Influence of island mobility on island size distributions in surface growth

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(Received 15 November 1995)

A computer-simulation study shows that mobility of islands (or clusters) during growth has strong effects on the statistics of layer growth. The exponent of the power-law relation between the maximum island density and the normalized flux changes as the degree of island mobility is increased. With increasing island mobility the island size distribution splits in an increased density of small islands and a peak that progressively shifts to larger sizes.

It is well known that the mobility of different species on the surface plays a crucial role in nucleation and growth.[1,2] For some years now, it has been known that small clusters or islands (containing less than 20 atoms) on a surface can be mobile and that their mobility decreases with size.[3–7] Recently, however, scanning tunneling microscope (STM) measurements have clearly shown that not only small groups of atoms, but also large islands, containing up to several hundred atoms can be mobile. Experiments on Ag(100) have shown that large Ag islands can diffuse with an unexpectedly large mobility.[8] Furthermore, it has also been shown that in heterogeneous systems large clusters, whether grown on the surface or deposited from a cluster beam source, can be extremely mobile.[9,10] Traditionally, however, growth theories assumed that islands on the surface were immobile.[1,11] In these models the size of the so-called ''critical nucleus,'' which is the maximum size of island on the surface that is unstable, was all-important. This size, together with the ratio (D/F) of the hopping rate D of the atoms on the surface and the flux F of atoms impinging on the surface, then determined all the statistical properties of the film.

More recent models have included more details of the processes occurring during growth, such as edge diffusion[12] and adatom exchange with the substrate.[13] Indeed, just this year Ratsch *et al.* have abandoned the concept of a critical nucleus completely.[14] However, in these models the islands are still considered to be immobile. The question now arises as to whether the mobility of nucleated islands affects the growth statistics and, if so, in what fashion. Villain *et al.* have derived the effect of dimer diffusion during growth on the power-law relation between the island density and *(D/F)*.[15] Recent simulations including the mobility of clusters[16] (or islands) or dimers[17] (with or without dimer dissociation) confirm the predictions by Villain *et al.* Furthermore, Jensen *et al.* have reported that cluster mobility during growth can lead to a wide variety of fractal structures on the surface.[16]

In this paper we present the results of computer simulations that show the extent to which island (cluster) mobility affects the growth statistics. We find that the magnitude of the power-law exponent χ that relates the island density to *D/F* increases as the island mobility is increased. Moreover, the scaled size distributions of the islands show a dramatic change as a function of island mobility. We observe that, as the island mobility is increased, the size distribution splits: the original maximum sharpens up and shifts to larger sizes while the density of small islands increases dramatically and a minimum develops for intermediate sizes.

In the computer model atoms are randomly deposited with a constant flux F on a lattice with square symmetry. Subsequently, all the atoms and islands, chosen at random, can move on the lattice. The chance that an island moves depends on its size. The mobility as a function of size is chosen to be $D_N = D_0(\xi)^N$, where N ($N \geqslant 1$) is the number of atoms in the island, D_0 is the hopping rate for a single atom, and ξ is a number between 0 and 1. Thus, for increasing island size the island mobility decreases and for increasing ξ the overall mobility of all islands increases. For simplicity we will use $\xi = 0$ to refer to growth with immobile islands. When a *single* atom is selected to attempt a move, it will always do so by one lattice unit. Thus the chance that an island moves is $(\xi)^N$. The ratio D_0/F is therefore determined by the average amount of hops per atom in the time between consecutive deposits. In our simulations D_0/F takes the role of *D/F* in growth without mobile islands. When a move results in two particles (atoms or islands) touching, they will stick. In the model, desorption of atoms and/or particles from already formed islands is not allowed. The model also does not include edge diffusion.

It has recently been shown that the presence or absence of interlayer diffusion is strongly related to growth shapes on the surface;[18] smaller fractal-shaped islands increase the amount of interlayer diffusion. Since we do not allow edge diffusion, the growth structures found in the simulations have a strong fractal character.[11,19,20] We have therefore included interlayer diffusion in the simulations: when an atom lands on top of an island, it moves directly to the nearest unoccupied lattice site.

The exponential dependence of the island mobility on size was chosen somewhat arbitrarily. The choice was made despite recent experiments that found a power-law dependence.[8] Different theoretical studies of cluster diffusion have also come up with power-law dependences.[21,22] However, it has also been shown that different size dependences exist. In some cases, for example, the mobility undergoes oscillations as a function of size with larger, albeit still small

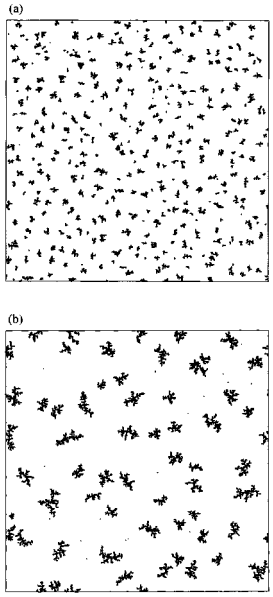

FIG. 1. Two typical snapshots of growth morphologies (area 500×500) at a coverage of 0.4 ML for growth with immobile islands (a); and for growth with mobile islands (ξ=0.8). (b) In both cases $D/F=1.6\times10^7$. It is clear that the increased overall mobility leads to a large reduction in island density.

