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Instability in a continuum kinetic-growth model with surface relaxation

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We study the recently proposed nonlinear kinetic-growth model with surface relaxation for molecular-beam epitaxy in both two (2D) and three (3D) dimensions. We find that the dynamics of the equation without noise is nonlinearly unstable, in contrast to the dynamics of the Kardar-Parisi-Zhang equation. Because of the large fluctuation exponent in 2D, one important consequence is that there exists a strong-coupling regime where the interface develops a local divergence in finite time. For 3D or higher dimensions, the fluctuation is not strong enough to drive the system towards divergence, and the scaling is correctly given by renormalization-group calculations.

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During the past ten years, kinetic-growth problems [1] have received a great deal of attention due to their application in material science, especially thin-film growth. Most of the growth models have scaling behavior characterized by the surface roughness or the interface width $\omega \sim t^{\chi} f(L/t^{z})$, where L is the system size, t is time. The scaling function $f(x)$ has the asymptotic behavior $f(\infty) = \text{const}$, and $f(x) \sim x^{\chi/z}$ as $x \to 0$. Continuum equations of Langevin type have been proposed to study the scaling behavior of these growth processes. For example, the Kardar-Parisi-Zhang (KPZ) [2] equation has been quite successful in describing a wide variety of growth models, including ballistic aggregation, Eden model, and vapor deposition [3]. Apparently, most of the growth models studied so far belong to the universality class of the KPZ equation.

Recently, an effort has been made towards the understanding of the critical fluctuation in "ideal" molecular-beam epitaxy system (MBE) [4–10]. There are two main differences between ideal MBE growth and the vapor deposition or Eden model. First, the MBE model is a solid-on-solid (SOS) model, the conservation of particles is enforced through the conservation of the height integrated over the substrate (apart from a constant growing part). Second, the relaxation force in MBE is due to difference in local bonding (surface diffusion) instead of difference in height (surface tension). Because of these differences, the scaling behavior of ideal MBE model should be different from that of the KPZ equation. Indeed, that has been confirmed by many atomistic simulations [5,6,9].

To understand the universality class of the critical fluctuation in ideal MBE growth, a continuum equation was proposed by Wolf and Villain [5] and later improved by Lai and Das Sarma [9] and Tang and Nattermann [10] (LDTN) by introducing a nonlinear term. The nonlinear equation is

$$\frac{\partial h}{\partial t} = \nabla^2[-\nu\nabla^2 h + \lambda(\nabla h)^2] + \eta(\mathbf{x},t)\,, \tag{1}$$

where $h(\mathbf{x})$ is the height of the interface in $d = d' + 1$ dimensions (d' is the substrate dimension) and η is the noise term with $\langle\eta\rangle = 0$ and

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

ν is the surface diffusion coefficient, and λ is the quadratic coupling constant. (For details of the physical meaning of the terms, the readers are referred to the original paper.)

This model was studied in [9,10] using renormalization-group (RG) theory. The upper critical dimension for this model is $d_c = 5$. But even below the critical dimension $d < d_c$, the exponents are exactly given from the RG argument as

$$\chi = \tfrac{1}{3}(5-d),\quad z = \tfrac{1}{3}(8+d) \tag{2}$$

in virtues of "Galilean" invariance of the equation, which implies $\chi + z = 4$, and the hyperscaling relation $2\chi = z - d + 1$, which results from the fact that the noise term is not renormalized.

It is the main purpose of this paper to show that the scaling behavior of Eq. (1) is more complicated than that given by Eq. (2). In fact, for $d = 1+1$, there exists a strong-coupling regime where the equation becomes divergent.

Also, for the LDTN model, $\chi = 1.0$ for $d = 1+1$, so terms with the form $\nabla^2[(\nabla h)^{2n}]$ $(n > 1)$ will be marginal by power counting. If one of the terms is relevant at the strong-coupling fixed point, the Galilean invariance will be broken, and the relation $\chi + z = 4$ will no longer hold exactly. For simplicity, we only include the next higher-order term, i.e., $\nabla^2[(\nabla h)^4]$. The model equation becomes

$$\frac{\partial h}{\partial t} = \eta + \nabla^2[-\nu\nabla^2 h + \lambda_2(\nabla h)^2 + \lambda_4(\nabla h)^4]\,. \tag{3}$$

Let us first simplify the above equation by introducing the transformations $h \to (D^{1/2}/\nu)^{1/2} h$, $t \to t/\nu$, and the effective coupling constants $\bar{\lambda}_2 = (\lambda_2^2 D^{1/2}/\nu^3)^{1/2}$, $\bar{\lambda}_4 = (\lambda_4^2 D^{3/2}/\nu^5)^{1/2}$. The equation becomes

$$\frac{\partial h}{\partial t} = \eta + \nabla^2[-\nabla^2 h + \bar{\lambda}_2(\nabla h)^2 + \bar{\lambda}_4(\nabla h)^4] \tag{4}$$

with $\langle\eta(\mathbf{x},t)\eta(\mathbf{x}',t')\rangle = \delta(\mathbf{x}-\mathbf{x}')\delta(t-t')$.

