Layer-by-layer epitaxy in limited mobility nonequilibrium models of surface growth

P. Punyindu Chatraphorn[1,2] and S. Das Sarma[1]
[1]*Condensed Matter Theory Center, Department of Physics, University of Maryland, College Park, Maryland 20742-4111*
[2]*Department of Physics, Faculty of Science, Chulalongkorn University, Bangkok 10330, Thailand*
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We study, using noise-reduction techniques, layer-by-layer epitaxial growth in limited mobility solid-on-solid nonequilibrium surface growth models, which have been introduced in the context of kinetic surface roughening in ideal molecular beam epitaxy. Multiple hit noise reduction and long surface diffusion length lead to qualitatively similar layer-by-layer epitaxy in $(1+1)$- and $(2+1)$-dimensional limited mobility growth simulations. We discuss the dynamic scaling characteristics connecting the transient layer-by-layer growth regime with the asymptotic kinetically rough growth regime.

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I. INTRODUCTION

Thin film growth, under solid-on-solid epitaxial conditions, from the vacuum vapor deposition of an atomic or a molecular beam (the so-called molecular beam epitaxy or MBE) is an important technological process used extensively to produce high quality thin films with smooth and flat surfaces and interfaces. It is also a growth process of considerable fundamental significance [1–6] in the statistical mechanics of nonequilibrium phenomena because MBE [at least in its ideal form [7], with no evaporation and vacancy or overhang formation at the growth front] in principle represents [7,8] a universality class of nonlinear surface growth outside the generic Kardar-Parisi-Zhang (KPZ) universality [9]. A great deal of attention has, therefore, focused over the last ten years on the statistical properties of kinetically rough (and, in principle, generically scale invariant) surface growth in low temperature (room temperature or below) MBE, following the suggestions [7,8,10–12] of the possible importance of ideal MBE in defining growth universality classes in kinetic surface roughening [1–6]. We note that the conserved surface current nature [7,8] of ideal MBE growth (i.e., solid-on-solid growth with no evaporation and vacancy or overhang formation) rules out a KPZ description of its growth dynamics.

It is interesting, perhaps even ironic, that MBE growth has played such a central role [1–6] in kinetic surface roughening phenomena because the primary materials science impetus for MBE growth is obviously to avoid kinetic roughening, as completely as possible in order to produce smooth and flat thin films of high surface quality with minimal amount of surface roughness. From the materials science perspective of producing high quality smooth (i.e., manifestly nonrough) thin films, therefore, MBE is typically carried out at elevated temperatures ($\sim$500–1000 K), where fast surface diffusion enables one to produce smooth thin films with very little surface roughness. Smooth MBE growth, as opposed to kinetically rough (low temperature) growth, is characterized by layer-by-layer growth oscillations [13,14], where each layer of the growing thin film (on a singular high symmetry substrate) essentially fills up completely before the next layer deposition begins (on the other hand, kinetically rough growth is, by definition, multilayer as many layers are partially filled at the growth front producing increasing surface roughness with *no* layer-by-layer oscillations), consequently the surface morphology and associated properties oscillate as growth proceeds. In the ideal layer-by-layer growth mode, therefore, the interface width (W, the root mean square fluctuation in the interface height) of the growing film oscillates (nominally between 0 and 1, as measured in lattice units, indicating an empty or a filled layer) whereas in the kinetically rough growth mode W increases monotonically as a power law in the average film thickness ($\langle h \rangle$). Layer-by-layer epitaxy is, however, an initial transient growth regime that eventually crosses over to the asymptotic kinetically rough growth regime as the shot noise intrinsic in the incident deposition beam fluctuations always wins out to damp the layer-by-layer oscillations, and at long enough time scales (and for large enough lateral system sizes) statistically scale invariant kinetically rough growth would always emerge. (In fact, the noise associated with the stochastic diffusion process also contributes to kinetic roughening, but the shot noise associated with the incident beam fluctuations is the most *relevant* roughening mechanism.) This is also the experimental observation: layer-by-layer growth oscillations, as studied for example through RHEED intensity oscillations monitoring the dynamical surface evolution [13,14], eventually always damp out as the stochastic deposition shot noise associated with incident particle beam fluctuations leads to kinetically rough multilayer growth after some characteristic time t_c. The damping time t_c, beyond which layer-by-layer growth dies out, depends on the growth temperature (which controls the surface diffusion rate), and is in general larger for higher temperatures because longer diffusion lengths at higher temperatures enhance layer-by-layer growth. (There is actually a complication, arising from the unaviodable vicinality in the starting substrate that can never really be precisely a high symmetry singular plane in real growth where layer-by-layer oscillations tend to disappear at both high and low growth temperatures—the low temperature behavior is from the multilayer kinetically rough growth as discussed above, but the high temperature disappearance arises from the so-called step flow growth mode that is caused by the very fast surface diffusion of deposited atoms at high temperatures leading to their moving directly to step edges, which must be present in any real substrate due to vicinality

