Growth-Induced Roughening of Crystalline Facets

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The dynamic roughening of a growing crystalline facet is reexamined by including nonlinear effects. A new and unusual fixed point controls the crossover between rough and faceted growth. Unlike linear treatments, the dynamics is no longer diffusive, and an anomalous decrease in mobility is predicted. Implications of these results for recent numerical simulations of growth and experiments on He films are discussed.

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Crystalline facets in equilibrium undergo a *roughening transition*:[1] Smooth facets exist at low temperatures and become rough at higher temperatures. In many cases one is interested in the morphology of a *growing interface*. It is therefore important to examine the dynamic (nonequilibrium) analog of the equilibrium roughening transition (ERT). Chui and Weeks[2] (CW) studied the linear response of a crystalline interface to a small driving force. They found a transition in mobility from activated growth (by formation of steps) in the flat phase to nonactivated growth at high temperatures. In fact, nucleation of steps and islands destroys the flat interface, so that a sufficiently large facet is roughened by any small chemical potential favoring growth. This is clearly seen from numerical simulations, and analytical arguments,[3] applied to polynuclear growth models. A detailed explanation of the resulting crossover behavior near the ERT, and its application to observations of roughening[4,5] on ^{4}He, is provided by Nozières and Gallet (NG). Both CW and NG conclude that, at the ERT, the dynamics is "conventional," in that the mobility stays finite.

In the linearized models of CW and NG, the roughness of the growing interface is similar to a static one, i.e., described by capillary modes, and characterized by logarithmic growth of height fluctuations. However, extensive numerical simulations of various growth models over the past few years[6] have established that nonequilibrium effects introduce a relevant nonlinearity[7] which leads to anomalous power-law growth of height fluctuations. Recently, a number of investigators have looked for a phase transition (possibly the dynamic analog of ERT) in growth models.[8-13] Perhaps the most convincing is the work of Amar and Family[9] (AF), who find evidence for a temperature interval in which the algebraic increase in height fluctuations is replaced by a logarithmic one. This transition has been attributed to an accidental vanishing of the nonlinear term in the growth equation.[14] Curiously, the nonlinearity vanishes close to where ERT is expected.

Lattice effects may play a significant role in these simulations. We also expect growth-induced nonlinearities to be important in experiments on ^{4}He. We thus generalize the studies of CW and NG to include both lattice effects and nonlinearities. The resulting crossover phenomena in the vicinity of the ERT are now controlled by a new and unusual fixed point. Although the lattice effect is renormalized to zero at high temperatures, it persists long enough in the vicinity of ERT to significantly diminish the effective nonlinearity, so that height correlations at the transition point are logarithmic, as in ERT. However, the dynamics is no longer conventional, and an anomalous decrease in mobility is predicted. Unlike linear treatments which predict a discontinuous jump in the mobility, we find that it vanishes continuously at the transition. The mobility μ can be measured indirectly from the angular dependence of the growth velocity, $v(\theta)-v(0)\sim\mu\theta^2$, for small angles.

We describe the interface by its height $h(\mathbf{x},t)$ at time t, above the position $\mathbf{x}$. The simplest local equation describing the evolution of h is

$$\mu^{-1}\frac{\partial h}{\partial t}=F+\nu\nabla^2 h-y\sin(2\pi h)+\tfrac{1}{2}\lambda(\nabla h)^2+\eta(\mathbf{x},t)\,. \tag{1}$$

Here μ is the microscopic mobility of the interface, and F is the driving force proportional to the chemical potential difference. If only the first term is considered, the interface simply moves at a constant velocity $v=\mu F$. The second term describes the smoothening effect of surface tension, and the third term mimics a crystalline potential that prefers integer values of h. Thus h is measured in units of the lattice spacing normal to the surface. The latter two terms are obtained from variations of the sine-Gordon Hamiltonian[15] which describes the ERT. The fourth term is due to nonequilibrium effects,[7] and vanishes when the surface is stationary. (It may also vanish accidentally at isolated points.[9]) Finally, η is a random function describing thermal and growth fluctuations. We consider uncorrelated noise with

$$\langle\eta(\mathbf{x},t)\eta(\mathbf{x}',t')\rangle=2D\delta^2(\mathbf{x}-\mathbf{x}')\delta(t-t')\,.$$

Note that this analysis does not include nonlocal effects such as competitive shadowing or screening, so that this model applies specifically to cases of liquid or vapor

deposition where the overall surface morphology does not affect the local deposition rate.

