Far from equilibrium nonconserved growth under a surface diffusion bias

C. J. Lanczycki,* V. Jejjala, and S. Das Sarma
Department of Physics, University of Maryland, College Park, Maryland 20742
(Received 8 February 1996)

We study a generic one-dimensional atomistic model of interface growth under random ballistic deposition in the presence of a surface diffusion bias allowing for surface overhangs and bulk vacancies. We find that various diffusion bias induced surface instabilities recently found in the solid-on-solid approximation of kinetic growth are absent in the generic model with the usual statistically self-affine Kardar-Parisi-Zhang scaling dominating the surface morphology. For strong biases and high temperatures, the growing surface resembles the zero temperature ballistic growth without a diffusion bias. The growth front morphologies show intricate flamelike nonlocal structures not typically present in self-affine surfaces. This indicates that the standard coarse-grained single-variable description of the growing film by its local surface height coordinate misses an important qualitative feature, namely, a novel flamelike roughening behavior along vertical faces of the growth front. [S1063-651X(96)08511-X]

PACS number(s): 05.40.+j, 82.20.Mj, 68.55.Jk

It has long been known [1] that an atom diffusing across a terrace is affected by an additional energy barrier, or surface diffusion bias, when crossing a step edge: an atom finds it easier to hop to an in-plane kink site rather than an out-of-plane kink. A number of recent studies [2–5] incorporating this effect have demonstrated complex and interesting morphological instabilities in nonvicinal interfaces growing under conservative dynamics in the presence of an external particle flux. These conserved models analyze the solid-on-solid (SOS) growth situation in which the crystal is free of defects and has a continuum description [6] obeying the current continuity equation $\partial h/\partial t = -\nabla\cdot\mathbf{j} + \eta$. Here, j is a model dependent surface current and η is a stochastic shot (white) noise term modeling the spatiotemporal fluctuations in the incident atomic beam. The surface profile at time t is expressed as the single-valued function $h(x,t)$ giving the coordinate of the highest atom above substrate site x and denoting the surface height fluctuation with respect to the average height h at time t. The simplest possible instability arises when $\mathbf{j} = -\nu_2\nabla h$ with $\nu_2 < 0$ [5], although more complex forms are possible and have also been studied [2].

In general, both continuum and discrete SOS growth models [2–4] in the presence of a diffusion bias lead to morphological instabilities in contrast to the usual self-affine dynamic scaling behavior traditionally associated with kinetic surface roughening. We report herein a study of a generic interface growth model [7] in $1+1$ dimensions which permits overhangs and defects to occur in the presence of a diffusion bias, and is not restricted to the simplest SOS approximation. Evaporation from the growth front is neglected in our work. We find that the generic model in the presence of a surface diffusion bias does not exhibit the typical growth instabilities, and in fact, $h(x,t)$ crosses over to the asymptotically expected statistically scale invariant behavior *faster* than it does in the absence of a diffusion bias. This result has also been seen by Schimschak and Krug (SK) [8] recently in a somewhat simplified variant of our full generic model. For strong diffusion biases at finite temperature T, the surface appears to evolve quantitatively as does a $T=0$ K model lacking any diffusion bias. Additionally, the diffusion bias leads to an unexpected and intricate flamelike growth front with lateral roughening (i.e., roughening normal to direction of growth) appearing along vertical faces. These highly nontrivial features are ignored by a description of the film using a single dynamical variable $h(x,t)$. This is therefore an interesting example of a situation where employing a single-variable coarse-grained continuum description, typically successful in theories of kinetic surface roughening, overlooks an important aspect of the interfacial dynamics. Our work and the recent work reported in Ref. [8] definitively establish that various reported growth instabilities under a surface diffusion bias are features of the SOS approximation and thus can appear only when bulk defects and surface overhangs are dynamically inactive during the growth process.

