Dynamic Scaling of Surface Growth in the Influence of Small Clusters Mobility *

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The deposition, diffusion and coalescence processes appeared in the surface growth of compact or liquid-like clusters system are studied. By introducing a critical cluster size i_c, such that clusters of size $s \leq i_c$ are mobile while clusters of the size $s > i_c$ are immobile, the effects of mobility of small clusters on cluster size distribution are simulated. As i_c increases, the exponent γ which relates the maximum cluster density to the normalized deposition rate is increased. Numerical simulation results indicate that the scaled cluster-size distributions are independent of the deposition rate, but closely associated with the small clusters mobility. As i_c increases, it is found that the distributions become narrow.

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Island growth on surfaces during submonolayer deposition has received considerable attention because it is the basic process of thin film growth.[1-5] With recent development of scanning tunneling microscopy and high-resolution diffraction and scattering techniques which can effectively probe the morphology and the growth of microstructures at the surface, much physical understanding has been achieved, and many mechanisms have been proposed. Using various experimental techniques, it is now possible to study the submonolayer islands morphology, density, and size distribution in a variety of systems ranging from homoepitaxial systems such as Fe/Fe(100),[6] Ni/Ni(100) (Ref. 7) and Cu/Cu(100) (Ref. 8) to heteroepitaxial systems such as Pb/Cu(001).[9] It has been realized that the island morphology and size distribution make an important effect on the film growth. By studying the scaling properties of island size distributions, valuable information of microscopic processes can be obtained. Many recent models have included more details of the microscopic processes occurring during growth, such as edge diffusion and adatom exchange with the substrate.[10,11] However, in these models the clusters are still considered to be immobile. Jensen *et al.*[12] introduced a possibility of cluster diffusion and developed a deposition, diffusion and aggregation (DDA) model. They showed that the island mobility considerable changes the surface morphology and the dynamical scaling of the growth. In DDA model, the edge diffusion is expected to be negligible and islands are considered as fractals. However, in real thin film growth processes, as the temperature increases islands become more compact and in some case they become liquid-like droplets.[13]

In this letter, we present a model that describes the deposition, diffusion and coalescence (DDC) processes exhibiting in compact or liquid-like clusters system. In the DDC model, all the clusters are simplified as two-dimensional droplets, this is inspired by recent droplet pattern in the thin film growth experiments such as GaAs film[13] and Cu/Cu(100) film where Cu clusters are compact and have round corners.[8] Using Monte Carlo (MC) simulation we may have a detail understanding on the scaling behavior of cluster size distributions in the early stages of surface growth. We find that a power-law exponent γ that relates the maximum island density to the scaled deposition rate increases as the cluster mobility increases. Moreover, the scaled cluster size distributions exhibit a self-similarity for different deposition rates and show a change as a function of cluster mobility.

In computer simulations, the DDC model is defined as follows:

(1) Deposition: Monomers (particles or clusters with size $s = 1$) are deposited at randomly chosen positions on the $L \times L$ lattice with a constant deposition flux F per lattice site per unit time.

(2) Diffusion: All monomers and clusters are chosen at random and attempt to move north, east, south, or west by one lattice constant. We introduce a critical cluster size i_c as the size for which all clusters of size $s \leq i_c$ are mobile, while clusters of size $s > i_c$ are immobile. The probability that the clusters of size $s \leq i_c$ actually move is proportional to the mobility $D_s = D_0 s^{-\alpha}$, where s is the size of the cluster, D_0 is the diffusion coefficient for a monomer, and the parameter α characterizes the dependence of D_s on cluster size.

(3) Coalescence: If two clusters of sizes s_1 and s_2 come to touch each other due to their diffusion, they will merge into a single cluster of size $s_1 + s_2$ with a shape of droplet. If an incident monomer lands on surface of a cluster, it will merge into the cluster immediately. There is no evaporation in this model.

Figure 1 shows examples of typical growth morphologies obtained from two different deposition rates. The simulations were performed on a 100×100 lattice at a coverage of 0.4 monolayer(ML) for $i_c = 1$, and $\alpha = 1$. We see that in the case of $D_0/F = 10^7$ the total number of cluster is $N = 74$ and the average cluster size is about $\langle s \rangle \approx 54$ (see Fig.1(a)), when the value of D_0/F increases to 10^8, N decreases to 34, while $\langle s \rangle$ increases to 118 (see Fig.1(b)). This implies

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that decreasing the deposition flux F (increasing the value of D_0/F from 10^7 to 10^8) gives rise to fewer and larger clusters. Our simulations also show that the cluster density is associated with the coverage θ and the critical cluster size i_c.

