Evidence from the Surface Morphology for Nonlinear Growth of Epitaxial GaAs Films

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The mesoscale morphology of homoepitaxial GaAs surfaces is explained with an anisotropic and nonlinear Kardar-Parisi-Zhang (KPZ) model in which adatoms are incorporated into the film from a metastable surface layer. Evaporation-condensation between the film and the metastable layer is proposed as the microscopic physical origin of the KPZ description, as well as of the excess noise observed in the power spectral density. The parabolic mounds observed experimentally in films grown on rough substrates are in good agreement with the surface shape expected from the solution of the KPZ equation in the large amplitude limit.

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Under rather general conditions, one expects an atomically flat substrate to become progressively rougher during epitaxial film growth due to noise associated with the random arrival of atoms from the vapor. This phenomenon is known as kinetic roughening and has been the subject of extensive theoretical research [1–4]. It has turned out to be quite difficult to explore experimentally, even in the most well-understood epitaxial systems, due to a variety of complicating factors such as problems with achieving a sufficiently flat starting surface, contamination of the starting surface, and variations in morphology associated with different growth conditions.

Of particular interest is the observation of mound formation in GaAs homoepitaxy, usually attributed to an unstable growth phenomenon [5–7]. According to this theory, atoms deposited on GaAs preferentially incorporate at up-steps due to asymmetric potential energy barriers at the step edges. This creates a tendency for atoms to migrate uphill, which leads to progressively steeper surface slopes during growth [8]. In the case of GaAs the slopes eventually saturate at a limiting value of $\approx 1°$, and the surface of an epitaxial film ends up covered with mounds. Scaling models which predict self-affine behavior for the surface morphology are not applicable for unstable growth. The experimental evidence for unstable growth in GaAs comes from the observation of mounds on the surface of GaAs films grown by molecular beam epitaxy at 550 °C [5,6]. In this Letter we propose an alternate explanation for the mounds on GaAs, namely, as incompletely smoothed remnants of the initial condition of the surface. We further show that the surface morphology of GaAs can be explained by the Kardar-Parisi-Zhang (KPZ) equation [4]. Although this is one of the simplest nonlinear growth equations, to our knowledge no one has succeeded in making a quantitative interpretation of the surface morphology of a thin film in terms of this equation.

Epiready GaAs substrates were heated to 600 °C in UHV under an As_2 overpressure (beam equivalent pressure 3.5×10^{-6} torr) to thermally evaporate the surface oxide, as discussed previously [9,10]. This process is known to produce a surface covered with submicron pits, as shown in the atomic force microscope (AFM) image in Fig. 1(a). Subsequent growth by molecular beam epitaxy (MBE) of a 1-μm-thick GaAs film at 550 °C with an As_2/Ga ratio of 3.5 (beam equivalent pressure measured with an ion gauge) produces the surface structure with elliptical mounds shown in the AFM image in Fig. 1(b). The in-plane anisotropy of the surface is sensitive to the As overpressure. This surface morphology is similar to that reported in Refs. [5,6], where similar mounds were attributed to unstable growth. Note that the rms surface roughness on the mounded surface (1.1 nm) is significantly smaller than on the pitted starting surface shown in Fig. 1(a) (5 nm). This is consistent with the mounded surface being a smoothed version of the starting surface. A scan line through the AFM image in Fig. 1(b) is shown in Fig. 1(d). The fact that the scan line does not have inversion symmetry, or more specifically the top of the mounds are rounded whereas the valley bottoms are V-shaped, suggests that the continuum growth equation which describes the smoothing is nonlinear [1].

The KPZ equation [4] is a suitable nonlinear growth equation for modeling the surface morphology:

$$\frac{\partial h(\vec{x},t)}{\partial t} = \nu \nabla^2 h(\vec{x},t) + \frac{\lambda}{2}[\nabla h(\vec{x},t)]^2 + \eta(\vec{x},t), \tag{1}$$

where $h(\vec{x},t)$ is the surface height relative to a reference plane; ν and λ are constants, and $\eta(\vec{x},t)$ is the noise due to the random deposition of atoms.

