Kinetic super-roughening and anomalous dynamic scaling in nonequilibrium growth models

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We argue that recently introduced models of surface-diffusion-driven nonequilibrium growth that are characterized by critical roughness exponents (α) exceeding unity ("super-rough" growth) exhibit an "anomalous" form of dynamic scaling whose asymptotic behavior is different from the usual scaling behavior of self-affine kinetic growth models with $\alpha < 1$. We propose a generalized scaling function for super-rough ($\alpha > 1$) growth and demonstrate its applicablity to several discrete nonequilibrium super-rough models.

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Nonequilibrium growth of thin films, where atomistic deposition on a $(d-1)$-dimensional flat substrate generates a kinetically rough interface growing dynamically in time in the "height" direction (i.e., perpendicular to the substrate plane), is considered [1] to obey the generic dynamic scaling behavior given by

$$W^2(L,t) \sim L^{2\alpha} f(L/\xi(t)) , \tag{1}$$

where W is the interface width (measured as the root-mean-square fluctuation of the growing surface height around its nominal average value), L is the lateral system size, α is the so-called roughness exponent, $f(x)$ is the dynamic scaling function, and $\xi(t)$ is the lateral (i.e., along the substrate) correlation length, beyond which the height fluctuations become uncorrelated. The scaling function $f(x)$ is taken [1] to have the following asymptotic behavior:

$$f(x) \sim \begin{cases} \text{const} & \text{for } x \ll 1 \\ x^{-2\alpha} & \text{for } x \gg 1 , \end{cases} \tag{2}$$

and the correlation function $\xi(t)$ obeys the dynamic scaling behavior

$$\xi(t) \sim t^{1/z} , \tag{3}$$

where z is the so-called dynamical exponent. Combining Eqs. (1)–(3), which form the basic dynamic scaling hypothesis for nonequilibrium growth, one finds

$$\begin{aligned} &W(L, t \ll L^z) \sim t^{\beta} \quad \text{where } \beta = \alpha/z , \\ &W(L, t \gg L^z) \sim L^{\alpha} , \end{aligned} \tag{4}$$

and concludes that the interface width obeys power-law scaling with growth time, and the saturated interface width (at long times) scales with the lateral system size L. The dynamic scaling hypothesis for the interface width [Eqs. (1) and (4)] asserts that $W^2/L^{2\alpha}$ is a general scaling function $f(x)$ of the variable $x = L/t^{1/z}$:

$$W^2/L^{2\alpha} \sim f(L/t^{1/z}) . \tag{5}$$

Observation of scaling as in Eq. (5), or of power-law behavior as in Eq. (4), is necessary and sufficient to establish the existence of dynamic scaling in a particular growth model. A large number of nonequilibrium growth models are found [1] to obey the dynamic scaling properties described by Eqs. (1)–(5).

One of the key features [1] of *all* of these growth models (with the exception of the ones to be discussed in this paper) is that the roughness exponent α, which characterizes the morphology of the saturated interface fluctuation for a fixed substrate size via the relation $W_{\text{sat}} \equiv W(L, t \to \infty) \sim L^{\alpha}$, is always less than unity ($\alpha < 1$), implying that the kinetically rough surface is a self-affine fractal with the local fractal dimensionality $d - \alpha$. Note that $W_{\text{sat}}/L \sim L^{\alpha - 1}$, and therefore in the thermodynamic limit W_{sat}/L vanishes provided $\alpha < 1$. The kinetically rough self-affine interface is therefore smooth on macroscopic length scales and the large-scale long-time orientation of the surface is the same as the underlying substrate, again provided $\alpha < 1$. All of this, however, becomes invalid if $\alpha > 1$, when W_{sat}/L diverges in the thermodynamic limit. Recently, one continuum growth equation and several discrete atomistic growth models [2–7], where surface diffusion drives the incident atoms to the nearest maximally coordinated sites, have been found to have $\alpha > 1$ in $d = 1+1$ dimensions. This pathological feature of super-roughness associated with $\alpha > 1$ (we will refer to models with $\alpha > 1$ as "super-rough" to emphasize the fact that W_{sat}/L diverges in the thermodynamic limit and therefore the large-scale, long-time orientation of these kinetically super-rough surfaces may differ from the substrate orientation) may make the basic hypothesis of a growing self-affine interface obeying dynamic scaling inapplicable to super-rough surfaces. In fact, it has been argued [4] that the solid-on-solid approximation is invalid for super-rough surfaces. An important additional conceptual problem with these super-rough models is that the value of α obtained by measuring $W_{\text{sat}}(L)$ disagrees with that obtained from the asymptotic short-distance behavior of the height-height correlation function [8,9]. In this article, we propose a theoretical model to understand kinetic super-roughness by explicitly showing that, while the basic scaling laws defined by Eqs. (4) and (5) sill apply to kinetically super-rough surfaces, the substrate size L plays a subtle and fundamentally important role of a cutoff length scale in the

