Instability in Surface Growth with Diffusion

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(Received 12 November 1991)

A model for surface growth with some of the features of molecular-beam epitaxy is proposed and investigated. Particles are deposited randomly on a one-dimensional substrate and the surface relaxes through diffusion processes, which obey detailed balance. The model undergoes a phase transition from a rough phase to a grooved phase. Both phases display scaling in space and time, with equal exponents. We also propose a Langevin equation which should describe this growth process and show that this equation contains an infinite number of relevant terms.

PACS numbers: 61.50.Cj, 05.70.Ce, 68.35.Fx

During the last few years a number of nonequilibrium growth models have been proposed and investigated from the perspective of fundamental statistical mechanics (dynamic universality classes, generic scale invariance) and as possible models for molecular-beam epitaxy (MBE), vapor deposition, and other growth processes. These models generally involve deposition of particles onto a substrate according to rules which couple the local growth rate to the structure of the existing deposit. Invariably one finds that the surface of the deposit progressively roughens and, in the long-time limit, attains a steady state in which the width of the interface scales in a nontrivial way with the size of the substrate. The processes by which the surface relaxes play an important role in determining both the steady state and short-time properties of growing clusters. If desorption is the dominant relaxation process, the growth kinetics seem to be well described by the so-called Kardar-Parisi-Zhang (KPZ) equation [1]. Much less is known if desorption can be neglected and surface diffusion is the main relaxation process. These conditions usually apply to vacuum deposition and MBE [2]. In this Letter we discuss a model in which relaxation occurs entirely through surface diffusion. We find scaling behavior markedly different from that of the KPZ equation and, remarkably, a phase transition from a conventionally rough steady state to a grooved phase with broken translational symmetry.

Surface dynamics governed by diffusion conserves the number of particles so that in the continuum limit the normal velocity of the interface obeys a continuity equation

$$v_n(\mathbf{x},t) = -\nabla\cdot\mathbf{j}(\mathbf{x},t)+\tilde{\eta}_c(\mathbf{x},t)\,,$$

where $\tilde{\eta}_c$ is a noise term, $\langle\tilde{\eta}_c\rangle=0$. The current density $\mathbf{j}$ is proportional to the gradient of the chemical potential $\mu=\delta\mathcal{F}/\delta h$ and is tangent to the surface. The operator ∇ must be computed in a local coordinate system with axes parallel to the surface. The Ginzburg-Landau functional may be taken as the free energy of the drumhead model, $\mathcal{F}=\mathcal{H}_{\rm DH}=\int d^dx\,\sigma\sqrt{g}$, $g=1+(\nabla h)^2$, where σ is the surface tension and $h(\mathbf{x},t)$ the height of the d-dimensional surface. Therefore the Langevin equation describing surface diffusion is given by

$$v_n=\frac{\partial_t h}{\sqrt{g}}=\Lambda_c\underset{\sim}{\Delta}\frac{\delta\mathcal{F}}{\delta h}+\tilde{\eta}_c\,, \tag{1}$$

where

$$\underset{\sim}{\Delta}=\frac{1}{\sqrt{g}}\frac{\partial}{\partial x_i}\left[\sqrt{g}\left[\delta_{ij}-\frac{1}{g}\frac{\partial h}{\partial x_i}\frac{\partial h}{\partial x_j}\right]\right]\frac{\partial}{\partial x_j} \tag{2}$$

is the Laplace operator in curved space [3] and the noise correlation

$$\langle\tilde{\eta}_c(\mathbf{x},t)\tilde{\eta}_c(\mathbf{x}',t')\rangle=-2k_BT\Lambda_c\underset{\sim}{\Delta}\delta(\mathbf{x}-\mathbf{x}')\delta(t-t')$$

reflects the conservation law. Power counting in the corresponding Martin-Siggia-Rose (MSR) functional [4] shows, however, that all nonlinear terms in Eq. (1) are irrelevant and that the scaling properties of the model are given by the linear equation

$$\partial_t h=-D_c\Delta\Delta h+\eta_c\,, \tag{3}$$

with $D_c=\Lambda_c\sigma$ and $\langle\eta_c(\mathbf{x},t)\eta_c(\mathbf{x}',t')\rangle=-2k_BT\Lambda_c\Delta\delta(\mathbf{x}-\mathbf{x}')\delta(t-t')$. From (3) one easily obtains the exponents $\zeta=(2-d)/2$ and $z=4$ which determine the scaling properties of the width of the interface $\xi(t,L)=L^{\zeta}f(t/L^z)$, where L is the size of the substrate.

