Solid-on-Solid Rules and Models for Nonequilibrium Growth in 2+1 Dimensions

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On the basis of a detailed stochastic simulation we find that nonequilibrium growth in 2+1 dimensions, within the simple solid-on-solid approximation, is surprisingly rich with its dynamical universality depending sensitively on the local atomistic relaxation rules of the growth model. We establish connections between our computed dynamical growth exponents for various physically plausible local growth models with those given by a recently proposed fourth-order nonlinear continuum differential equation.

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In a solid-on-solid (SOS) growth model, incident atoms stick only to the tops of the already existing substrate atoms which are arranged in a lattice [1]. The resultant growing structure is, therefore, a lattice of columns whose heights increase as matter is added from outside. The SOS model has been extensively used in both equilibrium and nonequilibrium crystal growth studies. While being conceptually simple, it also describes well many real situations where vacancies and overhangs are relatively rare. In this Letter, we present detailed simulation results which demonstrate that nonequilibrium growth, even within the simple SOS approximation, is extremely rich in 2+1 dimensions (i.e., two-dimensional substrate, $d'=2$, with growth in the other direction), leading to many different growth universality classes depending upon the precise nature of the local growth rules. We find that the local atomistic relaxation rule and, in particular, its dependence on the atomic coordination number which defines the model determine the universality class of the growth model. This surprising richness in the SOS nonequilibrium growth is totally unanticipated, because the simplicity of the model makes one expect some simple universality independent of the details of the local growth rules. In fact, it is interesting to note that more complicated nonequilibrium growth models involving ballistic deposition (where impinging atoms may stick to the sides of the existing columns, thus allowing overhangs and vacancies) are all believed to belong to a single universality class, namely, the KPZ universality [2], whereas the simpler SOS growth models cannot be characterized by a single universality class.

We impose two additional restrictions on the SOS model, motivated both by practical relevancy and computational tractability, namely, neglect of desorption and, in some cases [3] as discussed below, of any upward atomic relaxation [4]. The continuum equation governing the dynamics of growth under these conservative (i.e., no desorption and SOS restrictions) conditions has the general form [5]

$$\partial h/\partial t = a\nabla^2 h - b\nabla^4 h + (\text{fourth-order nonlinear terms}) + \cdots + \eta, \quad (1)$$

where $\eta(\mathbf{x},t)$ is the Gaussian white noise satisfying the correlation $\langle \eta(\mathbf{x},t)\eta(\mathbf{x}',t')\rangle = D\delta(\mathbf{x}-\mathbf{x}')\delta(t-t')$ with $\mathbf{x}$ and $\mathbf{x}'$ the lateral coordinates. Since there is no evaporation [6], vacancies [7], or a tilted substrate [8], the $(\vec{\nabla}h)^2$ term of the KPZ equation does not appear, as the growth process is manifestly current conserving. In the presence of the KPZ nonlinearity, growth in the continuum model is described by the KPZ equation [2] and has been studied [9]. In what follows, we present simulation results for SOS models which we believe obey Eq. (1) in $d'=2$. In $d'=1$, we have studied four models, while in $d'=2$, correspondingly, six. In all the models, atoms are deposited randomly on the substrate of size $L^{d'}$. Immediately after deposition, the atom can make a conditional hop to an empty nearest-neighbor (nn) site within the rules specified by the given model. These rules are depicted in Fig. 1. We designate these models with the abbreviated forms

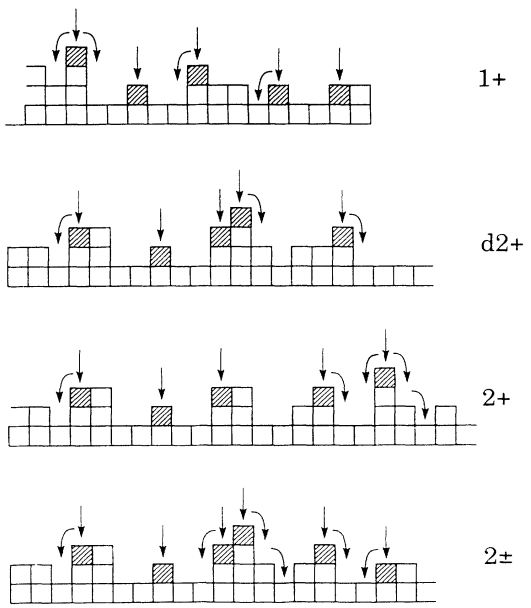

FIG. 1. Schematic various growth rules in $d'=1$. The shaded atom is the deposited one with the arrow on the top. Hops are denoted by other arrows. Note that two consecutive hops are possible for 2+ and 2±, while there is only a single allowed jump for 1+ and d2+.

of the rules used to describe the model. Our atomistic hopping, unless explicitly stated otherwise, is within the "no-upward jump" nn approximation.

