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EFFECTS OF ISLAND-EDGE EXCHANGE BARRIER ON 2D PATTERN FORMATION IN SURFACTANT-MEDIATED EPITAXY*

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The nucleation and growth of two-dimensional islands in a surfactant-mediated epitaxy system have been studied by computer simulation. To improve the recent results published in the literature, we use a configuration-dependent energy barrier for the exchange process at the island edge in our model. The simulations produce fractal islands at high temperatures or low deposition fluxes, and a transition to regular compact islands occurs at lower temperatures or higher fluxes, in good agreement with the recent experimental results. The barrier for the island-edge exchange has quite a strong effect on the island density as a function of temperature and flux. A small change of the island-edge exchange barrier induces a large variation of the island density in the low-temperature or high-flux region. The flux-dependent island density shows a clear scaling-law behaviour in the intermediate-flux region. The scaling exponent increases evidently as the island-edge exchange barrier increases.

Keywords: Monte Carlo simulation, epitaxy, growth, fractal
PACC: 6890, 6855

I. INTRODUCTION

Over the past few years, much effort has been devoted to the study of the influence of surfactant on semiconductor thin-film growth.[1−5] Different kinetic mechanisms have been proposed to describe the atomic process of the growth. For example, high density of small two-dimensional (2D) islands was observed in Si homoepitaxy on Si(111) with a monolayer of Sb as the surfactant, and the nucleation behaviour was attributed to a reduced surface diffusion length of the deposited adatoms by many researchers.[6−8] Kandel and Kaxiras[9,10] proposed a different mechanism based on first principles calculations: the deposited adatoms diffuse rapidly on the surfactant-passivated surface; adatoms exchange with surfactant atoms for nucleation or for attaching to the growing islands, and the exchange barrier is significantly higher than the diffusion barrier. Including these atomic processes in a kinetic Monte Carlo (KMC) simulation of the growth, they have indeed reproduced the observed high density of islands. More recently, the formation of 2D Ge islands grown on Si(111) with a monolayer of Pb as the surfactant has been studied in detail by Hwang *et al.*[11,12] They found that a morphological transition from fractal to regular compact islands occurs as the deposition flux is increased, or as the growth temperature is decreased. The result is completely contrary to what was expected from the traditional results. To explain this island shape transition, Liu *et al.*[13,14] proposed a model which includes three kinetic parameters related to the atomic processes: a small diffusion barrier on top of the surfactant layer, a high exchange barrier for nucleation, and a second highest barrier for the exchange of atoms to become incorporated at the island edges. Using the KMC method, the simple model can produce fractal islands at high temperatures or low fluxes, and a transition to irregular compact islands can be induced by either decreasing the growth temperature or increasing the deposition flux.

Because the surfactant layer saturates the surface dangling bonds and passivates the surface, the energy of the system is lowered when the surfactant layer is on top rather than buried under newly deposited atoms, and thus the surfactant tends to float on top of the growing film during deposition. Therefore, the decisive process which controls the nucleation and growth of 2D islands, is the place exchange between surfactant atoms and newly deposited atoms. Despite intensive experimental and theoretical studies, the information about the kinetic exchange process is still quite limited. In this paper, we simulate the 2D pattern formation in surfactant-mediated epitaxial growth by using a configuration-dependent exchange barrier to im-

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prove the previous simulation studies.[13,14] Our simulations produce surface patterns changing from fractal islands to regular compact islands as the deposition flux is raised or the temperature is lowered. We also find that in the case of compact growth, the island-edge exchange barrier has quite a strong effect on the density of 2D islands. By tuning the exchange barrier parameter, we can change the scaling behaviour in the temperature and flux dependence of the stable island density.

