Current-Induced Faceting of Crystal Surfaces

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A crystal surface can be destabilized by an external field which induces the directed migration of adatoms. We specify the conditions of instability in terms of the orientation dependence of the adatom mobility and determine the resulting faceted state as the stationary solution of a nonlinear evolution equation for the surface profile. Coarsening of the facet structure is studied using an atomistic solid-on-solid model, and the analogy to spinodal decomposition is emphasized.

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Introduction.—Crystal surfaces can undergo faceting transitions through a variety of mechanisms. As was first shown by Herring [1], a surface is thermodynamically unstable to the formation of a hill-and-valley structure if its orientation does not appear in the equilibrium crystal shape. The kinetics of this thermal faceting transition has been the subject of several recent experimental [2] and theoretical [3] studies. Other mechanisms for faceting include chemical absorption [4] and the deposition of material from an atomic beam [5]. In the latter case, the surface is destabilized by nonequilibrium surface diffusion currents induced by the external particle flux [6].

In this Letter we consider the nonequilibrium effects on the stability and morphology of a crystal surface which arise through the directed migration of adatoms under the influence of an external field. Our work is motivated by recent experiments, which have revealed the striking influence of a dc heating current in the bulk of a sample upon morphological transformations (such as reconstruction [7], faceting, and step bunching [8,9]) at the surface. These observations have been interpreted in terms of the surface electromigration of adatoms along the current direction [7,8], a phenomenon which is directly accessible to electron microscopy studies [10].

Previous theoretical studies of current-induced surface instabilities [11] have focused on the interaction of the (continuous) adatom density with surface steps and the resulting step dynamics. Here, we employ two complementary approaches. First, we formulate a continuum theory to describe the surface on scales large compared to the interstep distance. Within the continuum theory the precise conditions for surface stability are particularly transparent and can be derived from simple linear analysis. Not surprisingly, the stability of a given surface orientation is governed by the orientation dependence of the adatom mobility μ. To follow the morphological instability beyond the linear regime and predict the orientations that appear in the final, faceted state, the full nonlinear evolution equation for the surface has to be invoked. This will be detailed below. It turns out that the selection of well-defined stable facet orientations is possible only if certain conditions on the orientation dependence of the adatom mobility are met. Once the facet orientations are established locally, further evolution proceeds through a coarsening process which can be analyzed in the spirit of the continuum theory of spinodal decomposition [12,13].

In the second part of this paper we introduce an atomistic solid-on-solid model in which the electromigration forces appear as a bias in the jump rates of adatoms. This model goes beyond conventional step dynamical approaches [11] in that it includes the fluctuations which arise from the particulate nature of the adatom fluxes. One important fluctuation effect is the destabilization of surface orientations which, according to linear stability analysis, would be regarded as stable. By analogy with spinodal decomposition [13], fluctuations are also expected to affect the coarsening of the facet structure. We provide a link between continuum and atomistic modeling by showing how the facet structure selected by the solid-on-solid dynamics can be predicted from the continuum equation.

Since the external field singles out one direction in the plane, our discussion will be restricted to surface profiles which vary only in one dimension. Inclusion of the transverse dimension in the atomistic simulations leads to additional effects which are briefly mentioned at the end of the paper. In the interest of simplicity we further neglect the desorption of adatoms. While sublimation may play a role in some of the experimental systems [7–9], it is clearly not necessary for the appearance of current-induced instabilities [11]; moreover, it is straightforward in principle to include it in our analysis.

Continuum theory.—The one-dimensional surface profile is described by a height function $h(x,t)$, the external field being oriented along the x axis. Volume conservation implies that h satisfies a continuity equation

$$h_t + J_x = 0, \tag{1}$$

the subscripts denoting partial derivatives. In equilibrium the current J is proportional to the derivative of the surface chemical potential with respect to the arclength along the surface [14]. The external field E contributes a linear term Ex to the local chemical potential. The total

current is therefore

$$J = \frac{\mu(h_x)}{\sqrt{g}}\left[\sigma\left(\frac{h_x}{\sqrt{g}}\right)_{xx} + E\right], \tag{2}$$