The generic model in the presence of defects such as vacancies and overhangs [7,8] is expected to asymptotically obey the coarse-grained continuum equation proposed by Kardar, Parisi, and Zhang (KPZ) [9]:

$$\partial h/\partial t = \nu_2\nabla^2 h + \lambda(\nabla h)^2 + \eta. \tag{1}$$

The KPZ equation is nonconservative due to the nonlinear term $\lambda(\nabla h)^2$, which represents growth normal to the surface. The KPZ equation displays statistical scale invariant behavior, or equivalently, the KPZ interface is a self-affine object, although a more complex SOS regime has been observed in simulations prior to the crossover to the asymptotic KPZ scaling [7,8]. Characterizing the interface by its width $W(L,t) = \langle [h(x,t) - \bar{h}(t)]^2\rangle^{1/2}$, where we perform an ensemble average and an average over the L substrate sites, one can show that $W(L,t) \sim t^{\beta}$ for $t \ll L^z$ and $W(L,t) \sim L^{\alpha}$ when $t \gg L^z$. For the one-dimensional interface we consider $(d=1)$, the exact exponents of Eq. (1) are given by [9]: $\beta = 1/3, z = 3/2$, and $\alpha = z\beta = 1/2$.

* Permanent address: Computational Biosciences and Engineering Laboratory, Division of Computer Research and Technology, The National Institutes of Health, Bethesda, MD 20892.

The KPZ equation is usually considered for the case $\nu_2 > 0$, so one can question how a $\nu_2 < 0$ linear instability, as arising from a step-edge diffusion bias, might influence the KPZ dynamic scaling behavior. The long wavelength behavior in the presence of the instability is *asymptotically* expected to remain in the KPZ universality because upon renormalization the nonlinear coupling generates a positive ν_2 which eventually overcomes the initial negative ''bare'' ν_2. Physically, the lateral growth inherent in the generic nonconservative case will ultimately suppress incipient instabilities through the creation of localized overhangs, rendering the full multivalued nature of the interface invisible to the single-valued function $h(x,t)$.

The KPZ equation with $\nu_2 < 0$ is an important simplification of the noisy Kuramoto-Shivashinsky (KS) equation, recently analyzed by Cuerno and Lauritsen [10] in the context of interface growth phenomena:

$$\partial h/\partial t = \nu_2 \nabla^2 h - \nu_4 \nabla^4 h + \lambda (\nabla h)^2 + \eta, \tag{2}$$

where $\nu_2 < 0$ and $\nu_4 > 0$. We point out that the fourth order term $\nu_4 \nabla^4 h$ represents to leading order the surface diffusion of atoms to highly coordinated sites [6,13]. Without the noise term ($\eta = 0$), Eq. (2) is the standard KS equation of flame front propagation and chemical turbulence [14], while the noisy version has previously been applied to ion sputtering processes [10,11]. An anisotropic KS equation, with and without noise, has also been studied in the context of growth and erosion applications [12]. All these versions of the KS equation asymptotically display KPZ universality for $d = 1$ [10–12,15]. The exact correspondence between the KPZ equation and the noiseless KS equation is, in fact, quite nontrivial and has been convincingly demonstrated only in $d = 1$. The noiseless KS equation [$\eta = 0$ in Eq. (2)] is a deterministic equation that at very late times manifests a form of deterministic chaos which mimics the properties of white noise [14,15], and a direct derivation of Eq. (1) from the KS equation in the hydrodynamic regime has recently been accomplished [16]. The asymptotic KPZ scaling of the KS equation, however, appears only after an extremely long non-KPZ transient associated with the linear, conservative terms [15]. The corresponding crossover to the asymptotic KPZ scaling in the noisy KS equation has not yet been investigated and whether there is a long-lived preasymptotic transient in the noisy KS equation, similar to that of the noiseless case, is not in general known. In the sputtering models, for which the noisy KS equation has been proposed as a coarse-grained continuum description, such a long transient to the asymptotic scaling has been suggested [11]. In a growth model with solid-on-solid type deposition and Arrhenius-activated hopping of only singly bonded atoms that permits defect formation [8], SK found no instability and rather rapid development of KPZ asymptotics [8]. We address the robustness of these results by analyzing the transient (and experimentally relevant) crossover behavior in various regimes of temperature and diffusion bias strength, and utilizing the generic model which possesses several significant differences from that of SK. In particular, we go beyond the SOS approximation both for deposition and diffusion, and our activated hopping process is not arbitrarily limited to singly bonded atoms.

