Dynamic scaling in a (2+1)-dimensional limited mobility model of epitaxial growth

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We study statistical scale invariance and dynamic scaling in a simple solid-on-solid (2+1)-dimensional limited mobility discrete model of nonequilibrium surface growth, which we believe should describe the low temperature kinetic roughening properties of molecular beam epitaxy. The model exhibits long-lived *transient* anomalous and multiaffine dynamic scaling properties similar to that found in the corresponding (1+1)-dimensional problem. Using large-scale simulations we obtain the relevant scaling exponents, and compare with continuum theories. [S1063-651X(97)13905-8]

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A key issue in kinetic surface roughening [1,2] is making a connection between theoretical growth universality classes, as defined by continuum growth equations, for example, and experimentally observed rough growth in real surfaces that generally depends on many details such as growth conditions (e.g., temperature, surface orientation, and growth rate) and atomistic rules controlling attachment, detachment, evaporation, and, most importantly, diffusion of the deposited adatoms at the growth front. The concept driving much of the kinetic surface roughening research is that only a few universality classes [1,2] describe the *asymptotic* growth properties in many seemingly different nonequilibrium surface growth problems as most of the details are *irrelevant* from a renormalization group viewpoint and do not affect the asymptotic behavior. Much recent work has gone into building simple atomistic discrete nonequilibrium growth models that catch the essential aspects of a complicated growth problem and include only the *relevant* dynamical processes determining the asymptotic growth behavior. One such nonequilibrium growth model was introduced by one of us in Ref. [3] in the context of one dimensional molecular beam epitaxy. This growth model has since been extensively studied [4], and it seems to describe [4] well the low temperature growth properties of realistic stochastic Monte Carlo simulation results of molecular beam epitaxy. Although the growth model introduced in Ref. [3] has been fairly extensively studied in the literature [4], almost all of the existing work is in $1+1$ dimensions where a *one* dimensional substrate roughens as it grows. We present in this paper results of a systematic study of the growth model of Ref. [3] in the physically relevant $2+1$ dimensions.

In our growth model [3], atoms are randomly deposited on an $L\times L$ (we have studied system sizes up to $L=10^3$ with a maximum of 10^7 deposited layers, which amount to the deposition of up to 10^{13} atoms) flat substrate under solid-on-solid deposition and growth conditions. If a randomly deposited atom has *at least* one lateral nearest-neighbor bond (i.e., if its initial coordination number is 2 or more), then it is incorporated at the deposition site and stays there forever. Otherwise the atom could move to a nearest-neighbor lateral site (with no restriction on the number of vertical sites it moves in the growth direction) for incorporation provided it can *increase* (but not necessarily *maximize*) its coordination number at the final site. If no such nearest-neighbor lateral site is available (which would increase the atom's coordination number), the atom is again incorporated at the site of deposition. If more than one final site could increase its coordination number, the atom randomly moves to any one of these final sites with equal probability and is incorporated there permanently. The motivation for this manifestly nonequilibrium growth model is that during low temperature molecular beam epitaxy it is unlikely for atoms to break lateral bonds, and only deposited adatoms without any lateral bonds can move with appreciable mobility. This is a typical example of a limited mobility growth model where atoms at kink and trapping sites (i.e., those with at least one lateral bond) simply do not move. A typical example of a saturated growth morphology resulting from these rules is shown in Fig. 1.

