

Influence of edge diffusion on the growth mode on vicinal surfaces

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Growth on vicinal surfaces typically occurs via nucleation and growth of islands or via step flow. In this letter, we present computational results based on an island dynamics model that employs the level-set technique that study the transition between these two competing mechanisms. Our results demonstrate that the rate of diffusion of atoms along step edges is an additional parameter that controls the competition between these mechanisms. We make suggestions about how one might be able to exploit these results experimentally. © 2005 American Institute of Physics.

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The morphology of the interfaces between different layers in a layered semiconductor structure can crucially affect the electronic or optical properties of the device. It is therefore important to reliably control the morphology of a film during epitaxial growth. Smooth films develop if the film grows layer by layer. This occurs on vicinal (or stepped) surfaces either via step flow, or when nucleation of islands in higher layers is delayed until the previous layer is essentially filled in. The latter is commonly referred to as nucleation and growth, or birth-and-spread mechanism. While both of these mechanisms lead to smooth surfaces, there is a qualitative difference between them: During nucleation and growth, there will always be some islands present on the terraces. It is therefore important to understand and potentially control the transition between these two growth modes.

An experimental quantity that is typically used to characterize the growth mode is the intensity of reflection high energy electron diffraction (RHEED) experiments. The common assumption is that the RHEED signal is correlated to the step edge density,^{1,2} even though there is also some controversy about this assumption.³ Thus, if growth proceeds via step flow, the step edge density is constant, and the RHEED signal remains constant as well. During the nucleation and growth mechanism, on the other hand, the number of islands (and thus the total length of the step edge) is minimal when a layer starts to grow. When the islands cover roughly half of the terraces, the total step length reaches a maximum. Thus, the total step length and accordingly the RHEED signal oscillate, with a period that is essentially correlated to the deposition time of one atomic layer.

The growth parameters that can be used experimentally to control the transition from nucleation and growth to step flow are the deposition rate or the growth temperature. If the temperature increases, the diffusivity of adatoms on the surface increases. As a result, adatoms are more mobile, and are more likely to reach a step edge within a given time. If the time between deposition of atoms is of the order (or larger) than this time, adatoms will typically reach a step edge before another atom is deposited, and thus no nucleation will occur on the surface. Thus, increasing the temperature will move a system from the nucleation-and-growth regime to the step-flow regime. A similar argument applies to the dependence on the growth rate: If one decreases the growth rate,

adatoms have more time to reach a step-edge (before the next atom is deposited). Another parameter that influences the transition between the two growth mode is the terrace width, which is related to the miscut. A smaller terrace width increases the chance for an adatom to reach a step edge, and therefore promotes step flow. But unlike temperature or deposition flux, this is a parameter that cannot be changed during the deposition process.

In this letter, we will discuss another parameter that influences the transition from step-flow growth to growth via nucleation and growth of islands on vicinal surface, and will make some suggestions how one might potentially control this parameter in an experiment. We will show that an inhibition of the rate of diffusion of atoms along a step edge can drive the system toward growth via step flow. The basic explanation is that decreasing edge diffusion makes the edges of the steps less smooth, decreasing the effective average distance to the next step edge, and thus reducing the effective terrace width.

We model growth on a vicinal surface using the level set (LS) approach to epitaxial phenomena.^{4,5} In our implementation of the LS method, islands and step edges are resolved as atomistic in height but continuous in the lateral dimensions. The boundaries of islands and step edges of height $k+1$ are represented by the set of points \mathbf{x} where a level set function $\varphi(\mathbf{x},t)=k$. Adatoms are represented by an adatom density $\rho(\mathbf{x},t)$, which is updated by solving a diffusion equation

$$\frac{\partial \rho}{\partial t} = F + D \nabla^2 \rho - 2 \frac{dN}{dt}. \quad (1)$$

In Eq. (1), F is the deposition flux, D is the surface diffusion constant, and the last term represents the rate of nucleation of new islands on the surface, where N is the island density. The time of nucleation of a new island is chosen deterministically according to this rate. The position of the new island is chosen by properly taking account of the explicit stochastic nature of the nucleation process.⁶ Nucleation is then realized within the model by raising φ by one level at a certain number of points of the numerical grid (corresponding to the size of the new island). For all results presented in this letter, we solve the diffusion equation with the boundary condition for irreversible aggregation, $\rho(\mathbf{x},t)=0$ for all points along the boundary. We note that other boundary conditions that represent detachment of atoms from the step edge can also be