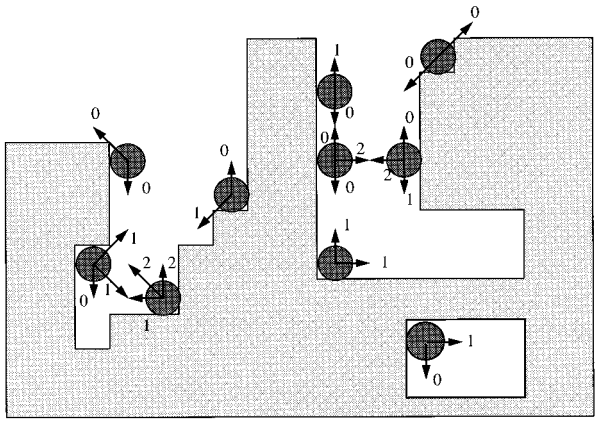

FIG. 1. For selected atoms (circles) on the surface, the possible diffusional moves are indicated, together with a number δ which is the decrease in next-nearest neighbors when $\delta \geq 1$ and $\delta = 0$ otherwise. The selected process overcomes the diffusion bias with probability p^{δ}.

We study a one-dimensional Monte Carlo growth model with random ballistic deposition and atomistic diffusion via activated hopping which possesses the following properties: vacancy and overhang formation ($\lambda \neq 0$) [7,8], surface diffusion ($\nu_4 > 0$) [13], an external atomic beam ($\eta \neq 0$), and a diffusion bias ($\nu_2 < 0$) [5]. Based on these properties and the usual coarse-graining procedure to go from a discrete model to the continuum limit, our model can be qualitatively mapped onto the noisy KS equation [17], Eq. (2). Atoms deposit one by one normally onto an initially flat (singular) one-dimensional substrate at the rate of 1 layer/sec following the rules of random ballistic deposition [7]. Periodic boundary conditions are employed. Surface diffusion occurs stochastically according to the local coordination dependent Arrhenius rates $R_n = (kT/h)e^{-E_n/kT}$. The activation energy for the diffusion of an atom with bonds to n occupied nearest neighbor (NN) sites is $E_n = 1.0\text{eV} + (0.3\text{eV})n$. Any atom may be selected to diffuse at any time subject to these average rates. The diffusing atom randomly chooses a landing site from the set of eight NN and next-NN sites which are unoccupied, and all hops are successful when no diffusion bias is present. However, a landing site is rejected unless it provides at least one NN bond (see Fig. 1), and the motion of that atom must not leave behind a disconnected atom or pair of atoms. Thus our simulation explicitly disallows the evaporation of single particles or two particle clusters; frequently, this logic also prevents clusters of three (or more) particles from becoming disconnected. All hops which lead to a disconnectivity of the crystal require a doubly or triply bonded atom to hop: at low temperatures, these events are rare while at high temperatures the surface tends to be compact enough so as to not generate configurations in danger of breaking free. If a large atomic cluster breaks off, it diffuses so slowly from the surface (being composed of many doubly and triply bonded atoms and due to its large size) that it is certain to rapidly reattach to the growth front. Hence, while not rigorously forbidden, in the temperature regime of interest previous experience [7,18] has established that our algorithm seems to eliminate evaporation for all practical purposes from our simulations.

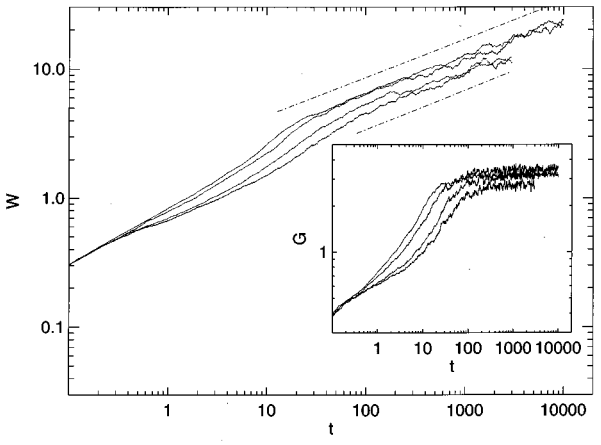

FIG. 2. The width and average slope (inset) for $T=600$ K, for diffusion biases $p=1.0,0.5,0.1,10^{-4}$ (bottom to top) with $L=1000$; average of 15 runs. The slope of the upper fit line ($p=10^{-4}$) is $\beta=0.294\pm0.005$, and for the lower line ($p=1.0$), $\beta=0.304\pm0.10$. The saturation of G confirms KPZ scaling at a time which decreases with p.