where $g = 1 + (h_x)^2$, and the surface tension σ has for simplicity been assumed to be isotropic. Expanding (1) and (2) around a uniformly tilted surface $h = mx$ with $m = \tan\theta$, we find the linear growth rate for a perturbation with wave number q,

$$\omega(q) = -\frac{E}{\sqrt{1+m^2}}\left(\frac{\mu(m)m}{1+m^2} - \mu'(m)\right)q^2 - \frac{\mu(m)\sigma}{(1+m^2)^2}q^4, \tag{3}$$

with $\mu' = d\mu/dm$. The stability is determined by the sign of the q^2 term. Under isotropic conditions $\mu' \equiv 0$, and we see that surfaces with a negative tilt ($m < 0$) are unstable. A surface oriented parallel to the field ($m = 0$) would appear to be stable; however, we expect that thermal fluctuations will tilt portions of the surface into the unstable regime, thereby inducing a faceting transition also in this case.

In general, the stability of a surface parallel to the field is seen to depend on the sign of $E\mu'(0)$. Clearly $\mu'(0)$ is expected to be nonzero for vicinal surfaces; an explicit expression for a simple step train model was derived by Stoyanov [11]. For singular orientations $\mu'(0) = 0$ by symmetry, and the $m = 0$ surface is stabilized by the surface tension term in (3). As argued above, fluctuations will nevertheless generate slightly misoriented regions which then become unstable. The instability affects negatively tilted ($m < 0$) regions if $E[\mu(0) - \mu''(0)] > 0$ and positively tilted ones otherwise.

It is worth pointing out that the external field has an important effect on the fluctuations of the surface even when it does not cause any instability. We recognize from (3) that a stabilizing field changes the small q behavior of the dispersion relation from the $\omega \sim q^4$ law characteristic of thermal equilibrium [14] to $\omega \sim q^2$. Adding a thermal noise term $(J_T)_x$ to the right-hand side of (1) and analyzing the resulting linear Langevin equation one finds that the field completely suppresses the thermal roughness of the surface [15]. Since the orientations that appear in the faceted state are in the *stable* regime of (3), the facets are, therefore, smooth.

We now ask what stable surface profiles can evolve beyond the linear instability. To answer this question it is convenient to introduce the field $\phi = h_x/\sqrt{g} = \sin\theta$, $-1 \le \phi \le 1$. The stationarity condition $J \equiv J^* = \text{const}$ then becomes equivalent to Newton's equation for the "particle coordinate" ϕ moving in "time" x, the "potential" $V(\phi)$ being determined by the orientation dependence of the mobility, through

$$\sigma\phi_{xx} = \frac{J^*}{\mu(\phi)\sqrt{1-\phi^2}} - E \equiv -V'(\phi). \tag{4}$$

To describe the faceted state we look for periodic trajectories of large period L, subject to the additional constraint

$$h(L) - h(0) = \int_0^L dx\,\phi(1-\phi^2)^{-1/2} = 0, \tag{5}$$

which implies that on average the surface is parallel to the field. Solutions of (4) with period $L \to \infty$ exist only if the particle spends most of its time at a position ϕ^* where $V'(\phi^*) = 0$; one readily verifies that in order to correspond to a stable facet orientation, ϕ^* has to be a local maximum, $V''(\phi^*) < 0$.

There are, in general, two conceivable types of solutions. In the first case, the particle starts at a local maximum ϕ^*, rolls downhill toward one of the boundaries $\phi = 1$ or $\sigma = -1$, where it is reflected in a (short) time of order $(\sigma/E)^{1/2}$, and returns to ϕ^*. This *bounce* solution corresponds to a single facet orientation $\theta^* = \arcsin\phi^*$ coexisting with a macrostep, i.e., a localized region where the surface profile is almost vertical (on a real surface these regions would manifest themselves as disordered step bunches [9]). In the second case the particle moves between two degenerate maxima $\phi^*_{1,2}$, a *kink* solution corresponding to the coexistence of two well-defined facet orientations. The stationary current J^* is fixed by the requirement that $V(\phi^*) = V(1)$ or $V(-1)$ for the bounce and $V(\phi^*_1) = V(\phi^*_2)$ for the kink, and it always satisfies $J^* = E\mu(\phi^*)\sqrt{1-(\phi^*)^2}$.

