Kinetic Roughening in Surfaces of Crystals Growing on Disordered Substrates

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Substrate disorder effects on the scaling properties of growing crystalline surfaces in solidification or epitaxial deposition processes are investigated. Within the harmonic approach there is a phase transition into a low-temperature (low-noise) superrough phase with a continuously varying dynamic exponent $z > 2$ and a nonlinear response. In the presence of the Kardar-Parisi-Zhang nonlinearity the disorder causes the lattice effects to decay on large scales with an intermediate crossover behavior. The mobility of the rough surface has a complex dependence on the temperature and the other physical parameters.

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Much progress has been achieved recently in the understanding of kinetic roughening in nonequilibrium surface growth [1–13]. In particular, spatial and temporal scaling properties have been predicted for surfaces of crystals growing either by solidification or from epitaxial deposition [1,2]. The simplest analytical models neglect the discrete structure of the crystal and the scaling properties are determined from the university classes of the continuous kinetic equations which govern the growth process [3–5].

The lattice structure [6–13], however, has crucial effects on the behavior with a kinetic phase transition mirroring the equilibrium roughening transition (ERT) [14]. The transition is governed by the noise, i.e., thermal and from the inherent stochasticity, of the growth process [5–7,13]. In the presence of a strong noise the surface is rough (in analogy with the rough equilibrium phase for $T > T_r$, where T_r is the ERT temperature). In this phase the lattice effects are unimportant and the dynamic properties are the same as in the respective continuum models. In the low-noise phase the surface is smooth on an intermediate scale (which becomes larger the smaller the average growth rate is) with drastically different dynamic properties [6,7].

In view of the crucial importance of the discrete structure at the low-noise regime, the following question should be addressed: What if the substrate on which the solid grows is not perfectly smooth? While all previous studies have assumed a perfectly flat substrate, in many potential realizations that will not be the case. The effects of quenched disorder in the substrate on the kinetic scaling properties of the growing crystalline surface are the subject of the present Letter. As detailed below we find that in the presence of substrate disorder the low-noise (or low-temperature) regime has dramatically different kinetic properties [15].

The scaling properties are manifested in the height-height correlation function:

$$C(L,\tau)=\langle \Delta h^2(L,\tau)\rangle=\langle [h(\mathbf{x}+\mathbf{L},t+\tau)-h(\mathbf{x},t)]^2\rangle \sim L^{2\alpha} f(\tau/L^z), \quad (1)$$

where $h(\mathbf{x},t)$ is the height of the surface at point $\mathbf{x}$ and time t. The roughness exponent α characterizes the long-time limit of the self-affine fractal structure as represented by the "surface width" $w(L,\tau)=\langle \Delta h^2(L,\tau)\rangle^{1/2} \approx L^{\alpha}$. At an early stage of the growth ($\tau \ll L^z$) $w \sim \tau^{\beta}$, where $\beta=\alpha/z$ and z is the dynamic exponent. Theoretical studies of kinetic surfaces have been conducted within two approaches: the "harmonic" approach [3,6,7] and the "nonlinear" approach [4,13]. While the latter takes into account the lateral growth of an oblique surface, the former applies to discrete systems for which the nonlinear effects are negligible [16–18]. Our subsequent analysis follows this tradition.

I. Harmonic approach.—The kinetic equation for the time evolution of the height within this framework is

$$\mu^{-1}\frac{\partial h(\mathbf{x},t)}{\partial t}=F+\nu\nabla^2 h(\mathbf{x},t) - \gamma_0 y \sin(\gamma_0[h(\mathbf{x},t)+d(\mathbf{x})])+\zeta(\mathbf{x},t), \quad (2)$$

where μ is a microscopic kinetic coefficient, F is the driving force proportional to the difference in chemical potential for solidification or determined by the rate of deposition for epitaxial growth, ν is the surface tension, y is the coefficient of the leading term due to the discreteness (higher harmonics are less relevant [6]), $\gamma_0=2\pi/b$ where

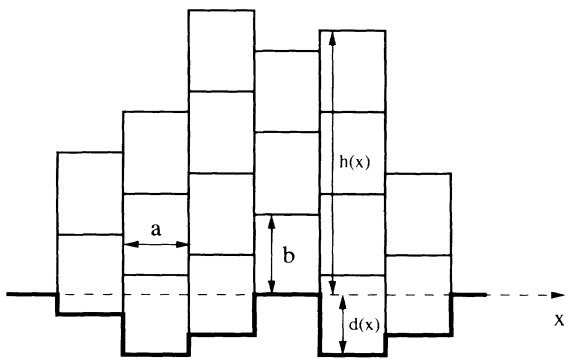

FIG. 1. A two-dimensional cut (along a lattice plane perpendicular to the disordered substrate) of the three-dimensional system.

