Inhomogeneous Growth Processes

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(Received 8 June 1990)

It is proposed that inhomogeneities in the deposition rate can be a powerful tool for investigating properties of growing films. The macroscopic shape of the resulting surface deformation is discussed analytically for the growth equation proposed by Kardar, Parisi, and Zhang [Phys. Rev. Lett. **56**, 889 (1986)]. Computer simulations for a single-step growth model confirm the predictions based on this equation and give explicit values for its parameters. It is argued that inhomogeneous deposition also provides a new method for measuring the roughness exponent.

PACS numbers: 68.55.−a, 05.40.+j, 05.70.Ln, 68.90.+g

If particles are deposited onto a substrate, the surface of the growing solid film will, in general, be rough due to fluctuations in the deposition rate. Such kinetic roughening[1] is usually stronger than the one due to thermal fluctuations alone; i.e., the roughness exponent ζ is larger under growth than in thermal equilibrium. Starting from a flat surface, one typically encounters a power-law dependence of the height-height correlation function

$$\langle |h(\mathbf{r},t)-h(0,t)|^2\rangle \sim |\mathbf{r}|^{2\zeta} f(\mathbf{r}/t^{1/z}), \tag{1}$$

where the scaling function f expresses that a time $t\sim|\mathbf{r}|^z$ is needed until the roughness on a given distance $|\mathbf{r}|$ is fully developed.

Though the scaling behavior (1) is one of the fundamental features of a growth process, and the values of the exponents provide an ultimate test to concepts like universality, it has proven difficult in many situations to measure these exponents directly. This may be due to the strong crossover effects during the early stage of growth,[2] or simply to the lack of a good experimental method. It is thus of practical interest to study other features of the problem which can be tested independently.

Identifying relevant phenomenological aspects is also of considerable theoretical importance. Like the Ginzburg-Landau approach to critical phenomena, a coarse-grained description of growth processes should yield a continuum theory with a few parameters which in essence represents the universal features responsible for the asymptotic scaling. In general it is difficult to derive such a continuum theory from a microscopic model. Edwards and Wilkinson[3] illustrated the procedure for their model of sedimentation. However, for more complicated models it is a fruitful strategy to devise the continuum theory simply on grounds of symmetry and other phenomenological aspects. In this way Kardar, Parisi, and Zhang[4] (KPZ) were led to the following equation:

$$\partial h/\partial t = \nu\nabla^2 h + v + \eta, \tag{2}$$

where the growth velocity depends on the tilt of the surface,

$$v = \kappa + \lambda|\nabla h|^2/2, \tag{3}$$

and η is the white noise due to fluctuations in the growth rate. The Laplacian term is frequently misinterpreted as describing surface diffusion (which actually leads to a fourth-order derivative[5,6] of h). Without explaining its microscopic origin this term was introduced phenomenologically as the simplest way to express a smoothing effect on the surface.

A phenomenological theory can be put to the test by measuring the system's response to externally controlled perturbations. For instance, the elasticity theory of a solid can be verified through a strain experiment. In this Letter we propose inhomogeneous growth as a general method for the experimental verification of phenomenological growth theories. Spatial inhomogeneities will in general induce a macroscopic deformation of the average surface profile. We show that measurement of this profile provides a test to assumptions such as (3), and yields values for the phenomenological constants. We examine the effect of surface fluctuations on the average profile, and present an example where the roughness exponent ζ can be determined without actually measuring the height-height correlation function or the surface width.

For simplicity we consider the case where the deposition rate

$$\kappa = \kappa_0 + \kappa_1 \sum_n \delta(x - L/2 - nL) \tag{4}$$

is uniform everywhere except on regularly spaced parallel lines along the substrate. One then expects that the noise-averaged profile $H(x,t) = \langle h(\mathbf{r},t)\rangle$ depends only on the coordinate x.

To illustrate the idea, we discuss in detail the phenomenological growth equations (2) and (3). It follows from these equations that $H(x,t)$ should satisfy

$$\frac{\partial H}{\partial t} = \nu H'' + \frac{\lambda\langle|\nabla\delta h|^2\rangle}{2} + \frac{\lambda H'^2}{2} + \kappa, \tag{5}$$

while the fluctuations $\delta h = h - H$ obey

$$\frac{\partial \delta h}{\partial t} = \nu \nabla^2 \delta h + \lambda H' \delta h' + \frac{\lambda}{2}(|\nabla \delta h|^2 - \langle |\nabla \delta h|^2 \rangle) + \eta . \tag{6}$$

Here a prime denotes a partial differentiation with respect to x. With (4), Eq. (5) can be alternatively formulated as a boundary value problem in the region $|x| \leq L/2$ with

$$H(L/2,t) = H(-L/2,t) , \tag{7a}$$

$$s \equiv H'(L/2,t) = -H'(-L/2,t) = \kappa_1/2\nu , \tag{7b}$$

where s is the left slope of the profile at $x = L/2 \pm nL$, where the deposition piles have cusps.