For large amplitude surface roughness, $h_{\mathrm{rms}} > 2\nu/\lambda$, the nonlinear term in Eq. (1) dominates [11]. In this case it is easy to show by direct substitution that the surface morphology should have the form of downward facing piecewise continuous paraboloids separated by cusps: $h(\vec{x},t) = h_0 - |\vec{x} - \vec{x}_0|^2/2\lambda t$. This limit is expected to apply early in the growth on the (rough) thermally cleaned substrates before the surface smooths. The mounded surface formed after growth on the thermally cleaned substrate shown in Fig. 1(b) does indeed consist of parabolic

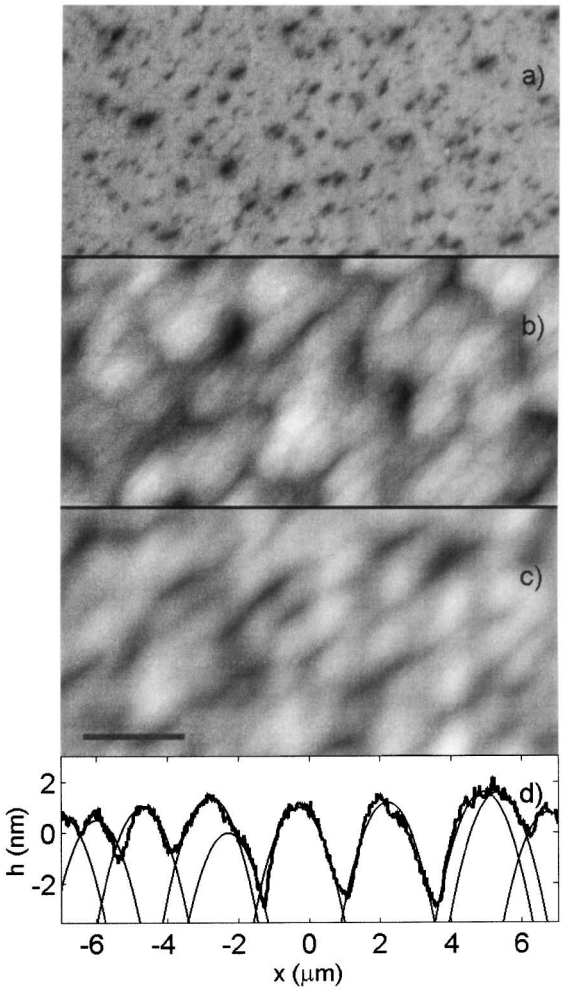

FIG. 1. (a) AFM image of a GaAs wafer after thermal evaporation of the surface oxide (5 nm rms roughness). (b) AFM image with a different gray scale (1.1 nm rms roughness) after 75 min growth on a surface prepared identically to the one in (a). (c) Simulated surface morphology based on Eq. (1) after 75 min growth. The initial condition coincides with the surface in (a), while the gray scale is the same as in (b). The scale bar is 2 μm. (d) Cross section of the surface in (b) along the elongated direction.

cross sections separated by cusplike valleys, as shown in Fig. 1(d). The cross section in Fig. 1(d) is a diagonal scan line through the AFM image in Fig. 1(b), oriented parallel to the long axis of the surface mounds. The nonlinear term in the KPZ equation is associated with a geometric effect in which growth along the local surface normal produces a net vertical growth rate that is higher on sloping surfaces. In this case $\lambda = F$, where F is the average growth rate of the film.

In MBE the atoms arrive ballistically from a source point, and at 550 °C all the deposited Ga atoms stick. The growth rate is thus set by the atom flux and not by the surface shape, and in general a conservative growth equation would be expected. Although the nonlinear term in the KPZ equation is needed to explain the surface profile, it is nonconservative and is therefore, strictly speaking, unphysical. A conservative nonlinear growth equation with similar properties could be obtained by replacing the nonlinearity in Eq. (1) with the fourth-order term $\nabla^2(\nabla h)^2$. If the fourth-order nonlinear term were dominant, one would expect the surface mounds to be quartics ($\sim|\vec{x}|^4/t$) which are inconsistent with cross section data such as that shown in Fig. 1(d). However, the presence of a linear term of the form $\nabla^2 h$ or $\nabla^4 h$ will once again tend to favor a parabolic surface profile. For this reason the parabolic shape of the mounds in Fig. 1(d) is not conclusive evidence for the KPZ type of nonlinearity. However, the up-down asymmetry confirms that nonlinear terms are essential for describing the surface morphology.

