Groove instabilities in surface growth with diffusion

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The existence of a grooved phase in linear and nonlinear models of surface growth with horizontal diffusion is studied in $d=2$ and 3 dimensions. We show that the presence of a macroscopic groove, i.e., an instability towards the creation of large slopes and the existence of a diverging persistence length in the steady state, does not require higher-order nonlinearities but is a consequence of the fact that the roughness exponent $\alpha \geq 1$ for these models. This implies anomalous behavior for the scaling of the height-difference correlation function $G(x)=\langle |h(x)-h(0)|^2\rangle$ which is explicitly calculated for the linear diffusion equation with noise in $d=2$ and 3 dimensions. The results of numerical simulations of continuum equations and discrete models are also presented and compared with relevant models.

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Recently, there has been considerable effort in understanding the dynamics of growing surfaces [1]. Much of this interest and activity is based on the recognition that surface fluctuations exhibit scaling behavior in both time and space. In particular, assuming an initially flat interface, the scaling of the interface width is expected to be of the form [2] $w(L,t)=L^{\alpha}f(t/L^{z})$, where $w(L,t)$ is the interface width on length scale L at time t, $z=\alpha/\beta$ is the dynamic exponent, and the scaling function $f(x)\sim x^{\beta}$ for $x \ll 1$ and $f(x)\rightarrow$const for $x \gg 1$. In an effort to understand the early-time morphology of thin-film growth such as molecular-beam epitaxy (MBE), considerable effort has been concentrated on the study of models in which surface diffusion as well as deposition (shot) noise were included [3–6].

Recently, Siegert and Plischke [7] considered a model corresponding to surface diffusion with deposition noise for which the Langevin equation [for a $(d-1)$-dimensional surface in d dimensions] may be written,

$$\frac{\partial h}{\partial t}=\Lambda_c\sqrt{g}\,\Delta\frac{\delta F}{\delta h}+\eta\ , \tag{1a}$$

where

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

is the Laplacian operator along the surface, and the Ginzburg-Landau functional was taken as the free energy of the drumhead model, $F=\int d^{d-1}x\sigma\sqrt{g}$, $g=1+(\nabla h)^2$ with $\langle \eta(x,t)\eta(x',t')\rangle=2D\delta^{d-1}(x-x')\delta(t-t')$. For this model they pointed out that all nonlinear terms (powers of $|\nabla h|$) are relevant for $d=2$ and are marginal in $d=3$. Following this observation, they studied a discrete model of surface deposition with horizontal diffusion, which they believed corresponded to Eq. (1). For the case in which a fourth-order nonlinearity ($|\nabla h|^4$) was included in the Hamiltonian for this model ($g_4\neq 0$) in $d=1+1$ dimensions, an interesting breakdown of scaling of the structure factor $S(k)$ was observed in addition to the formation of a grooved surface for which the exponent α governing the scaling of saturation width with system size L was approximately 3.6. This behavior was compared with that of the $|\nabla h|^2$ case ($g_4=0$, $\alpha\simeq 1.2$) for which no breakdown of structure factor scaling was observed, which was characterized as being in a "rough" rather than a grooved state.

In this article, we point out that while there is no breakdown in the scaling of the structure factor as in the nonlinear model, a grooved state at saturation (defined as an instability towards the creation of large slopes as well as a diverging persistence length) already occurs for the $g_4=0$ Siegert-Plischke diffusion model as well as for the linear equation,

$$\frac{\partial h}{\partial t}=-\kappa\nabla^4 h+\eta \tag{2}$$

in $d=2$ and 3 for which it is known that $\alpha=(5-d)/2$ and $\beta=(5-d)/8$. In $d=2$, one has $\alpha=\frac{3}{2}$ while in $d=3$ $\alpha=1$. The existence of a groove instability at saturation is a consequence of the fact that a self-affine surface cannot exist with roughness exponent $\alpha\geq 1$. In addition we show that this implies anomalous behavior for the scaling of the height-difference correlation function $G(x;t)=\langle [h(x;t)-h(0,t)]^2\rangle$. Finally, we contrast the behavior of these models with that of the discrete "horizontal diffusion" model previously studied by Wolf and Villain [3,5], as well as the $g_4\neq 0$ model of Siegert and Plischke.