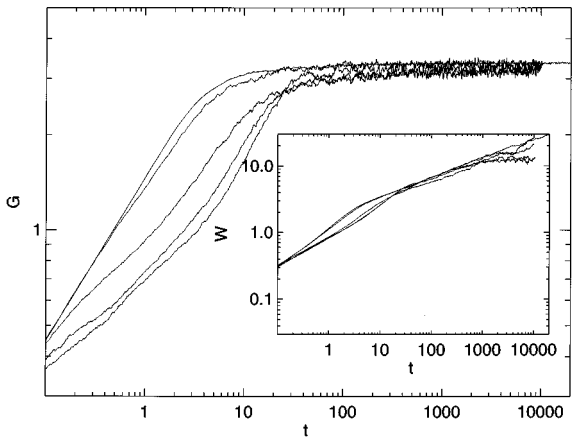

FIG. 3. The average slope and width (inset) for $p=10^{-4}$ at $T=625(L=500$; five runs), $600(L=1000;15$runs), 550 ($L=600$; ten runs), and 500 ($L=600$;ten runs) K and for $T=0$ K ($L=5000;20$ runs) from bottom to top. The growth at each T behaves as if $T=0$ K after defects are formed.

A step-edge diffusion bias is imposed upon this general scheme (following Ref. [19]) such that the total rate of a hopping process is $R=p^{\delta}R_n$. The parameter $p=e^{-E_s/kT}$ represents the hopping probability over the additional step-edge diffusion barrier of size E_s. If the number of *next*-nearest neighbors is reduced by the hop, the exponent δ gives this decrement; if the number of next-NNs increases or remains unchanged then $\delta=0$ for that particular process (i.e., there is no diffusion bias and $p=1$ for that situation) and R reduces to R_n. A random number decides if the barrier is surmounted, but upon failure the attempt is aborted and the atom remains in the initial position. Figure 1 demonstrates all possible diffusion moves for some selected atoms, along with the value of δ associated with each hop. This implementation, which we have called a ''reflection barrier'' [4], generally makes it difficult for atoms to reach the step edge. As a result, configurations conducive to disconnectivity are discouraged and we have observed no free detached clusters in our simulations. Using the step-edge barrier model of SK in our simulations instead of the reflection barrier, however, does lead to significant disconnectivity problems being observed due to the formation of straight chains of atoms favored by that rule [20]. The different ways in which the surface diffusion barrier enters growth simulations is a significant difference between our work and that in Ref. [8].

In Fig. 2 we show the evolution of W and of the rms surface slope $G(t)=\langle[h(x,t)-h(x-1,t)^2]\rangle^{1/2}$ for $T=600$ K with no barrier ($p=1.0$) and for progressively stronger diffusion biases of $p=0.5,0.1$, and 10^{-4}. Without a bias the asymptotic KPZ scaling regime ($t>100$), most easily identified by the saturation of $G(t)$, is preceded by a SOS transient ($t<20$) and a defect formation regime ($t=20$–100) [7]. Rather than extending the SOS transient associated with the conservative terms in Eq. (2), with increasing bias (decreasing p) the SOS transient in fact completely disappears with defect formation being the only precursor to the asymptotic KPZ scaling. We contrast this with the very long conservative transient in the noise-free KS equation [15]. At fixed T Fig. 2 shows that both W and G possess very notable changes with p within the KPZ scaling regime. This is most clearly seen in the inset where the earlier saturation of $G(t)$ signifies an earlier onset of KPZ scaling with increasing diffusion bias. A reduction in the crossover scale for KPZ scaling was noted by SK in a gentler model using a SOS deposition rule and for smaller barriers of $p\geqslant0.4$ [8], while it is explicitly observed here out to a very strong diffusion bias of $p=0.0001$ where one might naively expect instabilities to be strong. Based on these studies and a comparison to the noise-free KS equation, we conclude that the explicit shot noise present in Eq. (2) ($\eta\neq0$) can overwhelm pre-KPZ transients associated with the linear growth terms, and cause the asymptotic KPZ scaling to emerge much more readily than in the original noiseless KS equation itself.

