

Emergence of macroscopic surface structure from atomistic dynamics in epitaxial film growth

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A nonlinear differential equation is derived for the surface shape evolution in epitaxial growth, from a transport equation for the adatoms. A negative Ehrlich-Schwöbel barrier is assumed to be present at atomic steps, favoring downhill migration of adatoms. Expressions for the coefficients in the growth equation are obtained in terms of the deposition rate, step density, step edge potential barrier, and adatom release rate from step edges. The analytical model is tested by comparison with a kinetic Monte Carlo simulation of a solid-on-solid model, which includes the same physical phenomena.

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The time evolution of the surface shape is one of the most basic phenomena in epitaxial growth and has been studied extensively.¹⁻⁴ Although the underlying atomistic physics is conceptually simple, the aggregate dynamical behavior of many adatoms during crystal growth is difficult to model analytically. In the simplest case, vapor-deposited adatoms diffuse on the surface until they are absorbed by a step edge and incorporate into the surface.⁵ Adatoms also desorb from the step edges, but at a lower rate. An adatom located at the top edge of a step may experience an asymmetric potential, known as an Ehrlich-Schwöbel (ES) barrier,⁶ that will tend to either enhance or reduce the rate of transfer to the lower terrace. In this paper we describe a method for deriving a growth equation starting with an adatom transport equation, for the case in which the step edge potential barrier enhances the rate of transfer to the lower terrace.

Considerable effort has gone into identifying universality classes in crystal growth, from the scaling behavior of the interface width.⁷ If one can match the scaling behavior of the system to that of a suitable partial differential equation, then one can argue that the system can be described by this dynamical equation. However, this method provides little guidance on how to relate the coefficients in that model to microscopic physical processes. In addition, the system of interest could easily be in a crossover regime, which is not amenable to a scaling analysis.

In this paper, we derive an equation for the time dependence of the shape of a patterned surface directly from the adatom transport equation. The analytical model is compared with a kinetic Monte Carlo (KMC) simulation of a solid-on-solid (SOS) model of crystal growth. Since we understand the physics of this computational model completely, it is a better test of the analytical model than experimental data, for which the physical mechanisms are uncertain. The model in this paper was developed to describe measurements of surface shapes during epitaxial growth of GaAs on patterned substrates, and therefore the parameter values are designed to reproduce experimental parameters for GaAs.⁸ In particular, the GaAs experiments show a downhill bias in the adatom drift that we model with inverse (negative) ES barriers at step edges.

The transport equation for adatoms with a density n (with dimension nm^{-2}), crystal lattice constant a , and diffusion constant D is as follows:^{2,8}

$$\partial_t n = F + (Ka - \beta Dn)S^2 + D\nabla^2 n - \nabla \cdot J_{es}. \quad (1)$$

In this equation, F ($\text{nm}^{-2} \text{s}^{-1}$) is the flux of adatoms deposited from the vapor and S (nm^{-1}) is the density of surface steps per unit area. S is a function of the surface slope ∇h , where h is the surface height above a flat reference plane. K is the thermal release rate of adatoms from step edges ($\text{nm}^{-1} \text{s}^{-1}$) and β is a dimensionless geometrical constant. The second term on the right-hand side of Eq. (1) describes the net incorporation rate of adatoms into the substrate at step edges, namely, the difference between the capture and release rates. The third term describes adatom diffusion with coefficient D . The last term depends on the slope-dependent adatom current J_{es} , associated with the ES barrier. We assume that the potential barriers favor downhill migration of adatoms, as discussed above. Clearly, there are also systems for which the adatoms tend to migrate uphill, although we do not consider this case here. The various terms in Eq. (1) are explained in more detail below.

We neglect kinetic roughening associated with the random motion of adatoms. We also neglect island nucleation as a mechanism for adatom capture; however, nucleation is implicitly assumed to be present in order to maintain a steady-state step density.