b is the vertical lattice spacing, and $\zeta(\mathbf{x},t)$ is the noise term with $\langle\zeta(\mathbf{x},t)\zeta(\mathbf{x}',t')\rangle=2D\delta(\mathbf{x}-\mathbf{x}')\delta(t-t')$. Note that this equation of motion can be derived from a Hamiltonian $\mu^{-1}\partial h/\partial t=-\partial\mathcal{H}/\partial h+\zeta$, detailed balance is obeyed, and at (or slightly off) equilibrium the temperature of the system is $T=D\mu$. Equation (2) controls the lattice growth upon any generic rough surface, whatever the origin of the imperfections is. We consider the finite quenched fluctuations $d(\mathbf{x})$ of the substrate height (for a schematic description see Fig. 1) to have only short-range correlations and to be at least of the order of the lattice spacing of the bulk solid b. [For $|d(\mathbf{x})|\ll b$ the disorder is irrelevant.] Defining $\theta(\mathbf{x})=\gamma_0 d(\mathbf{x})$, the phaselike variables $\theta(\mathbf{x})$ obey $\langle e^{i\theta(\mathbf{x})}e^{-i\theta(\mathbf{x}')}\rangle=a^2\delta^2(\mathbf{x}-\mathbf{x}')$, where a is the lattice spacing in the horizontal planes.

In the absence of disorder [$d(\mathbf{x})=0$], lattice effects have been studied within the harmonic framework by Chui and Weeks (CW) [6] and later by Nozières and Gallet (NG) [7]. Their important findings were the following: For $D\mu=T>T_r$, the kinetic (as the static) behavior is unaffected by the lattice potential, $C(L,\tau)\sim(\ln L)f(\tau/L^2)$ [corresponding to $\alpha=\frac{1}{2}(3-d)=0$, $z=2$] and the macroscopic mobility $\mu_M=\lim_{F\to 0}v/F$ ($v=\langle\partial h/\partial t\rangle$, the average growth rate) is finite. For $T<T_r$, in the smooth phase, the surface tends to be pinned at the periodic minima, and $\mu_M=0$ (with a finite jump at $T=T_r$). For finite F, therefore, the growth is "activated" with nucleation [19,20] of higher "islands."

To apply the renormalization-group (RG) approach to Eq. (2) with the substrate disorder, we use the Martin-Siggia-Rose (MSR) formalism [21]. In addition to $h(\mathbf{x},t)$, an auxiliary field $\tilde{h}(\mathbf{x},t)$ is introduced as well as their conjugate "sources" $J(\mathbf{x},t)$ and $\tilde{J}(\mathbf{x},t)$. The thermal noise and the quenched disorder are averaged upon to yield the following averaged generating functional:

$$Z[J,\tilde{J}]=\int\mathcal{D}\tilde{h}\,\mathcal{D}h\exp\left\{\int\int d^2\mathbf{x}\,dt\left[Jh+\tilde{J}\tilde{h}+D\mu^2\tilde{h}^2-\tilde{h}\left(\frac{\partial h}{\partial t}-\mu\nu\nabla^2 h\right)\right]\right.$$
$$\left.+\frac{\mu^2\gamma^2 g}{2a^2}\int\int\int d^2\mathbf{x}\,dt\,dt'\tilde{h}(\mathbf{x},t)\tilde{h}(\mathbf{x},t')\cos(\gamma[h(\mathbf{x},t)-h(\mathbf{x},t')])\right\}$$
$$+\frac{1}{2}\mu^2\bar{\nu}\int\int\int d^2\mathbf{x}\,dt\,dt'\nabla\tilde{h}(\mathbf{x},t)\nabla\tilde{h}(\mathbf{x},t')\,, \tag{3}$$

with $g=\nu^2a^4$. The last term has been included since it is generated under renormalization.