Figure 3 shows growth simulation results under a strong bias of $p=10^{-4}$ at $T=500$, 550, 600, and 625 K, along with the $T=0$ K diffusionless curve for reference. For $t\geqslant20$ both G and W display the same quantitative behavior for the full range of temperatures (finite size effects in W for $t>1000$ arise due to the L dependence of the saturated width). Not only do the absolute values of G and W exhibit temperature independence once the asymptotic KPZ regime is attained, but the slope of the width curve also remains roughly unchanged, indicating an essentially temperature independent growth exponent ($\beta=0.295\pm0.015$). The only T dependence arises in how defects form and propagate in the early time regime ($t<20$). Unlike in the zero barrier case ($p=1$), where W and G have temperature dependent values [7], under a strong diffusion bias nonconservative growth appears to lose all temperature dependence within the KPZ regime and behaves quantitatively as it does at $T=0$ K. Interestingly then, a strong diffusion bias, in fact, tends to lead to *simpler* growth in that temperature effects in the surface height function $h(x,t)$, arising from thermally activated hopping at the growth front, appear to be suppressed in the asymptotic regime. Thus finite temperature ballistic growth under a strong diffusion bias is surprisingly similar to zero temperature ballistic growth without any diffusion.

Our results as well as those in SK clearly establish that the various growth instabilities obtained in the SOS model in the presence of a diffusion bias are specific features of the

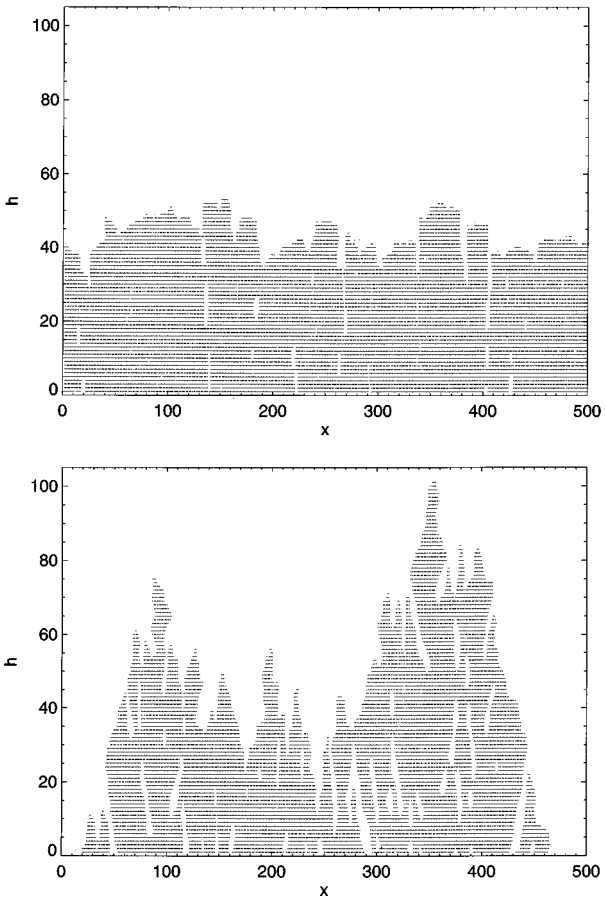

FIG. 4. Crystal morphologies with $p=1.0$ (top) and $p=10^{-4}$ (bottom) at $T=625$ with $L=500$ after 10 000 layers of growth. The vertical scale has been shifted for clarity. Note the flamelike character due to the diffusion bias induced destabilization of the flat vertical surfaces.

SOS approximation, which are rapidly renormalized away by vacancies and overhangs in the generic model. Except for the details of defect formation, the parameter p proves inconsequential in determining the surface dynamics from this viewpoint, i.e., one finds statistically scale invariant dynamic (KPZ) scaling for all values of p. This simple picture, based on a coarse-grained single-valued height variable description, misses the full role of the diffusion bias in determining a novel pattern formation behavior in nonconservative growth, however. The top panel of Fig. 4 shows growth at $T=625$ K with no bias and the single-valued function h is adequate in representing the interface. For a strong diffusion bias of $p=10^{-4}$ (lower panel), h is a poor representation of the richness of the growth morphology, which now exhibits many flamelike undulations on vertical columnar portions of the surface. This type of pattern formation due to a surface diffusion bias in ballistic growth can be considered a form of nonlocal kinetic roughening which is not amenable to a single-variable description. The nonconservative nature of the growth rules in our generic model, especially at high T and for strong biases, allows atoms to roughen the flat vertical segments seen for $p=1.0$ into the curved features for small p. A strong barrier in our model prevents the fastest atoms from reaching edge sites and thereby turning corners, and therefore an atom in column x can be readily trapped on

