Numerical test of the damping time of layer-by-layer growth on stochastic models

S. Park,[1] H. Jeong,[2] and B. Kahng[1]
[1]*Department of Physics and Center for Advanced Materials and Devices, Konkuk University, Seoul 143-701, Korea*
[2]*Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556*
(Received 17 December 1998)

We perform Monte Carlo simulations on stochastic models such as the Wolf-Villain (WV) model and the Family model in a modified version to measure the mean separation ℓ between islands in a submonolayer regime and the damping time $\tilde{t}$ of layer-by-layer growth oscillations in one dimension. The stochastic models are modified, allowing for diffusion within interval r upon deposition. It is found numerically that the mean separation and the damping time depend on the diffusion interval r, leading to the fact that the damping time is related to the mean separation as $\tilde{t}\sim\ell^{4/3}$ for the WV model and $\tilde{t}\sim\ell^{2}$ for the Family model. The numerical results are in excellent agreement with recent theoretical predictions. [S1063-651X(99)06505-8]

PACS number(s): 68.35.Ct, 81.15.−z

Recently the problem of surface growth by molecular beam epitaxy (MBE) has been attractive in statistical physics [1]. From the point of view of statistical physics, it is intriguing how various stochastic processes involving many atoms, such as shot noise, diffusion, and nucleation, are described in terms of scaling structures. These cooperative phenomena depend crucially on the ratio between the diffusion constant D and the deposition rate F. The deposition rate is defined as the number of atoms landing on a surface per unit area and unit time. Atoms deposited on a surface diffuse until they meet one another to form dimers which then grow into islands of monoatomic height. The mean distance of nucleation events, corresponding to the mean separation between islands, is determined by the ratio between the diffusion constant D and the deposition rate F as [2–8]

$$\ell\sim(D/F)^{\gamma}. \tag{1}$$

The exponent γ depends on the diffusion process of adatoms and island shape. It is also a function of the critical island size i^*, defined by the size i^*+1 of the smallest island which is stable enough that it never decays before capturing the next adatom. When only adatoms can move and desorption can be neglected, the exponent γ can be predicted using the kinetic theory as [6]

$$\gamma=\frac{i^*}{2i^*+d+d_f}. \tag{2}$$

However, recently the formula was corrected for the case of $d=1$ and $i^*\geqslant 2$ as [9]

$$\gamma=\frac{i^*}{2i^*+3}. \tag{3}$$

As the number of adatoms increases in the submonolayer, islands grow in size and coalesce, forming bigger islands, and eventually cover a monolayer. If adatoms are allowed to diffuse to stable sites such as kink sites or step edges before other adatoms are deposited and if interlayer diffusion is not inhibited by the Ehrlich-Schwoebel barrier [10,11], a smooth surface with a minimum of defects is grown. In this case, the surface exhibits a layer-by-layer growth [12]. As islands evolve, the density of atomic steps oscillates, which is a hallmark of layer-by-layer growth. The oscillating behavior can be monitored through reflection high-energy electron diffraction (RHEED) intensity in the laboratory, which is useful for measuring film thickness easily. Layer-by-layer growth is important for the fabrication of microelectronic devices. It is known that the oscillation persists perfectly when system size is smaller than the layer-coherence length $\tilde{\ell}$ such that for two sites within the coherence length $\tilde{\ell}$, they grow coherently, and beyond which they are out of phase [13]. The coherence length is related to the separation between island as $\tilde{\ell}\sim\ell^{4/(4-d)}$. The coherence length is much larger than other length scales such as the island separation ℓ or the characteristic length $\ell_0\sim(D/F)^{1/(2+d)}$, where ℓ_0 is formed by a dimensional combination of D and F, meaning that if system size is smaller than ℓ_0, it can accommodate at most one island. When system size is larger than $\tilde{\ell}$, the oscillation of step density is damped due to various types of fluctuations. It is recently found [13] that there exists a characteristic time $\tilde{t}$, beyond which the damped oscillation disappears. The characteristic time is scaled as

$$\tilde{t}\sim(D/F)^{\delta}, \tag{4}$$

meaning the critical time for the transition from layer-by-layer growth to kinetic roughening growth.