The downhill drift current and step edge capture rates in Eq. (1) can be estimated with reference to the solution of the diffusion equation on a terrace bordered by parallel steps, a distance L apart. In this case, the step density, or step length per unit area, is $S=1/L$. The up and down steps capture adatoms with capture velocities C_u, C_d , respectively, and release them back onto the terrace at a rate $K/2$ per unit length. Therefore, we can write the gradient boundary conditions

$$D \nabla n|_{u,d} = \pm C_{u,d} n|_{u,d} \mp K/2 \quad (2)$$

at the step edges.⁵ The diffusion coefficient $D=a^2\omega_0/4$, where $\omega_0=2k_B T/h \times \exp(-E_{sub}/k_B T)$ is a temperature-dependent hop rate with substrate bonding energy E_{sub} . Solving the steady-state diffusion equation for the adatom density with these boundary conditions, we find the net step edge capture rate to be $(12Dn_{av}^2 - Ka)S^2$ where n_{av} is the average adatom density. This reproduces the second term on the right-hand side of Eq. (1) with $\beta=12$. This analysis ignores the fact that, in general, the steps will not be parallel lines,

but rather will have a random shape. We wish to find out whether the observed surface shapes can be described without detailed knowledge of the shape of the step edges.

The net downhill flux for a single terrace is the difference in the rate of adatom absorption at the down step and the up step. This is readily obtained from the solution of the diffusion equation in the parallel step configuration and is given by

$$J_S = \zeta(F + KS) \quad (3)$$

where $\zeta = \frac{1}{2}D(1/C_u - 1/C_d)$ is the Ehrlich-Schwöbel parameter. In our case, the absorption rate at the downhill step is greater than the rate at the uphill step ($C_d > C_u$) so that $\zeta > 0$. In the case of a low surface slope, many of the terraces contain islands that are bordered by opposite-sign steps (down on both sides) and will not contribute to the net downhill flux associated with the macroscopic topography. In this case we prorate the downhill flux by the fraction of terraces that have the same sign steps (up on one side, down on the other). In the low-slope limit $\nabla h < aS$ and

$$J_{es} = J_S \times \nabla h / aS. \quad (4)$$

(This result differs from results reported earlier in that we have also included the rate of adatom release from step edges.⁹) The step density S also depends on the surface slope. For example, in the high-slope limit, when the surface resembles a staircase, $aS \sim \nabla h$. A suitable interpolation formula for the step density can be obtained from the incoherent sum of the random local slope associated with kinetic growth phenomena and the deterministic slope ∇h associated with the macroscopic surface shape,^{8,9}

$$S = \sqrt{S_0^2 + (\nabla h/a)^2} \quad (5)$$

where S_0 is the step density at zero slope. This expression has the expected asymptotic step density at large slopes. Substituting Eq. (5) into Eq. (4), we obtain the downhill adatom flux J_{es} for both large and small slopes.

The time derivative in Eq. (1) is approximately equal to zero, because the adatom dynamics (diffusion, step attachment) are fast compared with the rate at which the macroscopic shape of the surface changes. The transport equation [Eq. (1)] can now be solved for the adatom density n . Using Eqs. (3)–(5) and Eq. (1), we get

$$F - \beta DS^2 n + KaS^2 + \zeta \left(\frac{F}{S} + K \right) \nabla^2 h + D \nabla^2 n \approx 0. \quad (6)$$

In the limit of weak surface topography, the spatial variation in the adatom density is small compared to its average value. In this case, the last two terms in Eq. (1), which describe the spatial variations, are small compared to the first three terms and we can solve Eq. (6) for n to lowest order in the ∇^2 operator as follows:

$$n = \frac{\zeta}{\beta DS^2} \left(\frac{F}{S} + K \right) \nabla^2 h + \frac{F + KaS^2}{\beta DS^2 - D \nabla^2} \approx \frac{\zeta}{\beta DS^2} \left(\frac{F}{S} + K \right) \nabla^2 h + \frac{F + KaS^2}{\beta DS^2} - \frac{2F \nabla^2 S}{\beta^2 DS^5}. \quad (7)$$

The last term in Eq. (7), with the ∇^2 moved to the numerator, is obtained by solving Eq. (6) for the Fourier transform of n to second order in the spatial frequency, and then inverse transforming.