We first study the *global* scaling behavior of the surface by considering the dynamical surface width W, which is the root-mean-square height fluctuation of the growing surface, defined as

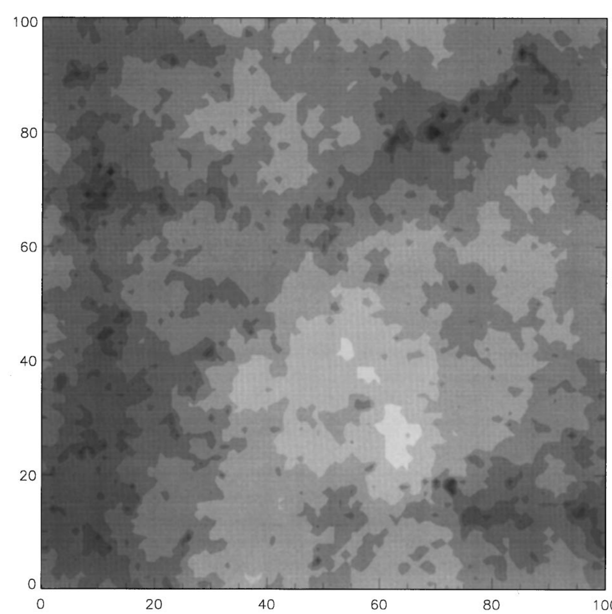

FIG. 1. The saturated growth morphology on a 100×100 substrate at 3×10^6 ML. Darker (lighter) shades represent lower (higher) points on the surface.

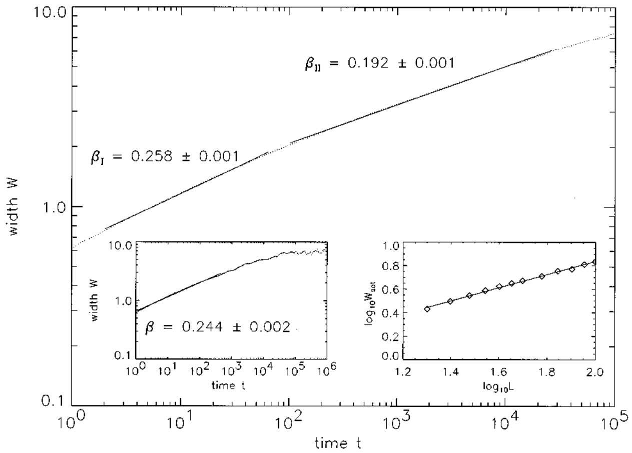

FIG. 2. The interface width W as a function of deposition time t in the 500×500 system (left inset: 100×100 system). Right inset: The saturation width W_{sat} vs the substrate size L.

$$W(L,t)=\langle (h-\langle h\rangle)^2\rangle^{1/2}. \tag{1}$$

If there is statistical scale invariance in the problem, then W scales with the lateral system size L and time t as [1,2]

$$W(L,t)\sim L^{\zeta}f(L/\xi(t)), \tag{2}$$

where the scaling function $f(y)$ is

$$f(y)=\begin{cases}\text{const} & \text{for } y\ll 1\\ y^{-\zeta} & \text{for } y\gg 1.\end{cases} \tag{3}$$

Here ζ is the *roughness* exponent, and $\xi(t)$ is the lateral correlation length obeying the dynamic scaling relation $\xi(t)\sim t^{1/z}$ where z is the *dynamical* exponent. Time in the simulation is measured in the average number of deposited layers. Combining these scaling relations the interface width W can be written as $W(L,t\ll L^z)\sim t^{\beta}$ and $W(L,t\gg L^z)\sim L^{\zeta}$, where $\beta=\zeta/z$ is the *growth* exponent. Our calculated results for $W(L,t)$ are shown in Fig. 2 for a substrate size $L=500$ and 100 (inset). The calculated growth exponent (for $L=500$) clearly shows a crossover from an initial value of about 0.25 to an asymptotic value of about 0.2. We find the same crossover behavior in other large systems (L = 500–1000) we have studied — in smaller systems ($L=100$), however, the crossover to the asymptotic exponent ($\beta\approx 0.2$) is not seen and β remains around 0.25 as can be seen in the inset of Fig. 2. In a second inset, the values of the saturation width $W(L,t\rightarrow\infty)$ are plotted as a function of the system size $L=20$–100. (The small L values in the saturation plot reflect the high value of $z\sim 3$ in the problem: $L=100$ requires more than 10^6 layers to saturate.) The slope yields the *global* roughness exponent $\zeta=0.56\pm 0.1$.