For concreteness, let us consider a singular surface of a crystal with fourfold symmetry, such that $\mu(\theta) = \mu_0 + \mu_1\cos 4\theta$. In the isotropic limit, $\mu_1 = 0$, the potential has at most one maximum (for J^* such that $0 \le J^*/E \le \mu_0$), so only bounce solutions are possible; an explicit calculation shows, however, that due to the divergence of $V'(\phi)$ at $\phi = \pm 1$, the constraint (5) cannot be satisfied for $L \to \infty$. Thus, for small anisotropy *no stable inhomogeneous stationary solutions exist.* This is confirmed by the numerical integration of the full time-dependent problem (1,2), which shows the development of macrosteps of ever increasing height [16]. A finite amount of anisotropy has to be introduced in order to produce several local maxima in $V(\phi)$. For $\mu_1 < 0$, two maxima appear when $\mu_1/\mu_0 < -\frac{1}{17}$; this condition turns out to be *sufficient* for the existence of a kink solution. The time-dependent numerical simulation confirms that this solution is indeed dynamically selected. In contrast, for $\mu_1 > 0$, kink solutions are viable only for μ_1/μ_0 close to unity [16].

Atomistic modeling.—The microscopic approach is based on a standard solid-on-solid (SOS) model for surface diffusion. The (one-dimensional) surface is represented by integer height variables $h(i)$ with $i = 1, \ldots, L$ and periodic boundary conditions. The energy of a configuration is given by the Hamiltonian

$$\mathcal{H} = \sum_{i=1}^{L} |h(i+1) - h(i)|. \tag{6}$$

In an elementary surface diffusion step, an atom moves from a randomly chosen site i to one of the neighboring sites $i \pm 1$, e.g., $h(i) \to h(i) - 1$ and $h(i+1) \to h(i+1) + 1$. This occurs at rate $\Gamma = \min(1, e^{-\Delta\mathcal{H}/T})$, where $\Delta\mathcal{H}$ is

the energy difference between the final and the initial state of the surface, and T denotes temperature. In equilibrium, the direction of an attempted move is chosen with equal probability to the left ($i \to i - 1$) or to the right ($i \to i + 1$). In the presence of an external field the moves are biased: Moves to the right are attempted with probability p, to the left with probability $1 - p$, where $p > \frac{1}{2}$.

Figure 1 shows the result of a simulation of the biased dynamics. One observes the formation and coarsening of an array of bounces—facets of a well-defined orientation separated by macrosteps (or step bunches). To understand this result, we need to somewhat modify the continuum theory formulated above. The central observation is that, in the SOS geometry, the surface diffusion current is driven by *horizontal* chemical potential differences rather than differences measured along the arclength of the surface (vertical potential differences have no meaning in these models). Therefore the factor $\sqrt{g}^{-1}$ in front of the square bracket in (2) should be omitted when describing an SOS model. This has two important consequences. First, the prefactor of the q^2 term in the dispersion relation (3) becomes simply $E\mu'(m)$, showing that the external field has a (stabilizing or destabilizing) influence on the dynamics only if the mobility is explicitly orientation dependent. Second, the potential that appears in the particle equation (4) is now defined through $V'(\phi) = E - J^*/\mu(\phi)$, which remains finite at $\phi = \pm 1$; this guarantees the existence of bounce solutions in cases where $V(\phi)$ has only a single maximum.