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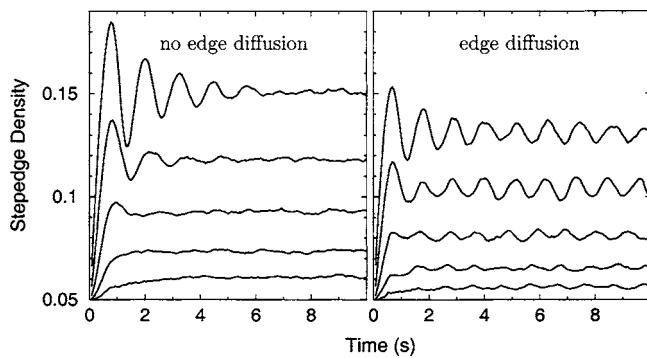


FIG. 1. Step edge density for different values of surface diffusion D without edge diffusion (left panel), and with edge diffusion (right panel). The values for D are 10^5 (top), $10^{5.5}$, 10^6 , $10^{6.5}$, and 10^7 (bottom). The magnitude for the step edge density decreases as D increases. The units are per (lattice constant)².

implemented in this model.⁷ However, the main conclusion of this study will be the same if detachment is included.

The level set function evolves according to

$$\frac{\partial \varphi}{\partial t} + v_n |\nabla \varphi| = 0. \quad (2)$$

The normal velocity v_n is computed as

$$v_n = a^2 D \left(\left. \frac{\partial \rho}{\partial n} \right|_- - \left. \frac{\partial \rho}{\partial n} \right|_+ \right) + D_{\text{edge}} (\kappa - \kappa_{\text{av}}), \quad (3)$$

where a is the lattice constant, n is the island boundary normal, and the two terms in the first parenthesis represent the flux of adatoms toward the step edge from the terrace above (+) and below (-) the step edge. The last term represents edge diffusion, where D_{edge} is related to the microscopic rate for edge diffusion, κ is the curvature of the step, and κ_{av} is the average curvature. We note that we choose a dependence on $(\kappa - \kappa_{\text{av}})$ instead of a dependence on the second derivative κ_{ss} because of numerical efficiency. The expression we chose is conservative, and tests revealed that during growth it captures the main effects of edge diffusion.

We start the discussion of our results by focusing on the transition from nucleation and growth to step flow in the absence of edge diffusion. For all results shown, we chose a system size of (100×100) , with five steps on the terrace (so that the average terrace width is 20), with periodic boundary condition. The deposition flux F is set to 1 ML s^{-1} , so that the remaining variable is D . All results presented would be similar (just shifted) for different values of terrace width and deposition rate. The left panel in Fig. 1 shows the time evolution of the step edge densities for different values of D without edge diffusion. Clearly, there are several oscillations present for $D=10^5$ (indicating nucleation and growth), while there are no oscillations at $D=10^7$ (indicating step flow). One might argue that the transition occurs somewhere around $D=10^{5.5}$ or $D=10^6$, where we see only one maximum in the step edge density.

We now examine the resulting step edge densities for simulations that are essentially the same, with the only exception that we also allow for diffusion along the step edge. This is shown in the right panel of Fig. 1. For all results, we choose a value of $D_{\text{edge}}=50$, but the results are qualitatively the same for higher or lower values of D_{edge} . The main observation is that the transition now occurs at much higher

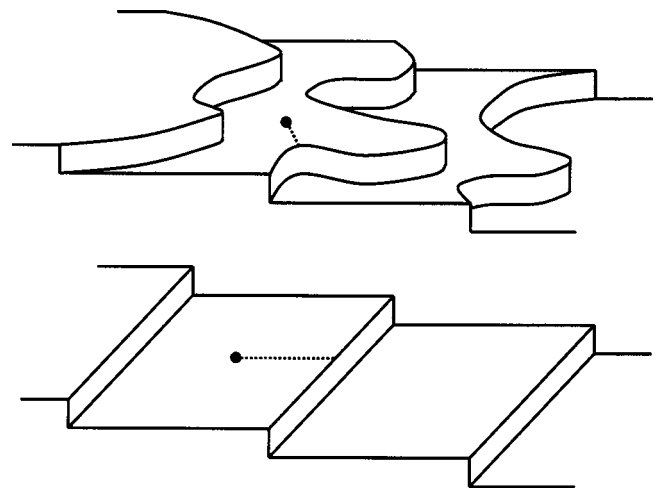


FIG. 2. Schematic representation of the increased effective terrace width l_{max} (dashed line) in the presence of edge diffusion. Without edge diffusion, the steps meander, and the longest distance from any point on the terrace to the next step edge is rather short (top panel). In the presence of edge diffusion, this length increases (bottom panel).

values of D : For $D=10^{5.5}$, we see many oscillations. Even for $D=10^6$, we can clearly identify an oscillating step edge density all the way to a time of 10 s. Only at $D=10^7$ do oscillations (almost) disappear. Thus, we conclude that increasing the rate of edge diffusion can prolong the regime of nucleation and growth (as D increases). Alternatively, one can say that the step-flow regime is increased if one is able to inhibit edge diffusion.