For the case of the linear equation (2), we can explicitly understand the existence of a grooved state by considering the exact solution for the height-difference correlation function $G(x;t)=\langle [h(x;t)-h(0;t)]^2\rangle$ which may be written (starting from a flat interface at $t=0$) [8],

$$G(x;t)\simeq\frac{2D}{\pi^{d-1}\kappa}\int_{\Lambda_L}^{\Lambda_a}\frac{dk}{k^{6-d}}[1-\cos(kx)](1-e^{-2\kappa k^4 t})\ , \tag{3}$$

where the cutoffs are $\Lambda_L\sim 1/L$ and $\Lambda_a\sim 1/a$, where a is the short length-scale cutoff and L is the system size. Changing variables to $u=kx$, this may be rewritten,

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$$G(x;t) \simeq \frac{4D}{(2\pi)^{d-1}\kappa} x^{5-d} \int_{cx/L}^{cx/a} \frac{du}{u^{6-d}}(1-\cos u) \times [1-e^{-2\kappa u^4 t/x^4}] . \quad (4)$$

For $t/L^4 \gg 1$, the exponential may be neglected, and for the case $x/L \ll 1 \ll x/a$, limits of the integral may be taken to be zero and ∞, assuming that the integral converges. One thus has $G(x;\infty) \sim x^{2\alpha} \sim x^{5-d}$ in agreement with the general expression for α. For $d=2$ and 3 dimensions, however, the integral diverges at $u=0$ so that this is not the case. In particular, for $d=2$ one obtains $G(x;\infty) \sim Lx^2$, while for $d=3$ one obtains $G(x;\infty) \sim x^2 \ln(L/a)$. From inspection of Eq. (4) one also obtains in the early-time regime $x^4 \ll \kappa t \ll L^4$, $G(x;t) \sim t^{1/4} x^2$ in $d=2$, and $G(x;t) \sim \ln(t/x^4) x^2$ in $d=3$. Thus, for $d=2$ and 3 the scaling of the correlation function $G(x;\infty)$ disagrees with the known scaling of the saturation width as a function of system size, which is already a sign that the surface at saturation is not self-affine. Our results indicate that in $d=2$, the surface at saturation consists essentially of linear pieces with slope of order $\pm\sqrt{L}$ [since the average slope over a distance x is $\sqrt{G(x)}/x$], which corresponds essentially to a set of grooves. Assuming a single groove whose width is the system size, this implies that the saturation width scales as $L\sqrt{L} = L^{3/2}$, in agreement with the known results for this case.

For the linear case, the average slope may also be directly calculated in $d=2$ as

$$\langle (\nabla h)^2 \rangle \sim \begin{cases} \int_{\Lambda_L}^{\Lambda_a} \frac{dk}{k^{4-d}}(1-e^{-2\kappa k^4 t}) \sim L \quad \text{as } t \to \infty & (5a) \\ t^{1/4} \int_0^\infty \frac{du}{u^2}(1-e^{-2\kappa u^4}) \quad \text{for } t/L^4 \ll 1 . & (5b) \end{cases}$$

In $d=3$ one obtains $\langle (\nabla h)^2 \rangle \sim \ln(L)$ at saturation and $\sim \ln(t)$ at early time. Inspection of Eq. (4) also indicates that after a time t in $d=2$ the scale over which the "linear" behavior $[G(x) \sim x^2]$ of the surface occurs grows as $x_\parallel \sim t^{1/4}$. Thus one may conclude that a crude description of the interface at time t is that it consists of grooves of width $x_\parallel \sim t^{1/4}$, and slope $t^{1/8}$, so that the surface width grows as $t^{3/8}$ as expected. As time goes on, these grooves coarsen and become steeper until finally at saturation one has a single groove of slope $\sqrt{L}$.

The fact that there is a single groove at saturation for the linear equation may be seen more clearly by considering the correlation function of the slope $m = \nabla h$ at saturation,

$$\begin{aligned} G_{\Delta m}(x) &= \langle [m(0)-m(x)]^2 \rangle \\ &\sim x \quad \text{in } d=2 , \\ &\sim \ln(x) \quad \text{in } d=3 \quad \text{for } x/L \ll 1 . \end{aligned} \quad (6)$$