When particle deposition (normal to the substrate) is included, the interface moves with a mean velocity modulated by a shot noise $\eta(\mathbf{x},t)$ with $\langle\eta(\mathbf{x},t)\eta(\mathbf{x}',t')\rangle=2D\delta(\mathbf{x}-\mathbf{x}')\delta(t-t')$. In the comoving coordinate system the resulting Langevin equation reads [5]

$$\partial_t h=\Lambda_c\sqrt{g}\underset{\sim}{\Delta}\frac{\delta\mathcal{F}}{\delta h}+\eta\,, \tag{4}$$

where the conserved noise $\tilde{\eta}_c$ has been omitted because it is irrelevant in the presence of nonconserved noise. Although the particle beam breaks the up-down symmetry, the resulting equation is nevertheless invariant under the transformation $h(\mathbf{x},t)\to-h(\mathbf{x},t)$. In particular, a previously proposed term [6] proportional to $\Delta(\nabla h)^2$ does not appear in Eq. (4). This is a consequence of the assumption that deposition does not change the nature of the diffusion process, i.e., it is still driven by energy

differences and can be described using a surface Hamiltonian $\mathcal{F}$ [7].

In contrast to the case of pure diffusion (1), the nonlinearities in (4) are important. Power counting in the MSR functional reveals that ∇h has the scaling dimension $(D/D_c)^{1/2}q^{d/2-1}$, where q is an inverse length. All nonlinear terms in (4) differ from the linear terms by powers of ∇h. Thus, *all* nonlinear terms are relevant (marginal) for $d<2$ ($d=2$) and a renormalization-group analysis would seem to be a formidable task. We note that even an anisotropic surface tension $\sigma=\sigma(\nabla h)$ is a relevant perturbation. At this point it is not known if a single nontrivial fixed point determines the scaling properties of the growth process or if the microscopic details of the model are paramount.

We have performed Monte Carlo simulations for a one-dimensional model designed to reproduce the scenario described above. In particular, diffusion is driven by energy changes and obeys detailed balance. The surface energy is defined through the Hamiltonian of an *unrestricted* SOS model,

$$\mathcal{H}=J\sum_n[(h_n-h_{n-1})^2+g_4(h_n-h_{n-1})^4 +g_6(h_n-h_{n-1})^6], \quad (5)$$

where h_n is the (integer) height variable at site n and periodic boundary conditions are used. The role of the coefficients g_4,g_6 of the higher-order terms will be discussed in more detail below. At this point we simply require $\mathcal{H}$ to have a single minimum at $h_n-h_{n-1}=0$.

In the simulations a lattice site n is randomly selected and a particle is deposited at that site with a probability τ. A diffusive move is attempted with probability $1-\tau$. The direction of diffusion is chosen randomly and the move is accepted with probability $w_{n\to m}=[1+\exp(\beta\Delta\mathcal{H}_{n\to m})]^{-1}$, where m is a nearest-neighbor site and $\Delta\mathcal{H}_{n\to m}$ is the energy change associated with the move of the particle. In the limit of deposition rate $\tau=0$ this algorithm produces the correct exponents $\zeta=\frac{1}{2}$ and $z=4$ of the linear theory (3).