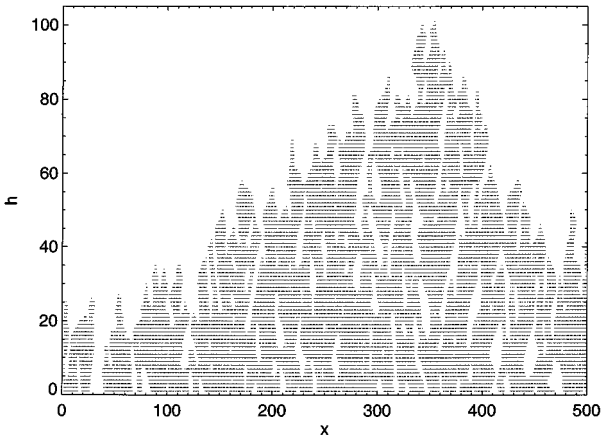

FIG. 5. Crystal morphology for $p=10^{-4}$ at $T=600$ with $L=1000$ after 10 000 layers of growth. The vertical scale has been shifted for clarity. The flamelike character persists at this lower temperature.

a vertical surface, diffusing along it until encountering other atoms with the same substrate coordinate x. Such processes build up the ''lateral'' roughness which collectively produces the flamelike character in the $p=10^{-4}$ case. This implies a novel nonlocal instability of the flat vertical columns in the presence of the diffusion bias. The flamelike undulations also lead to large tilted voids in the bulk rather than the extremely narrow vertical voids seen [7,8] for the no-bias $p=1.0$ case. In Fig. 5 we show a morphology grown at $p=10^{-4}$ and $T=600$ K. A similar flamelike morphology is apparent, and we conclude that this nonlocal flamelike roughening pattern along vertical faces of the surface can arise whenever the ''reflection barrier'' type diffusion bias [4] is strong in a nonconservative growth model. For the highest barrier in the SK model ($p\sim 0.05$ in our language) a more angular set of peaks emerges [8], consistent with their use of a barrier akin to the ''edge barrier'' which produced such morphologies even in a purely SOS model [4]. The details of atomic motion near a step edge thus appear to be quite important in determining the full morphology of nonconservatively grown films. While the ''reflection barrier'' type diffusion bias produces the flamelike pattern, the step edge barrier model used by SK does not.

Our generic model with a reflection barrier is a unique example in which a coarse-grained description of growth in terms of the single-valued height function $h(x,t)$ becomes inadequate and a more complete theory is needed to capture the lateral roughness observed. The nonlocal flamelike pattern formation we find is manifestly a nonconservative growth phenomenon, totally distinct in nature from the various diffusion bias induced SOS growth instabilities (which are all local phenomena) [2–6] and is qualitatively different from the SK results [8]. For nonconservative nonequilibrium growth in the presence of a step-edge diffusion bias, new nonlocal methods must be developed to characterize the full morphological features of the resulting crystals. The most obvious and simplest extension of current formalism is to consider the perimeter of the crystal, given as a line $s(t)$ in two dimensions, rather than the height $h(x,t)$. Due to the incident flux, this stringlike object would be elastic as its length must be nonconserved, and subject to particular equa-

tions of motion whose elucidation remains a topic for future research. One interesting avenue to investigate this novel type of pattern formation, which manifestly requires going beyond the single-valued height description, may be the recently introduced ''reparametrization invariance'' [21] idea in stochastic continuum growth equations. To the extent one insists upon a single-valued coarse-grained description where the surface is defined by the height maxima, the KPZ equation continues to describe asymptotic nonconservative growth, even with a strong destabilizing diffusion bias. While both our generic model and that of SK contain all the continuum elements of the noisy KS equation, it appears that the shot noise plays an important role in bringing about a very rapid renormalization of the $\nu_2<0$ instability and the consequent emergence of the statistical scale invariance associated with KPZ universality. For very strong biases (e.g., $p=0.0001$), growth in the KPZ regime is essentially independent of temperature although intricate flamelike morphologies develop at the growth front, suggesting other non-KPZ ordering phenomena are present. A theoretical understanding of this new pattern formation at the growth front must await further investigation.