Two limits of the above equation are well known: For a stationary interface, $\lambda = F = 0$, Eq. (1) is the Langevin equation for the sine-Gordon problem.[15] It describes the equilibrium roughening transition[1] which is dual[2] to the Kosterlitz-Thouless (KT) transition.[16] CW and NG observe that the dynamics is conventional, in that on approaching the transition from the rough side the mobility μ stays finite, and drops to zero only in the flat phase. With $F \neq 0$, nucleation and growth of steps leads to a small, activated mobility for the flat phase.[3] Once the interface acquires a finite velocity $v(F)$, the lattice potential in Eq. (1) averages to zero[4] over a time scale $\tau \sim 1/v(F)$. For longer times the interface is rough, and the true roughening transition is destroyed by a finite force F. However, in many situations (e.g., molecular-beam epitaxy) the characteristic size of facets before roughening becomes evident can be macroscopically large, giving the appearance of a sharp transition from amorphous to layer-by-layer growth. The other limit of Eq. (1) with $y = 0$ and $\lambda, F \neq 0$ describes the dynamic fluctuations of a growing interface.[7] The nonlinearity changes the logarithmic increase of height fluctuations to algebraic.[6]

Crossover between rough and flat growth, as calculated by CW and NG, is governed by the dynamics of the sine-Gordon equation. We include nonlinear effects in a dynamic renormalization-group (RG) approach by simply adding up the recursion relations obtained in the above two limits[4,7] in the vicinity of ERT ($y = \lambda = 0$, $\mu D/\nu = 2/\pi$). Under a change of scale, $x \to e^{l}x$, $t \to e^{2l}t$, $h \to h$, the parameters in Eq. (1) renormalize as

$$\frac{d\ln D}{dl} = \frac{\lambda^2}{4\pi^2\nu^2} + c_1\frac{y^2}{\nu^2}, \quad \frac{d\ln y}{dl} = 2 - \frac{\pi\mu D}{\nu} + c_3\frac{y^2}{\nu^2},$$
$$\frac{d\ln\lambda}{dl} = 0, \quad \frac{d\ln\nu}{dl} = c_2\frac{y^2}{\nu^2}, \quad \frac{d\ln\mu}{dl} = -c_1\frac{y^2}{\nu^2}. \tag{2}$$

Formally, Eqs. (2) are obtained by treating y and λ perturbatively, and including all terms to *second order*. For $y = 0$, the only $O(\lambda^2)$ correction is to D, which signals the marginal relevance[7] of λ. Terms proportional to λy are excluded by symmetry of Eq. (1) under $y \to -y$ or $\lambda \to -\lambda$. Although $O(y^2)$ corrections depend somewhat on the precise RG scheme, the flow diagram is only sensitive to relative values of these coefficients at ERT. We use $c_1 \approx 4.6$, $c_2 \approx 1.9$ from NG, and $c_3 = 5/4\pi$ from Ref. 15. Two new terms could have arisen, at $O(\lambda^2)$ to y, and at $O(y^2)$ to λ, but we calculate both to be zero.

Finally, the recursion relation $dF/dl = 2F + \pi\lambda$ indicates that a finite F is always relevant, and ultimately averages y to zero at sufficiently long length scales. NG use a "stopped RG" procedure in which y is simply set to zero at this scale. Following NG, we focus on the behavior of Eqs. (2) only at smaller distances. We examine the RG flows near the ERT point by setting $t = 2\pi\mu D/\nu - 4$, $\bar{y} = 2(c_2)^{1/2}y/\nu$, and $\bar{\lambda} = \lambda/\pi\nu$. Then to lowest order in $(t, \bar{y}, \bar{\lambda})$, we have

$$\frac{dt}{dl} = \bar{\lambda}^2 - \bar{y}^2, \quad \frac{d\bar{y}^2}{dl} = -\bar{y}^2[t + \beta\bar{y}^2], \quad \frac{d\bar{\lambda}^2}{dl} = -\alpha\bar{\lambda}^2\bar{y}^2, \tag{3}$$

with $\alpha = 0.5$ and $\beta \approx 0.4$. For $\bar{y} = 0$ and finite $\bar{\lambda}$, the temperaturelike variable $t \approx T - T_c$ goes to infinity indicating relevance of the nonlinearity. For $\bar{\lambda} = 0$, we have the usual KT equations,[16] with a phase transition governed by the fixed point at $t = \bar{y} = 0$.