or miscut, without any layer-by-layer growth oscillations; we neglect considerations of such a step flow growth mode in this paper assuming all growth to be occuring strictly on singular high symmetry substrates.)

The ideal MBE growth (on flat singular high symmetry substrates) can be thought of as composed of two regimes—an early time ($t<t_c$) transient (fast diffusion driven) regime of layer-by-layer growth followed by the asymptotic ($t>t_c$) kinetically rough (deposition beam fluctuation driven) growth regime (with no layer-by-layer oscillations) characterized by power law evolution [1–12] in surface roughness. At low enough temperatures, when surface diffusion is extremely slow, t_c could be less than the time it takes to grow one monolayer of deposit on the average, and in that situation layer-by-layer transient growth regime is invisible with the kinetically rough growth regime being dominant essentially right from the beginning. Conversely, sufficiently high temperature growth on a small substrate could continue in the layer-by-layer mode for a very long time although some damping of the growth oscillations is inevitable with time as distant spatial regions on the substrate must lose coherence due to the inherent shot noise fluctuations associated with the discrete deposition process in the incident beam. Thus layer-by-layer growth us purely a ''finite size'' (both spatially and temporally) transient phenomenon—if the substrate is made sufficiently large and/or if one waits for sufficiently long growth time, layer-by-layer epitaxy must necessarily cross over to kinetically rough growth. It is important to emphasize, however, that the surface diffusion length is typically an exponentially activated function of growth temperature, and therefore, a small change in temperature could cause a sharp large change in the growth mode (layer-by-layer to rough and vice versa depending on whether the growth temperature is decreased or increased) for a given substrate, leading to the empirical concept [15] of an epitaxial growth temperature T_c with growth being layer-by-layer (rough) for $T>T_c$ ($T<T_c$)—clearly T_c is a loosely defined concept because it must be a weak (sublogarithmic) function of the effective substrate (or the terrace) size for a given material [15]. In general, ''good'' MBE growth aiming toward producing high quality smooth epitaxial thin flims is carried out at the highest possible growth temperature (within the constraint that evaporation or desorption from the growth front should be negligible so that the growth temperature cannot be arbitrarily high) so as to make atomic mobility at the growth front to be very high leading to large ''surface diffusion length'' l. Here l is taken to be the linear size over which the surface is smooth due to atomic diffusion. Assuming the deposition process to be a random Poisson process it is then easy to see that the typical surface roughness over terraces of size l would only grow as $\sqrt{\langle h\rangle}/l$, where $\langle h\rangle$ is the thickness of the grown film (and we measure all lengths in lattice units). Thus for large l, one would have to grow a very thick film of thickness l^2 before the surface roughness reaches even one monolayer fluctuation. One can, therefore, grow MBE thin films of very high smoothness and quality, without worrying at all about the kinetic surface roughening by properly adjusting the growth temperature [15] to make l large.

