Kinetic Roughening in Molecular-Beam Epitaxy

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We extend a continuum model recently proposed by Villain [J. Phys. I (France) **1**, 19 (1991)] to study equilibrium and nonequilibrium diffusion on a high-symmetry surface under a fluctuating particle beam. Exponents characterizing dynamic scaling in various regimes are derived explicitly in all dimensions, as well as the relevant lengths which separate these regimes. Different surface morphologies are correlated with experimentally accessible parameters such as substrate temperature and deposition rate.

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In vacuum deposition experiments, epitaxial growth of a crystalline film is usually achieved through surface migration of newly arrived atoms to the energetically more favorable ledge and kink sites.[1] The morphology of the film surface is thus sensitive to the beam intensity which controls the nonequilibrium adatom population, and to the substrate temperature which influences the rate of surface diffusion. Even in the case of stable growth, one encounters different forms of kinetic roughness, ranging from mere terrace width fluctuations, as in step-flow growth on a vicinal surface at elevated temperatures, to the nucleation of islands on islands, which occurs at lower temperatures when the mobility of the surface atoms is much reduced.[2,3]

Recent theoretical studies of driven interfaces have focused on elucidating the role of symmetry and conservation laws in determining dynamic universality classes.[4-6] These interfaces or surfaces typically exhibit self-affine structures both in space and in time: The height fluctuation across a distance L in the surface scales as L^ζ, and has a lifetime proportional to L^z.[7] Here ζ and z are known as roughness and dynamic exponents, respectively. In molecular-beam epitaxy (MBE) the primary relaxation mechanism—surface diffusion—conserves the mass of the film. If the vacancy concentration in the film does not vary during growth, mass conservation translates into volume conservation described by a continuity equation:[8-11]

$$\partial Z/\partial t+\Omega\nabla\cdot\mathbf{j}=F\,. \tag{1}$$

Here $Z(\mathbf{x},t)$ is the film thickness above a substrate site $\mathbf{x}$ at time t, $\mathbf{j}$ is the particle-number current density within the surface, Ω is the atomic volume, and F is the beam intensity which we take to be independent of Z. Equation (1) distinguishes surface diffusion from other relaxational processes such as desorption. Indeed, the generic continuum description in the latter case is given by the Kardar-Parisi-Zhang (KPZ) equation[4,12] which, however, is incompatible with the conservation law imposed by (1).

Villain[10] recently proposed a continuum model of the form (1) which includes nonequilibrium terms that are consistent with in-plane isotropy and a continuous translational symmetry in the growth direction. These terms were shown to reduce the extreme roughness induced by the shot noise (beam fluctuation) if the surface were to relax via equilibrium surface diffusion[13] alone. The surface is nevertheless rough on all length scales at and below (2+1) dimensions. In this Letter we extend Villain's model by including two additional terms which are likely to influence small-length-scale features: (i) a lattice-pinning term which reduces the continuous translational symmetry to a discrete one; (ii) a conserving component in the noise arising from the stochastic nature of surface diffusion on the atomic level. In the limit of vanishing beam intensity, we arrive at the equilibrium sine-Gordon model[14-18] with a conserving dynamics. As in the case of nonconserving dynamics,[16,17] a finite growth velocity diminishes lattice pinning on sufficiently large length scales, at which point the model proposed by Villain is recovered. The roughening behavior of the latter model is analyzed in a renormalization-group (RG) treatment which yields the exponents proposed by Villain.[10] On smaller length scales and below the thermal roughening temperature T_R of the equilibrium model, the lattice-pinning term may become dominant, leading to the formation of faceted areas on the surface. We show that, under a nonconserving noise, faceting on arbitrarily large length scales is not possible at and below (2+1) dimensions, even if the surface is not moving.

For a sufficiently weak beam, it appears reasonable to write

$$\mathbf{j}=\mathbf{j}_{\rm eq}+\mathbf{j}_{\rm neq}\,, \tag{2}$$

where $\mathbf{j}_{\rm eq}$ is the (quasi)equilibrium part driven by the gradient of the chemical potential[13]

$$\mu=\Omega\,\delta\mathcal{F}/\delta Z\,, \tag{3}$$

and $\mathbf{j}_{\rm neq}$ the nonequilibrium correction to $\mathbf{j}_{\rm eq}$. In the following we shall consider (3) on a semimicroscopic level by postulating a Ginzburg-Landau free energy of the form[14-17]

$$\mathcal{F}=\int d^dx\left[\tfrac{1}{2}\Gamma(\nabla Z)^2+V(Z)\right], \tag{4}$$