Model 1+.—All deposited atoms with the coordination number one ($N_c=1$) are allowed to hop if they can increase N_c. (Deposited atoms with $N_c>1$ do not hop.) The maximum possible number of hops (N_m) for an atom in this case is one [10].

Model d2+.—When $N_c\leq 2$ *on deposition*, the atom may hop if N_c increases. $N_m=1$ [11].

Model 2+.—If $N_c\leq 2$ *on deposition or after a hop*, the atom may hop again if N_c increases. With these rules $N_m=2$.

Model 2±.—In this model, if $N_c\leq 2$ *on deposition or after a hop*, the atom may hop again provided a nn empty site is available with no restriction on N_c (it may increase, decrease, or remain the same). $N_m=2$ in this case.

In $d'=2$, there are many more possibilities for N_c and we have correspondingly six models.

Model 1+.—The same as 1+ above.

Model d4+.—This model corresponds to $d2+$ in $d'=1$. It follows the same rules except that N_c *after deposition* can be up to 4 compared to 2 in $d'=1$ for an atom to hop ($N_c=5$ and 3 denote the maximum values in $d'=2$ and 1, respectively). $N_m=1$. When upward hops are allowed, we designate the model $ud4+$.

Model 2+.—The same as 2+ above.

Model 4±.—The same as 2± above except that the maximum value of N_c can be up to 4 *after deposition or hopping* so that an atom may further increase or decrease N_c by additional hops. $N_m=4$. In the presence of restricted (unrestricted) upward hops, we designate the model $ru4\pm$ ($uu4\pm$).

Model 3+.—An atom becomes immobile for $N_c\geq 4$. For $N_c\leq 3$ *on deposition or hopping*, an atom will hop if it increases N_c. $N_m=3$.

Model 4+.—In this case, an atom becomes immobile for $N_c\geq 5$, otherwise it will hop if it can increase N_c. $N_m=4$.

The growth exponents α and β are computed using the relationships [7] $W(L,t)\sim t^{\beta}$ for $t\ll L^z$ where $z=\alpha/\beta$ and $W(L,t)\sim L^{\alpha}$ for $t\gg L^z$ from the log-log plots of the interface width W as a function of t (measured as average height) and of the system size L. W is computed from the simulation data using $W^2=\sum_{i=1}^{N}(h_i-\bar{h})^2/N$, where h_i is the height of the column at the site i and $\bar{h}$ is the average height. In $d'=1$, system sizes up to $L=100$ are used to obtain α, while for β, up to $L=10000$ are used. In $d'=2$, $L=10$ to 60 are used to find α, while L up to 1000 are used to determine β. The averaging is performed as necessary to reduce the statistical error below 0.2% in all the simulations. In order to confirm that model 1+ follows the fourth-order linear equation over several decades, massive parallel computation was performed using a CM-2 Connection Machine. In order to ascertain the dynamical universality of a model, we have carried out a direct least-squares fit of the simulated width W to the analytical finite-size solution of the linear equation

$$\partial h/\partial t=a\nabla^2 h-b\nabla^4 h+\eta(\mathbf{x},t)\,. \tag{2}$$

The details of these results will be published elsewhere.

TABLE I. Calculated growth exponents α and β obtained from the stochastic simulation of different models in substrate dimensions $d'=1$ and 2. The local rules for the various growth models are described in the text.

d'	Model	β	α
1	1+	0.375 ± 0.001	1.450 ± 0.011
	$d2+$	0.375 ± 0.002	1.422 ± 0.007
	2+	0.366 ± 0.001	1.461 ± 0.003
	2±	0.255 ± 0.003	0.501 ± 0.021
2	1+	0.237 ± 0.002	0.95 ± 0.01
	$d4+$	0.194 ± 0.006	
	2+	0.192 ± 0.002	
	4±	$<0.02\pm0.001$	$<0.101\pm0.001$
	3+	0.162 ± 0.002	
	4+	0.162 ± 0.002	
	$ud4+$	0.156 ± 0.002	
	$ru4\pm$	$<0.02\pm0.002$	
	$uu4\pm$	$>0.45\pm0.04$	

