Unstable Growth on Rough Surfaces

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We present experimental data for the morphological evolution of InAs buffer layers which are interpreted using continuum equations of motion and kinetic Monte Carlo simulations. Our analysis reveals the presence of an instability even as an initially rough surface smooths during growth. This instability is due to the step-edge barrier and causes a characteristic length to emerge while the surface roughness is decreasing, well before the formation of the mounds. The smoothing is well described by a linear continuum equation identical to that which describes the early stages of spinodal decomposition and has important practical implications for the growth of device buffer layers. [S0031-9007(98)07810-7]

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The formation of large-scale morphologies during epitaxial growth has received considerable attention from both the experimental and theoretical communities in recent years. Moundlike structures have been observed in semiconductor [1–3], metal [4,5], and other [6] systems under a variety of growth conditions. The observed structures range in height from tens to hundreds of angstroms and, in lateral extent, up to several microns. Most theoretical treatments of unstable epitaxial growth have assumed that the initial surface is smooth and that mound formation is evident from the onset of growth [7–12]. In particular, there have been studies that have yielded analytical results on several aspects of mound coarsening [10,12].

The experimental situation is not at all like this, however, especially during the growth of actual device materials. Semiconductor substrates, for example, are quite rough after oxide desorption and, in fact, become smoother during the growth of buffer layers. This important regime has received no theoretical treatment in spite of the fact that it clearly influences the initial stages of mound formation and leads to obvious questions concerning the mechanisms that are active during the smoothing of initially rough surfaces.

In this Letter, we present results from continuum equations of motion, kinetic Monte Carlo (KMC) simulations, and the growth of InAs buffer layers which indicate the presence of a *linear* instability during the smoothing of an initially rough surface. This instability is presumed to be due to the presence of an asymmetric step-edge barrier [13], but its effectiveness is mediated by surface diffusion. In particular, we find that long wavelength modes are unstable and short wavelength modes decay, with a critical wavelength that depends on material parameters and growth conditions. This linear instability leads to the formation of features we call "hillocks" and is qualitatively distinct from the instability which is operative during later stages of growth that leads to the appearance and subsequent coarsening of mounds.

Growth of InAs buffer layers was performed in a Fisons V80 molecular-beam epitaxy chamber using "epi-ready" InAs(001) $\pm$ 0.1° substrates obtained from Wafer Technologies. Following desorption of the oxide, InAs buffer layers of various thicknesses were grown at 500 °C in an As overpressure using a III/V ratio on the order of 10. Atomic force microscopy (AFM) measurements were performed on these buffer layers under ambient conditions using a Quesant Instruments QScope operating in contact mode with an etched silicon Ultralever tip.

Figure 1 shows AFM images of surfaces with three buffer layer thicknesses together with representative line scans of their heights. The surface at 600 Å [Fig. 1(a)] shows remnants of deep pits that commonly occur as a result of oxide desorption. At 1500 Å [Fig. 1(b)], the surface shows some signs of organized hillocks, with the largest hillocks no more than 20 Å high. Considerably later in the growth, at 2 μm [Fig. 1(c)], there is clear evidence of mounds on the surface with heights of up to 60 Å and lateral dimensions of up to 2 μm. Although hillocks eventually become mounds (see below), we distinguish between them because the height of hillocks *decreases* with buffer layer thickness, but that of mounds *increases* even though the lateral size of both increases.

Large-scale features are believed to form during epitaxial growth as a result of asymmetric barriers for surface diffusion across or near steps [13,14]. During smooth layer-by-layer growth, atoms that are deposited on top of existing two-dimensional islands diffuse onto lower terraces, so that one layer is completed before a new layer starts to grow. An enhanced step-edge barrier prevents atoms from diffusing downwards, and it is more likely for a new nucleus to form on top of existing islands *before* the layer underneath is completed. Kinetic Monte Carlo simulations of simple models for epitaxial growth have confirmed that the inclusion of step-edge barriers indeed produces an instability that leads to the formation and growth of mounds [8,15,16]. The behavior shown in