Layer-by-layer MBE growth has been extensively studied [16–20] in the literature using computer simulations of MBE growth through the stochastic (or kinetic) Monte Carlo simulations, with the atomistic diffusion at the growth front assumed to be controlled by stochastic activated hopping process with the hopping rate determined by a local coordination number dependent activated Arrhenius hopping. Such activated diffusion Arrhenius hopping simulations (sometimes also referred to as ''full diffusion'' simulations to differentiate them from ''limited mobility'' growth models which are our main interest in this paper) involve continuous (possible) hopping of all surface atoms according to their local bonding configurations (which determine the activation energy for the hopping process). Such full diffusion simulations are obviously not well designed to study the kinetic roughening universality class of MBE growth because they are extremely time consuming and cannot really be carried out for large systems [particularly in the physically relevant $(2+1)$ dimensions] for long times, an essential requirement for ascertaining the asymptotic universality class of a growth model. Although there are some notable exceptions [21–23], the full diffusion Arrhenius activated kinetic Monte Carlo simulations of MBE growth have not been used with particular success for understanding statistical scale invariance properties of kinetic surface roughening. Instead, important insights into the MBE universality class of kinetic surface roughening have come primarily from nonequilibrium limited mobility growth models—mainly the so-called Wolf-Villain [11] (WV) and the Das Sarma-Tamborenea [10] (DT) model—which were introduced specifically for the elucidation of the MBE growth universality.

In this paper, we study the DT and the WV model (we emphasize that WV and DT models, in spite of their close similarity in growth rules, belong [24] to different asymptotic universality classes in both $(1+1)$ and $(2+1)$ dimensions although their preasymptotic scaling behavior is very similar which has led to considerable confusion in the literature) in the complementary layer-by-layer growth regime rather than the kinetically rough growth regime that motivated the introduction of these models. We mention in this context that some $(1+1)$-dimensional studies of WV and DT models in the layer-by-layer growth regime have recently been reported in the literature [25,26]. Our results, where applicable, agree with these earlier works [25,26], but our focus in this paper is $(2+1)$-dimensional growth and the effect of long surface diffusion length in $(1+1)$-dimensional growth, neither one of which has earlier been studied.

In limited mobility growth models (the models and growth rules used in this paper are described in Sec. II of the paper—see, for example, Fig. 1), in sharp contrast to full diffusion MBE growth simulations, the goal is to suppress crossover and transient effects as much as possible (so as to efficiently reach the asymptotic kinetic surface roughening regime) and as such only the most recently deposited atom is allowed to diffuse or relax instantaneously to the appropriate incorporation site following the mobility rules of the specific model. This allows suppression of crossover effects invariably present in the full diffusion simulations arising from many different diffusion rates corresponding to many differ-

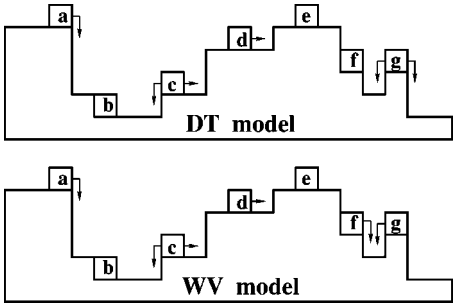

FIG. 1. Schematic plots showing the diffusion rules for the DT and WV models when the diffusion length l and noise-reduction factor m are both 1.

ent possibilities for local bonding configurations—the only time scale in the limited mobility growth models being the deposition rate, which defines the time unit for the problem. From now on we take the time unit (sometimes referred to as a ''second'') as the time to deposit one monolayer on the average. Thus the growth time in this paper also defines $\langle h \rangle$—the average thickness of the deposited film measured in units of monolayers or the lattice constant, which we take to be the unit of length throughout. (With no loss of generality we choose the lattice constant to be the same along the substrate and the growth directions.)

The limited mobility growth models [10,11] are by construction strongly dominated by the deposition shot noise because the goal is to study the scale invariant kinetic surface roughening behavior. This is particularly true in the original versions of the growth model where the surface diffusion length is choosen to be unity, $l=1$, i.e., the deposited atoms are allowed to move only to the nearest neighbor incorporation sites around the deposition site. The original DT and WV models, therefore, did not exhibit, by design, any layer-by-layer growth oscillations since the smoothing or the healing distance (l) was just one lattice unit. In order to manifest layer-by-layer epitaxy in limited mobility growth models one must, therefore, suppress the shot noise associated with the incident beam fluctuations.