where Γ is the surface stiffness constant, $V(Z+a) = V(Z)$ is a "lattice-pinning" potential which has its minima at integer multiples of the layer spacing a, and d is the *dimension of the surface.* Correspondingly, we take $\mathbf{j}_{\rm eq}$ to be of the Langevin type and write, in accordance with the Nernst-Einstein relation,

$$\mathbf{j}_{\rm eq} = -(\rho_s D_s/k_B T)\nabla\mu + \mathbf{j}_0 , \tag{5}$$

where D_s is the surface diffusion coefficient, ρ_s is the surface density of the diffusing species, and $\mathbf{j}_0$ is the stochastic current due to thermal fluctuations, with

$$\langle j_{0i}(\mathbf{x},t) j_{0k}(\mathbf{x}',t')\rangle = 2\rho_s D_s \delta_{ik}\delta^d(\mathbf{x}-\mathbf{x}')\delta(t-t') .$$

The appropriate form for the nonequilibrium current $\mathbf{j}_{\rm neq}$ in the context of MBE is less clear. In principle, $\mathbf{j}_{\rm neq}$ could contain terms of the same type as in $\mathbf{j}_{\rm eq}$, resulting in a redefinition of the equilibrium parameters such as the temperature T and the adatom concentration ρ_s. More importantly, $\mathbf{j}_{\rm neq}$ may contain *new* terms which cannot be obtained from (3)-(5), and which govern dynamic scaling on sufficiently large length scales. For a high-symmetry surface, Villain[10] proposed the following expression:

$$\mathbf{j}_{\rm neq} = -(\nu/\Omega)\nabla Z - (\sigma/2\Omega)\nabla(\nabla Z)^2 . \tag{6}$$

This choice is consistent with the translational symmetry $Z \to Z+a$ which excludes terms like ∇Z^2. Phenomenologically, the ν term in (6) indicates a downhill (or uphill) current for a surface tilted away from the high-symmetry direction.[19] Though absent in the equilibrium problem when gravity is negligible, such a current could be justified under the growth condition.[10] The σ term cannot be obtained from a free energy and it also behaves differently from the ν term under the transformation $Z \to -Z$.[20] Only stable growth ($\nu \geq 0$) will be considered here.

To complete the definition of the model, we write $F = F_0 + f$, where f represents fluctuations away from an average beam intensity F_0. For simplicity, we shall also assume $V(Z) = -V_0\cos(2\pi Z/a)$. The final equation of motion for Z takes the form

$$\frac{\partial Z}{\partial t} = \nu\nabla^2 Z - \gamma\nabla^4 Z + \frac{\sigma}{2}\nabla^2(\nabla Z)^2 + \upsilon\nabla^2\sin(2\pi Z/a) + F_0 + \eta . \tag{7}$$

Here $\gamma = \rho_s D_s \Omega^2\Gamma/k_B T$ and $\upsilon = 2\pi\rho_s D_s\Omega^2 V_0/a k_B T$. The noise $\eta = f - \Omega\nabla\cdot\mathbf{j}_0$ is assumed to be Gaussian distributed with zero mean and the following correlator:

$$\langle \eta(\mathbf{x},t)\eta(\mathbf{x}',t')\rangle = 2\mathcal{D}\delta^d(\mathbf{x}-\mathbf{x}')\delta(t-t') , \tag{8a}$$

$$\mathcal{D} = D_0 - D_1\nabla^2 + D_2\nabla^4 , \tag{8b}$$

where $D_0 \simeq \Omega F_0/4$ and $D_1 = \Omega^2\rho_s D_s$. The D_2 term is of technical importance only.

Equation (7) has sufficient complexity to allow for a number of different spatial scaling regions which presumably also exist in experiments. To illustrate the point let us consider the solvable case $\sigma = \upsilon = 0$.[10] Depending on the relative strength of ν and γ, two dynamical regimes are identified with

$$z = 4, \quad L \ll L_\nu , \tag{9a}$$

$$z = 2, \quad L \gg L_\nu , \tag{9b}$$

where $L_\nu = (\gamma/\nu)^{1/2}$ and L is the length of interest. A second length scale