The questions that we want to address here are whether the scaling behavior specified by Eq. (2) is independent of $\bar{\lambda}_2$ and whether the fixed point is stable against higher-order derivative terms. A perturbative RG calculation is

useless here because we are interested in a strong-coupling fixed point and $d=2$, 3 is too far away from the upper dimension $d_c=5$. So we have to rely on the numerical method.

We use a finite difference scheme to integrate Eq. (4) in time. In discrete form, Eq. (4) can be written as

$$h_i(t+\Delta t)=h_i(t)+\Delta t\nabla^2[-\nabla^2 h+\bar{\lambda}_2(\nabla h)^2+\bar{\lambda}_4(\nabla h)^4]_i + \sqrt{\Delta t}\eta_i(t). \quad (5)$$

The second term on the right-hand side of the above equation is expressed through the discretization scheme:

$$(dy/dx)_i=(y_{i+1}-y_{i-1})/2,$$

$$(d^2y/dx^2)_i=y_{i+1}+y_{i-1}-2y_i.$$

$\eta_i(t)$ is an independent random variable uniformly distributed between $-\sqrt{3}$ and $\sqrt{3}$, so that

$$\langle\eta_i(t)\eta_j(t')\rangle=\delta_{ij}\delta(t-t').$$

The width of the interface, $w=(\langle h^2\rangle-\langle h\rangle^2)^{1/2}$, is measured at different times. The initial condition of the interface is taken to be flat. Both periodic and free boundary conditions are used, and the results do not depend on the specific choice of boundary condition. The time step we use is $\Delta t=0.01$.

We first set $\bar{\lambda}_4=0$ and simulate the equation for various values of $\bar{\lambda}_2=0.5$, 1, 2, 3, for system size $L=32$, 64, 128, 256, and up to 512. For each data set, we average ten runs. In Fig. 1, we show the dependence of the width on time t on a log-log plot for $L=128$ for $\bar{\lambda}_2=0.5$, 1, 2, 3. We find that for small values of $\bar{\lambda}_2$, i.e., $\bar{\lambda}_2=0.5$, 1, 2, the dependence of the width on time t follows a power law $w\sim t^{\alpha}$ for $t<t_c(\bar{\lambda}_2)$, with $\alpha=0.32\pm0.02$. When $t>t_c(\bar{\lambda}_2)$, the width saturates to its equilibrium value, depending on the system size L, and also the coupling constant $\bar{\lambda}_2$. A simple scaling estimate of Eq. (4) gives that $w\sim\bar{\lambda}_2^{-1/3}$, which roughly agrees with our simulation.

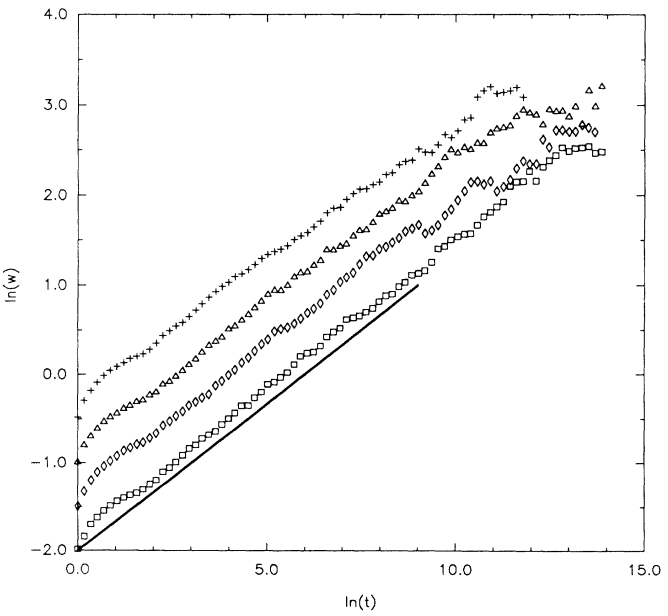

FIG. 1. Width vs time on log-log scale for $L=128$ and $\bar{\lambda}_2=0.5$, 1.0, 2.0, 3.0 (from bottom to top), each set of data is shifted upwards successively by 0.5 with respect to the previous set. The solid line has a slope equal to 1/3. The data set for $\bar{\lambda}_2=3.0$ ends where it develops singular behavior.