Our RG analysis of this effective field theory follows closely that of Goldschmidt and Schaub for the XY model with random anisotropies [22] (details of the calculation will be published elsewhere [23]). Under rescaling $x\to e^l x$ and $t\to e^{lz}t$ ($l=\ln b$ where b is the rescaling factor) the RG analysis yields, to lowest nontrivial order in g, the following recursion relations:

$$\frac{d\nu}{dl}=0\,, \tag{4a}$$
$$\frac{d\bar{\nu}}{dl}=\frac{\pi\gamma^2}{4\nu(D\mu)^3}g^2\,, \tag{4b}$$
$$\frac{dF}{dl}=2F\,, \tag{4c}$$
$$\frac{dD}{dl}=\left(2-z+\frac{g\gamma^2\sqrt{c}}{D\mu\nu}\right)D\,, \tag{4d}$$
$$\frac{d\mu}{dl}=\left(z-2-\frac{g\gamma^2\sqrt{c}}{D\mu\nu}\right)\mu\,, \tag{4e}$$
$$\frac{dg}{dl}=\left(2-\frac{\gamma^2 D\mu}{2\pi\nu}\right)g-\frac{2\pi}{(D\mu)^2}g^2\,. \tag{4f}$$

The last equation provides the other parameter of the expansion $\delta=\gamma^2 D\mu/4\pi\nu-1$ (the deviation from the critical point) and these equations are of the first order in δ. γ remains at its bare value $\gamma=\gamma_0$ and ν at ν_0 (we may rescale h such that $\nu_0=1$). We first look at the static properties by defining $T=D\mu$. It obeys $dT/dl=0$ and the last equation can be written as

$$\frac{dg}{dl}=2\left(1-\frac{T}{T_{\rm sr}}\right)g-\frac{2\pi}{T^2}g^2$$

with $T_{\rm sr}=\nu b^2/\pi$. We recognize the static equation of Cardy and Ostlund for the random-anisotropy XY model [24]. Toner and DiVincenzo have analyzed these equations in the context of equilibrium crystal surfaces with bulk disorder in a limit where their bulk disorder is equivalent to the substrate disorder considered here [25]. So their results directly apply: For $T>T_{\rm sr}$, $g\to 0$, the disorder is irrelevant, and $w(L)\sim(\ln L)^{1/2}$. For $T<T_{\rm sr}$, g approaches a finite value $g^*\sim-\delta=1-T/T_{\rm sr}$, disorder is relevant, and the surface becomes superrough [25] $w(L)\sim\ln L$.

We now turn to study the kinetic properties. In the high-noise regime ($\delta>0$) we find $z=2$. In the low-noise regime z increases (the width spreading becomes slower) continuously as $z=2+4\sqrt{c}\,|\delta|$ with $c=\frac{1}{4}e^{2E}\sim 0.7931$, where E is the Euler constant. The mobility μ_M far from the transition in the high-$D\mu(=T)$ noisy phase assumes a finite value. Approaching $T_{\rm sr}$ from above, however, μ_M vanishes with δ continuously. Integration of the recursion relation yields $\mu_M=(\mu_0/2\pi g_0)|\delta|^\eta$ with $\eta=2/\sqrt{c}\sim 1.58$. In the low-noise phase μ_M vanishes as

$$\frac{v}{F}\sim\frac{v_0}{F_0}e^{-(z^*-2)l}\sim e^{-4\sqrt{c}|\delta|l}.$$

The behavior of z and μ_M as a function of T is summarized in Fig. 2.

Physically the dynamic behavior in the superrough phase may be understood as follows: The superrough phase is a phase in which the disorder barely dominates over the thermal fluctuations [25]. The increased roughness is the result of the surface attempt to balance the tendency of the surface to adjust to the substrate (making $\mathbf{h}+\mathbf{d}$ an integer multiple of b) without paying too much in elastic energy. Although the pinning is not uniform as with a smooth substrate the effect of locally preferred locations is enough to slow the spreading of the surface width (as manifested by $z>2$) and to prevent it from moving with a uniform average velocity when an infinitesimal driving force F is applied. Naturally if a finite force is applied the surface will move, on the average, at a constant velocity. This motion will wipe out the pinning effect (as it does to the periodic potential in absence of disorder). If the force F is small the behavior described here will hold up to a scale $L<L^*\sim aF^{-1/2}$ and the effective mobility will be

$$\mu(l^*=\ln L^*)\sim\mu_0(L^*)^{-4\sqrt{c}|\delta|}\sim\mu_0F^{2\sqrt{c}|\delta|}.$$

It should be emphasized that this yields another important finding, namely, the nonlinear response to a small F in the presence of which the averaged velocity scales as $F^{2\sqrt{c}|\delta|+1}$. This explicit behavior was derived based on scaling near the critical point but is also consistent with activated dynamics over free-energy barriers [26] $E(L)$ given by $\epsilon(T)\ln L$ with $\epsilon(T)=4\sqrt{c}\,(T_{sr}-T)$.

