Kinetic Roughening of Vicinal Surfaces

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The description of growth at vicinal surfaces leads to an anisotropic generalization of the Kardar-Parisi-Zhang equation [Phys. Rev. Lett. **56**, 889 (1986)] which is investigated by a dynamical renormalization calculation. If the nonlinear terms have opposite signs parallel and perpendicular to the average step direction, the roughness is only logarithmic. This should be the case, e.g., for step-flow growth. As the temperature is lowered so that island formation on the terraces becomes significant, a sharp morphological transition to algebraic roughness is predicted.

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The phenomenon of kinetic roughening [1], i.e., the emergence of scale-invariant surface fluctuations in growth processes, has attracted a lot of interest for two reasons: It is of technological relevance and, at the same time, it provides a paradigm for the nonequilibrium generalization of dynamical scaling [2].

In order to be able to design surfaces with special properties, a roughness tuned by the mode of growth may be desirable [3]. However, the more typical situation, e.g., in thin-film growth, is that one wants to have as perfectly flat a surface as possible. So far most theories of kinetic roughening predict [1] that the root-mean-square surface fluctuations increase algebraically with deposition time, and that the morphology of the surface develops coarser and coarser features characterized by a length ξ parallel to the substrate with

$$(\overline{h^2}-\bar{h}^2)^{1/2}\sim\xi^{\zeta} \text{ and } \xi\sim t^{1/z}. \quad (1)$$

These asymptotic power laws define the roughness exponent ζ and the dynamical exponent z. Hence, in three dimensions, the kinetic roughness is predicted [4,5] to be much stronger than the thermal one which is only logarithmic or, below the roughening temperature, even nonexistent [6].

Being isotropic, none of the models applies to the growth of vicinal surfaces, where the tilt direction is special, at least as long as the formation of islands on the terraces does not render the excess steps determining the overall tilt dynamically unimportant. Therefore the question arises if anisotropy is relevant in the sense of giving rise to a different scaling behavior from that predicted previously.

From the theoretical point of view, kinetic roughening has recently been considered in the more general context of self-organized criticality [7]. This term means the emergence of generic scaling behavior in driven systems. It is well known that the lifetime of a perturbation diverges with the wavelength for systems with a conservation law or a spontaneously broken continuous symmetry [8]. Such a system may therefore be called marginally stable: Every perturbation decays, but the characteristic time involved may become arbitrarily long. Examples are sandpile models (mass conservation) or models of kinetic roughening (broken translational invariance perpendicular to the surface). If such a system has nonlinear dynamics and is driven into a steady nonequilibrium state, the perturbations organize themselves in a way which in general implies nontrivial scaling. In sandpile models, anisotropy has been shown to be relevant [9,10]—a hint that the growth of a vicinal surface may also be in a different universality class from ordinary growth models.

In this Letter I show that this is indeed the case, e.g., for growth in the step-flow mode where kinetic roughening turns out to be only logarithmic. If island growth on the terraces becomes important, a sharp morphological transition is obtained to the much stronger algebraic roughness.

The following model [5] will be considered. The growth starts from a vicinal surface with an equidistant array of parallel, monoatomic steps separating high-symmetry terraces. Atoms are deposited and diffuse on these terraces until they find a step or desorb. Only when an adatom reaches a step does it become bound and contribute to the growing film. Island formation on the terraces is neglected for the moment. This corresponds to an idealized step-flow growth mode [11]. The fraction of adatoms reaching a step is then proportional to the step density, if the surface is not tilted too much with respect to the high-symmetry orientation of the terraces, and it saturates for large tilt angles, where hardly any desorption occurs anymore, because most adatoms are readily absorbed by steps. Hence one expects a qualitative dependence of the growth velocity κ on the step density [12] or tilt angle s as in Fig. 1.

Ignoring the fluctuations, the vicinal surface with slope $\bar{s}$ and growth velocity $\kappa(\bar{s})$ is simply described by $H_0(\mathbf{x},t)=\bar{s}x_{\parallel}+\kappa(\bar{s})t$. Here, the height H_0 is measured perpendicularly, and the coordinates $\mathbf{x}=(x_{\parallel},x_{\perp})$ are measured parallel to a terrace, $x_{\parallel}$ ($x_{\perp}$) being parallel (perpendicular) to the tilt direction. In the presence of noise, the height H of the surface deviates from this reference surface by $h=H-H_0$, which evolves in time according to the stochastic differential equation

$$\partial_t h=\kappa[(\bar{s}+\partial_{\parallel}h)^2+(\partial_{\perp}h)^2]^{1/2} -\kappa(\bar{s})+\nu_{\parallel}\partial_{\parallel}^2 h+\nu_{\perp}\partial_{\perp}^2 h+\eta, \quad (2)$$