This work is supported by the U.S. ONR.

[1] G. Ehrlich and F. G. Hudda, J. Chem. Phys. **44**, 1039 (1966); R. L. Schwoebel and E. J. Shipsey, J. Appl. Phys. **37**, 3682 (1966).

[2] M. D. Johnson, C. Orme, A. W. Hunt, D. Graff, J. Sudijono, L. M. Sander, and B. G. Orr, Phys. Rev. Lett. **72**, 116 (1994); M. Siegert and M. Plischke, *ibid.* **73**, 1517 (1994).

[3] I. Elkinani and J. Villain, J. Phys. (France) I **4**, 949 (1994).

[4] C. J. Lanczycki and S. Das Sarma, Phys. Rev. Lett. **76**, 780 (1996).

[5] J. Villain, J. Phys. (France) I **1**, 19 (1991); J. Krug, M. Plischke, and M. Siegert, Phys. Rev. Lett. **70**, 3271 (1993).

[6] Z. W. Lai and S. Das Sarma, Phys. Rev. Lett. **66**, 2348 (1991).

[7] S. Das Sarma, C. J. Lanczycki, S. V. Ghaisas, and J. M. Kim, Phys. Rev. B **49**, 10 693 (1994); references therein.

[8] M. Schimschak and J. Krug, Phys. Rev. B **52**, 8550 (1995).

[9] M. Kardar, G. Parisi, and Y. C. Zhang, Phys. Rev. Lett. **56**, 889 (1986).

[10] R. Cuerno and K. B. Lauritsen, Phys. Rev. E **52**, 4853 (1995).

[11] R. Cuerno and A.-L. Barabasi, Phys. Rev. Lett. **74**, 4746 (1995); R. Cuerno, H. A. Makse, S. Tomassone, S. T. Harrington, and H. E. Stanley, *ibid.* **75**, 4464 (1995).

[12] M. Rost and J. Krug, Phys. Rev. Lett. **75**, 3894 (1995).

[13] D. E. Wolf and J. Villain, Europhys. Lett. **13**, 389 (1990); S. Das Sarma and P. I. Tamborenea, Phys. Rev. Lett. **66**, 325 (1991); J. M. Kim and S. Das Sarma, *ibid.* **72**, 2903 (1994); J. Krug, *ibid.* **72**, 2907 (1994); S. Das Sarma, C. J. Lanczycki, R. Kotlyar, and S. V. Ghaisas, Phys. Rev. E **53**, 359 (1996).

[14] Y. Kuramoto and T. Tsuzuki, Prog. Theor. Phys. **55**, 356 (1976); G. I. Shivashinsky, Acta Astronautica **6**, 560 (1979); S. Zaleski, Physica D **34**, 427 (1989); F. Hayot, C. Jayaprakash, and Ch. Josserand, Phys. Rev. E **47**, 911 (1993).

[15] K. Sneppen, J. Krug, M. H. Jensen, C. Jayaprakash, and T. Bohr, Phys. Rev. A **46**, R7351 (1992).

[16] C. Chow and T. Hwa, Physica D **84**, 494 (1995).

[17] The model analyzed by Schimschak and Krug (Ref. [8]) also has this general mapping property, although overhang and vacancy formation (ballistic deposition versus SOS deposition) and the diffusion barrier (in strength and implementation) are notably different in our generic model. The SK diffusion rates (given as $R\tau_{ML}=200$) are, however, roughly comparable to ours at $T=600$ for singly bonded atoms.

[18] C. J. Lanczycki and S. Das Sarma, Phys. Rev. B **51**, 4579 (1995).

[19] P. Smilauer, M. R. Wilby, and D. D. Vvedensky, Phys. Rev. B **47**, 4119 (1993).

[20] V. Jejjala, C. J. Lanczycki, and S. Das Sarma (unpublished).

[21] M. Marsili, A. Maritan, F. Toigo, and J. R. Banavar, Rev. Mod. Phys. (to be published).