Ⅱ. MODEL AND SIMULATION

We study the growth of a material B on the substrate A with a monolayer of S as a surfactant. As deposition starts, the deposited adatoms hop on the surfactant terrace at a rate of $h_{\rm d} = \nu\exp(-V_{\rm d}/kT)$, where ν is the attempt frequency, and $V_{\rm d}$ is the corresponding activation energy. Nucleation of a 2D island occurs when an adatom exchanges its position with the surfactant atom beneath it by overcoming an energy barrier $V_{\rm s}$ ($> V_{\rm d}$). Once an adatom undergoes place exchange and reaches the substrate, it is immobile (dead atom). A stable island consists of dead atoms (see ball model in Fig.1). We neglect the binding energy between the adatoms on the surfactant terrace in the present model, i.e. the clusters formed on top of the surfactant layer are unstable, and can dissociate. When a diffusing adatom hops to a site adjacent to a dead-atom island, it remains stuck there. The stuck adatom is still active, it finally exchanges position with the surfactant atom beneath it by overcoming an island-edge barrier and becomes part of the island. Because the exchange at an island edge is assisted by the dead atoms of the island, to which the stuck adatom is sticking, the island-edge exchange barrier is lower than the energy barrier for preventing an isolated adatom from exchanging down for nucleation. Hence the barrier for island-edge exchange can be expressed as $V_{\rm s}$-$nV_{\rm a}$ if a stuck adatom has n B-type second nearest neighbours beneath it. The rate at which an active adatom exchanges position with the surfactant atom for nucleation (n=0) or incorporation at the existing island edge (n=1,2,3) is determined by the configuration-dependent Arrhenius-type expression $h_{\rm e}=\nu\exp[-(V_{\rm s} - nV_{\rm a})/kT]$. Here, the attempt frequency ν is assumed to be the same for all processes. The three barriers used in the model satisfy the relations $V_{\rm a} \ll V_{\rm d} < V_{\rm s}$. $V_{\rm s}$ is the largest because the corresponding exchange process is related to the interlayer atomic motion. Diffusion barrier $V_{\rm d}$ is relatively small due to the chemical passivation of the surface. A very small $V_{\rm a}$ means a large island-edge exchange barrier, making the growth behaviour of the exchange rate of the 2D islands limited.

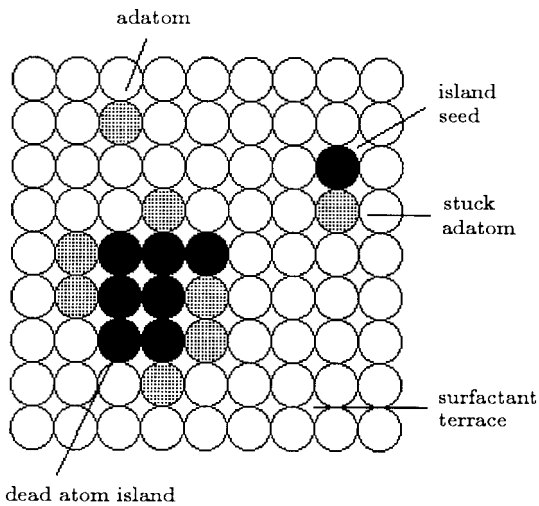


Fig.1. The ball model of SME growth. The white circles are surfactant atoms. The lightly shaded circles are B-type atoms adsorbed on the surfactant terrace. The black circles represent the buried B-type atoms after place exchange.

The actual simulations were carried out on a square 200×200 lattice with periodic boundary conditions. The growth of the island was initiated by the random deposition of atoms onto the surface at a rate F. Between two successive deposition events each diffusing atom can make $h_{\rm d}/F$ single site hops in one of four possible, randomly chosen directions. Additionally, for each hop an adatom can undergo place exchange with a probability $h_{\rm e}/h_{\rm d}$. We have set the parameters $V_{\rm d}$=0.6eV, $V_{\rm s}$=0.95eV, and ν=$10^{13}{\rm s}^{-1}$ with variable temperature, flux, and $V_{\rm a}$.