$\lambda = 0$.—In this case the average profile and the fluctuations around it decouple, and (5) assumes the form of a standard diffusion equation with an array of particle sources. Because of the symmetry $(H,\kappa) \rightarrow (-H,-\kappa)$, one only need to consider the case $\kappa_1 > 0$ (extra deposition). Starting from a flat surface, piles build up at $x = L/2 \pm nL$ until a parabolic stationary profile

$$H_\infty(x,t) = sL(x/L)^2 + (\kappa_0 + \kappa_1/L)t \tag{8}$$

is formed. The excess growth velocity is determined by κ_1 which, when combined with the information on the shape of the profile (parametrized by s), yields ν via (7b).

$\lambda \neq 0$.—A simultaneous sign change $(H,\lambda,\kappa) \rightarrow (-H,-\lambda,-\kappa)$ leaves (5) and (6) invariant. Therefore we restrict the following analysis to positive λ.

Let us first examine how the average profile H might influence the fluctuations due to the coupling term in (6). Obviously a uniform translation of H has no effect on δh. Less trivial is that a uniform tilt of H only amounts to a spatial shift of δh. This is because a term linear in δh with a constant coefficient c can be eliminated via the Galilean transformation $x \rightarrow x - ct$.[6] As the gradient $\delta h'$ which couples to H' is dominated by short wavelengths if $\zeta < 1$, we expect that δh is influenced by local variations of H only. From these observations one may plausibly write

$$\langle |\nabla \delta h|^2 \rangle \simeq d_0 + d_2 H'' , \tag{9}$$

provided that H is a sufficiently slow-varying function of x. Substituting (9) into (5) yields the deterministic Burgers equation with an effective coefficient for the Laplacian, $\nu_{\text{eff}} = \nu + \lambda d_2/2$. This equation can be solved analytically.[7,8] In the following we discuss various features of the solution which can be employed to determine the phenomenological constants.

The stationary profile for extra deposition (+) is given by

$$\tilde{H}_+ = \ln(\cosh q\tilde{x}) + q^2\tilde{t}, \quad 1 = q \tanh q\tilde{L}/2 , \tag{10}$$

where we have introduced the dimensionless variables

$$\tilde{H} = (H - v_0 t)/H_0, \quad \tilde{x} = sx/H_0, \quad \tilde{t} = \lambda s^2 t/2H_0 , \tag{11}$$

with v_0 being the growth velocity of a horizontal surface, and $H_0 = 2\nu_{\text{eff}}/\lambda$. The implicit equation for q determines the finite-size correction to the profile. It is very weak: $q = 1$ within 1% for $\tilde{L} \geq 6$. For lack of deposition (−) one gets

$$\tilde{H}_- = \ln(\cos q\tilde{x}) - q^2\tilde{t}, \quad 1 = q \tan q\tilde{L}/2 . \tag{12}$$

In contrast to (10) the $\tilde{L}$ dependence of q here is crucial.

The typical stationary profiles for positive and negative values of κ_1 are illustrated in Fig. 1. Solutions to the linear cases are also included for comparison. The latter are symmetric with respect to the sign of κ_1, and have an amplitude $H(L/2) - H(0) = sL/4$. This symmetry is absent in the nonlinear case: For $\lambda > 0$ extra deposition leads to a pile of amplitude $H(L/2) - H(0) \simeq sL/2$, while lack of deposition produces a groove of logarithmic depth $H(L/2) - H(0) \simeq -H_0 \ln L$. For $\lambda < 0$ the linear and the logarithmic L dependence are exchanged. Thus by employing the asymmetry and the L dependence of the amplitude one can determine the sign of λ and the value of s.

In the case $\lambda > 0$ and $\kappa_1 > 0$ the stationary profile essentially has a triangular cross section with a little rounding near $x = 0$. Though the extra deposition is in effect only at $x = L/2 \pm nL$, due to the internal mechanism which generates the nonlinearity, the surface acquires a *finite* excess growth velocity $\Delta v = v - v_0$ even in the limit $L \rightarrow \infty$. According to (3) the value of λ is given by $2\Delta v/s^2$. At $x = 0$, Δv must be accounted for by the curvature term in (5). Measurement of the curvature should yield ν_{eff} once Δv is known. Useful information can also be gained by studying the buildup and decay of the stationary profile.[8] In particular, starting from a flat surface at $t = 0$, (5) dictates a sideways growth of a pile of constant slope s, whose bottom size increases at a constant velocity λs until it reaches L.

