

Kinetic Growth with Surface Relaxation: Continuum versus Atomistic Models

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We introduce a geometrical interpretation to classify possible linear and nonlinear terms in the models for driven growth with surface relaxation. A nonlinear differential equation, distinct from the Kardar-Parisi-Zhang equation, is proposed as a relevant continuum model describing atomistic kinetic growth under conditions of chemical bonding. The scaling relations among growth exponents that we derive from a dynamic renormalization-group analysis are exact for a class of growth models with surface relaxation. The calculated exponents are in excellent agreement with our discrete atomistic growth simulation.

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There has been much recent interest in kinetic growth processes of surfaces far from equilibrium.¹ The growing surfaces naturally evolve into self-similar structures and the surface roughness or the interface width can be described by the dynamical scaling form, for a system of size L and at time t , $W(t, L) = L^\alpha F(t/L^{\alpha/\beta})$, with $F(\infty) = \text{const}$ [i.e., $W(L) \sim L^\alpha$ as $t \rightarrow \infty$] and $F(x) \sim x^\beta$ for small x [i.e., $W(t) \sim t^\beta$ before saturation]. Most of the growth models studied so far fall in the universality class of the Kardar-Parisi-Zhang (KPZ) equation,^{2,3} which is given by

$$\frac{\partial h}{\partial t} = v\nabla^2 h + \lambda(\nabla h)^2 + \eta, \quad (1)$$

where $h(\mathbf{x}, t)$ is the height of a growing interface in $d = d' + 1$ dimensions (d' is the substrate dimension) and η is the stochastic noise driving the growth.

The KPZ equation successfully describes many different growth processes such as the Eden model and ballistic deposition. Our motivation in this work is to develop a continuum growth model relevant for the technologically important molecular-beam-epitaxy (MBE) process,^{4,5} particularly at higher growth temperatures where relaxation of the incident atoms (via rapid surface diffusion) plays a dominant role in producing smooth growth. It is well known that in the temperature range of MBE growth desorption of atoms and formation of overhangs and bulk defects is negligibly small. Under *ideal* MBE growth conditions⁶ the growth equation must obey a mass conservation law, leading to the continuity equation $\partial h / \partial t = -\mathbf{V} \cdot \mathbf{j} + \eta$. Thus, the nonlinear term $\lambda(\nabla h)^2$ in the KPZ equation is inoperative in MBE growth.

In an effort to better understand growth under a chemical-bonding environment, Das Sarma and Tamborenea⁷ and Wolf and Villain⁸ have recently investigated a simple growth model in which the particles are randomly deposited onto a substrate and subsequently relax to nearby kink sites maximizing the number of saturated bonds. They found that the growth exponents are given by, in $d = 1 + 1$ dimensions, $\alpha \approx 1.5$, $\beta \approx 0.375$, and

$z = \alpha/\beta \approx 4$. These exponents are surprisingly consistent with the linear surface diffusion⁹ equation under a chemical-potential gradient,

$$\frac{\partial h}{\partial t} = -v_1 \nabla^4 h + \eta, \quad (2)$$

where the noise η has a Gaussian distribution and satisfies

$$\langle \eta(1)\eta(2) \rangle = 2D\delta^d(\mathbf{x}_1 - \mathbf{x}_2)\delta(t_1 - t_2). \quad (3)$$

The dynamical exponents associated with Eqs. (2) and (3) are $\alpha = (5 - d)/2$, $\beta = (5 - d)/8$, and $z = 4$.

However, as pointed out in Refs. 7 and 8, there is no obvious chemical-potential gradient in this manifestly nonthermodynamical problem. It is not clear why the growth process of the model is so close to this linear diffusion equation driven by the chemical-potential difference. Is there a possible nonlinear equation describing the MBE process? We also note that, in the atomistic model introduced by Das Sarma and Tamborenea,⁷ the atomistic diffusion follows an Arrhenius activation behavior. At high temperatures (but below the roughening temperature) the atoms at the kinks may break bonds and hop around (both "up" and "down" because gravity is negligible). What equation may describe this process?