The *local* scaling behavior is studied by calculating the height-height correlation function $G(\mathbf{r},t)$ defined as $G(\mathbf{r},t)=\langle |h(\mathbf{x}+\mathbf{r},t)-h(\mathbf{x},t)|^2\rangle_{\mathbf{x}}^{1/2}$. The conventional scaling form of the correlation function is

$$G(\mathbf{r},t)\sim r^{\zeta}\hat{f}(r/\xi(t)), \tag{4}$$

where $r=|\mathbf{r}|$ and the asymptotic behavior of the scaling function $\hat{f}(y)$ is the same as that of the function f in Eq. (3). It is, however, known that the corresponding *one dimensional* growth model exhibits anomalous [5] and multiaffine [6] scaling behavior in contrast to the simple conventional dynamic scaling defined in Eq. (3). We find the same to be true in the $(2+1)$-dimensional model also with the only difference being that the anomalous behavior is manifestly a long-lived (over three decades in time) transient in $2+1$ dimensions whereas in $1+1$ dimensions it lasts [4–6] for at least eight decades in time (and perhaps much longer; the true asymptotic limit may not have yet been reached in $1+1$ dimensions [7]). In order to study multifractality, we follow Krug [6] and define a generalized correlation function by considering higher moments of the equal-time height difference correlator:

$$G_q(\mathbf{r},t)=\langle |h(\mathbf{x}+\mathbf{r},t)-h(\mathbf{x},t)|^q\rangle^{1/q}. \tag{5}$$

These functions G_q [note that for $q=2$ one gets back the G of Eq.(4)] have q-dependent roughness exponents if the growing surface is multiaffine whereas they all scale with the same exponent in a self-affine statistically scale invariant surface.

In Fig. 3 we show the multifractal behavior of $G_q(\mathbf{r},t)$ in our model. We find exactly the same qualitative and quantitative multifractal behavior (with the same critical exponents within numerical accuracy) for the different system sizes ($L=100-1000$) we study. We summarize below (see Refs. [2,4–7] for details) the asymptotic behavior of the correlation function G_q in the anomalous and multiaffine scaling situation depicted in Fig. 3.

Multiaffine scaling (as opposed to statistically scale invariant self-affine scaling) signifies q-dependent dynamic scaling properties of G_q:

$$G_q(\mathbf{r},t)\sim\begin{cases} r^{\zeta_q} & \text{for } r\ll\xi(t)\\ t^{\beta} & \text{for } r\gg\xi(t).\end{cases} \tag{6}$$

In addition, the *local* and the *global* dynamic scaling properties are necessarily different in this anomalous scaling situation. Thus, $\hat{f}$ in Eq. (4) scales differently from f in Eqs. (2) and (3):

$$\hat{f}(y)=\begin{cases} y^{-\alpha_q} & \text{for } y\ll 1\\ y^{-\zeta_q} & \text{for } y\gg 1.\end{cases} \tag{7}$$

Using the asymptotic behavior of $\xi(t)$ we obtain the asymptotic behavior for the qth moment of the height-height correlation function as

$$G_q(\mathbf{r},t)\sim\begin{cases} r^{\zeta_q-\alpha_q}t^{\alpha_q/z} & \text{for } r\ll t^{1/z}\ll L\\ r^{\zeta_q-\alpha_q}L^{\alpha_q} & \text{for } r\ll L\ll t^{1/z}\\ t^{\beta} & \text{for } r\gg t^{1/z}.\end{cases} \tag{8}$$

Note that an additional scaling exponent α_q is needed to fully characterize the anomalous multiscaling situation. In the conventional scaling situation $\alpha_q\equiv 0$ and $\zeta_q\equiv\zeta$.