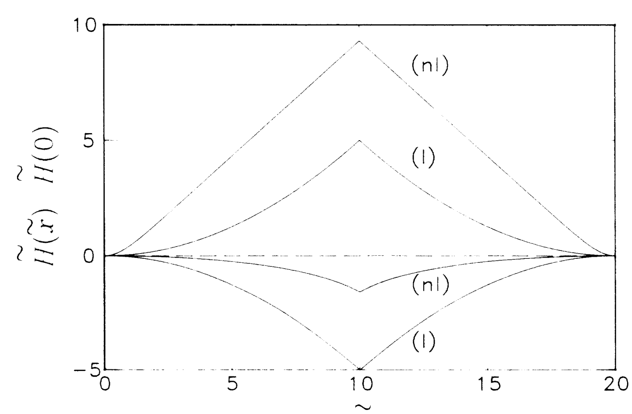

FIG. 1. Stationary profiles for the KPZ equation ($\lambda \geq 0$) scaled according to (11) with $H_0 = 2\nu_{\text{eff}}/\lambda$ in the nonlinear case (nl) and arbitrary H_0 for $\lambda = 0$ (l). The upper two curves are for extra deposition at $\tilde{x} = \tilde{L}/2 = 10$, the lower ones for lack of deposition.

Therefore the average excess velocity of the whole surface obeys

$$\bar{v} - v_0 \simeq (\lambda/2) s^2 g(\lambda s t/L) , \qquad (13)$$

where $g(\tau) \simeq \tau$ for $\tau \ll 1$ and approaches 1 for $\tau \gg 1$.

We have compared the above predictions of the linear and nonlinear theories with simulations of the single-step model in two space dimensions.[9,10] The growth starts from a flat ("zigzag") substrate parallel to the (11) direction of a square lattice. Particles (squares) can be added provided that the surface length remains constant. Periodic boundary conditions are imposed. To gain a computational advantage we employed a parallel updating scheme on the two checkerboard sublattices:[11] Eligible growth sites on a given sublattice are filled simultaneously with a probability $\frac{1}{2}$. Time is measured in units of sweeps of both sublattices, and lengths in units of $a/\sqrt{2}$ where a is the lattice constant. The growth velocity is reduced by including evaporation at sites observing the above constraint of constant surface length. The case $\lambda = 0$ is achieved by balancing deposition with evaporation.[10]

Before the growth inhomogeneity is switched on we let the surface evolve until its roughness is fully developed. Then the growth probability above a fixed substrate site is changed to a value of $p \neq \frac{1}{2}$ rendering the growth process inhomogeneous. As growth is only possible if the surface length remains constant, κ_1 is not simply related to $p - \frac{1}{2}$ in the present model.[8]

Here we only discuss our results for lack of deposition, $p < \frac{1}{2}$, without evaporation. We find the stationary profile to be of the type (10), meaning $\lambda < 0$. Figure 2 shows the average velocity of the whole surface versus the time t elapsed after the inhomogeneity was switched on. The data collapse at different values of L and s confirms the analytical result (13), and yields $\lambda = -0.70 \pm 0.02$, in good agreement with our mean-field result[8] $\lambda = -1/\sqrt{2}$.

Figure 3 shows the rounded part of the inverted stationary profile at three different values of s. Finite-size corrections are negligible for the L values considered. The x coordinate is scaled by s^{-1} in accordance with (11). The rounding extends to larger values of xs as s decreases, thus preventing the data collapse which would be expected if $\nu_{\rm eff}$ were constant. A data collapse is achieved by a different scaling of both x and $H(0) - H(x)$, in the way shown in the inset. All three curves fit well with (10) by assuming

$$H_0 = 2\nu_{\rm eff}/\lambda = (1.7 \pm 0.3)/s . \qquad (14)$$

The scaling found in Fig. 3 is due to the characteristic length

$$R(s) = a|s|^{1/(\zeta - 1)} \qquad (15)$$

which is obtained by equating the imposed asymptotic slope $|s|$ to the typical slope fluctuations $\delta h/R \sim (R/a)^{\zeta - 1}$ over the distance R. A data collapse for systems of different s is expected if one scales the x coordinate by R and the surface height by $sR = a(R/a)^{\zeta}$. This leads to a scaling ansatz for the average profile

$$H(x,L,t,s) - H(0,L,t,s) = sRF(x/R, L/R, t/R^z) . \qquad (16)$$

If there is no coupling between the fluctuations and the average profile as in the linear theory, H cannot depend on R so that the right-hand side of (16) becomes $sL\tilde{F}(x/L, t/L^z)$, in agreement with (8). For any *nonlinear* theory of surface growth, however, (15) and (16) offer a new way of determining the roughness exponent