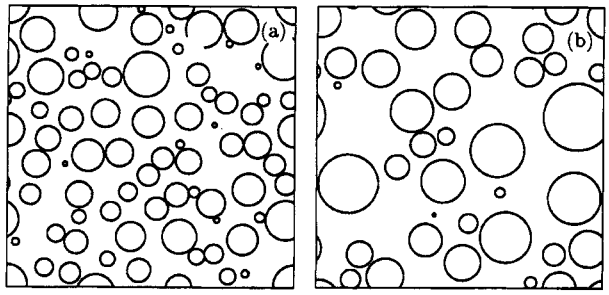

Fig. 1. Snapshots of surface morphologies obtained in MC simulations for two different deposition rates at the coverage of 0.4 ML: (a) $D_0/F = 10^7$, (b) $D_0/F = 10^8$. The simulation were carried out on a 100×100 lattice in the case of $\alpha = 1, i_c = 1$.

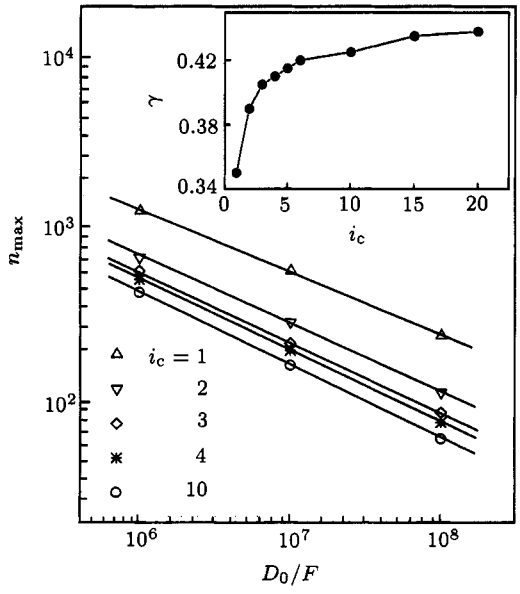

Fig. 2. A log-log plot of the dependence of the maximum cluster density on the scaled deposition rate D_0/F for different values of i_c in the case of $\alpha = 1$. The lines are power-law fits to the data. All the data exhibit a good power-law behavior of which the exponent γ becomes larger as i_c increases (see the inset graph).

The maximum cluster density versus the scaled deposition rate D_0/F is presented in Fig. 2 for several different critical cluster sizes i_c in the case of $\alpha = 1$. For each data the coverage is well within the quasi steady-state regime, in which the cluster density reaches its maximum. In this regime, scaling arguments indicate that in the asymptotic limit of low deposition rate, the maximum cluster density n_{max} exhibits a power-law behavior

$$n_{max} \approx \left(\frac{D_0}{F}\right)^{-\gamma}, \qquad (1)$$

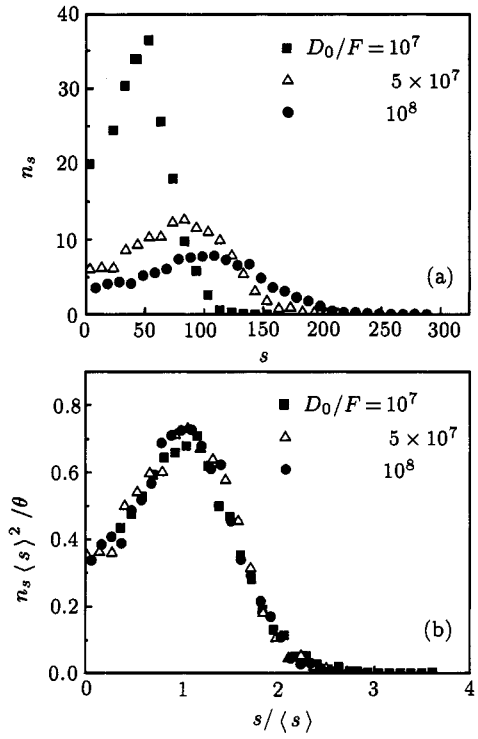

Fig. 3. (a) Unscaled cluster size distributions n_s resulting from the simulations with $i_c = 1, \alpha = 1, \theta = 0.1$ for various D_0/F on a lattice of 1000×1000. (b) Scaled cluster size distributions of (a) which shows a strong self-similarity.