Thus, in $d=2$, one has $G_{\Delta m}(x)/\langle m^2 \rangle \simeq x/L$ $[\simeq \ln(x)/\ln(L)$ in $d=3]$, which implies an orientational persistence length at saturation $\xi_P \sim L$. One may also calculate the product correlation function $G_{mm}(x) = \langle m(0) m(x) \rangle$ at saturation,

$$G_{mm}(x) \sim L \quad \text{for } x/L \ll 1 , \quad (7a)$$

$$G_{mm}(x) \to 0 \quad \text{for } x/L \approx 0.78 . \quad (7b)$$

In $d=3$ one obtains $G_{mm}(x) \sim \ln(L/x)$. The last result [Eq. (7b)] is obtained for the case of periodic boundary conditions in $d=2$ by converting the integral for $G_{mm}(x)$ to a discrete sum over k and summing numerically [9]. Thus, as already stated, there is an orientational persistence length at saturation of order L, corresponding to a single groove. This is in contradiction to the statement (made by Golubovic and Bruinsma [10] and again by Golubovic and Karunasiri [11]) that in the steady-state there is a persistence length which depends on D and κ as $\xi_P \sim \kappa/D$ (or in $d=3$ as $e^{\kappa/D}$). This statement was made under the assumption that the slope is of order 1, which is not the case for large L at late times.

Finally, we point out that once the groove is formed it may "move." This is analogous to the Goldstone mode described in Ref. [7]. Specifically, one may calculate the correlation function $G_{mm}(t)$ in the steady-state $(\kappa t' \gg L^4)$ in $d=2$,

$$G_{mm}(t) = \langle m(x,t') m(x,t'+t) \rangle < L \exp(-\kappa t/L^4) \quad \text{for } \kappa t/L^4 \gg 1 . \quad (8)$$

The fact that $G_{mm}(t)$ goes to zero for $\kappa t/L^4 \gg 1$ is due to the motion of the groove. A similar result may be obtained in $d=3$.

In order to test these predictions and obtain a clear picture of the interface in models with horizontal surface diffusion, we have numerically integrated Eq. (2) in $d=2$ and 3, using a finite-difference method on a lattice ($\kappa=1$, $D=0.1$, lattice spacing $\Delta x=1$, and time-step $\Delta t=0.01$) starting from a flat interface at $t=0$. Figure 1 shows the development of the groove as well as the final single groove at saturation for a system of size $L=128$ in $d=2$.

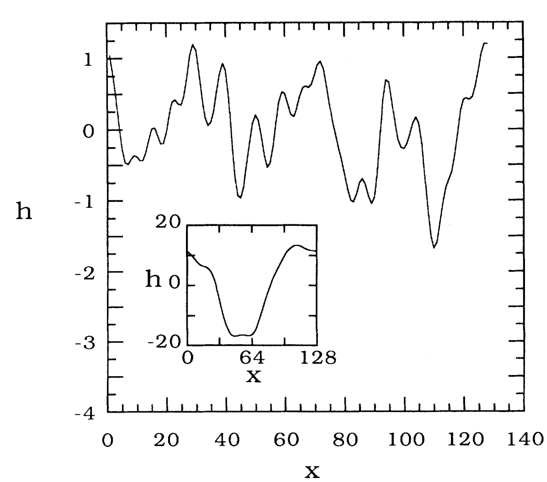

FIG. 1. Interface profile obtained from numerical solution of Eq. (2) with $D=0.1$, $\kappa=1.0$, $L=128$, at time $t=10.0$. Inset shows snapshot of single groove formed at time $t=100\,000$.

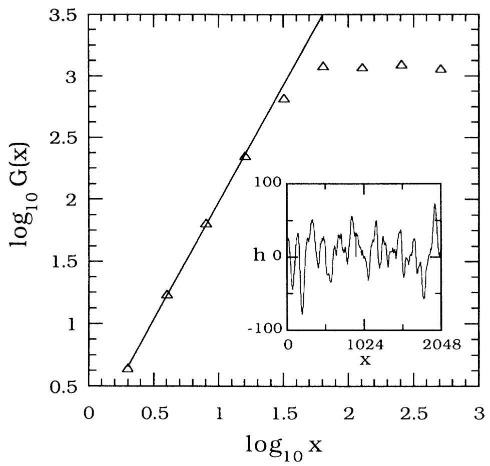

FIG. 2. Correlation function $G(x)=\langle[h(x)-h(0)]^2\rangle$ for Eq. (2) at "early time" for $L=1024$, averaged from $t=10\,000$ to 20 000. Fit to linear region has slope 1.9. Inset shows interface at $t=20\,000$.

Figure 2 shows the correlation function at "early" time for a larger system ($L=1024$) while the inset shows the corresponding picture of the interface. As predicted, the correlation function $G(x)$ scales approximately as x^2 (rather than x^3) at early-time as well as in the late-time grooved state. Figure 3 shows the surface obtained from numerical integration of Eq. (2) in $d=3$ for a system of size 32×32 at late time. We again see that the steady-state surface appears to consist essentially of a single groove, although this case is somewhat marginal.