The surface height increases whenever an adatom attaches to a step edge and decreases when an adatom is released from a step edge. Therefore, the time dependence of the surface height is

$$\partial_t h = a^3 S^2 (\beta D n - Ka). \quad (8)$$

From Eqs. (7) and (8) we then obtain the growth equation

$$\partial_t h = a^3 F + \nu \nabla^2 h - \lambda \nabla^2 (\nabla h)^2 \quad (9)$$

to lowest order in the derivatives of the linear and nonlinear terms in ∇h and to second order in h , where $\nu = a^3 \zeta (F/S_0 + K)$ and $\lambda = aF/(\beta S_0^4)$. This equation is of mixed order in the spatial derivatives and is expected to describe surface shape evolution for low-amplitude, long-wavelength surface topography. A similar expression for λ was obtained earlier by Krug⁷ and Politi *et al.*⁹ From Eq. (7) we conclude that the nonlinear term arises from the diffusion of adatoms from regions with low densities of attachment sites to regions with high densities.

The coefficient ν of the linear term in Eq. (9) reproduces the result of Vvedensky if we apply the constraints of the Wolf-Villain model.¹⁰ In that model, the ES parameter $\zeta = 1$, since the particle always moves down at a step edge; similarly, the step density is $\sim 1/a$ as every site acts as an adatom-immobilizing step site since there is no surface diffusion.

Equation (9) has the form of a conservative Kardar-Parisi-Zhang equation,¹¹ in good qualitative agreement with experimental data for GaAs.^{12,13} For a more rigorous test of the continuum model, we compare the predictions of Eq. (9) with numerical simulations of crystal growth using a full-diffusion, restricted SOS model.^{8,14} In the particular model that we use as a test case, adatoms are deposited at a rate F and bind to terrace sites with a activation barrier for hopping of 1.25 eV. At step edges, the adatoms bind laterally with up to three additional bonds, each with energy 0.35 eV. On the upper edge of an atomic step, there is a barrier lowering of 50 meV ($E_{es} = -50$ meV) for adatom hopping to the lower terrace, which simulates the ES barrier that favors downhill migration. A typical simulation involves 10^{10} events.

In Fig. 1, we show the steady-state density of step edge attachment sites on a vicinal surface as a function of temperature and surface slope in the SOS simulation. The initial condition for these simulations consists of regularly spaced steps on a 400×400 lattice with wrapped boundaries. As expected, the step density decreases with temperature and increases with surface slope. At 400 °C, the slope dependence of the step density matches our interpolation formula in Eq. (5) rather well. At higher temperatures, the step den-

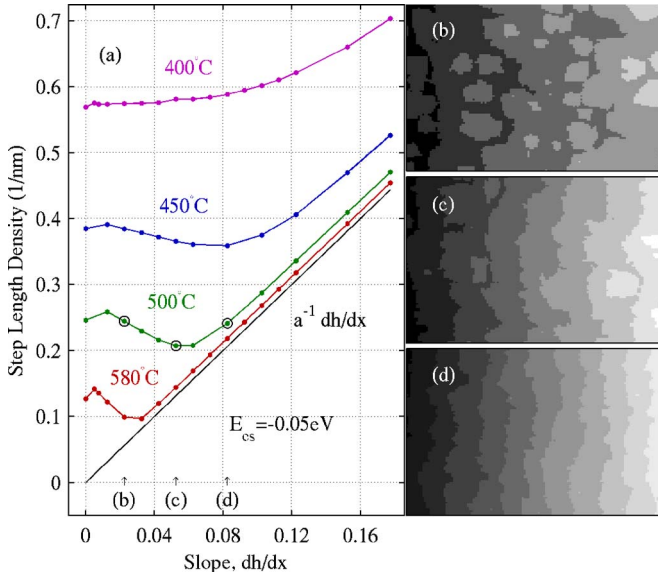


FIG. 1. (Color online) Steady-state step density from SOS KMC simulations of vicinal surfaces at four different temperatures. The line through the origin is the step density for a straight-stepped vicinal surface (staircase). (b)–(d) show sections of three surfaces from simulations at 500 °C taken (b) before, (c) at, and (d) after the step-density minimum, at slopes indicated in (a) by circles.

sity has a nonmonotonic dependence on surface slope. The reason for this behavior can be seen in Figs. 1(b)–1(d), which show the surface topographies for three different surface slopes for growth at 500 °C. At low slopes the steps have a closed-loop, island-type topology, while at higher slopes they have an open-loop, staircase-type topology. The minimum step density occurs at the transition between the two different step topologies.

