Roughening of Steps during Homoepitaxial Growth on Si(001)

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The two-dimensional kinetic roughening of steps on vicinal Si(001) has been analyzed with scanning tunneling microscopy for growth at different rates, doses, and temperatures. Growth exponents for the evolution of the step roughness are extracted. They suggest extremely ineffective relaxation mechanisms for the rough step.

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In the growth of thin films, a number of microscopic kinetic mechanisms compete to produce a film that has desirable electrical, optical, magnetic, or mechanical properties. A signature of the competition between these mechanisms is the evolving growth front morphology. For this reason, considerable effort, both theoretical and experimental, has recently been expended on understanding the dynamics of interface evolution. On the theoretical side, much of the interest revolves around discovering universality relationships in the kinetic roughening of thin films [1]. The technological driving force is to produce films that have a specified surface or interface morphology. The experimental effort attempts to bridge these two goals, on the one hand investigating conditions to create a particular morphology [2], on the other hand studying if universality relationships can in fact be developed for film growth.

The mechanisms that control the evolution of the growth front morphology include diffusion of adatoms on a terrace, diffusion over steps, and the sticking of atoms at steps or existing structures. The competition between them determines the morphology. A measure of this growth front roughness is the "interface width," the part of the growing film that consists of incomplete layers. This interface width is expected to increase in a systematic way as material is added [3]. In the simplest case, epitaxial growth, a set of conditions desired to produce the smoothest films (high temperature, low deposition rate, or small distance between steps) is "step flow." The flow of a step across the surface is the 1D analog of the evolution of the 2D growth front, with analogous mechanisms being diffusion along the step, diffusion over kinks, and sticking at kinks. In addition, however, there is a new feature: For a surface, flux can arrive only from one direction, the gas phase; for a step it can arrive from the terraces both above and below the step. In the same manner as for a surface, we can speak of kinetic roughening of a step: It can develop a morphology that reflects the competition between various kinetic factors. In this Letter, we describe scanning tunneling microscopy (STM) measurements of the evolution of the roughness of steps during Si growth on vicinal Si(001). We determine the growth laws for the evolution of this roughness. The exponents in these growth laws are surprising in that they imply that the stability of the morphological features that form in the step is greater than what we would expect from independent measurements of the mobility of adatoms at the step, suggesting that relaxation mechanisms for the step are extremely ineffective and contrary to simple expectations.

The Si(001) surface reconstructs to form dimers in order to reduce the free energy of the system. Because of the crystal structure, the dimerization direction rotates by 90° on alternating levels of the surface, resulting in two types of steps: S_A steps in which the dimer rows on the upper level run parallel to the step, and S_B steps in which the dimer rows on the upper level run perpendicular to the step. Such steps are "pure" on surfaces miscut toward [110]. For surfaces miscut towards [100], steps are composed of equal amounts of S_A and S_B segments, as the nominal step direction runs at 45° with respect to the dimerization direction. Self-diffusion on Si(001) is anisotropic, with motion parallel to the dimer rows about 3 orders of magnitude larger than that perpendicular to the dimer rows [4]. Adatoms are also much more likely to stick to the end of a dimer row (S_B step) than to the side (S_A step) [4]. The probability of crossing a step from one terrace to another has also been found to be quite different for S_A and S_B steps. Atoms approaching an S_A step from either above or below are reflected with high probability, while atoms arriving at an S_B step from the top cross over and have a high probability of being incorporated at the S_B edge [4].

We have made measurements of the evolution of [100] oriented steps on vicinal Si(001) with various miscuts. Samples were cleaned and annealed in the usual manner [5] to produce an equilibrium step structure [Fig. 1(a)]. Si was subsequently deposited at rates between 0.003 and 0.020 ML/sec (ML denotes monolayers). At these deposition rates and at temperatures above 500 °C the equilibrium step distribution shown in Fig. 1(a) replicates itself. If the temperature is lowered only slightly (or the deposition rate increased), the edge morphology undergoes drastic changes, forming dendriticlike fingers that are dependent upon time (dose), as shown in Fig. 1(b) and in Fig. 2. We have measured the time evolution of this fingering at different deposition rates and substrate temperatures. Fingers begin to grow out from the step and