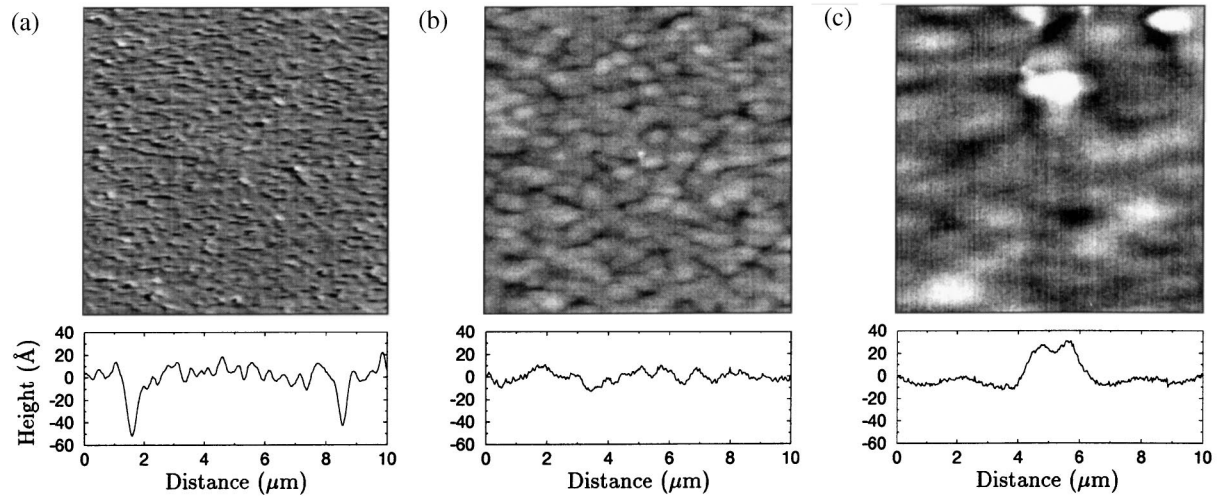

FIG. 1. Morphology and representative line scans of surface height in the $[0\bar{1}1]$ (horizontal) direction for InAs buffer layers of thickness: (a) 600 Å, (b) 1500 Å, and (c) 2 μm.

Fig. 1 is similar to that of GaAs(001) [2] and strongly suggests that there is an enhanced step-edge barrier in both systems.

Continuum theories have also been developed over the past several years to describe unstable growth of surfaces [7–9,12]. In these theories, a form for the mass current on the surface consistent with observations in KMC simulations is postulated. This current typically is of the form

$$\mathbf{j} = \nabla h(\alpha - \beta|\nabla h|^2) + \kappa\nabla|\nabla h|^2. \tag{1}$$

Here α, β, and κ are positive, material, and temperature dependent parameters, and the corresponding terms phenomenologically account for step-edge barriers, stabilization of height gradients, and surface diffusion, respectively. For the current assumed above, the evolution equation $h_t = \nabla \cdot \mathbf{j}$ is

$$\frac{\partial h}{\partial t} = \nabla^2 h(\beta|\nabla h|^2 - \alpha) + \nabla h \cdot \beta\nabla|\nabla h|^2 - \kappa\nabla^4 h\,. \tag{2}$$

Equations of this type have been used [7–9,12] only in the regime where mound coarsening is fully developed. Little or no attention has been paid to transient behavior from initial conditions even though our, as well as other [2], experiments suggest this is relevant. We show below that this continuum model qualitatively describes the experimentally observed behavior when growth is initiated on rough surfaces and, more important, that the evolution of hillocks is governed by different terms than those which drive the evolution of mounds at later times.

Figure 2 summarizes the results of integrating Eq. (2) with initial conditions corresponding to a rough starting surface [17]. Shown is the spherically averaged structure factor, $S(k) = \langle h_k h_{-k}\rangle$, at different times, obtained from the Fourier transform of the usual height correlation function, $\langle h_i h_j\rangle$. Also shown is the surface roughness, $W^2 = \langle (h - \langle h\rangle)^2\rangle$, as a function of time. Since W^2 is the integral of $S(k)$, the latter can be conveniently interpreted as the distribution of roughness over the various length scales k^{-1}. Two things are immediately apparent from the data. First, the surface roughness decreases for a considerable time period before eventually increasing at later times. Second, long wavelength modes are unstable and there appears to be a well-defined wave number separating stable and unstable modes that is independent of time.