Our results are summarized in Table I. On comparing the simulation results with the known theoretical [5] values of α and β, it is clear that in $d'=1$ all the models apparently follow linear growth equations, whereas in $d'=2$, only 1+ and 4± follow linear equations. We use our computed values of β, which are much more reliable than those for α, and the least-squares fitting to Eq. (2) in deciding which continuum equation applies to a particular simulation result. (Our least-squares fitting for the linear models reveals that α is always strongly affected by crossover effects.) Figure 2 shows the log-log plot of W vs t for all the models in $d'=2$ with $L=256$. The inset gives the same plot for models 2+, $d4\pm$, 3+, and 4+ for $L=1000$. The curves for 3+ and 4+ are overlapping because the distinct configurations in 4+ are rare. For $L\geq 50$ the power-law behavior sets in almost immediately as soon as one monolayer is completed. We find that the β values change with the system size for smaller values of L, but tend to saturate with increasing L. Thus, on comparing the inset with the main figure the slopes of the curves change by small amounts when L varies from 256 to 1000. The values displayed in Table I are for $L=1000$ for the models in the inset. For 1+, β saturates around 0.24 for $L\geq 70$. For 4±, β tends to zero showing logarithmic growth, consistent with the expected Edwards-Wilkinson universality [12].

Comparing these results with the theoretical exponents [5], the underlying growth equation for each model becomes apparent. Thus, 1+ follows

$$\partial h/\partial t=-b\nabla^4 h+\eta,\quad \beta=(5-d)/8\,, \tag{3a}$$

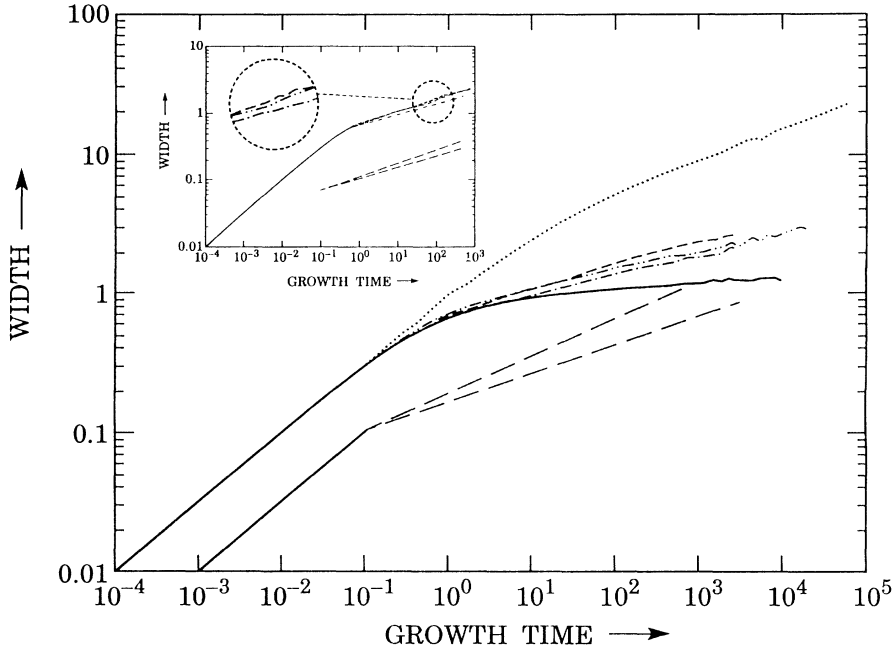

FIG. 2. Log-log plot for the simulated width W vs t (average height) for various models in $d'=2$, for the substrate size 256×256. (····) 1+, (---) 2+, (-·-·) $d4+$, (— · · — · ·) 3+ and 4+, and (——) $4\pm$. Inset: Results for 2+, $d4+$, and 3+ and 4+ for the system size 1000×1000. For comparing the slopes, straight lines with slopes $\frac{1}{4}$ and $\frac{1}{5}$ ($\frac{1}{5}$ and $\frac{1}{6}$) are shown in the main figure (the inset).

$4\pm$ and $ru4\pm$ follow

$$\partial h/\partial t = a\nabla^2 h + \eta, \quad \beta=(3-d)/4, \tag{3b}$$

$d4+$ and 2+ follow

$$\partial h/\partial t = -b\nabla^4 h + c\nabla^2(\bar{\nabla}h)^2+\eta, \quad \beta=(5-d)/(7+d), \tag{3c}$$

3+, $ud4+$, and 4+ follow

$$\partial h/\partial t = -b\nabla^4 h + d\bar{\nabla}\cdot(\bar{\nabla}h)^3+\eta, \quad \beta=(5-d)/2(3+d). \tag{3d}$$

The theoretical [5] (α,β) values in $d'=1$ are (1.5,0.375), (0.5,0.25) for Eqs. (3a) and (3b), respectively, while in $d'=2$, they are (1.0,0.25), (0.0,0.0), (0.666,0.2), and (0.5,0.166) for Eqs. (3a), (3b), (3c), and (3d), respectively.