In Fig. 3(a) we show our results for G_q for $L=500$ at 10^3 ML. It is obvious that the lines for different q have different *local* roughness exponents, $\zeta_q'\equiv\zeta_q-\alpha_q$, implying

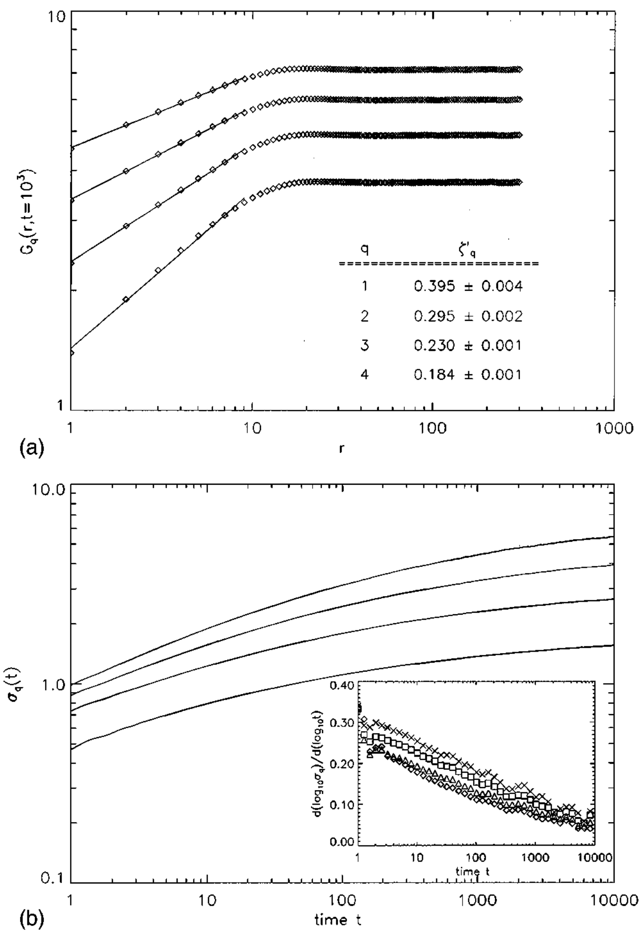

FIG. 3. (a) Height-height correlation function $G_q(r)$, $q=1-4$ from bottom to top. ($L=500$ at 10^3 ML). The solid lines indicate power law fits with slope ζ_q' as shown in the table at the lower right. (b) Nearest-neighbor height difference function $\sigma_q(t)$, $q=1-4$ from bottom to top ($L=1000$). Inset: The local derivative of $\log_{10}\sigma_q$ with respect to $\log_{10}t$. $\Diamond$, $\triangle$, $\square$, and $\times$ correspond to $q=1,$ 2, 3, and 4, respectively.

multifractality. The *local* roughness exponents ζ_q' vary from 0.184 for $q=4$ to 0.395 for $q=1$. Comparing $\zeta_2'=0.3$ with $\zeta=0.56$ (cf. Fig. 2) we conclude that the local and global roughness in our problem have different behavior (anomalous scaling [5]), which is one direct consequence of the spatial multiscaling. In order to gain more insight into the multiaffine scaling behavior we show in Fig. 3(b) the time evolution of the nearest-neighbor height difference and its higher moments $\sigma_q(t)\equiv G_q(\mathbf{r}=\mathbf{1},t)$ for $L=1000$. Substituting $r=1$ in Eq. (8), we get (following Ref. [6])

$$\sigma_q(t)\sim\begin{cases} t^{\alpha_q/z} & \text{for } t^{1/z}\ll L \\ L^{\alpha_q} & \text{for } t^{1/z}\gg L. \end{cases} \tag{9}$$