Usually the scaling behavior of the width $\xi(t,L)\sim t^{\zeta/z}$ for $t_0\ll t\ll L^z$ and $\xi(t,L)\sim L^\zeta$ for $t\to\infty$ is used to determine the exponents ζ and z. In our model we are forced to determine ζ from the steady-state width of relatively small systems because z is so large that the time the system takes to reach the steady state is extremely long. One can also obtain these exponents from the steady-state correlation function

$$S(k,t)=\lim_{t'\to\infty}\langle\hat{h}_k(t+t')\hat{h}_{-k}(t')\rangle=k^{-\gamma}s(k^z t) \quad (6)$$

with

$$\hat{h}_k(t)=L^{-d/2}\sum_n[h_n(t)-\bar{h}(t)]e^{ink},$$

$\bar{h}(t)=L^{-d}\sum_n h_n$, and $\gamma=2\zeta+d$. It has been shown [8,9] that extrapolating the exponent ζ from the small k behavior of the steady-state structure factor $S(k)=S(k,t=\infty)$ eliminates some of the finite-size effects due to the large k modes. This has the additional advantage that, in principle, simulations for only one (preferably large) system are needed. The exponent z then can be calculated from the condition [9] that $\Phi(k,t)=S(k,t)/S(k)$ be a function of the single argument $k^z t$. The simulations were performed for two temperatures $\beta J=1$ and 0.01 and two deposition rates $\tau=0.01$ and 0.1. We do not believe that the exponents depend on either of these parameters, although this cannot be definitely ruled out because of finite-size effects.

We first present the results for the discrete Gaussian model, $g_4=g_6=0$. Figures 1 and 2 show the steady-state structure factor $S(k)$ and the steady-state correlation function $\Phi(k,t)$ from which the exponents γ (respectively ζ) and z can be determined. We have also calculated ζ/z from the early-time behavior of the width. The least reliable value is probably the result for ζ. Because of the relatively small system size ($L=64$) crossover effects might influence the extrapolation of the small k data. Nevertheless, the Monte Carlo data indicate that the universality class of random deposition with surface diffusion is characterized by the exponents

$$\zeta=1.2\pm0.2,\quad z=3.6\pm0.3,\quad \zeta/z=0.35\pm0.01 \quad (7)$$

which differ from the values of the linear theory $\zeta=(4-d)/2$, $z=4$ obtained by neglecting all nonlinearities in (4).

In Fig. 3 we show the structure factor for $g_4=1$, $g_6=0$. In contrast to the simple power-law behavior seen in Fig. 1, $S(k)$, $k=2\pi m/L$, is orders of magnitude larger for odd values of m than for even m. Clearly $S(k)$ does not obey the scaling law (6). The behavior of the structure factor is reminiscent of phenomena like phase separation [10]. Translational invariance is spontaneously broken and the expectation value of the height itself becomes a nontrivial function $\langle h(x,t;L)\rangle=f(x+x_0(t),t;L)$, where

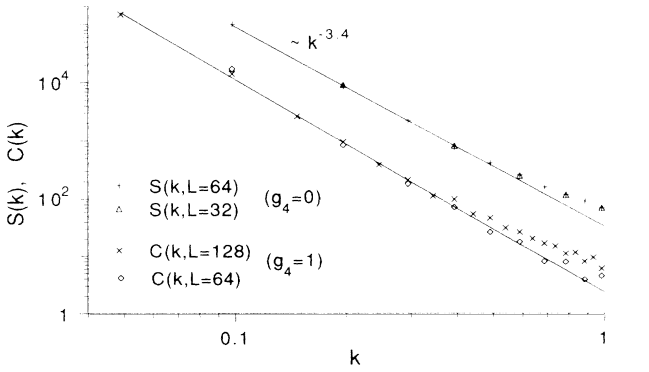

FIG. 1. Steady-state structure factors $S(k)$ for $g_4=0$ and $C(k)$ for $g_4=1$. The slopes, indicating an exponent $\gamma=3.4$, are approximately the same for $S(k)$ and $C(k)$ ($\tau=0.1$, $\beta J=0.01$, $g_6=0$).