We have also conducted long-time simulations of the $g_4=0$ version of the Siegert-Plischke model in $d=1+1$ dimensions. Figure 4 shows a snapshot of the surface configuration for a system of size $L=128$ at $t=128\times10^6$. As can be seen from the figure, there is a well-defined groove whose width is of the order of the

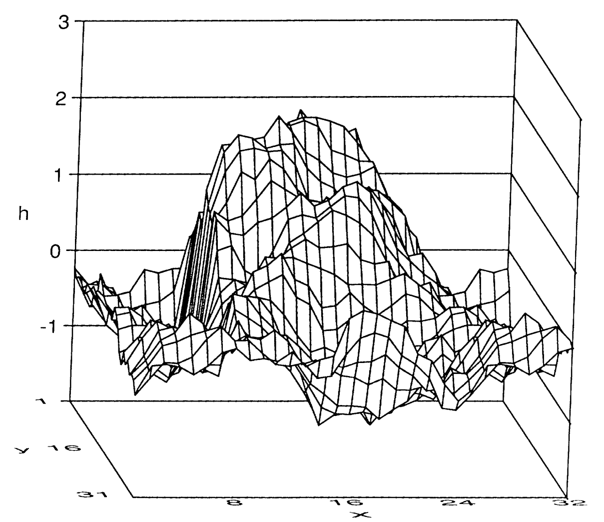

FIG. 3. Picture of interface obtained from numerical simulation of Eq. (2) in $d=3$ with $L=32$ showing a macroscopic "groove" at late time $t=10\,000$.

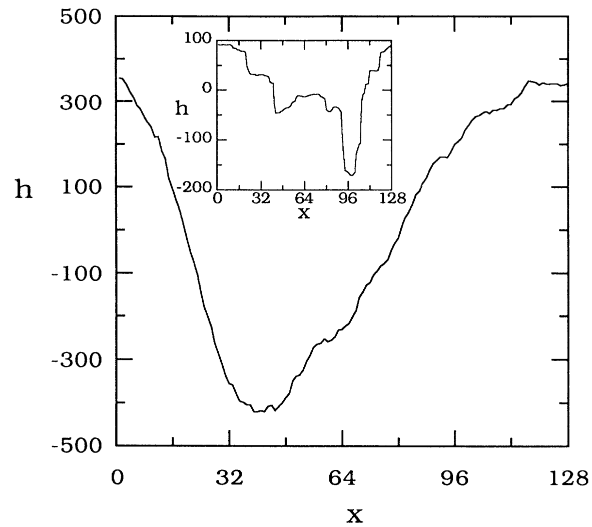

FIG. 4. Snapshot of interface profile for Siegert-Plischke model at $t=128\times10^6$ in $d=2$ with $g_4=0.0$, $\tau=0.1$, $\beta J=0.01$, and $L=128$. Inset shows snapshot for Wolf-Villain model with $L=128$ at $t=3.2\times10^6$.

system size as predicted. The inset shows a similar snapshot at late time for the Wolf-Villain model. In this case, however, the interface has much larger fluctuations and unlike our predictions for Eq. (2), the width of the groove is significantly less than the system size. In this regard we note that while previous data for the scaling behavior of the interface width had suggested [3,4] that this model is in the same universality class as Eq. (2), recent work [12,13] indicates that it actually has a small but finite surface-tension term $\nu\nabla^2 h$ in addition to the $\nabla^4 h$ term. Thus the existence of a crossover from the $\nabla^4 h$ to the $\nabla^2 h$ (Edwards-Wilkinson) [14] universality class (which has no groove since $\alpha<1$) at large length scales may account for the large fluctuations observed in the inset of Eq. 4.

We have also numerically simulated the nonlinear equation [4,6],

$$\frac{\partial h}{\partial t}=-\kappa\nabla^4 h+\lambda_2\nabla^2(\nabla h)^2+\eta\ , \tag{9}$$

for which it is known that $\alpha=(5-d)/3$. In $d=2$, $\alpha=1$ which is marginal. Figure 5 shows the steady-state configuration at two successive times after saturation, indicating the existence of a macroscopic groove as well in this case.