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$\alpha > 1$ situation, which gives rise to the unusual asymptotic behavior of the height-height correlation function. The novel scaling behavior that emerges from our analysis of the $\alpha > 1$ situation shows that dynamic scaling for super-rough surfaces [2–9] is "anomalous" in the sense that the scaling function for the height-height correlation function behaves quite differently from the well-studied $\alpha < 1$ situation. We emphasize that Eqs. (4) and (5) are sill obeyed by these super-rough models—the subtle scaling behavior manifests itself only in the asymptotic properties of the height-height correlation function which is described by an effective or apparent roughness exponent $\alpha' \leq 1$ resolving the pathology associated with the super-roughness of $\alpha > 1$.

We start by considering three atomistic solid-on-solid growth models in $d = 1+1$ dimensions. Two of these models are the $1+$ and $d2+$ models of Ref. [5], studied earlier by Das Sarma and Tamborenea [2] and by Wolf and Villain [3], respectively. The third discrete model we consider is the restricted-curvature (RC) model [6] of Kim and Das Sarma. The numerically estimated (in both $d = 2$ and 3) dynamical exponents based on measurements of $W(L,t)$ for all of these models agree with the theoretically calculated exponents $\alpha = (5-d)/2$ and $z = 4$ [i.e., $\beta = \alpha/z = (5-d)/8$] of the fourth-order Herring-Mullins linear diffusion equation [2–7] describing conservative growth and nonconserved noise:

$$\frac{\partial h}{\partial t} = -\nu_4 \nabla^4 h + \eta \ , \tag{6}$$

where η is the random noise (assumed to be Gaussian white noise) associated with the stochastic deposition process which is responsible for producing the kinetic roughness during growth [the term corresponding to the average deposition flux is not shown in Eq. (6), where $h(x,t)$ corresponds to the deviation from the average height]. Finite-size simulations of $W(L,t)$ have established that in $d = 1+1$ these three discrete atomistic growth models obey the dynamic scaling hypothesis as defined by Eqs. (4) and (5) with β and α given by those corresponding to the continuum linear growth equation given by Eq. (6). While this by itself came as a surprise [the fourth-order continuum growth equation, given by Eq. (6) and its nonlinear generalizations [7], have not earlier been discussed in the literature [1] because the theoretical expectation was that all physically reasonable atomistic growth models would correspond to second-order continuum-growth equations], the implication of having $\alpha > 1$ is troublesome from a conceptual viewpoint. In addition, the effective roughness exponent for these models [and for Eq. (6)] as deduced from the asymptotic behavior of the height correlation function differs from that obtained from the direct measurement [2,3] of $W(L,t)$.