To actually predict the facet orientation that is selected in Fig. 1, we would have to know the orientation-dependent adatom mobility. In general, this quantity is

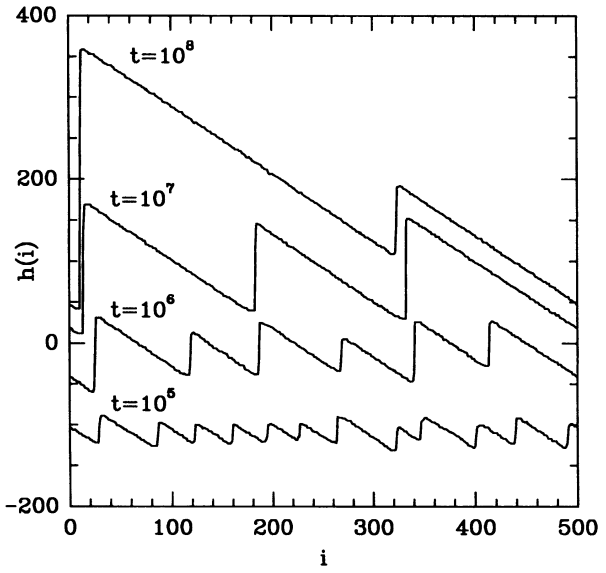

FIG. 1. Current-induced faceting in the biased one-dimensional SOS model at temperature $T = 0.5$ and bias $p = 0.7$. The times (in attempted moves per site) are indicated above the profiles. Subsequent profiles have been displaced by 100 lattice units in the vertical direction.

given by a Green-Kubo formula, which involves an integral over certain dynamical correlation functions, and is therefore difficult to explicitly evaluate [17,18]. The Green-Kubo formula shows, however, that the mobility is always bounded from above by the ensemble-averaged jump rate $\langle\Gamma\rangle$. It is straightforward to calculate $\langle\Gamma\rangle$ for the present, one-dimensional model, as a function of orientation and temperature. One obtains a symmetric function with a single minimum at zero orientation and the limiting value $\langle\Gamma\rangle = 1$ at large positive or negative slopes. Numerical measurement [18] of the adatom mobility shows that its qualitative behavior is well represented by $\langle\Gamma\rangle$. For actual calculations we use the form $\mu(\phi) = 1/(a - b\phi^2)$, with $a > b$; this has the advantage of producing a purely cubic potential $V(\phi)$, for which the bounce trajectory (in the limit $L \to \infty$) can be explicitly computed. One finds that the selected orientation is $\theta^* = \arcsin(-\frac{1}{2}) = -30°$ independent of a and b, which is rather close to the numerical value $\theta^* \approx -35°$.

We have argued above that a singular surface, while being marginal in the sense of linear stability analysis, will become unstable due to thermal fluctuations. This is clearly confirmed by the simulation shown in Fig. 1, which starts from a flat (singular) surface. It is natural to ask how far into the linearly stable regime this fluctuation-induced destabilization mechanism will be effective. Simulations of tilted surfaces [16] indicate a simple answer: At long enough times any surface with average orientation $\theta > \theta^*$ breaks up into an array of facets (of orientation θ^*) and macrosteps. In analogy with phase separation in equilibrium systems [2,3] we may therefore conclude that $\theta = 0$ plays the role of the *spinodal* for the faceting transition, while the *phase boundary* is located at θ^*.

Coarsening.—By inspection of Fig. 1, one estimates that the typical facet size l doubles when time increases by a factor of 10, corresponding to a coarsening law $l \sim t^n$ with $n \approx \ln 2/\ln 10 \approx 0.3$. A more quantitative analysis, involving measurements of both the average distance between macrosteps and the position of the first zero of the real space height-height correlation function, indicates that $n \approx \frac{1}{4}$. It is tempting [3] to relate this behavior to spinodal decomposition (SD) in one dimension [12,13], given that the dynamical equation for the "order parameter" $\phi(x,t)$ (written here in the form appropriate for the SOS geometry)

$$\phi_t = -(1 - \phi^2)^{3/2}[\mu(\phi)(\sigma\phi_{xx} + E)]_{xx} \qquad (7)$$

is similar, but not identical, to a conserved Ginzburg-Landau model [13] (note, in particular, that ϕ is not conserved, but $\phi/\sqrt{1 - \phi^2}$ is). Langer's [12] approach to SD is based on a linear stability analysis of the stationary inhomogeneous order parameter profile of given periodicity L; the lifetime of the profile provides the time scale for further coarsening when the typical domain size is of order L. In our context, this implies investigating the eigenmodes of (7) linearized around the stationary bounce