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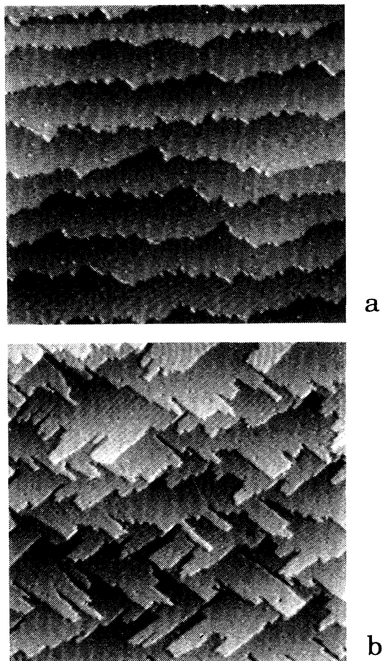

FIG. 1. STM images of Si(001) miscut 0.3° toward [100]. 3000×3000 Å images of the surface (a) at equilibrium and (b) after 500 sec. Deposition at 0.02 ML/sec at a substrate temperature of 350 °C. The mean terrace width in both images is ~300 Å.

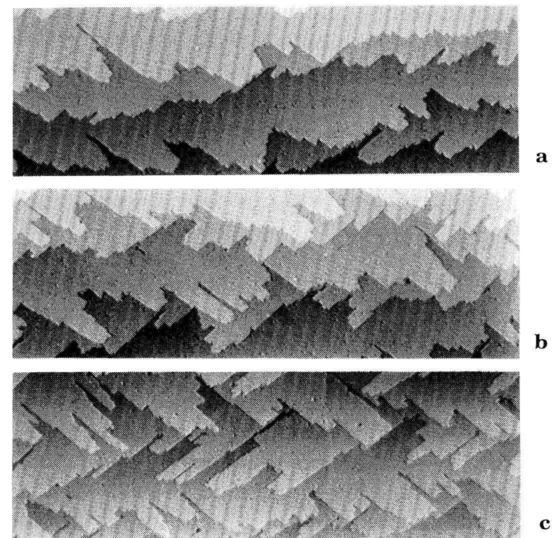

FIG. 2. 1000×3000 Å STM images of the Si(001) surface, miscut toward [100], after growth at 350 °C and 0.02 ML/sec for (a) 100, (b) 300, and (c) 500 sec.

become more prominent with continued deposition. For higher temperatures the onset of fingering is delayed to higher doses. Figure 2 illustrates the evolution of roughness with dose at a fixed temperature and deposition rate.

The origin of the fingering is not simply analogous to the roughness buildup that occurs in random deposition of atoms under a set of kinetic limitations [6–8]. Some instability in transport [9] across or sticking at the step, leading to positive feedback, is required to cause the formation of fingers. Simple one-dimensional examples of positive feedback have been described [10]. In these the barriers for "up" and "down" transport across steps are unbalanced. This concept has been extended to two dimensions [11] in a model in which an anisotropy in adatom attachment from above and below causes unstable growth in the form of protrusions extending out from the growth front.

In the present situation the cause of the fingering is more complex. A combination of anisotropic diffusion and the presence of segments of two types of steps (S_A and S_B) in each nominal [100] step leads to enhanced instability of the growth front and the creation of fingers. Because of the approximately 1000:1 anisotropy in diffusion [4], an adatom that lands on a particular dimer row will diffuse almost exclusively along the length of that row. These adatoms will eventually arrive at one of the two ends of the row, which are the steps. The down-step end of a row is always an S_B step; the up-step end, conversely, terminates at an S_A step (except for the few rows that align with the end of a row of the upper terrace, which terminate at an S_A/S_B kink). Therefore atoms falling on the terraces primarily approach the S_A step from the terrace below and the S_B step from the terrace above. An adatom arriving at S_B falls over the step and is incorporated into the step [4,9,10], while atoms arriving at S_A are preferentially reflected and will eventually diffuse to the down step. Thus adatoms deposited on a particular dimer row act to extend that row at its down-step end. The longer a row is the more it is able to collect deposited atoms. Fluctuations in the terrace size, either entropic [12] or strain induced [13–16], result in a distribution of dimer row lengths. These fluctuations, as well as local fluctuations in the flux, cause some rows to grow more quickly, and thus to collect even more atoms. This positive feedback causes the creation of fingers. It will exist for all temperatures for which the diffusion length is sufficient to prevent island formation.

Runaway growth of the fingers is prevented in our case by the next down step of the staircase. The most obvious manifestation is the fact that steps cannot cross; however, narrower confinement of the step can result as a consequence of entropic [12] and strain [13–16] mechanisms that limit the equilibrium meandering of steps. In our case, the equilibrium step confines its meandering to approximately 10% of the available terrace area, as shown in Fig. 1(a). At dimensions equal to some fraction of the mean terrace width, the growth of fingers will begin to slow because of this confinement.

