent of these parameters.

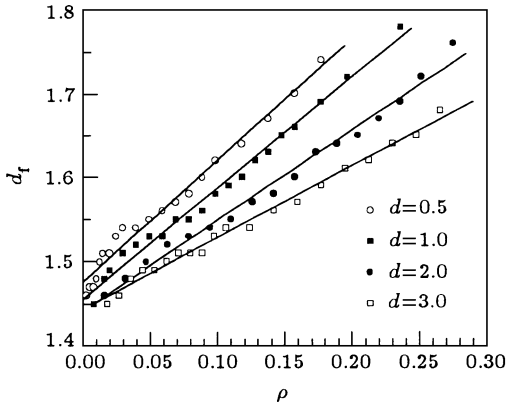


Fig.3. The fractal dimension d_f as a function of the surface coverage ρ for different disc diameters d.

Figure 3 shows the dependence of d_f on ρ for four systems with single kind of discs: $d = 0.5, 1.0, 2.0$ and 3.0, respectively. We find that all the fractal dimensions of the four systems increase linearly with the surface coverage when $\rho < 0.25$. The linear relation can be expressed approximately as

$$d_f = a + b\rho, \tag{2}$$

which is in excellent agreement with our previous finding.[13] The values of intercept a and slope b derived from the simulation data are listed in Table 1. Unfortunately, it is difficult to study the fractal dimension behavior at higher surface coverage since the simulation period increases with the surface coverage. Asymptotically, it is expected that the fractal dimension d_f reaches its limit value ($d_f = 2.0$) when the surface coverage ρ approaches 0.4 for the system $d = 1.0$.

Table 1. Values of the intercept a and slope b for different distributions of the disc sizes.

	a	b
$d = 0.5^{(1)}$	1.475	1.452
$d = 1.0^{(1)}$	1.455	1.322
$d = 2.0^{(1)}$	1.441	1.074
$d = 3.0^{(1)}$	1.443	0.851
$\bar{d} = 1.25^{(2)}$	1.448	1.220
$\bar{d} = 1.5^{(3)}$	1.442	1.181

(1) results obtained from a simulation with single kind of discs;
(2) result obtained from a simulation of discs with a truncated Gaussian distribution in size, with $\sigma = 0.37$ and $\Delta = 0.05$;
(3) results obtained from a simulation of the system containing two kinds of discs.

Shown in Fig.4 is the dependence of the fractal dimension on the surface coverage for three systems with a truncated Gaussian distribution of disc diameters. The standard errors of the three systems are $\sigma = 0.25, 0.37$ and 0.45, respectively. Here we select $\Delta = 0.05$ for these simulations. An interesting observation is that the lienar relation between the fractal dimension and the surface coverage is still available for all the systems and the fractal dimension is almost independent of the standard error σ. In Table 1, one notes that the data of slope b obtained from the simulation lie between those of the systems with $d = 1.0$ and 2.0. This result is quite reasonable since the mean disc diameter of the system is $\bar{d} = 1.25$, which just lies between $d = 1.0$ and 2.0. Therefore, we concluded that the relation between $d_{\rm f}$ and ρ depends on the mean diameter $\bar{d}$ rather than on the standard error σ.

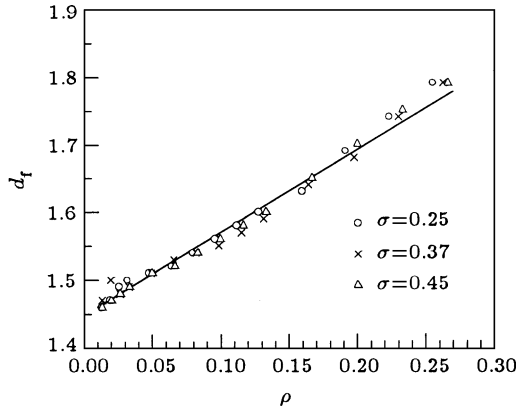


Fig.4. The fractal dimension $d_{\rm f}$ as a function of the surface coverage ρ for different standard errors σ. The solid line is a linear fit for the data of $\sigma = 0.37$.

The relationship between $d_{\rm f}$ and ρ with different diameter intervals $\Delta = 0.02, 0.05, 0.07$ and 0.1 is shown in Fig.5. In these simulations, the standard error of the size distribution is $\sigma = 0.37$. We find that the interval of diameter Δ does not obviously influence the relation between $d_{\rm f}$ and ρ.

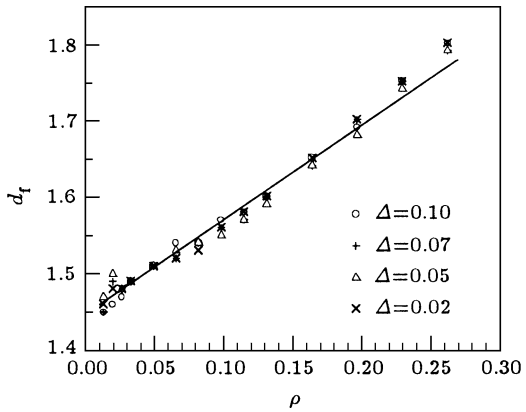


Fig.5. The fractal dimension $d_{\rm f}$ as a function of the surface coverage ρ for different diameter intervals Δ. The solid line is a linear fit for the data of $\Delta = 0.05$.