During MBE growth the deposited atoms are known to go first into a metastable precursor phase (which, for convenience, we will refer to as the "float") before they are permanently bonded into the growing film [12]. Given that the adatoms in the float are mobile and can diffuse over micron scale distances on the surface [13], we can offer the following physical rationale for the inclusion of the nonconservative KPZ nonlinearity. If the diffusion length of the adatoms is large, the float can act as a pseudovapor-phase from which atoms may be incorporated into the film uniformly for each unit of exposed surface area. The $(\nabla h)^2$ term then has the usual geometric interpretation [4], and the normal growth rate is determined by the atomic flux from the vapor divided by the total surface area. The density of atoms in the float will be determined by a balance between the rate of arrival of atoms from the vapor and the net rate of incorporation into the film, which will depend on the surface shape. During growth the adatoms move on the surface until they find a binding site, such as a step-edge kink, at which they can incorporate into the film. The exact nature of the species which diffuse on the surface is not important as long as it involves Ga. Since the As atoms are supplied in excess from the vapor, dissociation of Ga-As at one point on the surface can be made up by reassociation from vapor-supplied As after the Ga has diffused to another point on the surface.

The float concept also provides a natural explanation for the $\nabla^2 h$ term in the KPZ equation, without the need to invoke asymmetric step-edge potential barriers [8]. If there is a dynamic equilibrium between the surface incorporation sites, such as step-edge kinks, and the pseudovapor-phase (the float), then one would expect to have similar terms in the growth equation as for conventional evaporation-condensation from a vapor phase. The rate of "evaporation" of atoms depends on the local chemical potential of the surface, which is proportional to the curvature of the surface. This contributes a linear $\nabla^2 h$ term to the growth equation [2,8].

In order to describe the surface morphology in the region where both the linear and the nonlinear terms are important, it is necessary to solve the KPZ equation [Eq. (1)] numerically. In Fig. 1(c) we show a simulated surface computed by numerically solving the anisotropic KPZ

equation using the AFM image in Fig. 1(a) as the initial condition. Since the surface slopes are relatively small in the simulation, the effect of the nonconservative term on the growth rate is small. The simulated surface is in good agreement with the AFM image in Fig. 1(b). Since the starting surfaces in the simulation and in the experimental buffer layer are necessarily different, the two surfaces can only be compared in a statistical sense.

In the simulation we have used $\nu_{x,y} = 10$, 1 nm^2/s, and $\lambda_{x,y} = 1$, 5 nm/s. This anisotropy in the growth parameters suggests that in the x direction ($[1\bar{1}0]$) the ν term is dominant on the time scale of our experiments, whereas in the y direction the λ term dominates. There is a range of $\lambda_{x,y}$ values over which it is possible to achieve acceptable matches between the simulation and the data. Nevertheless we find that the $\lambda_{x,y}$ must be larger than the growth rate F ($\approx$0.3 nm/s) to obtain the observed cusplike valleys. It is not known why the $\lambda_{x,y}$ are large compared to the flux. Perhaps this is an indication that the higher-order nonlinear term $\nabla^2(\nabla h)^2$, which has a different physical origin, is important.

The thermally desorbed starting surface has a power spectral density (PSD) with slope ≈ -4 at large q, as shown in Fig. 2. After a short period of growth, the PSD drops rapidly and develops a smaller slope close to -2 at large q. The region with slope -4 is pushed to lower q values. A -4 falloff is expected for the PSD of a 2D function with a discontinuity in the first derivative, as embodied, for example, by the V grooves between parabolic mounds. This is consistent with our AFM data. Very long growths are required to eliminate all traces of the parabolic mounds left over at low q from the initial condition in the surface morphology.