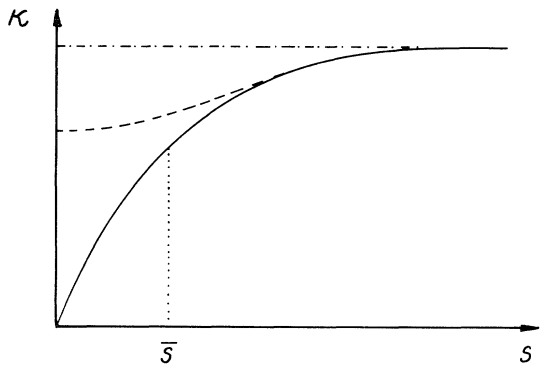

FIG. 1. Schematic dependence of the growth velocity κ on the tilt s. Solid line: only attachment to the steps taken into account. Dashed line: island formation on sufficiently large terraces included. Without desorption the growth velocity becomes independent of the tilt in this model (dot-dashed line).

where ∂_t, $\partial_\parallel$, and $\partial_\perp$ denote the partial derivatives with respect to t, $x_\parallel$, and $x_\perp$. The preference of the surface to have a uniform tilt is expressed by the curvature terms with positive coefficients [13] $\nu_\parallel$ and $\nu_\perp$. It is counteracted by fluctuations in the deposition rate giving rise to the Gaussian noise η with correlator

$$\langle \eta(\mathbf{x},t)\eta(\mathbf{x}',t')\rangle = 2D\delta(\mathbf{x}-\mathbf{x}')\delta(t-t') . \quad (3)$$

Expanding (2) for small deviations from the global tilt, one arrives at

$$\partial_t h = \tfrac{1}{2}\lambda_\parallel(\partial_\parallel h)^2 + \tfrac{1}{2}\lambda_\perp(\partial_\perp h)^2 + \nu_\parallel\partial_\parallel^2 h + \nu_\perp\partial_\perp^2 h + \eta , \quad (4)$$

where a term linear in $\partial_\parallel h$ has been transformed away by $x_\parallel \to x_\parallel - \kappa'(\bar{s})t$. The λ's are connected with the growth velocity $\kappa(s)$ through [5]

$$\lambda_\parallel = \kappa''(\bar{s}), \quad \lambda_\perp = \kappa'(\bar{s})/\bar{s} , \quad (5)$$

where κ' and κ'' denote the first and second derivatives with respect to the tilt s of the surface. From Fig. 1 it is immediately clear that $\lambda_\parallel$ is negative while $\lambda_\perp$ is positive. This was first observed by Villain [5] and turns out to be crucial for the kinetic roughening in step-flow growth.

If both λ's are zero, (4) is linear and equivalent to the Edwards-Wilkinson equation [14]. It can be solved exactly and gives rise to logarithmic roughness. In the following I shall only consider $\lambda_\perp \neq 0$. In the isotropic case, $r_\lambda \equiv \lambda_\parallel/\lambda_\perp = 1$ and $r_\nu \equiv \nu_\parallel/\nu_\perp = 1$, (4) reduces to the equation proposed by Kardar, Parisi, and Zhang [15] (KPZ) for describing the Eden model or ballistic deposition. This equation has some remarkable properties which only partly carry over to the more general case. The first property is the invariance under a simultaneous sign change of h and λ which obviously also holds for (4).

The second property of the isotropic equation is an invariance with respect to infinitesimal tilts which is usually referred to as Galilean invariance [16] because of the relation of the KPZ equation to the Navier-Stokes (or more precisely the Burgers) equation. Similarly, (4) does not change under the transformations

$$x_i \to x_i - \lambda_i \epsilon t, \quad h \to h + \epsilon x_i - \lambda_i \epsilon^2 t/2 , \quad (6)$$

where $i = \parallel$ or $\perp$, and ϵ is arbitrary [17].

Third, the KPZ equation can be mapped onto a linear equation for the partition function of a directed polymer in a random medium [15]. In the more general case (4), this works only for $r_\nu/r_\lambda = 1$. Then the Hopf-Cole transformation $Z(\mathbf{x},t) \propto \exp[\lambda_\parallel h(\mathbf{x},t)/2\nu_\parallel]$ maps (4) onto

$$\partial_t Z = \nu_\parallel\partial_\parallel^2 Z + \nu_\perp\partial_\perp^2 Z + (2\nu_\parallel/\lambda_\parallel)\eta Z , \quad (7)$$

which, up to an anisotropic scale transformation, again describes a directed polymer in a random medium.