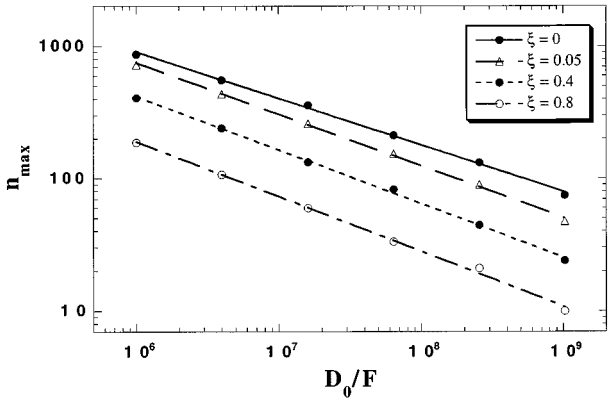

FIG. 2. Double logarithmic representation of the maximum island density vs D_0/F for different values of ξ. The lines are power-law fits to the data. All the data exhibit a clear power-law behavior of which the exponent χ becomes smaller as the island mobility is increased.

($N<20$), islands sometimes being more mobile than small islands.[23] The exponential size dependence used in this study is chosen to focus on the influence of island mobility on growth statistics in general, rather than to make claims about particular systems. However, through some preliminary simulations using a power-law dependence, we have verified that the trends observed in the shape of the island size distributions (see below) are general and independent of the precise functional form of D_N. The quantitative details of the simulation results do vary for different functional forms of D_N.

Figure 1 shows two examples of typical growth morphologies obtained at a coverage of 0.4 ML for $D_0/F=1.6\times10^7$ (for a 500×500 lattice). In the case of no island mobility [Fig. 1(a)] the density of islands is a factor of 5 larger than in the case with island mobility, ξ=0.8 [Fig. 1(b)]. Both figures show the typical dendritic structures expected for growth without edge diffusion. The increased mobility on the surface in the case of mobile islands decreases the nucleation probability and therefore results in a reduced island density.

The exponential dependence of D_N means that large islands are virtually immobile. As a consequence the island density as a function of coverage remains constant after the maximum island density has been attained. We found that for a coverage of 0.4 ML all the different simulations had reached and were maintaining their maximum island density. Figure 2 shows a double logarithmic representation of the maximum island density as a function of D_0/F for several values of ξ. For all values of ξ the island density shows a clear power-law dependence on D_0/F. With increasing island mobility the exponent of the power law, χ, changes from a value of -0.35 for ξ=0 to -0.41 for ξ=0.8. These values should be compared to theoretically derived values for χ of $-\frac{1}{3}$ for growth with immobile islands and a critical nucleus size of 1,[24] $-\frac{1}{4}$ for growth with anisotropic surface diffusion without mobile islands,[24] and $-\frac{2}{5}$ for growth with mobile dimers.[15] Recent computer simulations found values of $\chi=-0.35$ for mobile dimers[17] and $\chi=-0.42$ for growth with mobile islands obeying a power-law size dependence of their mobility.[16] We attribute the difference between the value χ of -0.35 obtained for immobile islands and the expected value $(-\frac{1}{3})$ to the interlayer diffusion. The interlayer diffusion has only a minor effect on the rest of the growth statistics. The reason for this is that the interlayer diffusion will cause islands to grow at a rate that is proportional to the product of the flux and their area. This rate should be compared to the rate at which they grow due to surface diffusion that is proportional to D/F, in most of the cases this is orders of magnitude larger.[12,13,24]

It has been proposed that the maximum island density is related to the fractal dimension as

$$n_{\max}\propto F^{2/(4+d_f)}, \tag{1}$$

where d_f is the fractal dimension and $n_{\max}$ is the maximum island density.[25] It is clear that, for the values of the exponent that we have found, this would mean a smooth transition from a d_f of 1.71 to the unphysical value of 0.88.

In conventional growth theory it is well established that size distributions obtained for different coverages and/or different values of D/F can be collapsed onto a single curve when scaled properly.[24] In the binary collision epitaxy model by Bartelt and Evans the shape of the curve will then depend only on the size of the critical nucleus.[24] In this scaling one computes $s_{\rm av}^2 n_s/\theta$ versus $s/s_{\rm av}$, where $s_{\rm av}$ is the average island size, n_s is the density of island of size s and θ is the surface coverage. Figure 3 shows the scaled island size distributions obtained for different values of D_0/F for ξ=0.05. It is clear that the size distributions resulting from growth with mobile islands exhibit the same self-similarity observed

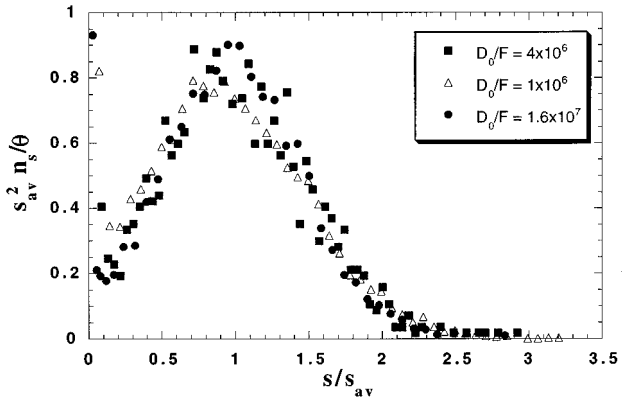

FIG. 3. Scaled size distributions for growth with $\xi=0.05$ for different values of D_0/F. The curves show a strong self-similarity.