The time evolution of an initially sinusoidal surface in the SOS simulation can be fitted by Eq. (9) with ν and λ as adjustable fitting parameters. The fitted parameters can then be compared with theoretical values computed from the expressions for ν and λ following Eq. (9). The time development of an initially sinusoidal surface cross section is shown in Fig. 2(a) for growth at 495 °C, along with the continuum model fit obtained by solving Eq. (9) numerically. The initially sinusoidal surface develops downward-pointing cusps, due to the nonlinearity in the growth process, as the surface amplitude decays. The best-fit values of ν and λ are obtained by matching the Fourier coefficients of the surface in the continuum model to the atomistic simulation according to $h(x, t) = \sum_{i=1}^3 a_i(t) \cos(2\pi i x/L)$, where L is the period of the starting surface and $a_1(0) = 6$ and $a_2(0) = a_3(0) = 0$. Examples of the time dependence of the Fourier coefficients are shown in Figs. 2(b)–2(d). The nearly exponential decay of the first Fourier coefficient is primarily determined by the linear term in Eq. (9). The growth of the second harmonic reflects the amplitude of the nonlinear term, and can be recognized in the broadening of peaks and the cusps developing between them. Figures 2(c) and 2(d) indicate that Eq. (9) matches the simulation very well for all three Fourier coefficients at low to medium growth temperatures. In Fig. 2(b) we see that high-temperature growths do not fit as well. This discrepancy may

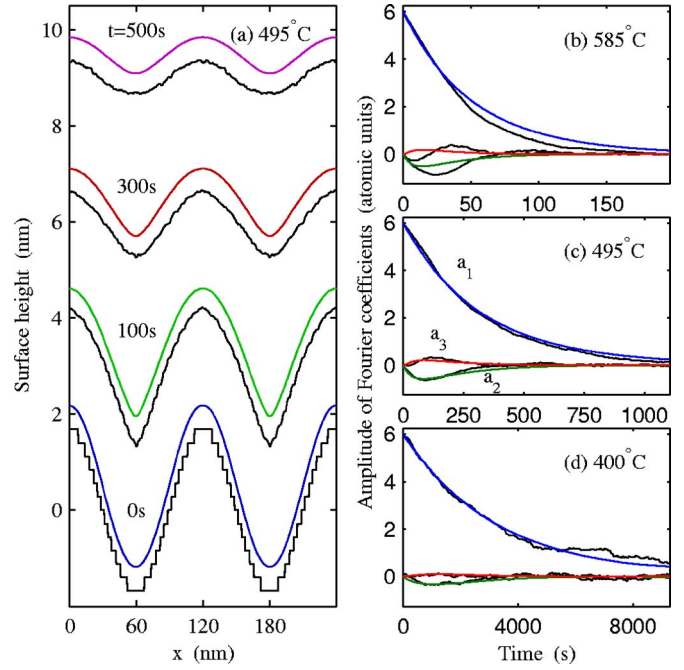


FIG. 2. (Color online) (a) shows the time evolution of an initially sinusoidal surface according to SOS KMC simulations at 495 °C and 1 monolayer (ML)/s growth rate, together with a best fit to the simulation with the continuum model Eq. (9), with $\nu = 1 \text{ nm}^2/\text{s}$ and $\lambda = 280 \text{ nm}^3/\text{s}$. The simulated surface and fits are offset for clarity. (b)–(d) show how the continuum model reproduces the first three Fourier coefficients (a_1 , a_2 , and a_3) in the Fourier series representation of the simulated surface topography as a function of time at 585, 495, and 400 °C, respectively.

be due to the fact that the step density does not increase monotonically at high temperatures as we have assumed [Eq. (5)], but shows instead a more complex behavior (Fig. 1). Support for this interpretation is that we observe cusp inversion for low-amplitude, high-temperature surfaces, as one would expect if the step density decreases with slope for low slopes.

Best-fit values for ν and λ for a wide range of growth temperatures and growth rates are shown in Fig. 3. We also show the values for ν and λ calculated from the theoretical expressions in the text following Eq. (9), using the growth parameters from the SOS simulations.¹⁵ The step density S_0 is obtained from the KMC simulation by summing over the three different types of step edge incorporation sites. The step density becomes constant after a few monolayers of growth, consistent with the earlier assumption. The average step edge release rate is calculated according to $K = \sum_{i=1}^3 k_i n_i / S_0$, where n_i (nm^{-2}) is the number density of sites on the surface with i lateral neighbors, and k_i is calculated from $D \exp(-E_i/k_B T)$. Throughout the range of the KMC simulations, K is constant as a function of time and closely approximated by $k_1 n_1 / S_0$, the release rate for adatoms with one lateral bond. For the material parameters we have chosen, the step edge release dominates the deposition flux F in the coefficient of the linear smoothing term in Eq. (9), at high temperatures and low fluxes.