The new feature in Eqs. (3) is that a finite lattice potential ($\bar{y} > 0$) diminishes the effective nonlinearity $\bar{\lambda} \sim \lambda/\nu$. The origin of this effect is that the lattice potential tends to stabilize the surface by increasing ν [see Eqs. (2)]. In the three-parameter space $(t, \bar{y}, \bar{\lambda})$, a two-dimensional separatrix, $\bar{y}^2 = \bar{\lambda}^2 + \bar{y}(t + \beta\bar{y}^2 - \alpha\bar{\lambda}^2) + O(\bar{y}^4)$, actually flows to the ERT fixed point ($t = \bar{y} = \bar{\lambda} = 0$). This is quite surprising since the fixed line at $\bar{y} = \bar{\lambda} = 0$ is unstable for $t < 0$ to $\bar{y}$ and to $\bar{\lambda}$ for all t. On the high-temperature side of the boundary, $\bar{y}$ itself is rapidly driven to zero, while $\bar{\lambda}$ persists, so that RG flows go to a sink at $t \to \infty$, $\bar{y} = 0$ (rough). On the other side, the opposite occurs and flows go to $t \to -\infty$, $\bar{\lambda} = 0$ (flat). Flows starting on the separatrix rapidly (i.e., exponentially) decay towards a special line, the "spine," given to lowest order by $\bar{\lambda}^2 = \bar{y}^2 = t/(\alpha - \beta)$. Along the spine, $\bar{y}$ decreases just slowly enough to drive $\bar{\lambda}$ to zero as $\bar{\lambda}^2 = \bar{y}^2 = \bar{y}_0^2/(\alpha\bar{y}_0^2 l + 1)$ ($\bar{y}_0$ is the value of $\bar{y}$ as the RG trajectory approaches the spine). Asymptotically $\bar{y}^2 \sim 1/l$, as compared to $\bar{y} \sim 1/l$ along the usual KT separatrix. Since $d\ln\mu/dl \sim -\bar{y}^2$, this slow convergence of $\bar{y}$ to zero causes the mobility μ to vanish at the transition. Also the true asymptotic behavior does not occur immediately, but is preceded by a long interval in which $\bar{y}(l)$ is approximately constant.

Figure 1 indicates how the roughening transition temperature (for a fixed initial $\bar{y}$) is modified by $\bar{\lambda}$ [for small

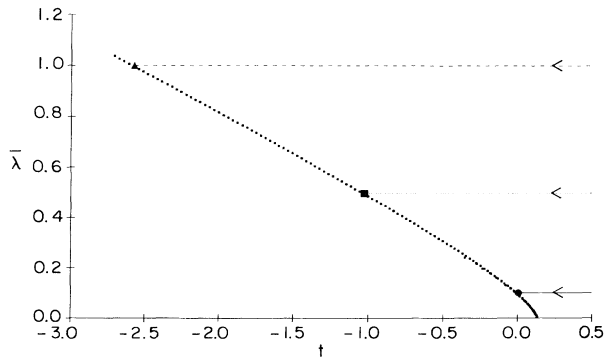

FIG. 1. The decrease in the roughening transition temperature with increasing $\bar{\lambda}$. The horizontal lines indicate the paths used in Fig. 2.