To proceed further we consider the height-height correlation function defined by (we explicitly consider only the $d = 1+1$ dimensions in this paper)

$$G(r,t) \equiv \langle [h(x,t) - h(x+r,t)]^2 \rangle \ , \tag{7}$$

where $\langle\ \rangle$ denotes an averaging over lateral sites and L is the system size. (x and r are one-dimensional vectors along the substrate "plane.") By a direct integration of the fourth-order differential equation (6) it is easy to show that the corresponding correlation function [10] behaves in the following way

$$G(r,t) \sim r^{2\alpha} f(r/\xi(t)) \ , \tag{8}$$

with $\alpha = (5-d)/2$ and $f(x)$ is a scaling function with the following asymptotic properties [10]

$$f(x) \sim \begin{cases} x^{-1} & \text{for } d = 2,\ x \ll 1 \\ \ln x & \text{for } d = 3,\ x \ll 1 \\ \text{const} & \text{for } d > 3,\ x \ll 1 \\ x^{-2\alpha} & \text{for all } d,\ x \geq 1 \ . \end{cases} \tag{9}$$

This d-dependent scaling function [whose asymptotic behavior for $x \ll 1$ is different in $d = 2,3$ from the usual scaling function of Eq. (2)] arises because the integral for $G(r,t)$ in $d = 2$ and 3 becomes divergent [10] in the thermodynamic limit for the super-rough growth models and therefore the system size L enters as an important cut off in the problem. The correlation length $\xi(t)$ of Eq. (8) follows the asymptotic behavior

$$\xi(t) \sim L g(t^{1/z}/L) \ , \tag{10}$$

where the scaling function $g(x)$ obeys

$$g(x) \sim \begin{cases} x & \text{for } x \ll 1 \\ \text{const} & \text{for } x \gg 1 \ , \end{cases} \tag{11}$$

so that

$$\xi(t) \sim \begin{cases} t^{1/z} & \text{for } t^{1/z} \ll L \\ L & \text{for } t^{1/z} \gg L \ . \end{cases} \tag{12}$$

Combining Eqs. (8)–(12) we conclude that the asymptotic behavior of $G(r,t)$ for super-rough growth models can be written as

$$G(r,t) \sim \begin{cases} r^{2\alpha-\kappa} t^{\kappa/z} & \text{for } r \ll t^{1/z} \ll L \\ r^{2\alpha-\kappa} L^{\kappa} & \text{for } r \ll L \ll t^{1/z} \\ t^{2\beta} & \text{for } r \gg t^{1/z} \ , \end{cases} \tag{13}$$

where $\kappa = 1$ for the specific analytical continuum equation (for $d = 2$) defined by Eq. (6). Note that the usual behavior of $G(r,t)$ for models with $\alpha < 1$ corresponds to $\kappa = 0$, and is consistent with Eq. (4) [which is very different from Eq. (13) in the short-distance regime $r \ll L, t^{1/z}$]:

$$G(r,t) \sim \begin{cases} r^{2\alpha} & \text{for } r \ll t^{1/z} \\ t^{2\beta} & \text{for } r \gg t^{1/z} \ . \end{cases} \tag{14}$$

We emphasize that the substrate size L plays no role in Eq. (14) (the "usual" behavior with $\kappa = 0$ and $\alpha < 1$) and a fundamental role [10] in Eq. (13) (the super-rough behavior with $\kappa \neq 0$ and $\alpha > 1$).

We hypothesize that the asymptotic scaling behavior defined by Eq. (13) with $\kappa \neq 0$ is the generic scaling behavior [11] of *all* super-rough kinetic growth models which have $\alpha > 1$. [For models with $\alpha = 1$ as measured

from $W_{sat} \sim L^{\alpha}$, one finds the exponent κ to be infinitesimal, implying a logarithmic $\ln L$ dependence of $G(1,t)$ on L.] Note that we are generalizing the $\kappa=1$ result obtained analytically for Eq. (6) to an arbitrary super-rough situation where κ ($\neq 0$) is not necessarily unity. We emphasize that the scaling exponents α and β (and, consequently, z) as extracted from the dependences of $W(t)$ on t, and, of $W_{sat}(L)$ on L are unaffected by this "new" scaling behavior of the correlation function. In particular, setting $r=L$ in Eq. (13), we conclude that $W(t) \sim t^{\beta}$ for small t, and $W_{sat}(t \to \infty) \sim (L^{2\alpha-1}L)^{1/2} \sim L^{\alpha}$, totally consistent with the "usual" asymptotic scaling behavior of $W(L,t)$ discussed in our introduction. The scaling function $f(x)$, however, behaves very differently in the small-x ($=r/t^{1/z}$) limit, where instead of going to a constant [Eq. (2)] as it should for the usual dynamic scaling behavior, the super-rough scaling function falls off as $x^{-\kappa}$ [with $\kappa=1$ for $d=2$ in Eq. (6), representing the analytic-surface-diffusion-equation case]:

$$f(x) \sim \begin{cases} x^{-\kappa} & \text{for } x \ll 1 \\ x^{-2\alpha} & \text{for } x \gg 1 \, . \end{cases} \tag{15}$$

From the viewpoint of dynamic scaling theory, Eq. (15) [as opposed to Eq. (2)] is the interesting anomalous behavior of super-rough models. Thus in super-rough models $G(1,t)$ increases as $t^{1/z'}$ in the intermediate-time range where $z'=z/\kappa$, in contrast to the "usual" ($\alpha<1$) case where $G(1,t)$ becomes a constant.

In the rest of this article we justify this generalized scaling ansatz (i.e., the existence of a finite κ) numerically for the three different kinetically-super-rough atomistic growth models which have recently been studied in the literature. The two discrete 1+1-dimensional models for which we present our numerical results in this paper are the Das Sarma–Tamborenea (DT) model [2,7] (referred to as the 1+ model in Das Sarma and Ghaisas [5]) and the RC model of Kim and Das Sarma [6]. Our numerical results for the $d2+$ model [5] (not shown here) are essentially the same as those for the DT model as presented in Figs. 1–3. Simulation studies of these models produce

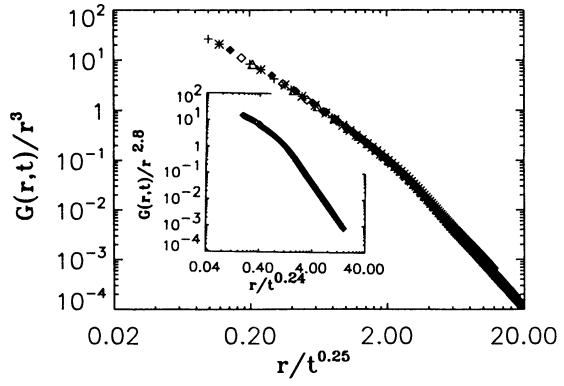

FIG. 1. Shows the scaling collapse of $f(x) \equiv G(r,t)/r^3$ against $x \equiv r/t^{0.25}$ in the DT model for five different values of t (=524, 1060, 2100, 4900, and 10 000). Inset shows the collapse with $\alpha \approx 1.4$, $z^{-1} \approx 0.24$ for the RC model for 20 different values of $t=50,100,\ldots,1000$. The power-law behavior of $f(x)$ for small values of x gives $\kappa \approx 1.6$ (inset: $\kappa \approx 1.0$). $L=10\,000$.

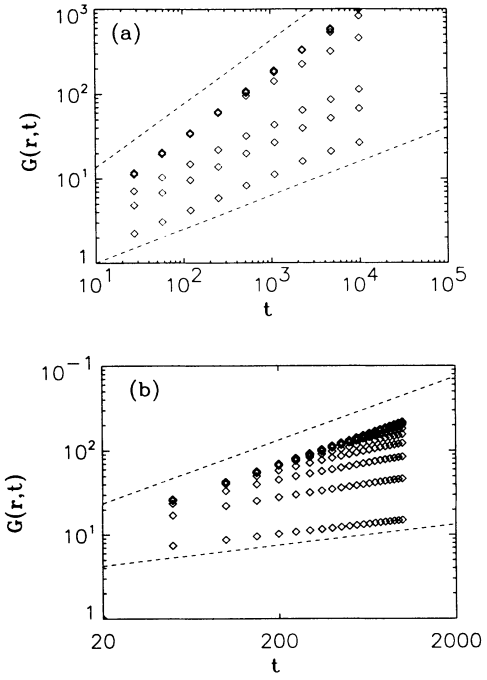

FIG. 2. Simulated $G(r,t)$ as a function of t for different fixed values of r ($=1,2,\ldots,40$ from the bottom to the top) for the DT model (a) and the RC model (b). Simulation results are consistent with Eq. (13). The upper dashed lines correspond to $2\beta=\frac{3}{4}$ and the lower dashed lines correspond to $\kappa/z=0.4$ (a) and 0.25 (b). $L=10\,000$.