The temperature and growth rate dependence of the theo-

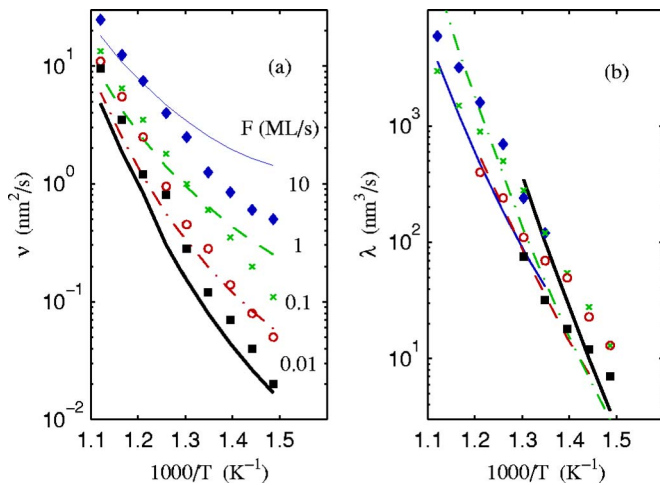


FIG. 3. (Color online) Temperature dependence of ν and λ at various growth rates obtained by fitting the SOS KMC simulations (solid squares 0.01; circles 0.1; crosses 1; and solid diamonds $F = 10$ ML/s) and the theoretical predictions based on the continuum model (thick solid line 0.01; dash-dotted 0.1; dashed 1; and thin solid line $F = 10$ ML/s). In the continuum model for λ we use $\beta = 6$.

retical values for ν, λ are in good agreement with the “measured” values from the KMC simulations over most of the range, as shown in Fig. 3. The agreement tends to break down at low temperatures and high growth rates. This is the regime where the step edges become highly irregular and fractal-like. Both the simulation and the theory show stron-

ger temperature dependence for λ than for ν , but a weaker flux dependence.

We can turn the problem around and use experimental values for the smoothing parameters to infer the atomistic growth parameters. For example, in the case of epitaxial growth of GaAs at $1 \mu\text{m/h}$ at 550°C , we find experimentally that $\lambda = 5 \times 10^5 \text{ nm}^3/\text{s}$ and $\nu = 1 - 10 \text{ nm}^2/\text{s}$, depending on the in-plane orientation.^{12,13} Setting $\beta = 6$ in the expression for the nonlinear prefactor λ , we find $S_0 = 0.03 \text{ nm}^{-1}$. This is consistent with step densities observed in atomic force microscope images of molecular beam epitaxy-grown GaAs surfaces which are in the $0.005 - 0.06 \text{ nm}^{-1}$ range. If we substitute $S_0 = 0.03 \text{ nm}^{-1}$ into the expression for the linear smoothing coefficient ν , use $E_{es} = -0.05 \text{ eV}$, and neglect K compared with F/S_0 , we find $\nu = 3 \text{ nm}^2/\text{s}$, which is also consistent with experiment.

In conclusion, we have obtained expressions for the linear and nonlinear smoothing coefficients for a generic model of epitaxial growth, in terms of atomistic parameters. The surface smoothing rate is controlled by the deposition rate and surface step density. The sign of the nonlinear term in the growth equation, which determines whether the surface develops up or down cusps, is determined by whether the step density increases or decreases with surface slope. Along with the Ehrlich-Schwöbel step edge barrier, the dependence of the step density on the surface slope is an example of a microscopic property that carries over to the macroscopic surface shape. The detailed step edge geometry on the other hand does not have a strong influence on the macroscopic surface shapes.

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- ¹⁵The Ehrlich-Schwöbel parameter in terms of the step edge barrier is $\zeta = (1 - 1/e)/2$ with $e = \exp(-E_{es}/k_B T)$. The barrier lowering also reduces the step edge release rate in the restricted SOS model by the factor $(1 + 0.5e)/(1 + e)$.