In the kinetic roughening growth, when the number of adatoms is conserved, surface growth may be described by the equation [11,13–15],

$$\partial_t h=-\nabla\cdot\mathbf{j}+\eta, \tag{5}$$

where h is surface height, $\mathbf{j}$ is adatom current, and $\eta(\mathbf{x},t)$ denotes shot noise satisfying $\langle\eta(\mathbf{x},t)\rangle=0$ and having the correlation

$$\langle\eta(\mathbf{x},t)\,\eta(\mathbf{x}',t')\rangle\sim\delta^{d}(\mathbf{x}-\mathbf{x}')\,\delta(t-t'), \tag{6}$$

with substrate dimension d. When adatom current is driven by the gradient of surface curvature and square of surface tilt with coefficients K and λ [11,13–15],

$$\mathbf{j}=\nabla[K\nabla^2 h-\lambda(\nabla h)^2]. \tag{7}$$

The continuum equation for surface growth is written as

$$\partial_t h = -K\nabla^4 h + \lambda\nabla^2(\nabla h)^2 + \eta(x,t), \tag{8}$$

which, called the conserved Kardar-Parisi-Zhang (CKPZ) equation, has been discussed in the context of MBE growth. For the CKPZ universality class, it was derived that the exponent δ for the damping time is related to γ for the island separation [13] as

$$\frac{\delta}{\gamma} = \frac{4d}{4-d}. \tag{9}$$

The derivation was based on the dimensional analysis on the continuum equation. The formula was checked numerically in one dimension by performing the coarse graining Monte Carlo simulations [13].

For the surface growth driven by downhill current with coefficient ν,

$$\mathbf{j} = -\nu\nabla h, \tag{10}$$

the continuum equation is written as

$$\partial_t h = \nu\nabla^2 h + \eta(x,t), \tag{11}$$

which is called the Edwards-Wilkinson (EW) equation [16]. In this case, adatoms tend to move in a downward direction on the surface, which is realized by exchanging a mobile atom with an adatom at a step edge as observed in homoepitaxial growth on Ir(111) [17]. In this case, the exponent δ for the damping time is related to γ [13] as

$$\frac{\delta}{\gamma} = \frac{2d}{2-d}, \tag{12}$$

which was also derived based on the dimensional analysis on the continuum equation, Eq. (11). However, successful numerical confirmation of Eq. (12) has not been reported yet. Our preliminary numerical result for MBE growth with downhill current does not fit well to Eq. (12), requiring a numerical check of Eqs. (9) and (12) on stochastic models belonging to the CFKPZ and EW universalities, respectively. In this report, we perform Monte Carlo simulations on the stochastic models, the Wolf-Villain (WV) model [18] and the Family model [19] with some modifications in their dynamic rules to check Eqs. (9) and (12).

Let us first consider the modified WV model, of which the dynamic rule is defined as follows. First, a site, say the ith site, is selected randomly on a one-dimensional flat substrate with system size L. Then we consider a subset of the system, composed of $2r+1$ sites, the randomly selected site, and its $2r$ neighboring sites on its right and left sides, respectively, within distance r. Among the $2r+1$ sites, the surface is advanced at the site offering the largest binding, that is, the most occupied neighbors. If there is more than one site offering the largest binding, then the site closest to the ith one among them is taken and its height is increased by one. The case of $r=1$ is reduced to the original version of the WV model. If the distance r is regarded as diffusion length, then the diffusion constant would be related to the distance as $D \sim r^2$. We measure the density of islands in the submonolayer regime and the surface fluctuation width [20],

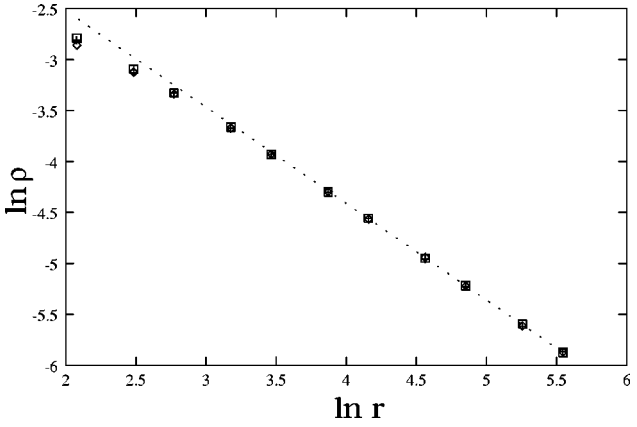

FIG. 1. Double logarithmic plot of the density of islands ρ versus the diffusion length r for the Wolf-Villain model in one dimension. The data are obtained from the system size $L=1000$ at different coverages, 0.3 ($\diamond$), 0.4 ($+$), and 0.5 ($\square$) from the bottom, and are averaged over 1000 runs. The dotted line with slope -0.95 is a guideline to the eyes.