$\beta \approx \frac{3}{8}$, $\alpha \approx \frac{3}{2}$, and $z \approx 4$ as obtained numerically by studying $W(L,t)$, and therefore both models belong to the super-rough category, having also the same critical exponents as those of Eq. (6). In Figs. 1–3 we show results for our numerical simulation of the height-height correlation in these two atomistic growth models. From Fig. 1 we conclude that both models show excellent dynamical scaling with $2\alpha \approx 3$ and $z \approx 4$ as $G(r,t)/r^3$ plotted against $r/t^{0.25}$ show perfect data collapse. Thus Eq. (8) is obeyed in both models—the scaling function $f(x)$, as can clearly be seen from Fig. 1, does not approach a constant for small values of x and, in fact, obeys the power-law scaling

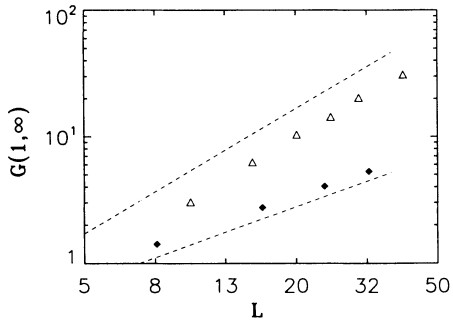

FIG. 3. Calculated $G(1,\infty)$ as a function of $\ln L$ showing that $\kappa \approx 1.6$ for the DT model (triangles) and $\kappa \approx 1.0$ for the RC model (diamonds). Dotted lines with slopes 1.0 and 1.65 are indicated.

$x^{-\kappa}$ with the numerical value of κ found to be $\kappa \approx 1$ for the RC model and $\kappa \approx 1.6$ for the DT model. In Figs. 2 and 3 we show the calculated $G(r,t)$ respectively as a function of t for various values of r (Fig. 2), and, for fixed $r=1$ and $t=\infty$ as a function of the system, size L (Fig. 3). From these results, it is clear that the discrete DT and RC Models obey the scaling ansatz defined by Eqs. (8)–(15) with $\kappa \approx 1.0$ (RC) and $\kappa \approx 1.6$ (DT).

Before concluding we emphasize that *all* the solid-on-solid stochastic growth models which have $\alpha \geq 1$ in $d=2,3$ dimensions, with the roughness exponent α measured from the dependence of the saturation interface width on the lateral substrate size, i.e., from the relationship $W(L,t\rightarrow\infty)\sim L^{\alpha}$, exhibit anomalous dynamical scaling behavior. The exponent κ is finite when $\alpha>1$ whereas κ is infinitesimal [indicating $G(1,t)\sim \ln t$ or $\ln L$ depending on whether $t^{1/z} \ll L$ or $t^{1/z} \gg L$, respectively] when $\alpha=1$. We have explicitly verified this for the discrete 1+ model ($d=2,3$), $d2+$ model ($d=2$), and $d4+$ model ($d=3$) of Das Sarma and Ghaisas [5], finding $\kappa=1.6$ ($d=2$), $0+$ ($d=3$) in the 1+ model, $\kappa=1.6$ ($d=2$) in the $d2+$ model, and $\kappa=0+$ ($d=3$) in the $d4+$ model, where $0+$ is an infinitesimal indicating logarithmic asymptotic behavior of the correlation function (instead of the usual saturation). We have also verified the anomalous dynamic scaling behavior in the $\nabla^4 h$ ($d=2,3$) and $\nabla^2(\nabla h)^2$ ($d=2$) continuum equations, finding $\kappa=1$ ($d=2$), $0+$ ($d=3$) in the $\nabla^4 h$ equation and $\kappa=0+$ ($d=2$) in the $\nabla^2(\nabla h)^2$ nonlinear Lai–Das Sarma equation [7]. All the other discrete and continuum growth models introduced in Ref. [5] have $\alpha<1$, and therefore they exhibit the usual dynamic scaling behavior.