We ascribe the difference in the manifestation of the nonlinear terms in $d'=1$ and 2 to the difference in the number of available configurations required to make use of further relaxations (i.e., $N_c \Rightarrow N_c^+$). In $d'=1$, there is an acute paucity of configurations which are needed for the nonlinearity to manifest itself even for system sizes as large as $L=10^4$. Note that the introduction of nonlinear terms in the growth equation causes the dynamical exponent $z=\alpha/\beta$ to decrease. Since the correlation length is given by $\xi\sim t^{1/z}$, at a given time during growth ξ increases as z decreases. Thus, the effect of relaxing the rules for hopping (i.e., as one goes from 1+ to 4+) is to make more and more configurations available for relaxation so that the height-height (h-h) correlations among the neighboring columns increase, making ξ grow faster. In $d'=1$, such configurations (i.e., configurations required to take advantage of the increased relaxation, as offered by the rules for 2+ and $d2+$) are far too few in number to significantly affect the surface morphology, even for $L=10^4$ and $h\cong10^4$. Our results in $d'=2$, however, unambiguously identify that the models 2+ and $d4+$ belong to the universality class defined by the growth equation (3c). This suggests that in $d'=1$ the coefficient c in Eq. (3c) is much too weak to overcome the crossover effect arising from the linear term even for the largest system sizes considered in the simulations. We note that the microscopic growth rules employed by Wolf and Villain [11] are those for $d2+$ and, for large enough system size and time, it should show the universality of Eq. (3c). Thus, Das Sarma Tamborenea [10] (model 1+) and Wolf and Villain [11] actually belong to different universality classes even though the difference does not manifest itself in $d'=1$ even for $L=10^4$.

One interesting aspect of these models is that it is possible to correlate the dynamical spreading of ξ ($\sim t^{1/z}$) with interlayer hops. We first note that in the absence of evaporation and vacancies h-h correlation is introduced only by interlayer hops, i.e., downward hops in this study. We have explicitly verified that the hops within the plane of deposition do not lead to any h-h correlation at long enough times—β for this purely intralayer hopping model

is 0.5. As one follows the rules for deposition sequentially from 1+ to 4+, more and more configurations are available for downward hops, reducing z correspondingly from 4 to 3. Even more such configurations are made available in $4\pm$ reducing z further to 2. Because of the flat substrate geometry, hops towards the substrate always lead to a smoother morphology. We have investigated the effect of the upward hops as well (to be published elsewhere) for the models $d4+$ and $4\pm$. With the inclusion of upward hops (model $ud4+$) in model $d4+$, β changes to 0.155 ± 0.001 which corresponds to the $\bar{\nabla}\cdot(\bar{\nabla}h)^3$ term in the growth equation with $z=3$, a lower value of z compared to 3.5 for the model $d4+$. For the model $4\pm$, when the upward hops are restricted by the condition that N_c must increase after the hop (model $ru4\pm$), logarithmic growth is obtained. On the other hand, when upward hops are unrestricted in model $4\pm$ (model $uu4\pm$), unstable growth ensues. These results suggest that the higher the probability for the *interlayer hops that tend to flatten the terraces, the lower* is the exponent z.

It is well accepted [1,9] that nonequilibrium growth *allowing* desorption or overhangs and/or vacancies is asymptotically described by the KPZ equation [2]. Ballistic deposition, where atoms are allowed to stick at the sides (leading to lateral growth) as well as on the tops of the substrate atoms, is a well-known example of such KPZ-type growth. Because some evaporation and bulk vacancy formation is, in principle, always present in any growth situation, the KPZ equation is always the asymptotic nonequilibrium growth equation (for long times and large systems). However, as a function of the substrate temperature or the growth rate, one would expect to observe various crossover effects corresponding to the various SOS hopping models discussed here as surface diffusion dominates the relaxation process for intermediate system sizes and growth times.

In conclusion, we have shown, using stochastic simulations of a number of growth models in $d'=1$ and 2, that within the SOS restrictions, the tendency to increase the coordination number through hopping leads to linear and nonlinear fourth-order dynamical growth equation depending upon the details of the local growth rules. Details of the local microscopic growth rules govern the universality class of SOS nonequilibrium growth, in contrast to ballistic deposition which is always described by the KPZ equation.

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