Finally, as a comparison between our results and those of Ref. [7], we show in Fig. 6 scaled plots of the surface profiles (averaged over several runs, and shifted so that the maximums are at the same location in x) for three different models including the nonlinear model of Siegert and Plischke [7]. As can be seen from the figure, the three grooves all have roughly the same shape. However, for the nonlinear ($g_4\neq0$) Siegert-Plischke model there is a fairly sharp cusp or slope-discontinuity at the bottom of the groove. This sharp discontinuity is due to a deterministic instability which occurs in this model and which accounts for the observation of pronounced oscillations

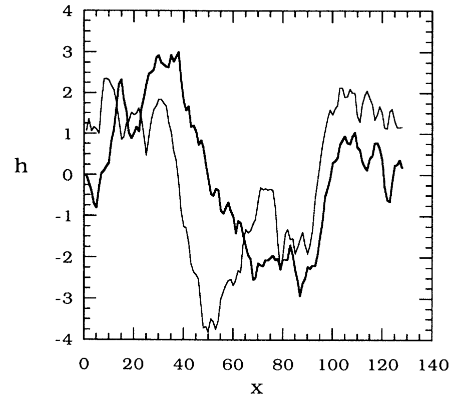

FIG. 5. Steady-state interface from numerical solution of Eq. (9) ($\kappa=1.0$, $\lambda_2=-1.0$, $D=0.1$) for system size $L=128$ showing groove at two different times ($t=200000$ and 600000) in saturation regime.

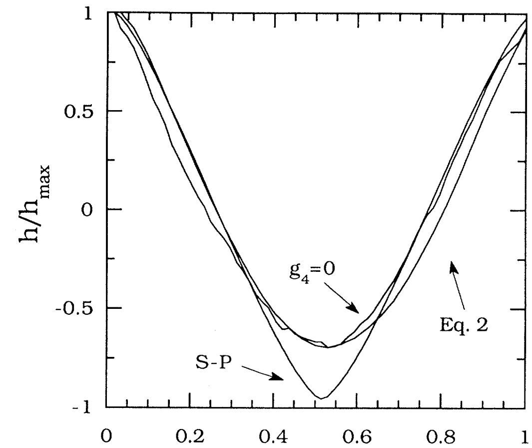

FIG. 6. Averaged scaled surface profiles for three different models at late time. Curve labeled Eq. (2) is linear model, averaged over 500 correlation times for $L=32$. Curve labeled $g_4=0$ is Siegert-Plischke model with $g_4=0$ and $L=64$ averaged over 20 runs. Lowest curve is for nonlinear Siegert-Plischke model with $g_4=1.0$, $\tau=0.1$, $\beta J=0.01$, and $L=64$, averaged over 20 runs.

and lack of scaling of the structure factor $S(k)$ [7]. Such an instability does not occur for the other two models and therefore the bottom of the groove is more rounded and oscillations in $S(k)$ either are not present or are significantly reduced for these models.

In conclusion, we have shown that the existence of a macroscopic grooved state with an instability toward the creation of regions of large slope and a diverging persistence length is not a specific feature of the nonlinear model studied in Ref. [7], but also occurs for the linear Eq. (2) as well as the $g_4=0$ version of this model. However, the lack of scaling of the structure factor $S(k)$ is a specific feature of the $g_4\neq0$ model and does not occur for the models discussed here. The existence of a macroscopic grooved state is most likely due to the fact that in all of these models the full diffusion along the surface is not taken into account.

In the case of the linear model Eq. (2) we have analytically and numerically demonstrated the existence of a macroscopic groove whose slope scales as $\sqrt{L}$ ($\sqrt{\ln L}$) in $d=2$ (3) and of a persistence length which diverges with increasing L at saturation. In addition, we have demonstrated the existence of anomalous behavior for the scaling of the height-difference correlation function $G(x)$. As we have pointed out, this is expected to occur for any model with roughness exponent $\alpha\geq1$. For the Wolf-Villain model, however, the situation appears to be different since the measured value of α is greater than 1 while the persistence length does not diverge with system size. This is an indication that, as suggested in Refs. [12] and [13], this model is crossing over to Edwards-Wilkinson ($\alpha<1$) scaling behavior at large length scales.

We note that recent suggestions [4,6] for the interface growth equation appropriate for molecular-beam epitaxy include the existence of a nonlinear term of the form $\nabla^2(|\nabla h|^2)$ as in Eq. (9) which results in a value of α less than 1 in the physically relevant case $d=3$. Thus a groove instability is not expected in this case. However, the derivation of this equation is based on the assumption of small slopes, and does not include the full nonlinearities due to curvature included in Eq. (1). Thus, it is still not clear whether or not this equation is really valid in the late-time regime for MBE growth. If, in fact, the nonlinear terms do lead to $\alpha\geq1$ in $d=3$, then the solid-on-solid approximation may not be valid either, as overhangs may be expected to form.

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