In this paper, we accomplish the noise suppression by two alternative techniques: The ''multiple hit'' noise-reduction technique [27–31] and the ''long surface diffusion length ($l>1$)'' noise-reduction technique [21]. These techniques, described in Sec. II of the paper, give rise to layer-by-layer growth (as monitored by an oscillatory surface roughness, i.e., $W(t)$ showing oscillations as a function of growth time t) in the limited mobility growth models as described in Sec. III of the paper.

The rest of this paper is organized as follows. In Sec. II we describe the limited mobility growth models and the noise reduction technique(s) employed by us. We also provide some theoretical background for our analysis of the simulation results. In Sec. III we present and describe our numerical simulation results for layer-by-layer epitaxial growth in DT and WV models. We also discuss in Sec. III various (approximate) scaling properties of our simulated layer-by-layer epitaxial growth results. We conclude in Sec. IV with a general discussion of our results making connections with some of the existing results in the literature and pointing out possible future directions as well.

II. MODELS, THEORY, AND BACKGROUND

The DT [10] and WV [11] models used in our simulations are shown in Fig. 1. We carry out growth simulations in both $(1+1)$ dimensions and $(2+1)$ dimensions (on 100 high symmetry substrates). Particles are dropped, obeying solid on solid constraint, sequentially (one by one) and randomly with an average rate of N per second, where $N=L$ for d $=(1+1)$ and $N=L^2$ for d$=(2+1)$, on a substrate of lateral linear size of L lattice units. We measure length in lattice units and time in inverse deposition rate (i.e., in units of monolayer filling since one average monolayer of deposition occures every ''second''). Each deposited atom is allowed to ''diffuse'' instantaneously to its incorporation site following the mobility rules of the specific model. The diffusion rules in the DT model are that a deposited atom can move only if it has no lateral nearest neighbor in the same layer (if it does, then the atom is incorporated at the deposition site)—if the deposition site has no lateral nearest neighbor then the incident atom may move instantaneously to a neighboring empty site (within a lateral diffusion length of l, where $l=1$ in the original model and in most existing simulations) provided the final incorporation site has a higher lateral coordination number (i.e., one or higher) than the deposition site. If several neighboring sites satisfy the diffusion rule then the atom will move randomly to any one of them with equal probability. The rules for the WV model are superficially similar to the DT rules: In the WV model all deposited atoms (and *not* just the ones with no lateral bonds) can, in principle, move provided they can increase their local lateral bonding and the deposited atom always moves to the site with the maximum local bonding environment. In both models, the deposited atom is incorporated at the deposition site if it cannot satisfy the diffusion rules (i.e., no sites with higher coordination available) within the diffusion length.

Both DT and WV models have been extensively studied in the literature (mostly within nearest neighbor, $l=1$, diffusion rules) in the context of their kinetic surface roughening universality classes. Recently, layer-by-layer epitaxy in the WV [25,26,30] and the DT [25] model have been investigated in $(1+1)$ dimensions using the multiple hit noise-reduction technique. The very first simulational observation of layer-by-layer growth in a limited mobility growth model was reported in the DT model in Ref. [21], where it was studied in $1+1$ dimensions using a long ($l>1$) surface diffusion length, but no details were investigated. We emphasize that the usual $l=1$ limited mobility growth model does *not* exhibit any layer-by-layer epitaxy by definition, and manifests kinetic roughening right from the beginning since for $l=1$ the layer-by-layer epitaxy regime is restricted to less than one monolayer coverage, i.e., in the standard limited mobility growth models [10–12] the layer-by-layer epitaxy regime does not exist.