$\bar{\lambda}$, not apparent in Fig. 1, $T_c(\bar{\lambda}) - T_c(0) \sim \bar{\lambda}^2$]. Figure 2 shows the renormalized mobility [the $l \to \infty$ limit of $\mu(l)$] as a function of temperature for selected values of $\bar{\lambda}$. The discontinuous jump[2,4] for $\bar{\lambda}=0$ is replaced by a continuous approach to zero, which becomes more apparent at larger $\bar{\lambda}$. Also included in Fig. 2 is the renormalized value of $\bar{\lambda}$, which vanishes at the transition. Asymptotically close to T_c, the correlation length ξ diverges as $\ln\xi \sim (\ln|T-T_c|)^2$. However, in numerically integrating Eqs. (3) we found that the true asymptotic behavior is observed only at very small reduced temperatures $|T-T_c| \approx \exp(1/\sqrt{a}\bar{y}_0)$. This follows from $\bar{y}(l)^2 = \bar{y}_0^2/(a\bar{y}_0^2 l + 1)$, since the asymptotic limit $\bar{y}^2 \sim 1/al$ is observed only for $l > 1/a\bar{y}_0^2$. For higher reduced temperatures, there is an apparent nonuniversal divergence, $\xi \sim |T-T_c|^{-1/\bar{y}_0}$. The dynamic crossover length between these two regimes, $\xi_{\rm dyn}$, is much larger than the corresponding static crossover length $\xi_{\rm st}$. In fact, $\xi_{\rm dyn} \approx \xi_{\rm st}^{\ln\xi_{\rm st}}$, which may be astronomically large in realistic cases, rendering the observation of true criticality impossible. The renormalized parameters in Eq. (1) have nonanalytic dependences on $\ln|T-T_c|$, so that, for example, μ vanishes as $\mu \sim (\ln|T-T_c|)^{-c_1/2c_2\alpha}$, and ν diverges as $\nu \sim (\ln|T-T_c|)^{1/2\alpha}$. At T_c, the height-height correlation function is

$$C(L,\tau) = \langle [h(\mathbf{x},t) - h(\mathbf{x}',t')]^2 \rangle = \frac{2}{\pi^2} \ln L\, g\left(\frac{\tau}{L^2 (\ln L)^{(c_1-c_2)/4ac_2}} \right), \quad (4)$$

with a similar expression for the surface width ($L = |\mathbf{x} - \mathbf{x}'|$, $\tau = |t-t'|$, and the scaling function g has the usual limits).[12] The static behavior is thus unchanged from the usual roughening transition,[1] while there are logarithmic corrections to the diffusive behavior predicted by conventional dynamics.[2,4,12]

In $d = 2+\varepsilon$ dimensions, the recursion relations are modified simply by the addition of $-\varepsilon$ [-4ε] to the first equation in (2) [(3)]. For $\varepsilon < 0$, the phase boundary is controlled by a fixed point at $\bar{y} = 2\sqrt{-\varepsilon}$, $\bar{\lambda} = t = 0$. Since $\bar{\lambda}$ is irrelevant at this fixed point, the analysis of the static roughening transition (dual to the Coulomb gas[17]) applies. At $O(\sqrt{-\varepsilon})$, $\xi \sim t^{-\nu}$, where $\nu = 1/2\sqrt{-\varepsilon}$, and $C(L,\tau) = (2/\pi^2) \ln L\, g(\tau/L^z)$ at the transition, with $z = 2 + \varepsilon(c_2 - c_1)/c_2$. For $\varepsilon > 0$, a line of critical points appears at $\bar{\lambda} = 2\sqrt{\varepsilon}$, $\bar{y} = 0$, $t \geq 0$. Since the $O(\bar{\lambda}^2)$ correction to ν in $d=2$ vanishes, $z=2$, and to lowest order the height-height correlation function decays logarithmically at T_c, but with a nonuniversal prefactor, i.e., $c(L,\tau) = (1/\pi K_{\rm eff}) \ln L\, g(\tau/L^2)$. This form was actually suggested for $d \geq 2$ by the numerical simulations of Forrest and Tang[12] (FT) in three dimensions ($\varepsilon = 1$). For the conjecture to be generally valid, all higher-order corrections to z must vanish. Since lattice effects are present in most numerical simulations,[10,12,13] and are immediately relevant upon entering the flat phase, the results should be interpreted accordingly. For example, FT observe[12] the onset of sublattice ordering below the transition, which is probably a lattice effect. They also note that the equilibrium width approaches its asymptotic value with a $1/L$ correction, though such a correction is cut off by lattice effects.