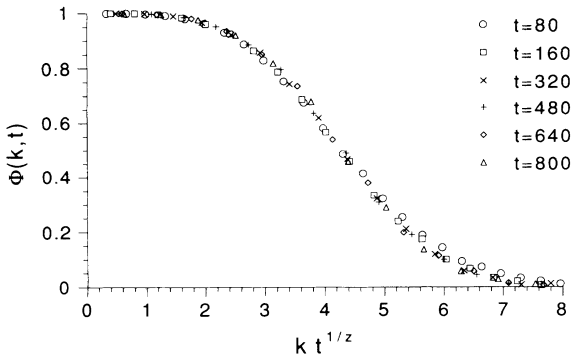

FIG. 2. Reduced time-time correlation function $\Phi(k,t)$ as a function of the scaled argument $kt^{1/z}$ for several times t showing a data collapse for $z=3.6$ ($L=64$, $\tau=0.1$, $g_4=g_6=0$, $\beta J=0.01$).

x_0 is, e.g., the position of the maximum of $\langle h(x,t;L)\rangle$ and reflects the Goldstone mode which translates $\langle h(x,t;L)\rangle$ on the lattice. To calculate $\langle h(x,t;L)\rangle$ in a simulation we eliminate this random phase x_0 by shifting the maximum of $h(x,t;L)$ to $x=0$ before averaging. The height function in the steady state seems to obey a scaling law $f(x;L)=L^{\kappa}g(x/L)$ as shown in Fig. 4. Finite-size effects are clearly visible in Fig. 4 and the exponent $\kappa\simeq 3.6$ calculated from the sizes $L=128$ and 64 is undoubtedly smaller than the true value. We note that $\langle h(x,L)\rangle$ is symmetric with respect to the x axis indicating that the corresponding Langevin equation is invariant under the transformation $h\rightarrow -h$ as is the case in Eq. (5). The breaking of translational invariance can be interpreted as an instability towards the creation of large slopes in the interface configuration. The slope is only limited by the periodic boundary conditions and the surface tension which suppresses large curvature. This explains the fact that the wavelength of the height configuration is always determined by the substrate size. To check that the occurrence of the instability is not related to the sign of the

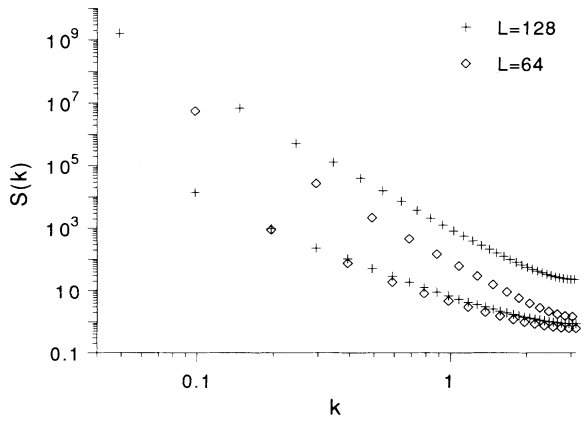

FIG. 3. Structure factor $S(k)$ for $g_4=1$ and two system sizes ($\tau=0.1$, $g_6=0$, $\beta J=0.01$).

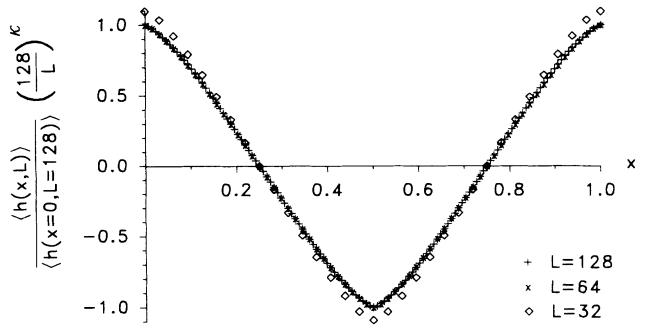

FIG. 4. Rescaled height configuration $\langle h(x,L)\rangle$ as a function of $x=n/L$, $n=0,1,\ldots,L$. The data are rescaled for the smaller system size $L=32$ and are shifted to larger values indicating finite-size effects and showing that the value of $\kappa\simeq 3.6$ is only a lower bound [$\langle h(x=0,L=128)\rangle=7.85\times 10^3$, $\tau=0.1$, $g_4=1$, $g_6=0$, $\beta J=0.01$].

coefficient g_4 in (6), additional simulations with $g_4=-\frac{1}{4}$ and $g_6=\frac{1}{8}$ were performed. These showed the same instability as for positive g_4.