To obtain a layer-by-layer growth regime in the DT and the WV model we use two distinct techniques to suppress noise and enhance diffusion, which enable our growth simulations to manifest strong layer-by-layer growth oscillations

before crossing over to the kinetic surface roughening regime with pure multilayer growth. One technique, referred to as the noise-reduction technique, has also been used by others [25–30] to produce layer-by-layer growth in various models, e.g., Eden model [28], single step model [29], and the WV and the DT model in $(1+1)$ dimensions [25,26,30]. We have earlier used this technique [31] to suppress corrections to scaling in the asymptotic kinetically rough surface growth regime in the DT and the WV model in order to accurately determine the dynamic scaling exponents and the associated growth universality class. In the noise-reduction technique, characterized by an integer number m (the usual growth model without any noise reduction is an $m=1$ model), a counter is put on each surface site and each discrete deposition event on a site advances the counter by unity. A deposition event at a particular site is accepted only when the counter reaches a predetermined number $m>1$. Thus, this technique is the multiple hit noise-reduction technique since $m(>1)$ deposition hits on a site are needed for a true deposition. After m hits on a site (i.e., after the acceptance of a deposition event) the counter at that particular site is set back to zero, and the whole multiple hit process begins all over again. The multiple hit noise reduction technique is a coarse-graining procedure which suppresses the deposition shot noise, and the noise reduction is enhanced for larger values of m. The second technique applied by us for obtaining layer-by-layer growth oscillations in limited mobility growth models is to use long surface diffusion lengths ($l>1$) in the growth simulations. Obviously long diffusion lengths enhance the layer-by-layer growth regime, and in particular, for $l>L$ i.e., the surface diffusion length exceeding the system size, the layer-by-layer growth may persist essentially indefinitely since each deposited atom may always be able to seek out a desired epitaxial site for incorporation. In some sense the multiple hit noise-reduction parameter m is equivalent to the dimensionless diffusion length parameter l/L because large values of both tend to enhance the layer-by-layer growth regime. In Sec. III, where we present our simulation results, we will see the precise nature of this correspondence between m and l/L in our two methods of obtaining layer-by-layer epitaxy in the DT and the WV model.

The central quantity of interest in layer-by-layer epitaxial growth is the characteristic time t_c at which layer-by-layer growth dies out, i.e., for the deposited average film thickness larger than $\langle t_c \rangle$, measured in lattice units or in monolayers, there are no discernible layer-by-layer growth oscillations. It has been found in earlier numerical simulations of layer-by-layer growth in a variety of contexts that t_c obeys an approximate scaling relation with the coarse-graining parameter m (or l/L as the case may be), and we are interested in investigating whether the following scaling relations hold in the limited mobility growth models

$$t_c \sim \begin{cases} m^{\mu} & \text{for the noise reduction method,} \\ (l/L)^{\delta} & \text{for the long diffusion length method.} \end{cases} \tag{1}$$

If such scaling relations do hold in our simulations we are interested in obtaining the relationship, if any, between the exponents δ and μ. Note that t_c depends on l/L (rather than just l) on dimensional grounds—in fact, the layer-by-layer growth mode is purely a finite size transient phenomenon; dynamical growth is, by definition, kinetically rough [32] in either the infinite size ($L\to\infty$) or the infinite time ($t\to\infty$) limit.

It is in fact fairly straightforward to obtain a relationship between the noise-reduction parameter m and the surface diffusion length parameter l/L using only dimensional arguments. In particular, we note that for a d'-dimensional substrate [$d'=2$ for real surfaces and $d'=1$ for the $(1+1)$-dimensional growth] one obtains a simple relationship between μ and δ by noting that a surface diffusion length l corresponds to $m\sim l^{d'}$ since there are $l^{d'}$ available sites for a particular deposited particle to incorporate at. This immediately leads to

$$\delta = \mu d'. \tag{2}$$

Within the limited accuracy of our growth simulations (see, Sec. III for the results), we find the scaling relation defined by Eq. (2) to be valid. We note that later in this paper, where we compare our simulation results with existing theories [33,34], it is the dependence of the damping time t_c on the diffusion length l that would be compared, the relationship between the noise-reduction parameter m and t_c (i.e., the exponent μ) would then follow using Eqs. (1) and (2).