Ⅲ. RESULTS

Figure 2 shows the evolution of the island shape as a function of temperature at 0.1ML coverage for F=0.003ML/s and $V_{\rm a}$=0.09eV. At a high temperature (370K), an adatom arriving at the edge of an island has a high exchange rate and can be easily incorporated into the existing island where it first attaches. The growing islands therefore have a fractal structure, similar to that of the hit-and-stick diffusion-limited aggregation (DLA) model (Fig.2(a)). When T=330K, an adatom stuck at an island edge can stay there for a relatively longer time before it exchanges down. The stuck adatoms act as shields and prevent incoming adatoms from sticking to the stable island

through their sites.[13] As a result, other free edge sites of the island have more chance to capture incoming adatoms and become active growth sites. Thus the growth leads to the formation of fractal islands with thicker branches (Fig.2(b)). As the temperature is further lowered, the decreased island-edge exchange rate makes the number of the adatoms stuck at the island edge increase; the shielding effect is enhanced, and the growing islands become more compact (Fig.2(c)). When the temperature is low enough (300K), any free island-edge site can capture adatom effectively, and the growing islands remain to be surrounded by the stuck adatoms as the deposition proceeds. Because the barrier for island-edge exchange depends on the number of dead B-type second nearest neighbours, an adatom stuck at a kink site (n=2, see Fig.1) exchanges down with a relatively higher rate than that at a straight edge (n=1). Therefore the edges of the islands tend to be straight with decreasing kinks. The growth finally leads to compact islands with rectangular shapes which reflect the symmetry of the underlying lattice (Fig.2(d)).

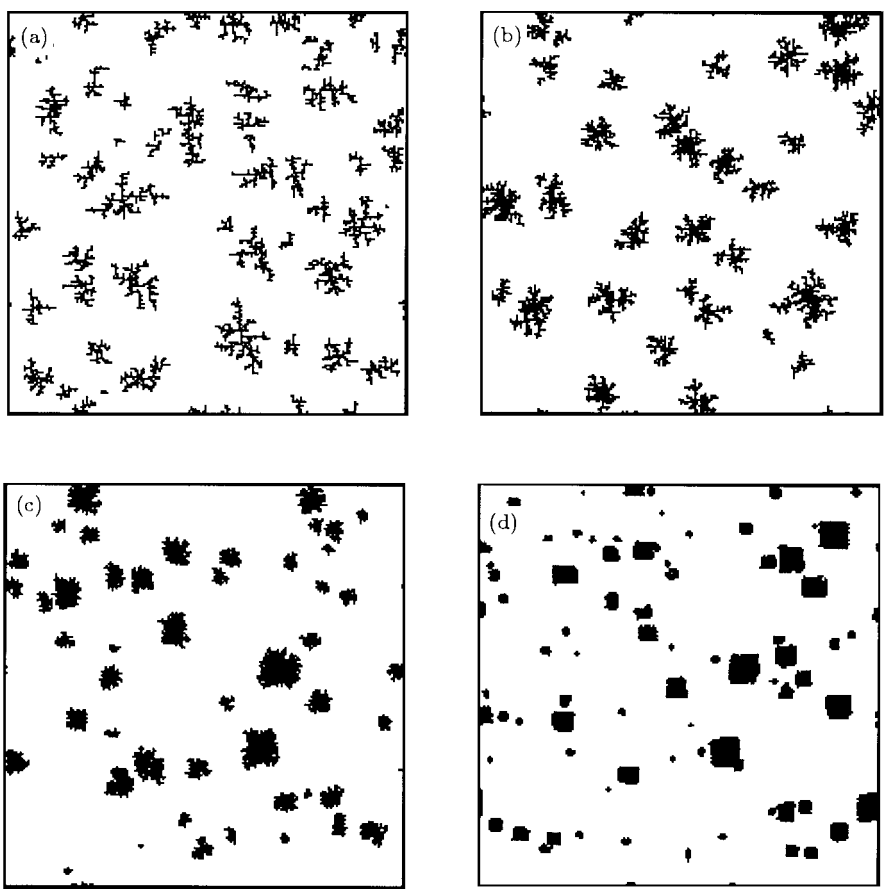


Fig.2. The simulated island shapes for F=0.003ML/s, θ=0.1ML, V_a=0.09eV: (a) T=370K, (b) T=330K, (c) T=310K and (d) T=300K.