Golubović and Karunasiri [10] (GK) studied a 1D Langevin equation similar to Eq. (4) with an additional term $\nu\partial_x(\partial_x h/\sqrt{g})$. This term yields an asymptotic behavior corresponding to the model of Edwards and Wilkinson [11] ($\zeta=\frac{1}{2}$, $z=2$). GK argued that the slope $\partial_x h$ of the surface profile behaves like the order parameter of Ising-like systems in spinodal decomposition [12]. Their argument depends crucially on ν being greater than zero and does not take into account that all nonlinear terms in (4) are relevant and that changing or omitting terms might have an important effect on the results. In such a case Monte Carlo simulations may be the only way to obtain a definite result. We also note that GK do not observe any real phase separation. On the other hand, our dynamical model yields a steady state, which is truly ordered even in *one* dimension.

Because of the breaking of translational invariance the correlation functions $S(k,t)$ defined in (6) are dominated by the behavior of $\langle h(x,t)\rangle$, i.e., the correlation functions,

$$S(k,t)\simeq\langle\hat{h}_k(t+t')\rangle\langle\hat{h}_{-k}(t')\rangle$$
$$=|\langle\hat{h}_k\rangle|^2\exp\{-k^2\langle[\Delta x_0(t)]^2\rangle/2\},$$

decay with a trivial dynamical exponent 2 because of the random walk of the height configuration in the steady state: $\langle[\Delta x_0(t)]^2\rangle=\langle[x_0(t+t')-x_0(t')]^2\rangle\sim t$. To compute the true fluctuations, which can be compared to $S(k,t)$ for $g_4=0$, the contributions of the height configuration are subtracted and the correlation functions for $g_4\neq 0$ are defined by

$$C(k,t)=\lim_{t'\rightarrow\infty}\langle\hat{h}_k(t+t')\hat{h}_{-k}(t')\rangle-\langle\hat{h}_k(t+t')\rangle\langle\hat{h}_{-k}(t')\rangle$$
$$=k^{-\gamma}c(kt^z). \qquad (8)$$

Although this subtraction amplifies the statistical errors the exponents γ (respectively ζ, see Fig. 1) and z are, within the error bars, the same as in the case $g_4=0$.

We interpret these results as a phase transition from a state with order parameter $\langle h_k\rangle=0$ to one with $\langle h_k\rangle\neq 0$. In the discrete model (5), the transition is driven by the control parameters g_4 and g_6. We believe that there will be an analogous phase transition in the continuum model (4), probably generated in this case by varying the anisotropy of the surface tension. Both phases show scaling behavior of spatial and temporal correlation functions in the steady state, remarkably, with the same exponents ζ and z. On the other hand, in contrast to the results of GK, the early-time behavior of the width for $g_4\neq 0$ does not show simple power-law growth in any time interval. Whether or not other correlation functions do is at this point an open question.

Solid-on-solid models become unphysical if the roughness exponent $\zeta\geq 1$. When the instability is present the situation is even worse because the height itself diverges faster than the system size. In the physically interesting dimension $d=2$ preliminary Monte Carlo simulations [13] indicate that the same instability occurs as in $d=1$. Although these results imply that the height configuration in the steady state is not relevant for real growth processes, these models provide valuable information on kinetic phase transitions and on the relation between discrete and continuum models. Indeed, preliminary numerical integrations of (4) indicate that instabilities similar to those found in our simulations are present in the Langevin equation and we will continue to explore this in both one and two dimensions.

One might also ask which processes prevent the divergence of the surface width in real materials. Desorption is certainly not exactly zero in MBE experiments and generalizing the model to allow some evaporation of particles might define a length scale for grooves on the surface. As far as the continuum model is concerned, a pinning term $\sim\cos 2\pi h$ should be included in the functional $\mathcal{F}$. Such a term is probably irrelevant in dimension $d=1$, but may be important in $d=2$. This will form the subject of a future investigation.

We thank Z. Rácz and R. K. P. Zia for helpful conversations. This research was supported by the Deutsche Forschungsgemeinschaft and the NSERC of Canada.

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