However, for larger values of $\bar{\lambda}_2$, for example, $\bar{\lambda}_2=3$, the simulation develops finite time singularities, i.e., the height of the interface at a certain point grows to infinity in finite time. We have checked our simulation with smaller time step $\Delta t=0.001$, it does not change the singular feature of the problem. Our simulation therefore strongly suggests that there exists a critical value of $\bar{\lambda}_2$, $\bar{\lambda}_2^*$. When $\bar{\lambda}_2>\bar{\lambda}_2^*$, the system blows up in finite time; when $\bar{\lambda}_2<\bar{\lambda}_2^*$, the RG flow leads the system towards a finite value fixed point $\bar{\lambda}_2^f$, where the exponents are given by Eq (4). The value of $\bar{\lambda}_2^*$ is found to be slightly less than 3.

We have also simulated the case with $\bar{\lambda}_4\neq0$ and small values of $\bar{\lambda}_2(<\bar{\lambda}_2^*)$, we find that there also seems to be a critical value $\bar{\lambda}_4^*(\bar{\lambda}_2)$, so that when $\bar{\lambda}_4<\bar{\lambda}_4^*$, the exponents remain approximately the same, meaning that the fixed point $\bar{\lambda}_2^f$ is linearly stable in the $\bar{\lambda}_4$ direction; when $\bar{\lambda}_4$ is larger than the critical value, the interface diverges in finite time. This divergence value of $\bar{\lambda}_4$ is found to be much smaller than that of $\bar{\lambda}_2$. According to our simulation, we can draw a rough illustration of the phase diagram in the $(\bar{\lambda}_4,\bar{\lambda}_2)$ plane, as shown in Fig. 2.

In order to understand the divergence of this model, we analyze the behavior of the dynamical equation without the noise term. We first review the case for the deterministic KPZ equation [2], which is basically Burger's equation [11]:

$$\frac{\partial h}{\partial t}=\nu\nabla^2 h+\lambda(\nabla h)^2.$$

Using the Hopf-Cole transformation $h\rightarrow(\nu/\lambda)\ln(\bar{h})$, the nonlinear equation for h can be transformed to a linear diffusion equation for $\bar{h}$. Therefore the deterministic KPZ equation is well behaved regardless of the initial condition.

For the deterministic LDTN equation in $d=1+1$ dimension,

$$\frac{\partial h}{\partial t}=-\nu\frac{d^4h}{dx^4}+\lambda\frac{d^2}{dx^2}\left[\frac{dh}{dx}\right]^2, \quad (6)$$

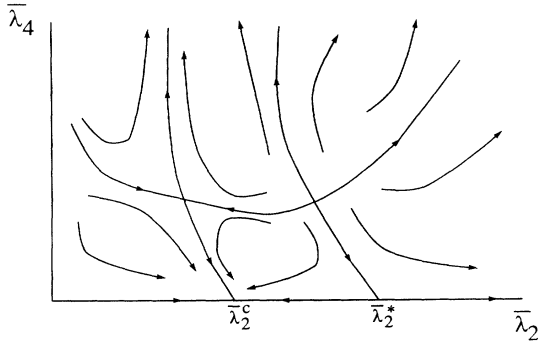

FIG. 2. The illustration of a plausible phase diagram in $(\bar{\lambda}_2,\bar{\lambda}_4)$ plane.

we first investigate the equation in the limit where $\nu \to 0$. Suppose the initial condition is a Gaussian; the nonlinear term will be maximal at the peak of the Gaussian, and the peak will become sharper and eventually diverge in finite time.

For finite surface diffusion constant ν, we have done an initial value simulation for the partial differential equation using an adaptive mesh size method. For initial conditions with large enough slopes and changes of curvature, the nonlinear term dominates the stabilizing linear term and the equation will also diverge [12]. This means that Eq. (6) is nonlinearly unstable.

In the usual growth model, the equilibrium state (when the width no longer depends on time) typically has a small expectation value of slope, which goes to zero as we go to larger length scale. So even though the dynamics could in principle develop singular behavior, the noise never drives the system to a state rough enough to be diverged by the nonlinear term. For example, the growth model proposed by Sun, Guo, and Grant [13], has exactly the same dynamics as the LDTN model, but with a much weaker noise term, and therefore does not develop any singularity.