II. The "nonlinear" approach.—Kardar, Parisi, and Zhang (KPZ) have pointed out that when the lateral growth of an oblique surface is accounted for the most relevant effect is the addition of a term of the form $\frac{1}{2}\lambda(\nabla h)^2$ to the growth equation [4]. In the absence of any lattice or disorder effects the KPZ equation is

$$\mu^{-1}\frac{\partial h}{\partial t}=F+\nu\nabla^2h+\frac{\lambda}{2}(\nabla h)^2+\zeta(\mathbf{x},t),$$

and the exponent α and z change [1] from $\alpha=0$ and $z=2$ for $\lambda=0$, to $\alpha\sim0.4$ and $z\sim1.6$.

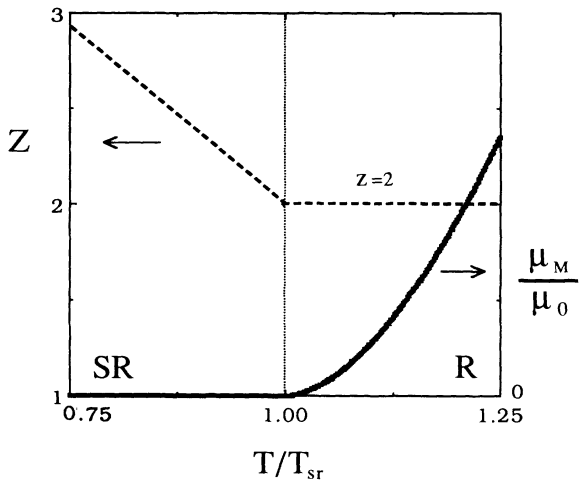

FIG. 2. The dependence of the linear response macroscopic mobility μ_M (bold line) and the dynamic exponent z (dashed line) on temperature for the harmonic model. The arrows indicate the appropriate scales on the vertical axis. (R, the rough phase $T>T_{sr}$; SR, the superrough phase $T<T_{sr}$.)

The question of whether a phase transition may occur in the presence of nonlinearity and a lattice with a perfectly flat substrate has been considered in a number of recent simulations of deposition (or growth) of discrete particles [8–12]. For example, the observed transition between logarithmic and power-law behavior of $w^2(L)$ by Amar and Family [8] has been attributed to an effective vanishing of the nonlinear term in their discrete model [16–18]. An extensive analytic study which includes both the lattice effects and the nonlinearity was performed by Hwa, Kardar, and Paczuski (HKP) [13]. Studying the intermediate scale $L<L^*$, HKP identified two phases. One is the high-temperature (strong noise) rough phase crossing over to KPZ scaling [13]. Approaching the transition from this phase the mobility vanishes as $(\ln|T-T_c|)^{-\zeta'}$. They also argue that for $T<T_c$ (lower noise) the surface is flat. This identification requires some caution since the generation of a term $y_2\cos(2\pi h/b)$ from the contraction of the terms $y_1\sin(2\pi h/b)$ and $\frac{1}{2}\lambda(\nabla h)^2$ was not considered. Combining both terms into the form $|y|\sin[2\pi h/b+\theta(l)]$ [with $y^2=y_1^2+y_2^2$ and $\theta(l)=\tan^{-1}y_2/y_1$], we find that for $T<T_c$ the flow is indeed toward $|y|\to\infty$ but the phase shift angle is rotating like $\theta(l)=\omega l$ with $l,\omega\sim\lambda/\nu$. Thus, this low-temperature phase is not characterized simply by an increase of the periodic potential. Higher-order terms in the recursion relations of y_1 and y_2 and that of ν will be required to identify with more confidence the nature of the low-temperature phase.