Since the relationship between $d_{\rm f}$ and ρ is independent of the standard error σ and the diameter interval Δ, then it is believable that this behavior will still be valid for the system containing several kinds of discs with a random diameter distribution. Consider a simple system which consists of two kinds of discs. The final ramified island is aggregated with n_1 discs of diameter $d_1 = 1.0$ and n_2 discs of diameter $d_2 = 2.0$ in a square of $L \times L = 200^2$. We vary the mean diameter $\bar{d}\left(\bar{d} = \dfrac{n_1 + 2n_2}{n_1 + n_2}\right)$ and surface coverage ρ by changing the values of n_1 and n_2. As expected, the linear relation between $d_{\rm f}$ and ρ is again observed. The intercept a and slope b for $\bar{d} = 1.5$ is listed in Table 1. One can see that the value b lies between those of $\bar{d} = 1.25$ and $d = 2.0$. Thus, we believe that the linear relation between $d_{\rm f}$ and ρ is generally valid at small surface coverage.

One can see, from Table 1, that the intercept a is nearly a constant. Therefore, at the low limit of the surface coverage, $d_{\rm f} \approx a = 1.44 \sim 1.48$, is quite close to that measured in the CCA model ($d_{\rm f} = 1.45 \sim 1.50$).[9] We then conclude that, when the surface coverage ρ is near zero, the fractal dimensions of the islands in all the systems mentioned above are nearly independent of the disc diameter and approach the result of the CCA model. However, the slope b drops with the increase of the mean disc diameter $\bar{d}$ and can be expressed approximately as

$$b = 1.55 - 0.237\bar{d}. \tag{3}$$

We define $\bar{d} = d$ for the system of single kind disc. Then the fractal dimension decreases with the increase of disc diameter at a fixed surface coverage. To establish a detailed theory for describing this phenomenon is out of the scope of this paper. However, we can

give a qualitative explanation for this interesting phenomenon below.

It was implicitly reported in the CCA model that the fractal dimension of the aggregating island increases with the number density of the discs.[9] Obviously, if the number density increases, the ramified island aggregated in the simulation will exhibit a more branched feature, which will result in the increase of the fractal dimension $d_{\rm f}$ since $d_{\rm f}$ increases with the degree of the branch abundance in the island. Thus, for the system with a fixed surface coverage, if we reduce the disc diameter, the number density (or number) of the discs in the system will increase and then the fractal dimension of the fractal island will go up. Therefore, the results shown in Figs.3, 4 and 5 are reasonable.

From the discussions above, we can conclude that, at small surface coverage ($\rho < 0.25$), the linear relation between $d_{\rm f}$ and ρ is a general phenomenon for the systems containing the discs with different size distributions and the fractal dimensions of all the systems approach the result of the CCA model as the surface coverage is near zero. For the system with fixed area and surface coverage, the key factor which affects the fractal dimension is the mean disc diameter $\bar{d}$, i.e., $d_{\rm f}$ decreases with the increase of $\bar{d}$ and the influence of $\bar{d}$ on $d_{\rm f}$ goes up as ρ increases.

IV. CONCLUSION

According to the MC study on the growth mechanism of the fractal islands on nonlattice squares, the following conclusions can be made:

1. For the ramified islands aggregated by the discs with different sizes, the fractal dimension of the islands is almost independent of the diffusion step length, mobility and rigid rotation angle of the islands.

2. The linear relation between the fractal dimension and the surface coverage of the system is generally valid for all the systems containing discs with different size distributions.

3. For the fixed system size and surface coverage, the fractal dimension is sensitive to the mean disc diameter but insensitive to the size distribution of the discs. At the low limit of the surface coverage, the fractal dimension of all the systems is around $1.44 \sim 1.48$, which is close to the result of CCA model. At a large surface coverage, the fractal dimension decreases with the increase of the mean disc diameter. And this influence of the mean disc diameter on the fractal dimension goes up as ρ increases.

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REFERENCES

[1] R. Q. Hwang, J. Schröder, C. Günther, R. J. Behm, *Phys. Rev. Lett.*, **67**(1991), 3279.
[2] M. Bott, Th. Michely, G. Comsa, *Surf. Sci.*, **272**(1992), 161.
[3] Th. Michely, M. Hohage, M. Bott, G. Comsa, *Phys. Rev. Lett.*, **70**(1993), 3943.
[4] R. Gomer, *Rep. Prog. Phys.*, **53**(1990), 917.
[5] H. Röder, E. Hahn, H. Brune, J. P. Bucher, K. Kern, *Nature*(*London*), **366**(1993), 141.
[6] J. M. Wen, S. L. Chang, J. W. Burnett, J. W. Evans, P. A. Thiel, *Phys. Rev. Lett.*, **73**(1994), 2591.
[7] S. C. Wang, G. Ehrlich, *Phys. Rev. Lett.*, **79**(1997), 4234.
[8] T. A. Witten, L. M. Sander, *Phys. Rev. Lett.*, **47**(1981), 1400.
[9] P. Meakin, *Phys. Rev. Lett.*, **51**(1983), 1119.
[10] P. Jensen, A. Barabási, H. Larralde, S. Havlin, H. E. Stanley, *Phys. Rev.*, **B50**(1994), 15316.
[11] G. X. Ye, Th. Michely, V. Weidenhof, I. Friedrich, M. Wuttig, *Phys. Rev. Lett.*, **81**(1998), 622.
[12] G. X. Ye, B. Yang *et al.*, *Acta Physica Sinica*, **47**(1998), 1900(in Chinese).
[13] M. B. Luo, G. X. Ye, A. G. Xia, J. S. Jin, B. Yang, J. M. Xu, *Phys. Rev.*, **B59**(1999), 3218.
[14] J. Z. Zhang, Fractal (Tsinghua University, Beijing,1995), p.122(in Chinese).