In the following the results of a dynamical renormalization-group investigation of (4) will be discussed. The calculation is a straightforward generalization of the one described in detail by Medina *et al.* [16]. In general, the morphology of an anisotropic surface involves two characteristic lengths, $\xi_\parallel$ and $\xi_\perp \equiv \xi$. In addition to the roughness and dynamical exponent introduced in (1) one needs an anisotropy exponent χ defined by [9]

$$\xi_\parallel \sim \xi^\chi . \quad (8)$$

One integrates out fluctuations with wave numbers $e^{-l}\pi/a \leq |k_\perp| \leq \pi/a$ and $e^{-l\chi}\pi/a \leq |k_\parallel| \leq \pi/a$, where a is a small-length-scale cutoff (lattice constant). This gives rise to renormalized coefficients in (3) and (4), e.g., $\tilde{\nu}_\perp(l,\chi)$. Here I only consider the renormalization in lowest order in the nonlinearity (one-loop approximation). Subsequently one rescales $x_\perp \to e^l x_\perp$ (and correspondingly $x_\parallel \to e^{l\chi}x_\parallel$, $h \to e^{l\zeta}h$, $t \to e^{lz}t$), thereby restoring the original lattice constant a. If the system is scale invariant this should also restore the old coefficients in (3) and (4), e.g., $\nu_\perp = e^{(z-2)l}\tilde{\nu}_\perp(l,\chi)$.

It turns out that the λ's are not renormalized. This is expected [16] if the symmetry (6) which contains them as parameters is preserved when the small-wavelength fluctuations are integrated out. Under the subsequent rescaling one has $\lambda_\perp \to e^{l(z+\zeta-2)}\lambda_\perp$ and $r_\lambda \to e^{l2(1-\chi)}r_\lambda$. Scale invariance therefore requires

$$z + \zeta = 2, \text{ and } \chi = 1 , \quad (9)$$

provided that $\lambda_\perp \neq 0$ and $r_\lambda \neq 0$. Then one obtains the following flow equations for the remaining coefficients:

$$\frac{\partial \nu_\perp}{\partial l} = \nu_\perp\left[z - 2 + \frac{g_\perp}{16\pi^2}\left(1 - \frac{r_\lambda}{r_\nu}\right)A(1,r_\nu)\right] , \quad (10)$$

$$\frac{\partial r_\nu}{\partial l} = -r_\nu\frac{g_\perp}{16\pi^2}\left[1 - \left(\frac{r_\lambda}{r_\nu}\right)^2\right]A(1,r_\nu) , \quad (11)$$

$$\frac{\partial g_\perp}{\partial l} = \frac{g_\perp^2}{32\pi^2}\left\{8\frac{r_\lambda}{r_\nu} - 3\left[1 - \left(\frac{r_\lambda}{r_\nu}\right)^2\right]\right\}A(1,r_\nu) , \quad (12)$$

with $g_\perp = D\lambda_\perp^2/\nu_\perp^3$ and

$$A(\chi, r_v) = [\arctan r_v^{1/2} + \chi \arctan(1/r_v^{1/2})]/r_v^{1/2}.$$

Obviously, (11) has two stable fixed points, $r_v^* = r_\lambda$ and $r_v^* = -r_\lambda$. As the stability of the surface requires that the ν's and hence also r_v be positive, the first (second) fixed point is adopted when the two λ's have equal (opposite) sign. As already explained above, at the first fixed point there exists a mapping to a directed polymer in a random medium, and the scaling properties are the same as predicted by the KPZ equation, i.e., one obtains algebraic roughness [18]. Equation (12) shows that the fixed point ($r_v^* = r_\lambda$, $g_\perp^* = 0$) is marginally unstable in the $g_\perp$ direction. This is in contrast to the other fixed point ($r_v^* = -r_\lambda$, $g_\perp^* = 0$), where any finite bare coupling $g_\perp$ is renormalized to zero. Hence, if the two λ's in (4) have opposite sign, the nonlinearity is irrelevant for the scaling properties, and $\zeta = 0$(log) and $z = 2$ as for the linear theory [14].