$$L_f = (D_1/D_0)^{1/2} \sim (\Omega\rho_s D_s/F_0)^{1/2} \tag{10}$$

separates two scaling regimes of the noise such that

$$z = 2\zeta + d + 2, \quad L \ll L_f , \tag{11a}$$

$$z = 2\zeta + d, \quad L \gg L_f . \tag{11b}$$

The combination of (9) and (11) yields explicit expressions for both exponents, applicable in respective regimes. For instance, in the regime where the shot noise dominates and is relaxed only by (quasi)equilibrium surface diffusion, i.e., $L_f \ll L \ll L_\nu$, we have $\zeta = (4-d)/2$, $z = 4$.[9-11] Asymptotically at $\nu \neq 0$, $D_0 \neq 0$, we recover the well-known Edwards-Wilkinson (EW) result $\zeta = (2-d)/2$, $z = 2$,[21] for which the critical dimension is $d = 2$. Other exponents are possible below L_f. In MBE, L_f may vary by many orders of magnitude due to the Arrhenius law $D_s \sim \omega_D \exp(-E_b/k_B T)$, where $\omega_D \sim 10^{13}$ sec^{-1} is the Debye frequency and E_b the activation energy for nearest-neighbor hopping. Taking $E_b = 1$ eV and $\rho_s \sim 1$ Å^{-2}, a deposition rate $F_0 \sim 1$ Å/sec yields $L_f \ll 1$ Å at room temperature but $L_f \sim 10^3$ Å at 500 °C.

Simple power counting shows that the ν term also dominates the two nonlinear terms in (7) in determining the asymptotic scaling, so that the *asymptotic result of the linear theory remains valid in general.* The remainder of the paper is devoted to the case $\nu = 0$. For a small but finite ν, our discussion is valid in the regime $L \ll L_\nu = (\gamma/\nu)^{1/2}$, keeping in mind that γ is subject to renormalization by the nonlinear terms.

Because of the conserving form assumed by (7), the average growth velocity is not renormalized by surface fluctuations.[22] By going to the comoving frame $Z \to Z + F_0 t$, the constant F_0 drops out, but a phase factor $2\pi F_0 t/a$ appears in the argument of the lattice potential. This suggests that growth averages out the effect of lattice pinning on modes whose lifetime exceeds $\tau_0 = a/F_0$.[16] By invoking dynamic scaling, one may define a length scale $L_F \sim \tau_0^{1/z}$, such that only fluctuations with wavelengths smaller than L_F feel a nonzero υ. The exponent z, of course, is determined by the dynamics below L_F. Equation (7) is now considered separately above and below L_F.

(a) $L > L_F$.—In this regime the continuous translational symmetry in the Z direction is restored. We analyzed Eq. (7) at $\nu = \upsilon = 0$ in a dynamic RG scheme similar to the one applied to the KPZ equation.[4,12] After in-

tegrating out fast modes in the momentum shell $e^{-l}\Lambda_0 \le |\mathbf{k}| \le \Lambda_0$ and performing the rescaling $\mathbf{x} \to e^l \mathbf{x}$, $t \to e^{zl} t$, $Z \to e^{\zeta l} Z$, the coefficients of various terms renormalize as, in a one-loop approximation,

$$d\gamma/dl = [z - 4 + (K_d/4d)g]\gamma , \tag{12a}$$

$$d\sigma/dl = (\zeta + z - 4)\sigma , \tag{12b}$$

$$dD_0/dl = (z - 2\zeta - d)D_0 , \tag{12c}$$

$$dD_1/dl = (z - 2\zeta - d - 2)D_1 , \tag{12d}$$

$$dD_2/dl = (z - 2\zeta - d - 4)D_2 + (K_d/4)(g_0 + g_1 + g_2)D\Lambda_0^{-4} , \tag{12e}$$

where $K_d^{-1} = 2^{d-1}\pi^{d/2}\Gamma(d/2)$, $D = D_0 + D_1\Lambda_0^2 + D_2\Lambda_0^4$, $g_i = D_i\Lambda_0^{d-4+2i}\sigma^2/\gamma^3$, $i = 0,1,2$, and $g = \sum_{i=0}^{2}(6 - d - 2i)g_i$. From (12e) we see that the D_2 term is generated by D_0 and D_1. This term influences the location of fixed points but not the exponents.

Although Eqs. (12) were obtained in a perturbative expansion up to first order in the g_i, it is evident from the RG scheme that (12c) and (12d) are free of higher-order corrections. We thus expect Eqs. (10) and (11) to hold generally (see also Refs. 5, 6, and 10). A more curious result is (12b).[23] Along with (11b) it provides, *within the one-loop approximation,* a confirmation of the exponents proposed by Villain,[10]

$$\zeta = (4 - d)/3, \quad z = (8 + d)/3 . \tag{13}$$

(b) $L < L_F$.—Equation (7) is now considered within the time interval τ_0 over which the surface advances by one layer on average. The strength of the v term on a given length scale is subject to renormalization due to fluctuations on smaller length scales. Here we discuss this effect in a perturbative RG scheme introduced by Nozières and Gallet,[16] including only the γ term, the v term, and the noise on the right-hand side of Eq. (7). This is justifiable since the remaining nonequilibrium terms in the equation could be expected to be less relevant in this regime, and in any case the simplified analysis serves as a starting point for further improvements.