$$W^2(L,t) = \left\langle \frac{1}{L}\sum_i (h_i - \overline{h})^2 \right\rangle, \tag{13}$$

with $\overline{h} = \Sigma_i h_i / L$ with varying diffusion length r. Note that the density of islands ρ is related to the mean separation ℓ as $\rho \sim \ell^{-1}$ in one dimension, and the damped oscillation in the step density occurs on the surface fluctuation width in the same way. In our simulations, we used the noise reduction method with the noise reduction parameter $m=5$ to get better data. However, we did not vary the parameter m in any case, so that the noise reduction parameter does not play the role of control parameter as considered in Refs. [21,22]. As shown in Fig. 1, the density of islands behaves as $\rho \sim r^{-0.95}$ for large values of r, suggesting that the exponent $\gamma \approx 0.48$. The numerical result is close to the theoretical result $\rho \sim r^{-1}$, which is obtained as follows. If we divide the system into the small cells with size $2r+1$, and a particle is deposited on one of the cells randomly, then one island would be formed on each cell, and the separation of islands in the system varies as $\ell \sim r$ for large r. Thus $\rho \sim r^{-1}$. However, the boundary between cells is virtual and could overlap in our simulations, but the correction due to fluctuation of the boundaries would be of higher order. Thus the density of islands would behave as $\rho \sim r^{-1}$ up to leading order, and $\gamma \approx 1/2$. We also count the number of nucleation events by ignoring monomers, and the result is the same as Fig. 1. Next, the surface fluctuation width W^2 exhibits a damped oscillation as shown in Fig. 2(a). The data for different diffusion lengths are well collapsed, when time is rescaled as $t/\tilde{t}$ with $\tilde{t} \sim r^{4/3}$ as shown in Fig. 2(b). The rescaled time suggests $\delta = 2/3$. Therefore, the ratio of the two exponents γ and δ is obtained as $\delta/\gamma \approx 4/3$, which is consistent with the theoretical prediction, Eq. (9) for $d=1$. At this stage, one may wonder how the modified WV model we consider is related to the CKPZ equation, because the original WV model is known as belonging to the EW universality class in the long time limit [23]. Thus, we measure the growth exponent β, defined by $W^2 \sim t^{2\beta}$, from the data of the case $r=8$ between $t=5000$ and 20 000 in Fig. 2. The growth exponent