Besides the temperature dependence of the island shape, the dependence of 2D island density on temperature is obtained simultaneously in the simulations. The curve of island density versus temperature (see Fig.3) is composed of a descending part and an ascending part, which correspond to the compact islands in the low-temperature region and the fractal islands in the high-temperature region respectively. The result is similar to that of the simulation by Liu *et al.*[13] In the present study, special attention has been paid to the exchange barrier effects at the island edges. From Fig.3 it can be seen that as the island-edge exchange barrier is increased (V_a is decreased from 0.11 to 0.07eV), the island densities in the low-temperature compact regime increase sharply while the variations of the island densities in the high-temperature fractal regime are small. The behaviour of island density on the island-edge barrier is closely related to the

shielding effect. During growth, if the shielding effect is strong, a mobile adatom can hit and diffuse back from stuck atoms many times. This process increases the chance for the adatom to nucleate rather than to incorporate into the island edge. In the case of fractal growth at high temperatures, the shielding effect is very weak; a small increase in the island-edge barrier would not significantly enhance the shielding effect. As a result, the variations of island density are less pronounced. In contrast, for compact growth at low temperatures, the shielding effect is strong, and the increase of island-edge barrier enhances the shielding effect further. This is the extreme case that the island growth is shielded, resulting in the sharp increase of the island density. This result that a higher island-edge exchange barrier is more favourable for the nucleation of the compact islands provides a reasonable explanation for the observed high density of small compact islands in surfactant-mediated Ge/Si heteroepitaxy [5,11,12] or other surfactant-mediated epitaxy (SME) systems.[15,16]

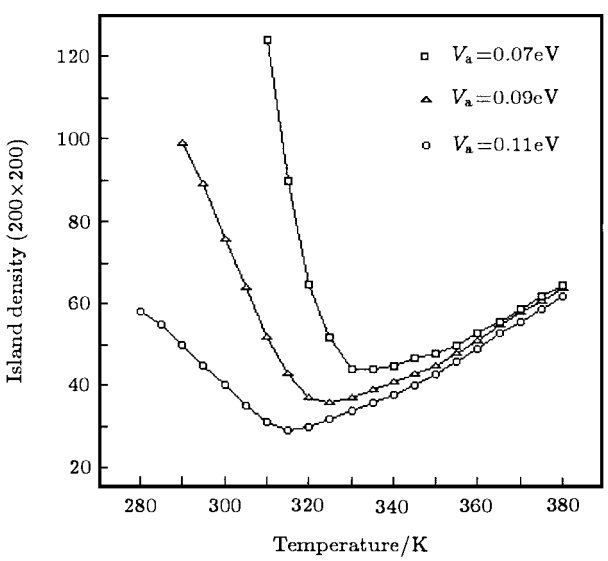


Fig.3. Dependence of the island density on temperature for three values of V_a at F=0.003ML/s and θ=0.1ML.

In order to characterize quantitatively these multiple fractal and compact island patterns, we introduce n_b defined as the average in-plane coordination number of B-type atoms in the pattern as a measure of the multiple island pattern. This quantity is plotted in Fig.4 as a function of temperature for three different values of V_a. At low temperatures, the islands are compact and n_b is larger than 3, but the compact island pattern grown with a higher island-edge barrier is associated with a relatively smaller n_b because of the higher island density. On the other hand, at high temperatures the data from three different values of V_a approach a single curve corresponding to the fractal island pattern. Figure 4 clearly shows the transition of pattern structures from compact to fractal with increasing temperature.