In this Letter, we elucidate the above issues by introducing a novel geometrical interpretation to study possible linear and nonlinear terms in the various models of stochastically driven growing surfaces. In particular, we address the issues of what physical process could conceivably produce a $\nabla^4 h$ term in the strongly nonequilibrium situation where the chemical-potential gradient is not a useful concept and what the possible nonlinear terms are to the lowest order. We identify a nonlinear differential equation, distinct from the KPZ equation, as a possible continuum model describing kinetic growth under *ideal* conditions of MBE. We establish the connection between the continuum model and the usual discrete atomistic growth model.

We assume that the surface height $h(\mathbf{x}, t)$ is *single*

valued in \mathbf{x} and its time dependence can be described by its local derivatives,

$$\frac{\partial h}{\partial t} = f(\nabla h, \nabla^2 h, \dots) + \eta. \quad (4)$$

The functional $f = -\delta H(h)/\delta h + f_1$, where the first part includes terms which can be derived from a ‘‘Hamiltonian’’ while f_1 is the rest. The noise η satisfies Eq. (3). Remembering that f is a scalar and considering only isotropic growth, we can write down the following general form for f :

$$f = \nu \nabla^2 h - \nu_1 \nabla^4 h + \lambda (\nabla h)^2 + \lambda_1 \nabla^2 (\nabla h)^2 + \lambda_2 \nabla \cdot (\nabla h)^3 + \dots \quad (5)$$

Note that the invariance properties of the growing interface ($h \rightarrow h + \text{const}$) disallow certain terms such as $\nabla^2 h^2$, etc.

To better examine the physical origin of each term in our growth equation [and, to establish a connection between the continuum growth equation as defined by Eq. (4) with the actual discrete atomistic growth] we have developed a geometric and graphical method as depicted in Figs. 1 and 2. We note that any coarse-grained surface morphology [with $h(\mathbf{x}, t)$ being single valued in \mathbf{x}] controlled by a general continuum equation of the type of Eq. (4) can be decomposed into the four topologically distinct basic building blocks shown as (a), (b), (c), and (d) in Fig. 1. These building blocks can be combined to form composite structures [e.g., Fig. 2(a)] such as peaks, valleys, and other structures. In Fig. 2 we show a typical surface morphology and various corresponding local derivatives, h , $(\nabla h)^2$, $\nabla^2 h$, $\nabla^4 h$, $\nabla^2 (\nabla h)^2$, and $\nabla \cdot (\nabla h)^3$ in $d = 1 + 1$ dimensions, which are the lower-order terms in Eq. (5). The qualitative features of higher-order derivatives in Fig. 2 are easily obtained from Fig. 2(a) if we write as a model $h = h_0 + h_1 \tanh(bx)$.

Based on the geometric forms of Figs. 1 and 2, we now discuss the physical interpretation of each term in Eq. (5). The term $\nu \nabla^2 h$ with $\nu > 0$ is positive near local minima and negative near local maxima as can be seen from Fig. 2(c). This implies that the valleys (peaks) will receive more (less) particles than the average flux. An equivalent description would be to say that the particles

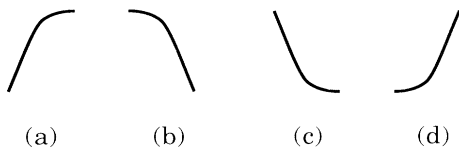


FIG. 1. Any small segment of the coarse-grained surface must have increasing, decreasing, or constant slopes and thus falls into one of the four topologically different basic building blocks as shown. The straight-line segments are just the topological extension of the two ends of the building blocks.