In the conventional scaling situation $\alpha_q=0$ for all q, and therefore $\sigma_q(t)$ quickly saturates to a constant value almost immediately. But in the anomalous scaling case $\sigma_q(t)$ grows with time [5,6,8] before saturating (at very long time) in the steady state when $\xi(t)\sim L$. The approximate crossover time for this saturation is $t_c\sim L^z$ and can be very large [5,6,8]. In Fig. 3(b) the nearest-neighbor height difference σ_q increases with time for all q in the small time region. For $1<t<20$, the slope (α_q/z) ranges from 0.22 ($q=1$) to 0.31 ($q=4$). It is absolutely clear, however, from Fig. 3(b) that $\sigma_q(t)$ is showing a crossover behavior where the *effective* exponent (α_q/z) of Eq. (9) is approaching zero long before the steady-state saturation regime where $\xi(t)\sim L$. It is obvious from the inset that the slopes of $\sigma_q(t)$ curves, instead of staying at constant values of α_q/z, are going down to zero around $t\sim10^5$, which is long before the expected saturation time $t_c\geqslant10^9$ ($L=1000$). [The corresponding $\sigma_q(t)$ results for a smaller $L=100$ system size show the vanishing of the effective exponents α_q/z around $t\approx10^3$, implying that multiscaling lasts for about three decades in time in a 100×100 system.] The bending of the $\sigma_q(t)$ curve as a function of t, which is glaringly apparent in our Fig. 3(b), can also be seen (in a much less pronounced fashion) in the corresponding (1+1)-dimensional results [4,6]. Our results presented in Fig. 3(b) make it obvious that the multiscaling observed in our (2+1)-dimensional simulations is a long-lived transient and is *not* the asymptotic behavior of the limited mobility epitaxial growth model introduced in Ref. [3]. Our results strongly indicate that the same must be true in $1+1$ dimensions also, which has already been suggested in Ref. [7], except that the multiscaling transient in $1+1$ dimensions lasts for at least 10^8 time steps and perhaps much longer. We have, in fact, studied a (1+1)-dimensional version of our problem for $L=10^4$ up to 10^8 ML of growth, and we find that α_q/z decreases from an initial value of $\alpha_q/z=0.195(q=1)$, $0.222(q=2)$, $0.262(q=3)$, $0.294(q=4)$ to $\alpha_q/z=0.107$, 0.143, 0.168, 0.179, definitively establishing that the multiscaling behavior [4–8] of the growth model introduced in Ref. [3] is an extremely long-lived transient, even in $1+1$ dimensions.

Having established that the anomalous multiscaling behavior of our limited mobility growth model, while being extremely interesting, is most definitely a *transient* (and *not* an asymptotic) behavior, we need to ask the obvious question: what is the universality class of the discrete growth model introduced in Ref. [3]? The asymptotic critical exponents $\beta\approx0.2$ and $\zeta\approx0.6$ in Fig. 2 are consistent with the nonlinear MBE growth equation introduced in Refs. [9] and [10]. We believe that the discrete growth model of Ref. [3], which we study here in $2+1$ dimensions, does indeed asymptotically belong to the universality class of the nonlinear growth equation [9,10]

$$\frac{\partial h}{\partial t}=\nu_4\nabla^4h+\lambda_2\nabla^2(\nabla h)^2+\eta, \tag{10}$$

which in $2+1$ dimensions has [10] the critical exponents $\beta=1/5$ and $\zeta=2/3$, which, within numerical errors, is consistent with the critical exponents we obtain in this paper [11]. The corresponding linear equation [with $\lambda_2=0$ in Eq. (10)] has $\beta=1/4$ in $2+1$ dimensions, explaining the crossover from $\beta\approx0.25$ to 0.20 seen in our Fig. 2. [It should be emphasized that in $1+1$ dimensions even the largest simulations of this model [3–7] obtain $\beta\approx0.375$, which is consistent with the linear growth equation ($\lambda_2=0$), and the anticipated crossover to the nonlinear model has not yet been definitely established.] Note that Eq. (10) breaks the up-down symmetry in the problem and is therefore a true nonequilibrium model of growth. The measured skewness

$S\equiv\langle(h-\langle h\rangle)^3\rangle/\langle(h-\langle h\rangle)^2\rangle^{3/2}$ and the effective fourth cumalant $Q\equiv\langle(h-\langle h\rangle)^4\rangle/\langle(h-\langle h\rangle)^2\rangle^2-3$ in our simulations are approximately -0.5 and 1.2, respectively, in the saturated regime.