The foregoing is precisely what would be expected from a completely *linear* evolution equation,

$$\frac{\partial h}{\partial t} = -\alpha\nabla^2 h - \kappa\nabla^4 h\,. \tag{3}$$

This is identical to the early-stage or linear theory of spinodal decomposition [18] and indicates that the nonlinear

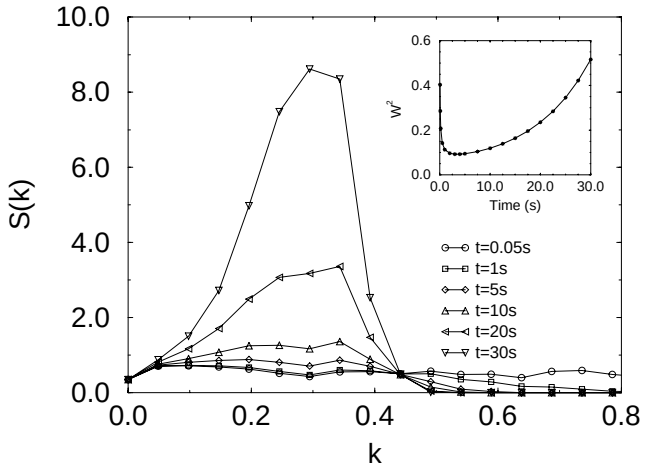

FIG. 2. Evolution of the structure factor $S(k)$ (in arbitrary units) and surface roughness W^2 for the continuum model described by Eq. (2). All simulations were performed on a 128×128 lattice with parameters $\alpha = 1.0$, $\beta = 50.0$, and $\kappa = 5.0$.

term in Eq. (2) plays almost no role in the evolution of the system until much later times. The key elements of this linear theory are the existence of a critical wave number, $k_c = \sqrt{\alpha/\kappa}$, below which all modes are unstable, and a maximally unstable wave number, $k_m = k_c/\sqrt{2}$, which defines a characteristic length $l_m = 2\pi/k_m$ [10]. For the simulation parameters used, the critical wave number $k_c = 0.45$, as is clearly evident in Fig. 2. The length l_m corresponds to the average distance between hillocks while the surface is smoothing. If Eq. (2) is integrated further, the peak of the correlation function $S(k)$ eventually shifts to longer wavelengths, an indication of nonlinearity in the system [18]. The morphology in this later regime exhibits coarsening, as seen in studies of similar equations [5,7–9,12].

One curious aspect of the early-stage behavior is that the nonlinearity plays such a small role in the evolution of the system. This occurs because, for a weak instability (α small) and strong surface diffusion (κ large), only a very *narrow* band of wavelengths is unstable. Since the amplitude of these unstable wavelengths grows like $e^{\alpha k^2 t}$, the system can evolve for a long time before the nonlinearity is manifested. This is in contrast to what is commonly observed in spinodal decomposition, where typically the instability is strong (α large) and the surface tension, which controls the width of the interface between phases, is weak (κ small). For this case, a *broad* band of wavelengths is unstable and, hence, the linear theory breaks down from the outset.

To clarify the dominant mechanisms in the early-stage regime, we have also performed KMC simulations of a solid-on-solid model previously employed to study coarsening and slope evolution during unstable epitaxial growth [16]. Atoms are deposited at random onto a surface at a rate F and are allowed to hop to a nearest-neighbor site at the rate $\tau^{-1} = (2k_BT/h)\exp[-(E_S + nE_N + \Delta m\Theta(\Delta m)E_B)/k_BT]$, where n is the number of nearest neighbors, Δm is the difference of next-nearest neighbors in the plane beneath *and* above the hopping atom before and after the hop, T is the substrate temperature, and $\Theta(x) = 1$ for $x > 0$, and 0 otherwise. The total energy barrier is comprised of a surface diffusion term, E_S, a contribution from each nearest neighbor, E_N, and an enhancement near step edges, E_B.