At first glance, this result might be somewhat surprising: The effect of edge diffusion is to smoothen the edge of the steps. But as a consequence, islands are more likely to nucleate on the terraces (moving the system away from step flow and toward nucleation and growth), which can be interpreted as an increase of the roughness. Thus, a smoothening mechanism leads to increased roughness. The explanation for this behavior is the following: As edge diffusion increases, the edges of the steps become more straight. This is illustrated in Fig. 2. As a consequence, the longest distance from any point on the surface to the next step edge increases. In an idealized scenario, it is one-half of the terrace width for perfectly straight steps, but (significantly) less for meandering steps. As explained above, this increase of the effective terrace width leads to more nucleation on the surface, and thus to an increase of the step-edge density.

The results reported here for vicinal surfaces can be compared to similar results that have been found for singular surfaces: Kalif–Michely–Comsa kinetic Monte Carlo (KMC) simulations have shown that enhanced edge diffusion for growth on singular surfaces leads to a faster roughening of the growing surface. It was argued that the underlying mechanism for this roughening might be an uphill current that is due to an asymmetry at corners along the step edge.^{8–10} But we have shown in earlier work that an alternative explanation that is similar to the results presented in this letter might be the more dominant effect that leads to roughening on singular surfaces.^{5,10} Faster edge diffusion leads to more compact island shapes, and as a result the residence time of an atom on top of compact islands is extended. This promotes nucleation at earlier times on top of higher layers, and thus enhances roughening.

Our results suggest that edge diffusion is an additional parameter that might be used to control the growth mode during an experiment for growth on a vicinal surface. In particular, we suggest several possibilities that might exploit the mechanism suggested in this letter to control the growth mode. The first suggestion is to introduce a surfactant. The requirement for this surfactant is that it has essentially no effect on diffusion on the surface. Rather, we suggest a surfactant that only acts on the step edge, suppressing (or enhancing) edge diffusion. The results of Kalff *et al.*¹¹ for the homoepitaxial growth of Pt/Pt(111) suggest that this is quite possible: They have shown that small impurity amounts of CO can change the energetics of different step types, thus changing the relative edge diffusion rates for the different step types. It is therefore quite plausible that such impurities, that are introduced deliberately and bind predominantly at steps, can act as surfactants to change the diffusivity along steps.

A second suggestion is particularly amenable to the growth of compound semiconductors, such as III/V semiconductors. It is well known^{12,13} that the surfaces of such systems reconstruct, and that the ratio of the group III to the group V species is a key factor in determining the surface reconstruction. Different surface reconstructions will lead to different microscopic structures along the step edge. In particular, it is well established that on (001) surfaces, two types of steps exist: III-terminated and V-terminated steps. For GaAs(001) under normal As-rich conditions [when a (2 × 4) reconstruction is observed], the Ga-terminated (A-type) steps are relatively straight, while the As-terminated (B-type) steps are more ragged.¹⁴ It is highly likely that the rates for edge diffusion are different along these different steps. We therefore suggest that a change of the III/V ratio might be used to change the growth mode on vicinal surfaces. Such a scenario has already been suggested in RHEED studies by Owen *et al.*,¹⁵ where it was also demonstrated that a change in the III/V ratio indeed influences the rate of decay of the RHEED oscillations. The mechanism of changing edge diffusion that is suggested here is in addition to the change of

growth modes due to different surface mobilities, which can also be achieved upon changing the III/V ratio.

One might also be able to exploit the properties of different material systems that are often used for multilayered structures to influence the growth mode. Recent results suggest that under essentially identical growth conditions GaAs grown on InP(100) exhibits much rougher step edges than AlAs grown on InP(100).¹⁶ These systems have essentially the same lattice mismatch. This suggests that diffusivity along the step might be slower for GaAs/InP(100), and that one might be able to prolong the step-flow regime for this system.

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