The anomalous scaling and multifractality in our results most likely arise from the existence of an infinite series of higher-order marginally irrelevant terms of the form $\Sigma_{n=2}^{\infty}\lambda_{2n}\nabla^2(\nabla h)^{2n}$ on the right-hand side of Eq. (10), as has recently been speculated in Ref. [4] and discussed extensively in Ref. [7]. Further discussion [7] of this issue is beyond the scope of our work, and we only note that this infinite series of higher order gradient terms should have a much stronger influence in $1+1$ dimensions where they are all *marginally relevant*, explaining why the observed multifractality is so much more pronounced in $1+1$ dimensions [6].

Finally, we note that the standard Laplacian term, $\nu_2\nabla^2 h$, of the Edwards-Wilkinson equation [12] is absent for the growth model introduced in Ref. [3] in contrast to other popular solid-on-solid growth models [13,14] studied in the literature in the context of epitaxial growth. The absence of the $\nabla^2 h$ term [and its fourth order counterpart [10], the $\nabla\,(\nabla h)^3$ term] in the growth model of Ref. [3] is, in fact, an exact result arising from a hidden symmetry [2] in the problem, which makes the inclination-dependent current on a tilted substrate [15] in this growth model vanish exactly [our calculated current on a tilted substrate in our (2+1)-dimensional simulation is $\pm 10^{-4}$, which is zero within our numerical accuracy]. We have also studied the closely related Wolf-Villain model [14], where the deposited adatoms (*independent* of their initial lateral coordination) seek out the nearest-neighbor sites of *maximum* coordination, which is now definitely known [15,16] to asymptotically belong to the Edwards-Wilkinson universality class, where our measured tilt-dependent surface current has a small negative (downhill) value (increasing in magnitude with inclination) of approximately -0.1 at a slope of 2. We see essentially no multiscaling behavior in our (2+1)-dimensional Wolf-Villain model simulations: α_q/z for $L=500$ is essentially q independent and decreases from a very small initial value of about 0.06 to zero at $t\approx 10^3$. Clearly, the Wolf-Villain model behaves very differently from the model of Ref. [3] in $2+1$ dimensions even though their effective behavior is known to be ''almost'' identical in $1+1$ dimensions. The fact that these two models, Refs. [3] and [14], have different universality classes was actually pointed out some time ago [17] although substantial confusion still seems to exist about their relationship. Results presented here strongly support the idea that the limited mobility nonequilibrium epitaxial growth model introduced in Ref. [3] is asymptotically described by the nonlinear MBE growth equation (10) although there are interesting nonasymptotic corrections associated with the anomalous multiscaling phenomena. The significance of this finding lies in the fact that most experimental investigations [18] of the kinetic surface roughening phenomenon in epitaxial growth obtain large growth ($\beta\approx 0.2-0.3$) and roughness ($\alpha\approx 0.5-1$) exponents, indicating that the linear and the nonlinear MBE growth equation may be playing important roles in real growth. The model of Ref. [3] seems to be the only known solid-on-solid epitaxial growth model that is asymptotically *not* described by the linear second-order Edwards-Wilkinson equation, but by the fourth-order nonlinear MBE growth equation [19].

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[18] See, for example, experimental references cited in Refs. [1,2,4] above, and in G. Palasantzas and J. Krim, Int J. Mod. Phys. B **9**, 599 (1995).
[19] It may be useful to point out in this context that the exponents obtained in this paper agree quantitatively with the corresponding exponents obtained in Ref. [4] for the *full activated diffusion* MBE growth simulations in 2+1 dimensions (see, for example, Table III in Ref. [4]) including the multiscaling exponents.