We now turn to the effect of substrate disorder in the presence of the KPZ nonlinearity: The RG analysis becomes much more complex. It turns out that a systematic expansion in the parameter $\delta=D\mu\gamma^2/4\pi-1$ requires the consideration of diagrams containing up to three nontrivial loops. Sophisticated techniques, based on dimensional regularization, were employed to extract their singular parts [23]. The following recursion relations for $x\to xe^l$, $t\to te^{2l}$ were derived ($\gamma=\gamma_0$):

$$\frac{d\nu}{dl}=0, \tag{5a}$$

$$\frac{d\bar{\nu}}{dl}=\frac{\pi\gamma^2}{4\nu(D\mu)^3}g^2, \tag{5b}$$

$$\frac{dF}{dl}=2F+\pi\lambda, \tag{5c}$$

$$\frac{dD}{dl}=\left(\frac{\lambda^2}{8\pi}D\mu+\frac{\gamma^2\sqrt{c}g}{D\mu}\right)D, \tag{5d}$$

$$\frac{d\mu}{dl}=\left(-\frac{\gamma^2\sqrt{c}g}{D\mu}\right)\mu, \tag{5e}$$

$$\frac{dg}{dl}=\left(2-\frac{D\mu\gamma^2}{2\pi}-\frac{\lambda^2c'}{\gamma^2}\right)g-\frac{2\pi}{(D\mu)^2}g^2, \tag{5f}$$

$$\frac{d\lambda}{dl}=0, \tag{5g}$$

where ν_0 is set to be 1 and $c'\sim 180.08$. Combining the equations for D and μ together we find ($T=D\mu$)

$$\frac{dT(l)}{dl}=\frac{\lambda^2}{2\gamma^2}T(l), \tag{6}$$

$$\frac{dg(l)}{dl}=\left(2-\frac{T(l)}{2\pi}\gamma^2-\frac{c'\lambda^2}{\lambda^2}\right)g(l)-\frac{\gamma^4}{8\pi^2}g^2(l). \tag{7}$$

The most important observation is that the nonlinearity increases the effective temperature $T(l)$ and, as a result, the effective coupling $g(l)$ becomes smaller. The only fixed point has $T\sim T_0e^{\lambda^2l/2\gamma^2}\to\infty$ and therefore $g\to 0$. The equation for $g(l)$ can be integrated exactly:

$$\frac{1}{g(l)}=\frac{1}{g^0}e^{s(l)}-\frac{\gamma^4}{8\pi^2}e^{s(l)}\int_0^l dx\,e^{-s(x)}, \tag{8}$$

where

$$s(x)=\left(\frac{\lambda^2c'}{\gamma^2}-2\right)x+T_0\frac{\gamma^4}{\pi\lambda^2}(e^{\lambda^2x/2\gamma^2}-1).$$

The behavior on long scales will have $g\to 0$ and the effective KPZ coupling $K=\lambda^2D\mu/\nu^3$ (note that to these orders in δ, g, and λ^2, the flow of this coupling is unaffected by g) will control the behavior.

However, since the scale associated with the increase of the KPZ coupling is exponentially large $L_K=ae^{8\pi/K_0}$ it is likely to be larger than L^* (the scale set by F). Therefore on scales smaller than L^* a rough surface will be observed but it will be in a crossover regime. If λ_0 and/or T_0 are small (especially with g_0 large), $g(l)$ will decay to zero quite slowly. This will be observable in the mobility which has the scale dependence

$$\mu(l)=\mu_0\exp\left\{-\frac{\gamma^4\sqrt{c}}{4\pi}\int_0^l dl'g(l')\right\}$$

with $g(l)$ given in Eq. (8). $\mu_M\sim\mu(l^*=\ln L^*)$ will be drastically reduced by the disorder effects (as they decay on intermediate scales) compared with its bare value μ_0; the same will hold for the width $w(l)$ when compared with the $g=0$ case.

To summarize, we have investigated how the scaling properties of growing crystalline surfaces are affected by disorder in the substrate. For $T<T_{sr}$ in the harmonic approach, the surface is superrough with anomalous dynamics. The height-height correlations are $C(L,\tau)\sim(\ln L)^2f(L/\tau^z)$ with $z=2+4\sqrt{c}(1-T/T_{sr})$. At the same time the response becomes nonlinear: $v\sim F^{\zeta+1}$ with $\zeta=2\sqrt{c}(1-T/T_{sr})$. In the presence of the KPZ nonlinearity a complex reduction in the width and the mobility of the rough surface have been obtained. These effects may be discernible in future precise measurements of solidification and epitaxial deposition processes.

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