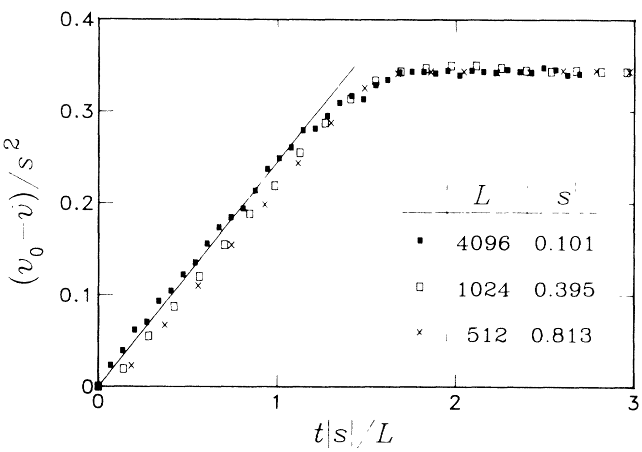

FIG. 2. Average excess growth velocity for the single-step model on the square lattice for different substrate sizes L and slopes s at the cusp of the profile. From the stationary value at large times t one can obtain $-\lambda/2$. The straight line with slope $\lambda^2/2$ is the early-time behavior predicted by (13).

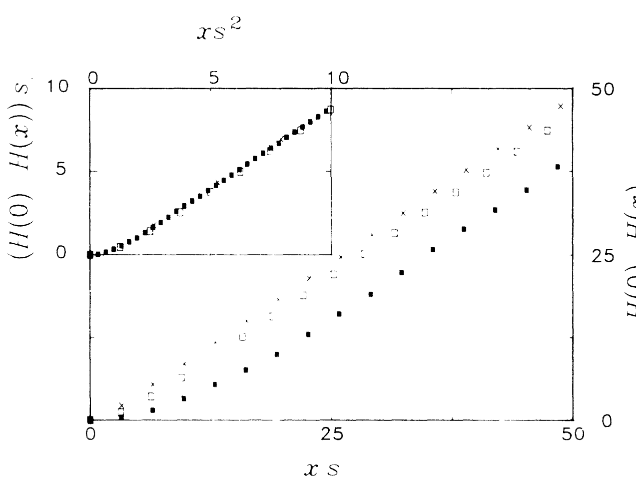

FIG. 3. Rounded parts of the stationary profiles scaled in two different ways for the same systems as in Fig. 2. A data collapse (inset) is obtained only if one assumes that $\nu_{\rm eff} \propto s^{-1}$.

through the introduction of a tunable characteristic length R. In the present case, R is simply a measure of the size of the rounded part of the average profile. The data collapse in Fig. 3 implies that $\zeta=\frac{1}{2}$, which is the known value for this model in two space dimensions.[9,10]

Our finding of the scaling (16) and the s dependence (14) of ν_{eff} is in perfect agreement with the scaling properties of the KPZ equation. Like any continuum theory describing the nonlinear behavior of a discrete model, the KPZ equation depends on the lower cutoff or coarse-graining length ξ. Hence there is a whole family of possible coefficients for a given lattice model,[4,12]

$$\nu(b)=b^{2-z}\nu(1),\quad \lambda(b)=b^{2-z-\zeta}\lambda(1) \tag{17}$$

(and corresponding equations for the other coefficients), depending on the choice of $\xi(b)=b\xi(1)$. By invoking this freedom, it is possible to relate two systems of different asymptotic slope through a scale transformation similar to (11), but using the scale-dependent coefficients (17) so that not only the slopes of the transformed systems match but also the cutoffs. The function F in (16) is then the solution of the scaled equation for a fixed ξ and asymptotic slopes ± 1. Equation (14) is a direct consequence of this rescaling. Of course, this argument is valid only if the coarse-graining length $\xi \ll R$.

In conclusion, we have shown that surface profiles obtained from inhomogeneous deposition provide useful information about the phenomenological equations governing the growth. If the KPZ equation applies to a given lattice model, one can easily obtain the value of λ.[12] The notion of a scale-dependent coefficient ν in the nonlinear case is corroborated in a numerical simulation, and a useful discussions. This work was supported by the Deutsche Forschungsgemeinschaft within Sonderforschungsbereich No. 341. Part of the computer simulations were done at the supercomputer center Hochstleistungsrechenzentrum, Jülich.

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