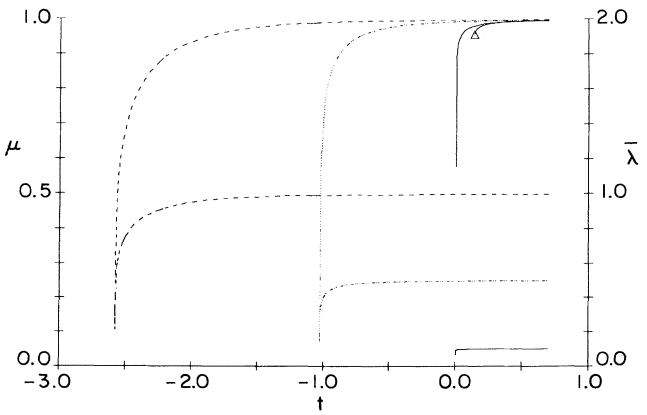

FIG. 2. The renormalized mobility and $\bar{\lambda}$ for the temperature scans indicated in Fig. 1.

We now discuss the broader implications of our results on numerical simulations of kinetic phase transitions, and the extent to which these transitions are related to ERT. The sharp morphological transition we have discussed blurs once the average motion of the interface is included, so that the transitions observed in $d=2$ in a number of recent simulations[10,11,13] are probably crossover remnants. As a "temperaturelike" control parameter is varied in the simulations, the coefficients ν, λ, y, and D in Eq. (1) change, so that their trajectory, probably, crosses the effective phase boundary discussed above. To better observe the predicted crossover phenomena, an independent "forcelike" parameter controlling the average velocity is useful. Crossover effects should become more pronounced as this parameter is reduced. Furthermore, by setting the forcelike parameter to zero, one can probe ERT by varying the temperaturelike variable. Thus, using both controls, a continuous sequence of nonequilibrium roughening transitions terminating at ERT can be obtained.

The work of Guo, Grossman, and Grant[11] (GGG) most closely relates to this theory since by setting their bias λ_a to zero, ERT is recovered. In the case of FT, $p^+ - p^-$ is the forcelike term, but an independent temperaturelike parameter is needed to see ERT. Since a signature of our predictions is an anomalous decrease in mobility, Ref. 13 is of particular interest. There, a new quantity, the compacity, which is proportional to the mobility, is found to exhibit singular behavior at the transition. It is important to note that in some simulations, the time axis is measured as the average height, which automatically scales out the mobility. Time measured in

units of trial updating steps, for example, is probably more realistic, and pertinent to our work. The model used by Amar and Family[9] is somewhat different in that the parameter λ in Eq. (1) has opposite signs at high and low temperatures,[14] and must vanish at some point. Since λ is not renormalized, this signals a true morphological transition, even in the presence of a finite force. However, since the critical surface is symmetric for the effective nonlinearity $\bar{\lambda} \rightarrow -\bar{\lambda}$, which is renormalized, it is possible that the trajectory of their scans crosses the phase boundary twice, rather than once. That is, on scanning λ from positive to negative values, a closed interval, intervening between two rough phases and surrounding $\lambda = 0$, exists where the mobility vanishes and the surface is flat. In fact, rather than occurring at an isolated point, the transition in AF appears to spread over an interval in temperature. However, more simulations are clearly necessary to clarify the above interpretations. Finally, although some simulations suggest that roughness exponents differ across the kinetic roughening transition, we note that on the flat side of the transition, characteristic length scales are quite large,[3] so that finite-size effects may lead to the appearance of a changing exponent.

Experimental data on the roughening transition of growing ^{4}He surfaces[5] have been fitted[4] by crossover forms obtained from Eqs. (2) with $\lambda = 0$. We suspect that including the effects of a finite λ in the "stopped RG" scheme of NG would improve the accuracy of these fits. A more complete calculation of crossover curves for mobility and $\bar{\lambda}$ will be left for the future. In our notation, the quantity $\lambda\mu$ can be determined from the dependence of the growth velocity on angle,[14] or from the parabolic shape of the growing crystals. Recent experiments[18] indicate that addition of very small concentrations of ^{3}He has dramatic effects on the roughening transition, and leads to pronounced irreversibility in growth and melting shapes. A macroscopic interpretation of this phenomenon could be an increased coefficient λ in Eq. (1). This system could thus provide a fruitful experimental ground for testing the above predictions for growth-induced roughening.

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