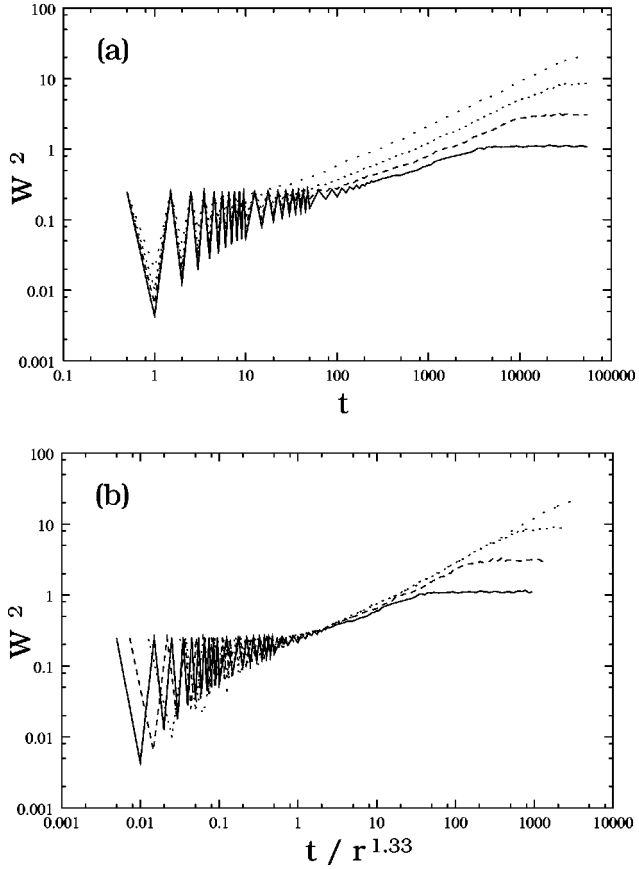

FIG. 2. Plot of the surface fluctuation width versus time t in double logarithmic scales for the case (a) and rescaled time $t/r^{4/3}$ for the case (b) in the Wolf-Villain model. The data are obtained from system size $L=1000$, and are averaged over 100 runs. The diffusion lengths $r=8$, 16, 24, and 32 were used from the top.

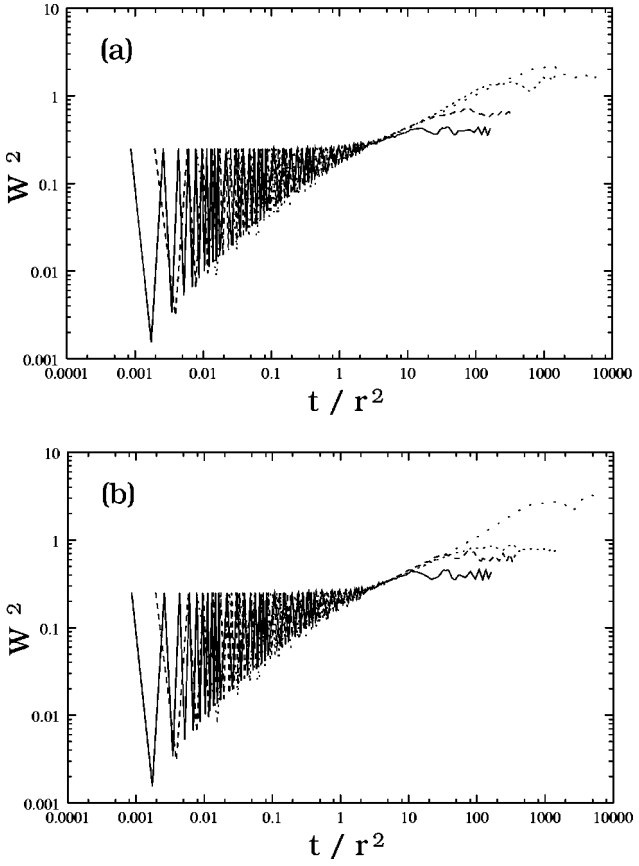

FIG. 3. Plot of the surface fluctuation width W^2 versus rescaled time t/r^2 in double logarithmic scales for the Family model. In the simulations, adatoms are allowed to run over humps for the case (a) and are not allowed to for the case (b). For both (a) and (b), various diffusion lengths, $r=4$, 8, 16, and 24 are used from the top. The data are obtained from system size $L=10^4$, and are averaged over 100 runs.

is measured as $2\beta\approx 0.62(1)$, which is close to the CKPZ value 2/3, rather than the EW value 1/2 or the value ≈ 0.73 for the original WV model without diffusion.

Next, we consider the modified Family model. In this case, we choose a site randomly, say the ith site, on a one-dimensional substrate with system size L, and consider a subset consisting of $2r+1$ sites as before. Among the $2r+1$ sites, the surface is advanced at the lowest site. If there is more than one site with the lowest height, the site closest to the ith site is taken. The case of $r=1$ is reduced to the original version of the Family model. We vary the distance r, and examine the density of islands and the damped oscillating behavior of the surface fluctuation width. The density of islands in the submonolayer regime behaves as the case of the WV model. For the surface fluctuation width, the data are well collapsed even for small values of r when time is rescaled as t/r^2 as shown in Fig. 3(a), implying that the damping time behaves as $\tilde{t}\sim r^2$. Thus the ratio between the exponents δ and γ is obtained as $\delta/\gamma\approx 2$ for the Family model, which is consistent with the theoretical prediction, Eq. (12). We also check the case involving a hump on the way to the lowest site in the subset. In Fig. 3(a), we considered the case that an adatom can run over the hump, and stay on the lowest site within the interval. In Fig. 3(b), the adatom cannot run over the hump, and is allowed to move only in a descending way or on the same height. We cannot find any difference between the two in the collapsing behavior, implying the assumption of the dephasing length $\tilde{\ell}$, within which the layer difference is at most one, is valid. Therefore, we conclude that the theoretical prediction is correct for the stochastic models, the WV model and the Family model, belonging to the CKPZ and EW universalities. However, the relevance of the theoretical prediction to MBE growth with downhill current is to be further investigated.