Theoretical work on the evolution of surface roughness

has been focused on the concept of dynamical scaling. It is predicted and has been observed [1] that growth can produce a self-affine surface that consequently is morphologically equivalent over a range of length scales. Self-affine growth results in simple, power-law scaling relationships for the surface roughness as a function of both lateral length scale and time,

$$W \propto t^{\beta}, \quad W \propto x^{\alpha}, \tag{1}$$

where W is the rms step roughness, t is deposition time at a given deposition rate (hence the dose), x is lateral distance along the interface, and α and β are the respective growth exponents.

In order to characterize quantitatively the evolving step morphology we have determined "height-height" correlation functions of the step roughness for different doses and step lengths. The correlation function is found by first performing a least-squares fit to the whole length of step we have imaged (typically 8500 Å) to provide the mean step position, and then measuring the height deviations along the dimer row direction relative to this mean step. The rms step roughness is the square root of the value of the correlation function at zero separation. In Fig. 3 we show the evolution of the roughness with deposition time. To determine the behavior of roughness with window size, we choose a window dimension (measured in the direction of the total mean step), make a dc adjustment to the line defining the mean step to place it at the average value for the step segment within the window, measure the height of each row of dimers in the segment relative to this average value, calculate the interface width from the zero crossing of the correlation function, displace the window to a nonoverlapping position, and repeat [17].

Finally all the values of the interface width for a given window size are averaged. The results are shown in Fig. 4 for several doses. The roughness increases with window size but slows as the window is widened sufficiently and the influence of adjacent steps becomes important. We obtain values of $\beta = 0.29 \pm 0.08$ (Fig. 3) and $\alpha = 0.45 \pm 0.1$ (Fig. 4).

In kinetic roughening, growth and relaxation processes compete. For the evolution of step morphology here, "growth" is determined by the arrival rate of the atoms at the step, through deposition rate, diffusion, and step crossing. "Relaxation" is determined by kinetic limitations for atoms to find equilibrium positions. Processes include desorption from the step, diffusion along the step, and crossing of kinks. The driving force for the smoothening of the step in the simplest picture is the reduction in line length and kink density. If relaxation processes dominate, the step will maintain its equilibrium configuration during growth. For very weak or zero relaxation processes, the limit of hit-and-stick growth (ballistic aggregation) is realized. Surprisingly, the exponents we obtain are in good agreement with the values of $\alpha = \frac{1}{2}$ and $\beta = \frac{1}{3}$ predicted by the ballistic aggregation model in $d = 1 + 1$ dimensions [18], which neglects growth front relaxation. More complicated models [19] that attempt to describe molecular beam epitaxy better by including activated diffusion along the 1D surface (step) predict values of 1 or greater for α, in considerable disagreement with our results. We conclude that the relaxation mechanisms for the rough steps are extremely ineffective.

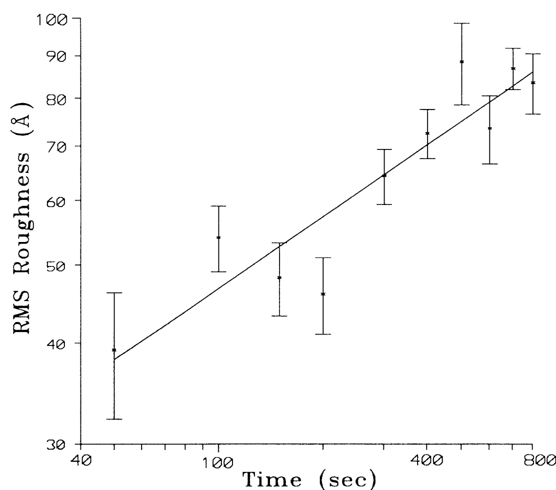

FIG. 3. Measured rms roughness as a function of deposition time for surfaces grown at 350 °C and 0.02 ML/sec. A window size of 100 Å is used. The slope of the line, β, is 0.29 ± 0.08.