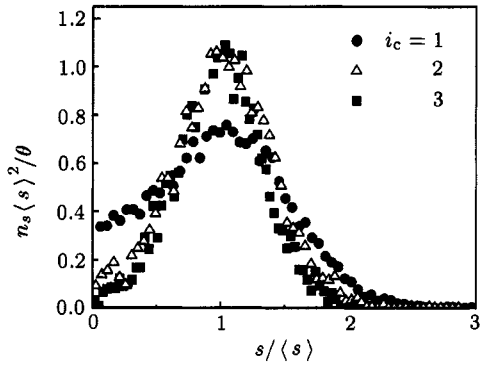

Fig. 4. Scaled cluster size distributions for three different i_c in the case of $\alpha = 1, \theta = 0.1, D_0/F = 10^8$. The simulation results were based on statistics collected from 20 runs on a 1000×1000 lattice.

where the exponent γ depends on microscopic properties of the system. From the figure we can see that for all values of i_c the maximum cluster densities show a clear power-law dependence on D_0/F. The exponent γ can be obtained from the slopes of the lines. With increasing small clusters mobility, the exponent of power-law γ changes from the value of 0.35 for $i_c = 1$ to 0.43 for $i_c = 15$. This result is in good agreement with Jensen *et al.*'s computer simulation result for DDA model.[12] They found the value of γ is 0.33 for only monomers diffusion and 0.42 for growth with mobile islands obeying a power-law size dependence of their mobility. The dependence of the exponent γ on the critical cluster size i_c is shown in the inset of

Fig. 2. In the region of small i_c the exponent γ changes rapidly, while in the region of large i_c the exponent γ changes very slowly (in the limit of $i_c \to \infty$, the simulation gives the result of $\gamma = 0.45$). This implies that the dynamic behavior of the early stage during surface growth is mainly dominated by the mobilities of small clusters.

The scaling properties of the cluster size distribution are further studied in our simulations. The cluster size distributions n_s can be rescaled with a scaling function $f(u)$ defined as[14]

$$n_s = \frac{\theta}{\langle s \rangle^2} f(\frac{s}{\langle s \rangle}) , \qquad (2)$$

where $\langle s \rangle$ is the average cluster size, θ the coverage and n_s the number of clusters with size of s. In conventional growth theory islands are considered as immobile and it is well established that this scaled size distributions for different coverages and different values of D_0/F can be collapsed into a single curve. Figure 3 shows the cluster size distributions for three different values of D_0/F in the case of $\theta = 0.1$ ML and the critical cluster size $i_c = 1$. Our simulations are performed on a 1000×1000 lattice, and in order to have a good fitting all data are obtained by the average of $10 - 50$ simulations. The dependence of the number of clusters, n_s, on their size s is shown in Fig. 3(a). As the value of D_0/F is decreased by increasing the deposition flux F, the number of small clusters grows and the number of large clusters reduces gradually. The position of the peak of the cluster size distribution also decreases while the peak height increases. The cluster size distribution n_s clearly depends on the deposition rate F, coverage θ and other parameters. When we use the scaling relation of Eq. (2) to scale the cluster size distribution n_s in Fig. 3(a), the scaling function $f(s/\langle s \rangle) = n_s \langle s \rangle^2 / \theta$ versus the scaled cluster size $s/\langle s \rangle$ can be obtained as shown in Fig. 3(b). Clearly, there is an excellent data collapse and all the distributions fall on a single curve. The scaled cluster size distribution is independent of the deposition rate. This implies that the cluster size distributions resulting from growth with deferent deposition flux F exhibit the same self-similarity.

However, the self-similarity vanishes when comparing scaled cluster size distributions for different cluster mobilities. Three different cluster mobility hypotheses have been made in our simulation model: (a) only monomers move ($i_c = 1$); (b) monomers and dimers diffuse ($i_c = 2$), and (c) not only monomers, dimers but also trimers do move ($i_c = 3$). Figure 4 shows the scaled cluster size distributions for different values of critical cluster size i_c. These results are based on statistics averaged from 20 runs on a 1000×1000 lattice with $\theta = 0.1$ ML and $D_0/F = 10^8$. We see that the scaled cluster size distribution function becomes narrower and has a higher maximum when larger clusters are allowed to diffuse. This result is corroborated by an experimental study by Bartelt *et al.*, which shows the sharpening effect of dimer diffusion on the island size distribution during epitaxial growth.[15] Our results can help to decide whether monomers or larger clusters can move on the surface by comparing the rescaled experimental size distribution and those predicted by the DDC model.

In summary, we present a model that describes the deposition, diffusion and coalescence processes exhibiting in the surface growth of compact or liquid-like clusters system. We have performed a study on the effect of mobility of small clusters for the scaling and morphology of surface growth. The exponent γ, which describes the dependence of cluster density on deposition rate, was examined with MC simulations. It was found that as i_c increases, γ increases rapidly in the region of small i_c. This reflects the fact that the mobility of small clusters plays a dominant role on the dynamic scaling of surface growth. Simulation results indicate that the scaled cluster size distributions are independent of the deposition flux F, but closely associated with the mobility of small clusters, and become sharp as i_c increases. Our simulation results can be of some help to experimentalists to understand the microscopic processes present in their experiments.

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