The borderline case, $r_\lambda = 0$, is more complicated, as one has to allow for $\chi \neq 1$:

$$\frac{\partial \nu_\perp}{\partial l} = \nu_\perp \left[z - 2 + \frac{g_\perp}{16\pi^2} \left(\frac{1-\chi}{(1+r_v)^2}(r_v+3) + A(\chi, r_v) \right) \right], \tag{13}$$

$$\frac{\partial \nu_\parallel}{\partial l} = \frac{\partial r_v \nu_\perp}{\partial l} = \nu_\parallel [z - 2\chi], \tag{14}$$

$$\frac{\partial g_\perp}{\partial l} = g_\perp \left[1 - \chi - \frac{g_\perp}{32\pi^2} \left(\frac{1-\chi}{(1+r_v)^2}(3r_v+13) + 3A(\chi, r_v) \right) \right]. \tag{15}$$

It follows from (14) and the fact that $\lambda_\perp$ is not renormalized that $z = 2\chi = 2 - \zeta$. It is easy to see that again $g_\perp^* = 0$ is a stable fixed point, implying logarithmic roughness and $z = 2$. In principle, (15) also has a fixed point $g_\perp^* \neq 0$ with $\chi \neq 1$, but (13) then yields that $g_\perp^*$ must be negative and hence unphysical.

In conclusion, I have shown that the anisotropic generalization of the KPZ equation has a rich and unexpected behavior in three dimensions (two-dimensional surface): If the coefficients of the nonlinear terms have opposite sign, they are irrelevant so that the roughness is only logarithmic. This remains true when one of the coefficients vanishes. However, if they have equal sign, the nonlinearity becomes marginally relevant, leading to the much stronger algebraic roughness known from the isotropic KPZ equation. All these results have been confirmed by computer simulations [17].

Recently, Tang, Nattermann, and Forrest [19] investigated the crossover behavior of the isotropic KPZ theory by taking the renormalized coefficients $\tilde{\nu}(l)$, etc., of the equation as the effective parameters governing the modes of wave vector $k = \exp(-l\pi/a)$. A similar approach is possible here, too, and should give information on, e.g., the amplitude of the logarithm and the crossover from logarithmic to algebraic roughness.

The transition from logarithmic to algebraic roughness is different from the two morphological transitions in three dimensions which have been known analytically before: In the PNG model there occurs a transition from a faceted to an algebraically rough surface [20], and if the sign of λ changes in the isotropic KPZ theory [21], one has the same type of algebraic scaling on either side of the transition, where the roughness is only logarithmic. However, the kind of transition discussed above is similar to the one postulated by Yan, Kessler, and Sander [22] based on a computer simulation of a discrete model, although it should not contain anisotropy of the type suggested here. It is also worth mentioning that the relevance of the anisotropy is in marked contrast to thermal roughening, which is not different for vicinal or high-symmetry surfaces, as far as the scaling is concerned [23].

The experimental relevance of this has been demonstrated for a simple model [5] of step-flow growth with a tilt-dependent growth velocity as given by the solid line in Fig. 1. If island formation can be neglected, the kinetic roughening is predicted to be only logarithmic. For sufficiently large terrace sizes (i.e., small tilts $\bar{s}$) and low temperatures the probability increases that adatoms form clusters before they reach a step, hence leading to the formation of islands on the terraces. Therefore the growth velocity should not vanish for $\bar{s} \to 0$ but should have a finite minimum as indicated by the dashed line in Fig. 1. Lowering the temperature, island formation becomes significant on smaller and smaller terraces. Thus the upwards curved part of $\kappa(s)$ should extend to larger tilts. Eventually, $\lambda_\parallel = \kappa''(\bar{s})$ will change sign, and this implies a sharp morphological transition from logarithmic to algebraic roughness.

If the surface initially is perfectly oriented, the regular array of steps gives rise to sharp Bragg peaks. As the surface becomes rough on increasing length scales the intensity of these Bragg peaks drops. Two main differences are expected on the two sides of the morphological transition: For logarithmic roughness the Bragg intensity drops roughly as a power law, i.e., more slowly than the stretched exponential on the algebraically rough side. At the same time, power-law peaks should occur at the Bragg positions if the roughness is only logarithmic, in analogy to thermal roughness. For algebraic roughness such peaks are absent. However, very slow crossovers as

known from the isotropic KPZ theory [19] may cause difficulties in locating the transition by a diffraction experiment. Other methods like inhomogeneous growth [24], which gives rise to macroscopically different surface deformations depending on the sign of $\lambda_{\parallel}$, might be more feasible. Little is known experimentally about transitions of the type described above, partly because step-flow growth cannot be monitored by reflection-high-energy-electron-diffraction or TEAS oscillations. This is in contrast to, e.g., the transition from layer-by-layer to multilayer island growth [25], and it would be interesting to see if this also implies a change of the roughness exponent.

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