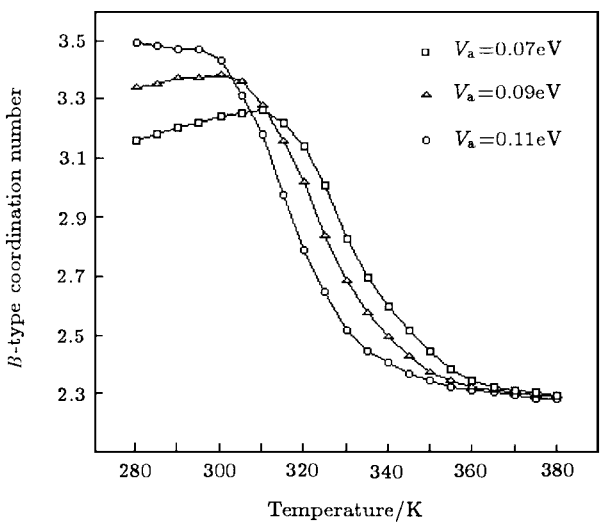


Fig.4. Average coordination number of B-type atoms as a function of temperature for three values of V_a at F=0.003ML/s and θ=0.1ML.

Dependence of the island shape on flux at 0.1ML coverage for T=320K and V_a=0.09eV is shown in Fig.5. The island shape transition from fractal to compact is also related to the shielding effect. When the flux is low (0.0001 ML/s), an adatom stuck at an island edge has enough time to be incorporated into the island before the arrival of one additional atom. Therefore the shielding effect is weak, and the deposition forms a fractal island (Fig.5(a)). As the flux is increased, the shielding effect is enhanced and the growing islands become more and more compact (Figs.5(b) and 5(c)). When the flux is high enough (0.02 ML/s), the growth leads eventually to regular compact islands (Fig.5(d)).

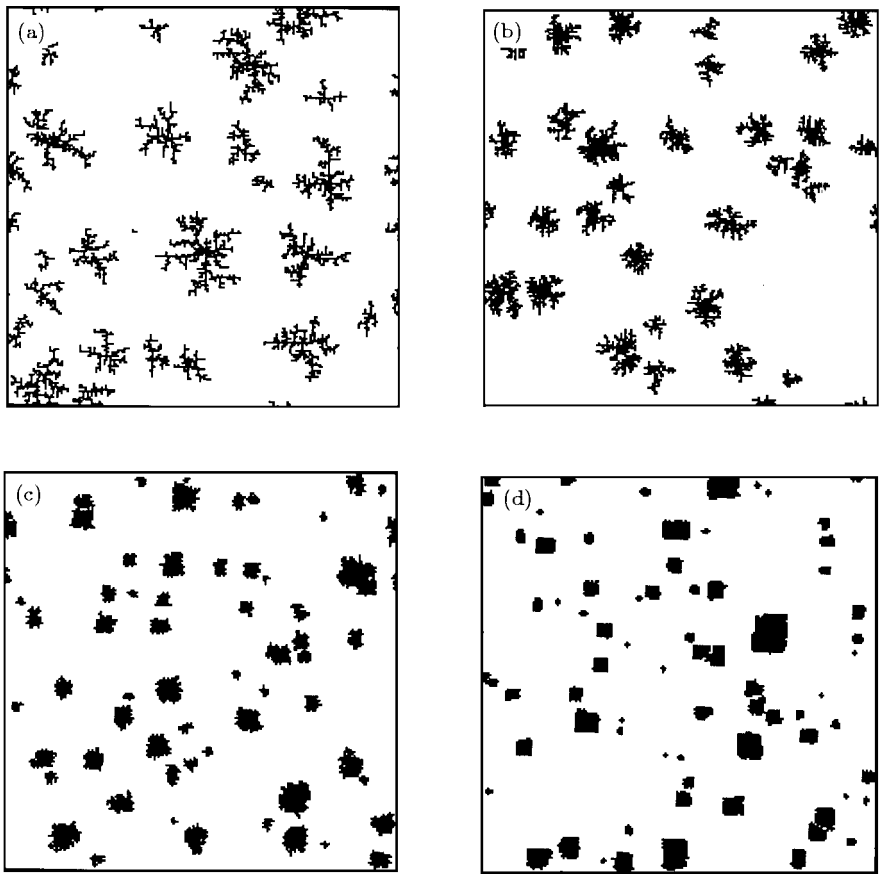


Fig.5. The evolution of the island shape as a function of flux at T=320K, θ=0.1ML, V_a=0.09eV, for (a) F=0.0001ML/s, (b) F=0.002ML/s, (c) F=0.01ML/s and (d) F=0.02ML/s.