Noise is included in the simulation by adding $\sigma\sqrt{(12\Delta t)}\,U(t)$ to each pixel in the image at each time step, where $\sigma^2 = 2D/(\Delta x)^2$, and $U(t)$ is a random number uniformly distributed between -0.5 and 0.5 [1]. We treat D as an adjustable parameter, although, in the case of flux noise, one expects $D = Fa^3$, where a is the monolayer height (0.3 nm). The simulation is in excellent agreement with the experimental PSD as shown in Fig. 2. The -2 power law at large q in the $[1\bar{1}0]$ direction in Fig. 2(b) is due to the linear Edwards-Wilkinson term which dominates in this direction. In the orthogonal direction, [110], in which the nonlinear term dominates we note that the slope at large q, in Fig. 2(a) and in other data (not shown), is consistently larger than -2, and close to the theoretically predicted value of -2.76 for the full nonlinear KPZ equation [1]. The peak in the PSD at low spatial frequencies reflects the presence of quasiperiodic mounds in the surface morphology.

To explore the effect of the morphology of the starting surface on the structure of the film, we have also grown GaAs on GaAs substrates that have been cleaned with atomic hydrogen in UHV. This cleaning procedure produces a relatively smooth starting surface [Fig. 3(a)] compared with the standard thermal cleaning process [Fig. 1(a)]. As shown in the AFM image in Fig. 3(b), the buffer layer grown on this surface has a smaller rms surface roughness (0.2 nm) than the buffer layer grown under identical conditions on a thermally cleaned surface (1.1 nm), and does not show mounds. This supports our conclusion that the mounds are due to the initial condition, rather than an unstable growth process, in agreement with Coluci *et al.* [14] who reached a similar conclusion for GaAs films grown by chemical beam epitaxy. The PSD for the layer grown on the H-cleaned substrate is also shown in Fig. 2. At large q the power law behavior closely matches the results for the thermally cleaned substrate, indicating that the same equation governs the growth after both types of cleaning.

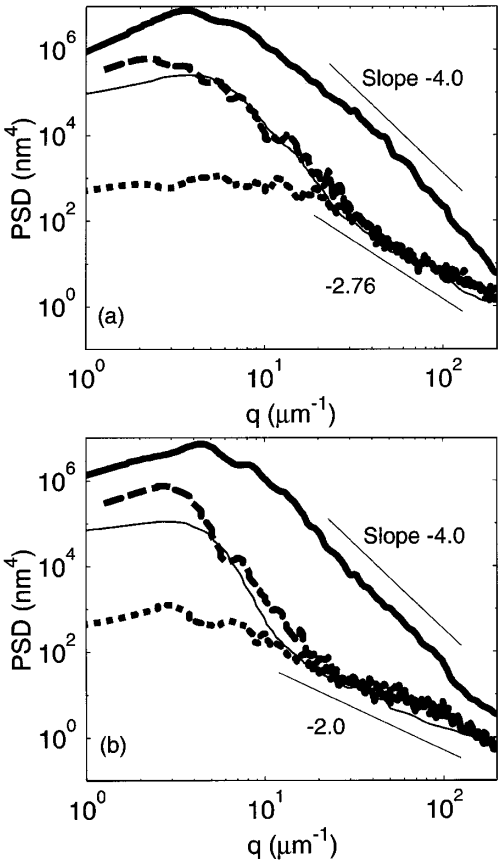

FIG. 2. PSD along (a) the [110] and (b) the $[1\bar{1}0]$ directions. The thick solid, dashed, and dotted lines correspond to the surfaces shown in Figs. 1(a), 1(b), and 3(b), respectively. The thin solid lines represents the PSD of the simulation in Fig. 1(c).

A PSD with an exponent of -2 would also be expected for a fourth-order nonlinear growth equation with conservative noise. This interpretation cannot easily be rejected on the basis of the data presented here alone. However, light scattering data on the time dependence of the surface roughness during GaAs growth on H-cleaned substrates is consistent with a dynamical exponent $z = 1.58$ expected for the KPZ equation [15], and is not consistent with a