In summary, we have performed Monte Carlo simulations on the modified version of the Wolf-Villain model and the Family model involving the diffusion length r on one dimension. We measure the density of islands ρ in the submonolayer regime and the damping time $\tilde{t}$ for the oscillation of layer-by-layer growth with varying diffusion length. We obtained numerically that the exponents δ and γ describing the damping time and the density of islands are related to each other as $\delta/\gamma\approx 4/3$ for the Wolf-Villain model and $\delta/\gamma\approx 2$ for the Family model. This result is in excellent agreement with recent theoretical predictions.

One of the authors (B.K.) would like to thank D.E. Wolf for helpful discussions. This work is supported in part by the KOSEF (Grant No. 971-0207-025-2), and in part by the Korean Research Foundation (Grant No. 98-015-D00090).

[1] J. Villain and A. Pimpinelli, *Physique de la Croissance Cristalline* (Éditions Eyrolles et CEA, Paris, 1995).
[2] K. Thürmer, R. Koch, M. Weber, and K. H. Rieder, Phys. Rev. Lett. **75**, 1767 (1995); J. A. Stroscio, D. T. Pierce, M. Stiles, A. Zangwill, and L. M. Sander, *ibid.* **75**, 4246 (1995); J. E. Van Nostrand, S. J. Chey, M.-A. Hasan, D. G. Cahill, and J. E. Greene, *ibid.* **74**, 1127 (1995).
[3] G. Zinsmeister, Thin Solid Films **2**, 497 (1968); **4**, 363 (1969); **7**, 51 (1971).
[4] S. Stoyanov and D. Kashchiev, in *Current Topics in Material Science*, edited by E. Kaldis (North-Holland, Amsterdam, 1981), Vol. 7, pp. 69–141.
[5] J. A. Venables, G. D. Spiller, and M. Hannbrucken, Rep. Prog. Phys. **47**, 300 (1984).
[6] A. Pimpinelli, J. Villain, and D. E. Wolf, Phys. Rev. Lett. **69**, 985 (1992).
[7] D. E. Wolf, in *Scale Invariance, Interfaces, and Non-Equilibrium Dynamics*, edited by A. McKane *et al.* (Plenum, New York, 1995), pp. 215–248.
[8] M. Schroeder and D. E. Wolf, Phys. Rev. Lett. **74**, 2062 (1995); H. Jeong, B. Kahng, and D. E. Wolf, Physica A **245**, 355 (1997).
[9] H. Kallabis, P. L. Krapivsky, and D. E. Wolf, e-print cond-mat/9806140.
[10] 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).
[11] J. Villain, J. Phys. I **1**, 19 (1991).
[12] E. Bauer, Z. Kristallogr. **110**, 372 (1958).
[13] H. Kallabis, L. Brendel, J. Krug, and D. E. Wolf, Int. J. Mod. Phys. B **11**, 3621 (1997).
[14] Z.-W. Lai and S. Das Sarma, Phys. Rev. Lett. **66**, 2348 (1991).
[15] L.-H. Tang and T. Nattermann, Phys. Rev. Lett. **66**, 2899 (1991).
[16] S. F. Edwards and D. R. Wilkinson, Proc. R. Soc. London, Ser. A **381**, 17 (1982).
[17] G. Ehrlich, Surf. Sci. **331**, 865 (1995).
[18] F. Family and T. Vicsek, *Dynamics of Fractal Surfaces* (World Scientific, Singapore 1991).
[19] D. E. Wolf and J. Villain, Europhys. Lett. **13**, 389 (1990).
[20] F. Family, J. Phys. A **19**, L441 (1986).
[21] P. Punyindu and S. Das Sarma, Phys. Rev. E **57**, R4863 (1998).
[22] L. Brendel, H. Kallabis, and D. E. Wolf, Phys. Rev. E **58**, 664 (1998).
[23] P. Smilauer and M. Kotrla, Phys. Rev. B **49**, 5769 (1994).