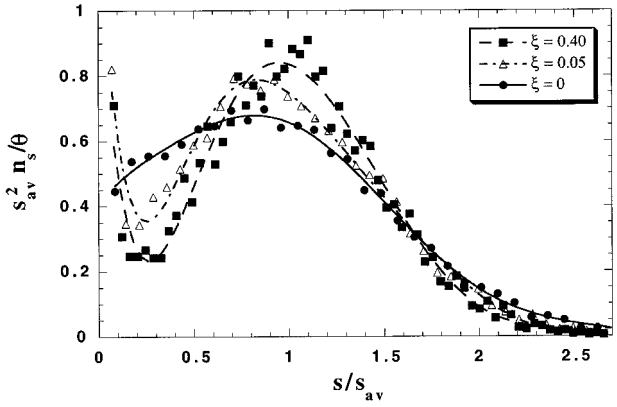

FIG. 4. Scaled size distributions for different values of ξ. The curves are guides to the eye. As the island mobility is increased the distribution splits up into two parts separated by a pronounced minimum.

for growth without island mobility. In order to obtain self-similarity, we have excluded the contribution made by monomers from the calculation of s_{av}.

However, the self-similarity vanishes when comparing scaled size distributions obtained with different island mobilities. Figure 4 shows the scaled island size distributions for different values of ξ. As the island mobility is increased from the case of immobile islands to more mobile islands, the scaled size distribution changes shape and splits into two parts. The first part is the broad maximum obtained without island mobility, which becomes higher and sharper while shifting to larger sizes as the island mobility increases. The second part arises from a dramatic increase in the density of small islands. In between the two branches of the distribution a pronounced minimum develops. The increased density of small islands in the case of mobile islands is a direct result of the increased overall mobility, which reduces the total number of islands; the smaller islands have less time to ''find'' the larger islands and aggregate. Growth with mobile islands therefore appears to lag behind growth without mobile islands at all coverages, despite the fact that in all cases presented here the maximum island density has been reached. We also note that the large increase in density of small islands is similar to the ''shoulder'' in the size distributions observed for growth including atom exchange with the substrate, but the minimum between the two parts of the distribution is much more pronounced in the case we are considering.[13]

The fact that the shape of the size distributions depends on the mobility of the islands should affect the analysis of size distributions in homoepitaxial growth obtained with the STM.[26,27] The sharp distribution at large sizes obtained for deposition of Fe on Fe(001) at high temperatures (compared to a broad distribution at lower temperatures) has been attributed to a critical nucleus size larger than 1.[26] We suggest that the change in peak shape might also be caused by the onset of island mobility at higher temperatures, rather than a change in size of the critical nucleus. This suggestion is corroborated by a recent study by Bartelt *et al.*, which shows the ''sharpening'' effect of dimer diffusion on the island size distribution during epitaxial growth.[28] Their obtained island size distributions do not exhibit the increased density of small islands that we observe. With the different exponents of the power-law relation between maximum island density and *D/F*, it unfortunately becomes hard to extract information about the growth mechanism from such exponents, except in the most obvious cases, such as, for example, an exponent of 0.27, which can only be due to anisotropic diffusion.[27]

Our computer simulations are complementary to recent calculations that first showed the effect of cluster mobility on the scaling between the island density and D_0/F.[16] We have now shown the profound effect that island mobility has on the size distributions. We have also reported how the exponent of scaling gradually changes as the island mobility is increased, making the relation between the fractal dimension and the maximum island density [Eq. (1)] more and more unphysical.

The shape of the scaled island size distributions found for growth with mobile islands (Figs. 3 and 4) is reminiscent of the size distributions derived for vapor condensation in, for example, a cluster source. We speculate that this should not be too surprising, since such condensation is well described by Smoluchowski-type rate equations in which the densities of all the different-sized species in the vapor are coupled to each other.[29] This type of equation also describes growth with mobile islands, albeit in two dimensions rather than in three dimensions.

We have shown, with simple computer simulations, that the introduction of mobile islands (clusters) in thin-film growth has profound effects on the growth statistics. As the mobility of islands is increased the scaling relation between island density and *D/F* changes. In addition, the size distribution splits in an increased density of small islands while the rest of the distribution shifts to larger sizes.

The authors would like to express their gratitude to H. M. van Pinxteren for providing computational facilities. L.K. acknowledges financial support from the EU Human Capital and Mobility Programme (ERBCHICT941736).

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