Figure 3 shows results of KMC simulations performed on an initially rough surface [19]. The evolution of the structure factor and surface roughness are qualitatively similar to that seen in the continuum model. The correlation function clearly exhibits a long-wavelength instability with $k_c \simeq 0.16$ and a prolonged regime in which the linear theory holds. The surface reaches a point of minimum roughness after approximately five layers are deposited, at which point it is organized into hillocks with feature size l_m. We expect, in general, that the point of minimum roughness is reached when the hillock size and l_m are comparable. We have checked that this point is indeed attained

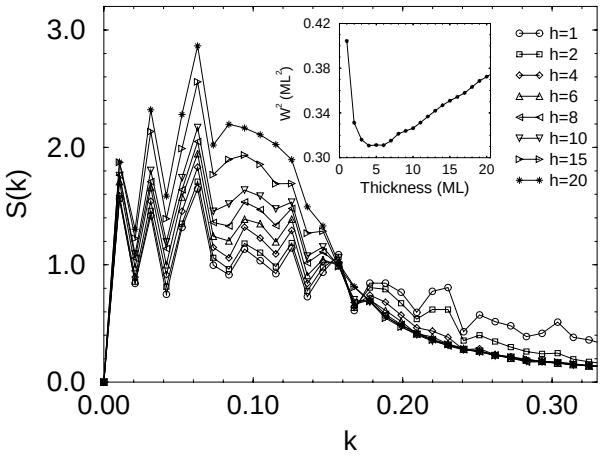

FIG. 3. Evolution of the structure factor $S(k)$ (in arbitrary units) and surface roughness W^2 for the KMC model. All data were averaged over 16 simulations performed on a square lattice of size 600×600 with parameters $E_S = 1.54$ eV, $E_N = 0.23$ eV, $E_B = 0.03$ eV, $F = 1/6$ monolayers (ML) s^{-1}, and $T = 800$ K.

at later times when E_B is decreased because the larger l_m is reached only at a later time. As in the continuum model, the simulations eventually produce mounds that arise from the hillocks already created by the linear instability mechanism which coarsen with time [16]. In that regime, the nonlinear effects become important, and we observe (not shown) that the peak of the structure factor shifts to smaller values of k.

The continuum theory and simulation results make two definite predictions: The structure factor exhibits an instability even in the early stages of growth, and the surface roughness decreases in the presence of this instability until reaching a minimum. These predictions can be checked by further analysis of the experimental results. Figure 4

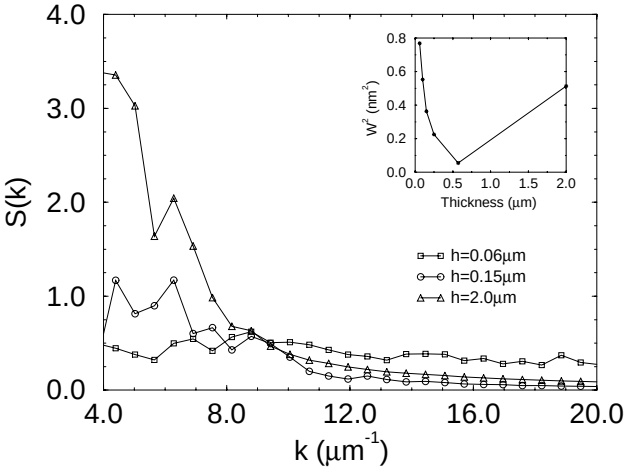

FIG. 4. Evolution of the structure factor $S(k)$ (in arbitrary units) and surface roughness W^2 for InAs buffer layers. Each data point represents an average over at least four independent AFM images of lateral size 10 μm.

shows $S(k)$ and W^2 for InAs buffer layers of different thicknesses. The data show an instability with $k_c \simeq 9.0\ \mu\text{m}^{-1}$. This corresponds to a characteristic length $l_m \simeq 1.0\ \mu$m between hillocks which is indeed observed in Fig. 1(b). In addition, the surface roughness exhibits a minimum that lies between 1500 Å and 2 μm and mounds are evident at later times, as shown in Fig. 1(c).

In summary, we have demonstrated that the formation of large-scale features on initially rough surfaces occurs in two distinct stages. At early times, a linear instability drives the formation of hillocks while the surface roughness is decreasing. These hillocks form a template on which mound evolution and coarsening take place at much later times [2–5]. Since experimental surfaces are typically rough before deposition, both regimes are important to surface morphological evolution. There are reports [20] in the experimental literature of the existence of "mounds" on various surfaces, some of which might well be structures evolving in the linear regime described here. A result of potential practical consequence is that the evolution of surface roughness depends on the length scale of interest. Although a point of minimum roughness exists, the entire distribution of roughness as described by $S(k)$ should be taken into account when considering the optimal thickness of device buffer layers.

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