landing on the peaks would tend to go down to the valleys. This is precisely the smoothening effect of gravity as in the Edwards-Wilkinson¹⁰ model which is Eq. (1) with $\lambda = 0$. (Desorption has a similar smoothening effect to the lowest order as well.) In fact, this term can be derived from the Hamiltonian $H_{gr} = \int d^d x (\nu/2) (\nabla h)^2$ by noting that $-\delta H/\delta h = \nu \nabla^2 h$. The physics of the gravitational term is that any height difference costs ‘‘energy,’’ acting, therefore, as a smoothening effect on surface roughness. From Fig. 2(b), we see that the $\lambda (\nabla h)^2$ term (with $\lambda > 0$) of KPZ allows the steepest points in the growth front to grow at faster rates by allowing bulk vacancies¹¹ since $\lambda \int d^d x (\nabla h)^2 > 0$. We emphasize that neither gravity nor desorption plays any role in MBE growth, where the energy scale is set by atomistic chemical bonding so as to make gravity completely negligible, and the growth temperature is usually low enough to make desorption an exponentially weak process. Thus we conclude that $\nu = 0$ for MBE growth, which is also consistent with the existing numerical simulations^{7,8} in $d' = 1$ dimension. We have already argued that $\lambda = 0$ for MBE growth because bulk vacancies and overhangs are not allowed. We, therefore, contend that the ν and the λ terms in f are absent for MBE growth.

The physical interpretation for the $\nu_1 \nabla^4 h$ term [Fig. 2(d)] is more complicated because it has additional structures with small positive (negative) regions near the valley (peak). This is precisely the feature mimicking the strong chemical-bonding situation of MBE where the atoms can stick to kink sites rather than simply rolling down to the local height minima (as in the $\nabla^2 h$ term). The corresponding Hamiltonian is $H_{ch} = \int d^d x (\nu_1/2) (\nabla^2 h)^2$. The $\lambda_1 \nabla^2 (\nabla h)^2$ term [Fig. 2(e)] can similarly be seen (for $\lambda_1 > 0$) to correspond to the situation where

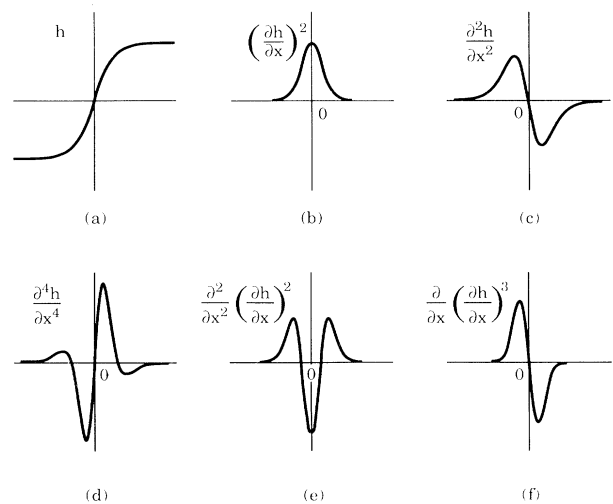


FIG. 2. (a) A typical segment of surface by combining building blocks (a) and (d) of Fig. 1. (b)–(f) Various derivatives of (a) as labeled.

the particles landing at high steps (large derivatives) relax to lower steps (smaller derivatives). We believe this term may correspond to the “high-temperature” regime of the atomistic model where the surface atoms at the kink sites can break bonds and hop with larger probability to steps with smaller height. This term does not appear in Refs. 7 and 8, where particles at kink sites are not allowed to move. Finally, from Fig. 2(f) we note that the $\nabla \cdot (\nabla h)^3$ term is very similar to the $\nabla^2 h$ term. In fact, the corresponding Hamiltonian is $H = \int d^{d'}x (\lambda_2/4)(\nabla h)^4$. We believe this term to be a higher-order correction to the $\nabla^2 h$ process. Thus, although this term is a mathematically relevant nonlinear term to the $\nabla^4 h$ term in our growth equation, we neglect it in the rest of our consideration because we have been unable to identify any physical process given by the growth equation

$$\frac{\partial h}{\partial t} = -v_1 \nabla^4 h + \lambda_2 \nabla \cdot (\nabla h)^3 + \eta \quad (6)$$

without generating the $\nabla^2 h$ term.

Based on the above discussion for the physical interpretation of various growth terms, we now propose the following as the relevant ideal MBE growth equation at intermediate to high temperatures:

$$\frac{\partial h}{\partial t} = -v_1 \nabla^4 h + \lambda_1 \nabla^2 (\nabla h)^2 + \eta. \quad (7)$$

Equation (7) represents a meaningful (if highly simplified) physical model for *ideal* MBE growth which catches some essential aspects of real MBE growth. Since the noise η is given by Eq. (3), Eq. (7) represents a situation of *conserved current* and *nonconserved noise*. Thus, our growth model is fundamentally different from the model studied by Sun, Guo, and Grant¹² with *both* the current and the noise conserved. In their model the total volume under the interface is conserved whereas in our model atoms are continually coming onto the surface from the incident flux as in the actual MBE growth.