In conclusion [12], we have introduced a generalized dynamic scaling ansatz for super-rough ($\alpha>1$) kinetic growth models which resolves the puzzle of how a physically meaningful growth model may have $\alpha>1$, by showing that the effective or the apparent roughness exponent [13] α' for the height-height correlation function [cf. Eq. (13)] becomes [13] $\alpha' \equiv \alpha - \kappa/2$, which for all the known super-rough models satisfies $\alpha' \leq 1$ (namely, $\alpha' \approx 1$ for the surface diffusion equation and the RC model, and $\alpha' \approx 0.7$ for the DT and $d2+$ models). Thus, while the real α measured from the dependence of $W_{\rm sat}(L)$ on L may be larger than unity, the system size acts as a cutoff to ensure that the apparent α' [measured by studying the behavior of $G(r,t)$ as a function of r for $r \ll L$] in the correlation function never exceeds unity, resolving the conceptual crisis of super-roughening. The issue of whether κ is a true dynamical exponent or just a very slow crossover effect remains unresolved at the present time.

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[10] It is easy to show that $G(r,t)$ corresponding to Eq. (6) is given by

$$G(r,t)\sim \int_{\pi/L}^{\pi} dk\{1-\cos(kr)\}\left[\frac{1-e^{-2\nu_4 k^4 t}}{k^{6-d}}\right],$$

which diverges in $d=2,3$ in the $L\rightarrow\infty$ limit (the divergence in $d=3$ being only logarithmic)—an asymptotic analysis produces Eq. (9).

[11] Note that Eq. (13) is an ansatz which cannot be theoretically proven for the discrete atomistic-growth models, but is consistent with our numerical simulation as presented in Figs. 1–3. For the continuum model defined by Eq. (6), $\kappa=1$ in $d=2$.

[12] Our conclusion differs from that in Ref. [8], which concludes that the surface-diffusion equation [our Eq. (6)] and the $d2+$ model leads to a groove instability. We also disagree with Ref. [9], which concludes that the discrete DT and $d2+$ models do not exhibit conventional scaling behavior (or, if they do, have the Edwards-Wilkinson scaling behavior with $\alpha \approx 0.5$, $\beta \approx 0.25$, and $z=2$ in $d=2$).

[13] Note that we can define apparent or effective exponents α' and z' based on Eq. (13) given by $\alpha'=\alpha-\kappa/2$ and $z'=z/\kappa$, with $G(r \ll t^{1/z} \ll L)\sim r^{2\alpha'} t^{1/z'}$ and $G(r \ll L \ll t^{1/z}) \sim r^{2\alpha'} L^{\kappa}$. For our discrete DT (RC) models $\alpha' \approx 0.7$ (1.0) and $z' \approx -2.5$ (4.0)—a direct simulation of the structure factor, related to the Fourier transform of G, gives us $S(k)\sim k^{-2.5}$ (k^{-4}) for small k in the DT (RC) model, consistent with the theoretical expectation that $S(k)\sim k^{-z'}$, again showing that $\kappa \approx 1.6$ (1) in the DT (RC) model. Note that the real exponents satisfy the known scaling relation $2\alpha+d-1=z$ whereas the effective exponents satisfy a different relation $2\alpha'+d-1=\kappa(z'-1)$. The effective exponents α' and z' for the various super-rough models studied by us are $\alpha' \approx 0.7$, $z' \approx 2.5$ for 1+ and $d2+$ models in $d=1+1$ dimensions.