solution $\phi_b(x)$. The eigenvalue problem becomes

$$-[1-\phi_b(x)^2]^{3/2}\frac{\partial^2}{\partial x^2}\mu[\phi_b(x)] \times\left[-\sigma\frac{\partial^2}{\partial x^2}+W(x)\right]\psi=\gamma\psi, \quad (8)$$

where, for our choice $\mu(\phi)=1/(a-b\phi^2)$, $W(x)=-2bJ^*\phi_b(x)$. By translational invariance, $\psi_b=d\phi_b/dx$ is an eigenfunction with eigenvalue $\lambda=0$. However, in contrast to standard SD, ψ_b has a node and is, therefore, not the ground state of the Schrödinger operator in the square brackets in (8), which peaks at the minima of $W(x)$. Consequently, the coarsening of a periodic array of bounces does not proceed through a translation of domain walls but rather by the evaporation of single peaks (i.e., macrosteps), the surviving part of the structure remaining essentially fixed in space. This is clearly seen in a numerical solution of (7) (Fig. 2). To determine the actual coarsening law one has to analyze the full linear operator (8), which has not yet been accomplished. In standard one-dimensional SD, the coarsening is logarithmic at zero temperature [12], while thermal fluctuations give rise to a power law $\ell\sim t^{1/3}$ [13]; at present, we are unable to decide whether a similar scenario applies here.

Conclusion.—In this paper, we have explored several aspects of the morphological instability of crystal surfaces induced by electromigration currents. Much remains to be done. Most importantly, the modeling should be extended to two-dimensional geometries. On the basis of symmetry arguments, one expects that the field will induce an inclination-dependent current also in the transverse direction, in analogy to the effect of the particle flux in deposition processes [6]. Preliminary simulations [16] of a two-dimensional atomistic model indicate that this leads, in addition to the step bunches obtained in the one-dimensional case (Fig. 1), to a macroscopic tilting of the surface in a direction *perpendicular* to the external field; this type of behavior has in fact been observed on the Si(111) surface [9]. A second open question concerns the behavior of real surfaces in situations where the continuum equations do not permit any stable, faceted solution. Finally, our somewhat naive approach of describing the heating current through a constant, homogenous external field may have to be modified, once the microscopic mechanism that allows the adatoms to become entrained with the current is better understood.

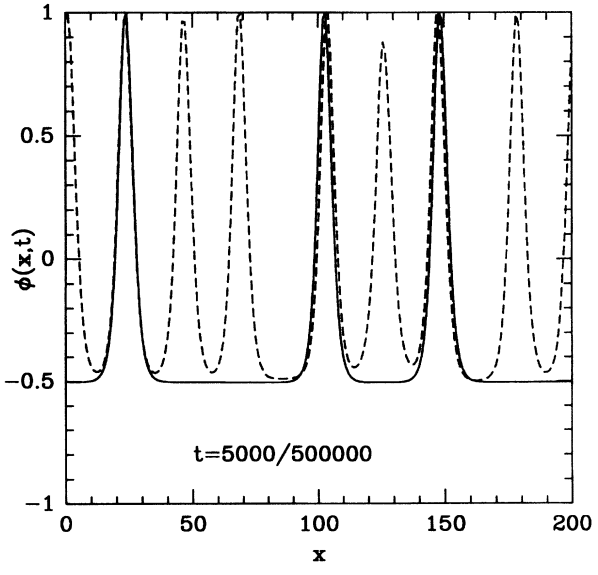

FIG. 2. Coarsening in the continuum model (7) with parameters $E=0.5$, $\sigma=1$, and $\mu(\phi)=1/(2-\phi^2)$. The equation was discretized and integrated forward in time with a time step of 0.05. The initial condition was random with average $\langle\phi\rangle=0.3$, inside the unstable regime. The figure shows the order parameter profile at times $t=5000$ (dashed) and $t=500\,000$ (full). The peaks correspond to macrosteps, the flat regions $\phi=\phi^*=-0.5$ are the facets.

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