The scaling behavior of the growth equation (7) can be analytically treated via the dynamical renormalization-group technique.^{2,13} Under a change of scale, $x \rightarrow e^l x$, $t \rightarrow e^{z l} t$, and $h \rightarrow e^{a l} h$, the coefficients in Eq. (7) renormalize as, to the lowest order,

$$\frac{dv_1}{dl} = v_1 \left[z - 4 + K_{d'} \frac{\lambda_1^2 D}{v_1^3} \frac{6-d'}{4d'} \right], \quad (8a)$$

$$\frac{dD}{dl} = D[z - 2\alpha - d'], \quad (8b)$$

$$\frac{d\lambda_1}{dl} = \lambda_1[z + \alpha - 4], \quad (8c)$$

with $K_{d'} = S_{d'}/(2\pi)^{d'}$ and $S_{d'}$ being the surface area of a d' -dimensional unit sphere. The effective coupling constant $\bar{\lambda}_1 = (\lambda_1^2 D/v_1^3)^{1/2}$ then flows under rescaling as

$$\frac{d\bar{\lambda}_1}{dl} = \frac{4-d'}{2} \bar{\lambda}_1 + K_{d'} \frac{3(d'-6)}{8d'} \bar{\lambda}_1^3. \quad (9)$$

The critical dimension is $d'_c = 4$ substrate dimensions ($d_c = d'_c + 1 = 5$ spatial dimensions). The exponents α and z for $d \leq d_c$ are then given by

$$z - 2\alpha - d' = 0, \quad (10a)$$

$$z + \alpha - 4 = 0, \quad (10b)$$

i.e.,

$$\alpha = (4 - d')/3 = (5 - d)/3, \quad (11a)$$

$$z = (8 + d')/3 = (7 + d)/3, \quad (11b)$$

which gives the exponent $\beta = \alpha/z = (5-d)/(7+d)$. It can be shown¹⁴ that the hyperscaling relation (10a) is exact (for $d \leq d_c$) for any growth equations with conserved current and nonconserved noise (totally independent of the detailed form of the current), whereas (10b) is exact only for the specific form of Eq. (7) as a consequence of the invariance of the equation under a transformation.¹² We mention that (10a) was earlier obtained from a physical argument by Wolf and Villain⁸ and (10b) was derived by Sun, Guo, and Grant.¹²

The physically interesting dimension for MBE growth is $d = 2 + 1$ where we obtain $\alpha = \frac{2}{3}$, $\beta = \frac{1}{5}$, and $z = \frac{10}{3}$. The situation with $d = 1 + 1$ is also physically realizable in the context of adatom motion on vicinal surfaces with steps and the corresponding exponents are $\alpha = 1$, $\beta = \frac{1}{3}$, and $z = 3$. (Note that $\beta = \frac{1}{3}$ for $d = 1 + 1$ in our growth model is fortuitously the same as the corresponding KPZ case.) We have verified (Fig. 3) our analytic results for the exponents in $d = 1 + 1$ dimensions by carrying out an atomistic one-dimensional growth simulation within a solid-on-solid model. The growth model involves random deposition of particles at lattice sites. If the deposited particle has two nearest neighbors it does not move any