Values of island densities versus flux are shown in the logarithmic scale in Fig.6 for three different values of V_a. The plot indicates the existence of three characteristic regimes: the island density is first nearly constant for low fluxes, then increases rapidly with increasing flux, and finally saturates when the flux is high. It can be noticed that a small change of island-edge barrier induces large variations of the island density in high-flux compact regime, while it shows little effect on the island density in low-flux fractal regime. In the intermediate-flux regime, the island densities show a scaling-law dependence on flux, $N \sim F^{\beta}$; the scaling exponent β can be obtained from the slopes of the lines. With the increasing island-edge barrier, the scaling exponent β changes its value from 0.35 for V_a=0.11eV, to 0.51 for V_a=0.09eV, and then to 0.65 for V_a=0.07eV. The trend that β increases as the island-edge barrier increases is consistent with the qualitative expectation although the exponents obtained are relatively small as compared with that found in experiments.[11,12]

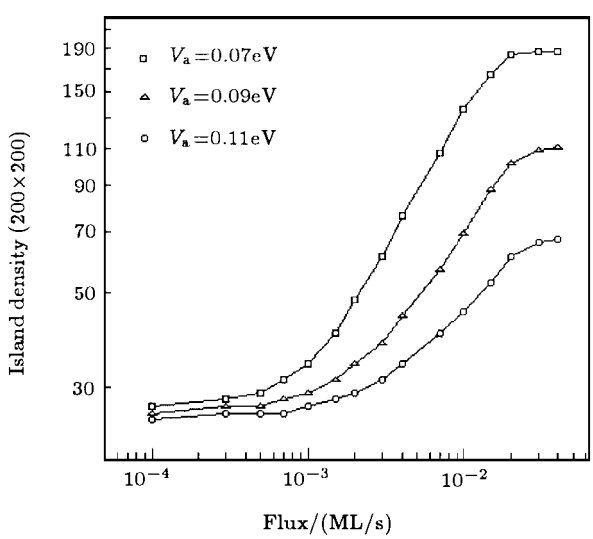


Fig.6. Dependence of the island density on flux for three values of V_a at T=320K and θ=0.1ML.

IV. DISCUSSION AND CONCLUSION

A series of surface morphologies of 2D islands in surfactant-mediated epitaxial growth ranging from

fractal structures to regular compact shapes have been generated by using a simple model including only three kinetic parameters. The obtained dependence of the island shape on temperature and flux is quite different from the case of the aggregation of metal atoms on metal surface.[17−20] In many metal-on-metal epitaxy systems, the observed island structures have traditionally been attributed to diffusion-limited aggregation and the edge diffusion:[21−23] at low temperature, atoms stick to the island edge where they first attach, and a fractal island is formed; as the temperature is increased, the activated edge diffusion makes the islands more compact; at high temperatures, the atom reaching an island remains mobile along island edge until it finds a relatively stable kink site, generating regular compact islands. In our model, the nucleation and growth of islands are controlled by the kinetic exchange process, and the shape of islands depends on the exchange rate at the island edge to a great extent. At high temperatures, the high island-edge exchange rate leads to fractal islands. With lowering temperature, the decreasing island-edge exchange rate induces a transition towards compact islands. If the temperature is low enough, at an island edge, except the kink sites, the exchange rate is very low. The regular compact growth at low temperatures is a direct result of the relatively low barriers (corresponding to a higher exchange rate) for place exchange at kinks. In a previous model, Liu and co-workers[13,14] used a constant barrier for the island-edge exchange. Although the effect of n dead B-type second nearest neighbours on the exchange process was considered, n enters the rate equation only linearly. Therefore the difference in exchange rate at various island-edge sites is less pronounced, and the regular compact islands at low temperatures or at high fluxes cannot be obtained.

The work described in this paper has been carried out, for convenience, using a cubic lattice. However, it is expected that similar results would be obtained using other lattices such as the hexagonal one.

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