To second order in v, the RG flow equations for the renormalized coefficients after the scaling transformation take the form

$$dv/dl = (z - \zeta - 2 - n)v + O(v^3) , \tag{14a}$$

$$d\gamma/dl = [z - 4 + (\pi^2/2)A_d(n,\kappa)y^2]\gamma , \tag{14b}$$

and $da/dl = -\zeta a$. Here $n = 2\pi^2 K_d(D_0 + D_1)/\gamma a^2$, $y = v/\gamma a$, $\kappa = D_0/D_1$, and $A_d(n,\kappa)$ is a positive function of n and κ in the range of interest. For simplicity we have set $\Lambda_0 = 1$. The two coefficients D_0 and D_1 in the noise correlator renormalize the same way as in (12c) and (12d). In terms of the dimensionless parameters y, n, and κ, we have

$$dy/dl = y(2 - n - cy^2) , \tag{15a}$$

$$dn/dl = n[2 - d_{\rm eff} - (\pi^2/2)A_d y^2] , \tag{15b}$$

$$d\kappa/dl = 2\kappa , \tag{15c}$$

where $d_{\rm eff} = d - 2\kappa/(1+\kappa)$. The exact form for (15c) requires the knowledge of the third-order term in (14a).

Equations (14) and (15) have the familiar look of the Kosterlitz-Thouless RG flow equations.[18] Indeed, by setting $\kappa = 0$ results for the equilibrium sine-Gordon model are correctly reproduced,[15-18] though the dynamics here is purely conservative. Recall that in the equilibrium model, an arbitrarily small v grows under the RG transformation above $d = 2$, so that the surface is faceted at all temperatures. For $d = 2 - \epsilon$, $\epsilon \ge 0$, there is a roughening transition at a finite temperature T_R. As readily seen from Eqs. (15), the nonconserving noise can be accounted for qualitatively (but not quantitatively) by the varying effective dimension $d_{\rm eff}$, such that $d_{\rm eff} = d$ for $\kappa \ll 1$ ($L \ll L_f$) and $d_{\rm eff} = d - 2$ for $\kappa \gg 1$ ($L \gg L_f$). As the length scale is varied, the RG flow samples the phase diagram of the equilibrium problem at various dimensions.[18] In particular, for $d \le 4$, there is the possibility that the RG trajectory starts out in the faceted phase, implying a faceted regime on small length scales, and ends up in the rough phase, meaning that facets of linear size larger than L_f are destroyed by the shot noise.

The faceted regime discussed above in (2+1) dimensions coincides with large terraces and two-dimensional islands observed in MBE experiments. The following argument shows that, no matter how strong the lattice pinning, the surface cannot remain faceted on arbitrarily large length scales under a nonconserving noise at and below (2+1) dimensions. Suppose that the contrary is true. Fluctuations of sufficiently long wavelengths should be limited to the bottom of a particular well of the lattice potential. It then makes sense to expand the sine term in (7) around such a minimum, say $Z = 0$. Neglecting irrelevant nonlinear terms, the resulting equation is the EW type which, however, predicts unbounded height fluctuations for $d \le 2$, inconsistent with our assumption.

It is interesting to speculate on what actually limits the size of faceted areas in MBE. Under the growth condition, the strength of lattice pinning is dramatically weakened on the length scale L_F, at which point surface fluctuations proliferate. Thus, at sufficiently high mobility of adatoms and well below T_R, we expect L_F to be the limiting length for the size of faceted areas on the surface. In the opposite case, the v term never becomes sufficiently strong to produce pinning, as seen from Eqs. (15), so that the faceted regime is altogether absent. In reality, the roughness of the substrate may also influence faceting of the film surface at early times.

To summarize, we presented a detailed analysis of a continuum model which includes all relevant terms compatible with the symmetries and the conservation law for the surface diffusion dynamics in MBE. Explicit expressions for the roughness and dynamic exponents in various scaling regimes were derived, as well as the relevant lengths which separate these regimes. In the physically interesting case $d=2$, fluctuations in the beam intensity were shown to destroy arbitrarily large facets which exist for an equilibrium surface below the thermal roughening temperature.

The behavior of the surface in the faceted regime is not properly described by Eqs. (15). By expanding the pinning potential at $Z=0$, one obtains a diffusive relaxation of fluctuations on the terrace with $z=2$ (see Grinstein and Lee, Ref. 6). On the other hand, such a treatment is inadequate for fluctuations across the wells, i.e., whose amplitude exceeds the layer spacing a.[24] The dynamics in this regime, and in particular how the size of faceted areas diverges as $F_0 \rightarrow 0$, remain as challenging open problems.

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