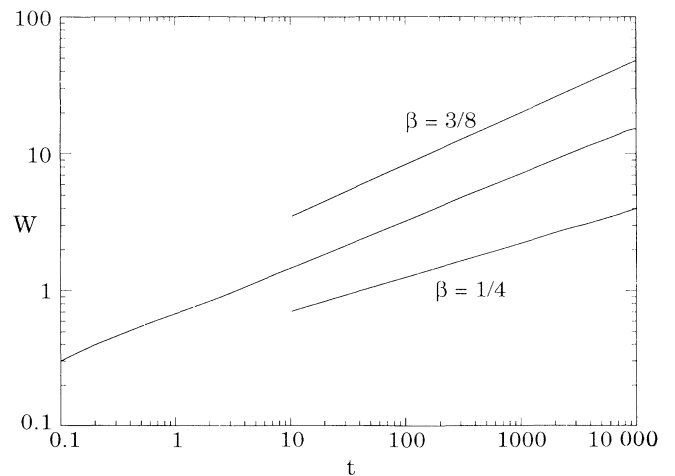


FIG. 3. Simulation results (middle curve) of the surface width W vs time t in logarithmic scale. System size $L = 10000$. Time t is equal to number of layers of growth. Top ($\beta = \frac{3}{8}$) and bottom ($\beta = \frac{1}{4}$) curves are drawn for reference.

more (and stays at that site forever). If the particle is at a kink site (i.e., with only one neighbor either to the left or to the right), it moves to the nearest kink site with a smaller step height (with the particle being allowed to move either up or down as needed). This is in contrast to Refs. 7 and 8, where particles at kink sites are not allowed to move. If the particle is at a site with no nearest neighbors, it diffuses to the nearest kink site within a finite distance l ($\ll L$). In Fig. 3 we show our simulated surface width W against time on a logarithmic plot. Our best estimates of the exponents from the simulation results are $\beta = 0.340 \pm 0.015$, $\alpha = 1.05 \pm 0.10$. Simulations for larger system sizes and for more complicated growth rules are currently under investigation.

Usually the situation with the roughness exponent $\alpha > 1$ is considered unstable because, for large enough system sizes, this implies that the surface will eventually develop arbitrarily large vertical fluctuations with $W \gg L$. We believe this may be an unavoidable feature of atomistic growth within the solid-on-solid model under the chemical-bonding environment at low temperatures (which severely restricts the ability of atoms to hop around). Thus one may have to allow vacancies and overhangs at low temperatures. We do point out, however, that at low temperatures where bulk vacancies and overhangs become important (and the solid-on-solid model may become invalid), the whole continuum approach based on a single-valued height function $h(\mathbf{x}, t)$ becomes inapplicable and processes such as bulk diffusion (which cannot be incorporated into the current class of continuum models) have to be taken into account.

For the nonlinear equation (7), $\alpha = 1$ in $d = 1 + 1$. Dimensional analysis shows that terms such as $\nabla^2(\nabla h)^4$ cannot change the exponent and hence are irrelevant. For the linear equation (2), $\alpha = \frac{3}{2}$ in $d = 2$. A number of nonlinear terms could bring the exponent α very close to 1 in $d = 2$. However, these terms may not be consistent with the chemical-bonding model within the solid-on-solid restrictions or their coefficients are perhaps so small that one may need very large system sizes and very long simulation times to see the crossover. Finally, if there were some processes corresponding to the nonlinear equation (6), dimensional analysis¹⁵ gives the exponent $\alpha < 1$ for any physical dimension $d \geq 2$. This is the *only* nonlinear term with fourth-order derivatives which can give $\alpha < 1$ for $d = 2$. These issues are currently under further investigation.

In conclusion, we have introduced a geometrical construct to physically understand the features of various growth terms in the models for driven surface growth. This enables us to identify a growth equation which may be relevant to the atomistic MBE process under the chemical-bonding environment at intermediate to high

temperatures. We have obtained the exact growth exponents $\alpha = (5 - d)/3$, $\beta = (5 - d)/(7 + d)$ in d dimensions. Our preliminary numerical simulation of an atomistic growth model is in agreement with these predictions in $d = 1 + 1$ dimensions.

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¹⁴For any equations of conserved current and nonconserved noise, the corrections to Eq. (8b) are of higher order and irrelevant. Thus the noise strength D is not renormalized.

¹⁵In this case, Eq. (10a) still holds, while dimensional analysis changes Eq. (10b) to $z + 2\alpha - 4 = 0$. Thus we obtain $\alpha = (5 - d)/4$, $z = (3 + d)/2$, and $\beta = (5 - d)/2(3 + d)$. For $d = 2$, $\alpha = \frac{3}{4}$ and $\beta = \frac{3}{10}$.