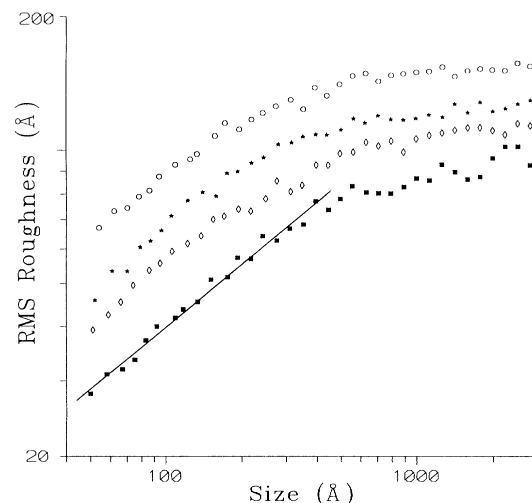

FIG. 4. Measured rms roughness as a function of window size for surfaces grown at 350 °C and 0.02 ML/sec for 50 sec (■), 100 sec (◇), 300 sec (★), and 700 sec (○). The average slope of the initial region (150–300 Å) of each curve is 0.45 ± 0.1; the solid line shows this slope. The turning-over point moves to lower values of x for greater rms roughness.

Recent real-time STM experiments [20] of the equilibrium fluctuations in S_B local step morphology at temperatures of $\sim 250\,^{\circ}\mathrm{C}$ allow an estimation of the rate at which a morphological feature should anneal away. Extrapolated to our growth temperatures ($\sim 350\,^{\circ}\mathrm{C}$), fluctuations of the local step morphology are almost an order of magnitude larger than the mean arrival rate of adatoms at the step in the growth experiment. With the assumption that this local picture can be applied to the whole finger, one would expect fluctuations produced by the growth to anneal out as quickly as they form. The continued existence of fingers under the present circumstances (once they have formed) suggests that there is a stabilizing mechanism, an effective trap for atoms at the ends of fingers (S_B steps). For example, an energetic barrier may exist for atoms to turn the corner of a finger or a strain potential similar to that responsible for domain structure on vicinal Si(001) [13-16,21,22] may act as an effective trap. A trap for atoms at the end of fingers reduces the influence of diffusion in edge relaxation, and may explain in part why the growth exponents we measure are more similar to ballistic-aggregation models than to models that include activated diffusion. Experiments are in progress to investigate the relaxation, after deposition has been halted, of rough steps produced by growth to understand the decay rates.

In summary, we analyze the growth morphology of dendritelike fingers that form at steps on vicinal Si(001) during Si epitaxy at temperatures between those for island formation and for step-flow growth. The evolution of edge roughness during growth suggests dynamical scaling in the stages before steps begin to interfere with each other. The values of the exponents correspond to those of a ballistic-aggregation model and imply that relaxation mechanisms of the step are extremely ineffective and that an effective trap exists for atoms at the ends of fingers, hindering the motion of atoms from the "hills" to the "valleys" in the step. We speculate on possible mechanisms for the stability of fingers in view of other evidence suggesting that they should decay more rapidly.

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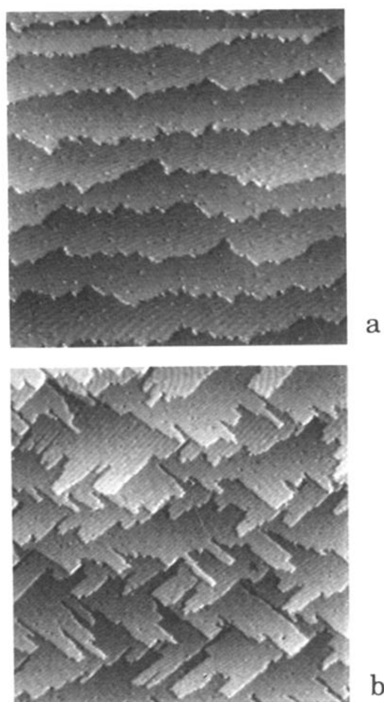

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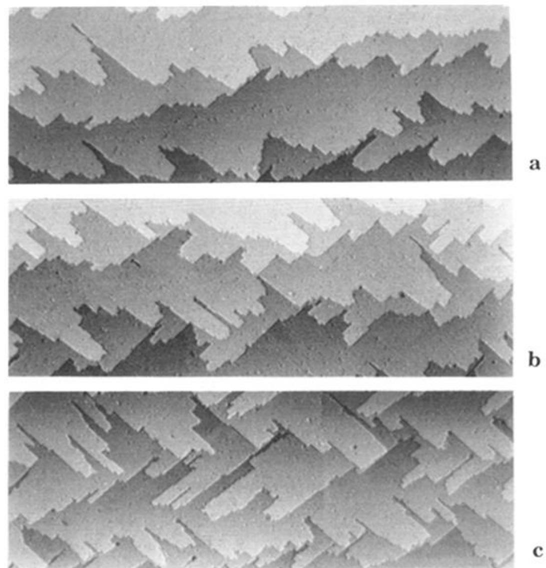

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