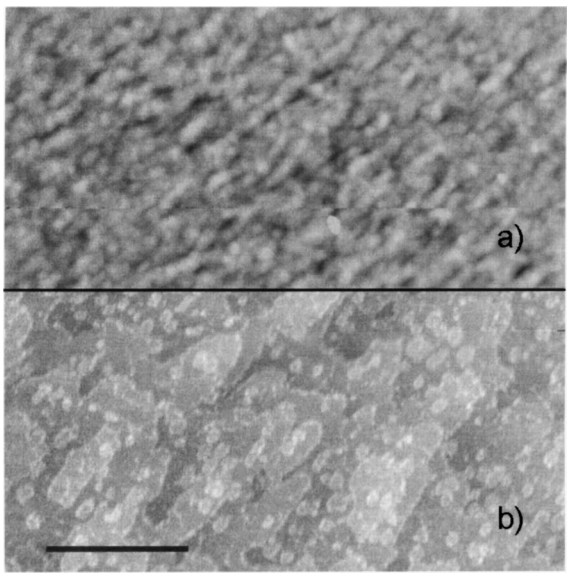

FIG. 3. (a) AFM image of a hydrogen etched GaAs surface (rms roughness 0.24 nm). (b) AFM image after 75 min growth on a hydrogen etched starting surface (rms roughness 0.23 nm), showing individual atomic steps. The scale bar is 500 nm, and the gray scales in (a) and (b) are identical. Note the absence of mounds on the surface.

dynamical exponent of 4, corresponding to the fourth-order equation.

In the numerical fit to the observed surface morphology we find that $D \approx 50Fa^3$, about 50 times larger than would be expected from deposition noise alone. This result can be understood if the deposited surface atoms incorporate into the film and re-evaporate back into the float a number of times, in a dynamic equilibrium, before being permanently incorporated into the film. Both the incorporation and subsequent evaporation will contribute nonconservative flux noise to the morphology of the growing surface.

In conclusion, we have shown for the first time that the mesoscopic surface morphology of homoepitaxial GaAs can be explained by a stable, but nonlinear, continuum growth equation, namely, the anisotropic KPZ equation. We are able to explain key features in the growth equation by assuming that the deposited atoms first enter a metastable and mobile surface phase before being permanently incorporated into the film. The initial condition of the substrate and the nonlinear term in the KPZ equation explain the mounds that are observed in the surface that had previously been attributed to unstable growth.

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[1] A.-L. Barabási and H. E. Stanley, *Fractal Concepts in Surface Growth* (Cambridge University Press, Cambridge, United Kingdom, 1995).
[2] J. Krug, Adv. Phys. **46**, 139 (1997).
[3] T. Halpin-Healy and Y.-C. Zhang, Phys. Rep. **254**, 215 (1995).
[4] A. Pimpinelli and J. Villain, *Physics of Crystal Growth* (Cambridge University Press, Cambridge, United Kingdom, 1998).
[5] M. D. Johnson, C. Orme, A. W. Hunt, D. Graff, J. Sudijono, L. M. Sander, and B. G. Orr, Phys. Rev. Lett. **72**, 116 (1994).
[6] A. W. Hunt, C. Orme, D. R. M. Williams, B. G. Orr, and L. M. Sander, Europhys. Lett. **27**, 611 (1994).
[7] G. Apostolopoulos, J. Herfort, L. Däweritz, K. H. Ploog, and M. Luysberg, Phys. Rev. Lett. **84**, 3358 (2000).
[8] J. Villain, J. Phys. (France) I **1**, 19 (1991).
[9] M. Adamcyk, A. Ballestad, T. Pinnington, T. Tiedje, M. Davies, and Y. Feng, J. Vac. Sci. Technol. B **18**, 1488 (2000).
[10] M. Adamcyk, T. Pinnington, A. Ballestad, and T. Tiedje, Mater. Sci. Eng. B **75**, 153 (2000).
[11] T. J. Newman and A. J. Bray, J. Phys. A **29**, 7917 (1996).
[12] T. Shitara, J. Zhang, J. H. Neave, and B. A. Joyce, J. Appl. Phys. **71**, 4299 (1992).
[13] S. Koshiba, Y. Nakamura, M. Tsuchiya, H. Noge, Y. Nagamune, T. Noda, and H. Sakaki, J. Appl. Phys. **76**, 4138 (1994).
[14] V. R. Coluci, M. A. Cotta, C. A. C. Mendonça, K. M. I.-Landers, and M. M. G. de Carvalho, Phys. Rev. B **58**, 1947 (1998).
[15] C. Lavoie, Ph.D. thesis, University of British Columbia, 1994.