But in the two-dimensional LDTN model, the equilibrium state has a finite expectation value of slope, i.e., finite $\langle (dy/dx)^2 \rangle^{1/2}$ independent of system size. So if the nonlinear coupling constant $\bar{\lambda}_2$ is large enough, the growth of some local peak region will be dominated by the nonlinear term and the nonlinear term will drive the peak towards infinity in finite time.

For the real SOS type atomistic block dropping models, because of the existence of a short distance cutoff, i.e., the block size of the particle, the interface will not generate singular behavior. But the growth can be dominated by individual columns for large enough nonlinear coupling. The height of these columns will be much higher than that of immediate surrounding columns, and they will not be relaxed down because of the nonlinear effect. This is exactly what a recent atomistic simulation by Kessler, Sander, and Levine [14] has found. When they put in realistic isotropic surface diffusion into the relaxation process, they observe that for small surface diffusion, the width versus time curve follows the power law $\omega \sim t^{\alpha}$ with $\alpha \sim 1/4$–$1/3$ for a certain time period depending on some details of the model, and then suddenly turns into a much faster growing process. Examination of the profile shows that the later stage of growth corresponds to independent columnar growth. As they increase the surface diffusion, this behavior disappears. The physical reason behind forming large peaks is the insufficiency of the relaxation mechanism, namely, the surface relaxation. For example, if the height difference between two sites which are close to each other (on the substrate) becomes very large, the surface diffusion mechanism will not reduce the difference sufficiently because these two points are far apart along the interface. It would be very interesting to extract from the atomistic simulations the quantitative information about the parameters ν and λ in the continuum model, and compare this transition with our study. Until now, we have not been able to do that.

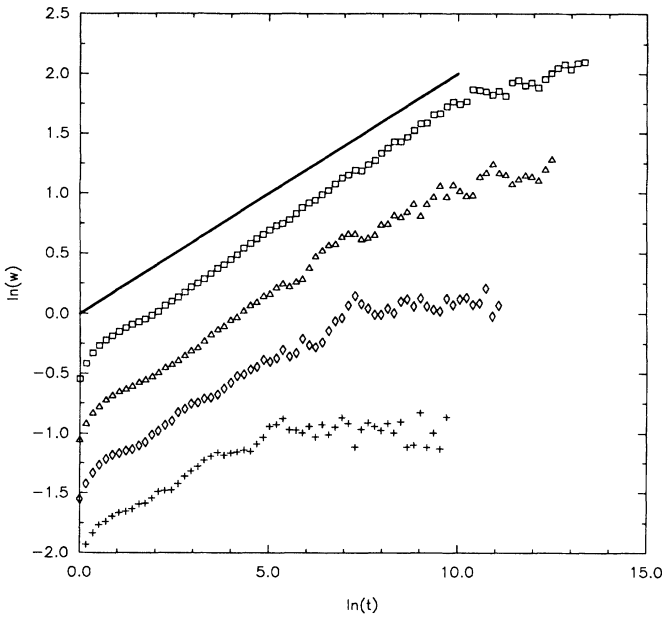

FIG. 3. Width vs time for different systems sizes, $N = 8\times8$, 16×16, 32×32, and 64×64 (from bottom to top) for $d = 2+1$ dimensions, $\bar{\lambda}_2 = 1.0$ each set of data is shifted upwards successively by 0.5 with respect to the previous set. The solid line has a slope of 1/5.

We conclude our paper by briefly discussing the results for $d = 2+1$ dimensions. We have simulated Eq. (4) in $d = 2+1$ dimensions for systems sizes $N = 8\times8$, 16×16, 32×32, and 64×64 at various values of $\bar{\lambda}_2 = 1$, 3, 5, 7. The results for $\bar{\lambda}_2 = 1.0$ are shown in Fig. 3. The time-dependent part of the width follows a power law $w \sim t^{\alpha}$, with $\alpha = 0.21 \pm 0.03$, which is in good agreement with the predicted value of $\chi/z = 1/5$, and the static exponent χ is found to be approximately 0.71, also in good agreement with the predicted value of 2/3. For the largest value of $\bar{\lambda}_2$ in our simulation, we do not observe any singular behavior of the interface. We also simulate the equation with nonzero $\bar{\lambda}_4$, but it does not change the exponents, meaning that the higher-order terms are irrelevant. We think even though the deterministic equation could also be nonlinearly unstable here, the fluctuation is not large enough to drive the system to divergence. This will be true also for higher dimensions.

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