One secondary theoretical goal of our work is to investigate the extent to which layer-by-layer growth oscillations obey scaling with respect to growth time (or equivalently, the average film thickness). Using Eq. (1) as the theoretical ansatz one can ask whether the surface roughness W, defined as the ensemble averaged (over many growth simulations) root mean square fluctuation in the interface height, which is oscillatory (and $W<1$ implying little roughness) in the layer-by-layer growth regime is a general scaling function of the layer-by-layer growth parameter m or l/L through a dependence of the form

$$W(t) \sim f_m(t/m^{\mu}) \quad \text{or} \quad f_l(t/(l/L)^{\delta}). \tag{3}$$

If a scaling form such as Eq. (3) holds in the layer-by-layer growth regime, then Eq. (1) for t_c trivially follows from it—t_c being the value of time where layer-by-layer oscillations cease to exist. We could go further in our scaling analyses and ask whether the scaling defined by Eq. (3) continues to hold (perhaps approximately) well beyond ($t>t_c$) the layer-by-layer growth regime establishing an approximate scaling relationship between the layer-by-layer growth regime ($t<t_c$) and the kinetically rough growth regime ($t>t_c$). Our results presented in Sec. III indicate that such an approximate scaling relation does indeed exist between the layer-by-layer growth regime and the kinetically rough growth regime.

Finally, we note that there have been recent attempts [33,34] at developing a theory for layer-by-layer growth oscillations starting from continuum growth equations underlying the coarse-grained long wavelength behavior of MBE growth. A simple dimensional argument [33], later followed

up [34] by a renormalization group approach, leads to the following results for MBE growth:

$$\mu = \begin{cases} 4/3 & \text{for } d' = 1, \\ 2 & \text{for } d' = 2. \end{cases} \tag{4}$$

The exponents defined by Eq. (4) correspond to the so-called [1–6] conserved fourth order nonlinear growth equation or the nonlinear MBE growth equation [which is sometimes also referred to as the conserved KPZ equation with nonconserved noise or the Lai-Das Sarma-Villain [7,8] (LDV) equation] which is given by

$$\frac{\partial h}{\partial t} = -\nu_4 \nabla^4 h + \lambda_{22} \nabla^2 (\nabla h)^2 + \eta, \tag{5}$$

where $h(\mathbf{x},t)$ is the dynamical height fluctuation variable relative to the average interface $\langle h \rangle$ at the substrate site $\mathbf{x}$ (with $\mathbf{x}$ is the lateral substrate coordinate), $\nabla \equiv \partial/\partial \mathbf{x}$ is the gradient operator along the surface, η is the deposition shot noise (which causes the kinetic surface roughening), and ν_4, λ_{22} are coefficients which in general depend on surface diffusion rate, deposition rate, etc. Since the continuum description of the DT and the WV model are actually quite complex [23,31,35–38], and are likely to be different in different dimensions [38], it is by no means clear that the exponents [given in Eq. (4)] corresponding to Eq. (5) apply without any qualifications to the DT and the WV model (as will be discussed in Sec. III, where we present our numerical simulations). We, therefore, also provide below the exponent μ for the linear second order Edwards-Wilkinson (EW) growth equation [39] which applies to the limited mobility Family (FM) growth model [40] and may also have significant relevance to the DT and the WV model [31,38,41]:

$$\mu = \begin{cases} 2 & \text{for } d' = 1, \\ \infty & \text{for } d' = 2. \end{cases} \tag{6}$$

The EW equation, whose layer-by-layer growth exponent in $(1+1)$ and $(2+1)$ dimensions is given in Eq. (6), is the following:

$$\frac{\partial h}{\partial t} = \nu_2 \nabla^2 h + \eta. \tag{7}$$

We note here that both sets of exponent values given by Eqs. (4) and (6) will be relevant in our discussion of our simulation results to be presented in the following section.

III. RESULTS AND DISCUSSIONS

We now present our $(1+1)$ (i.e., $d'=1$) and $(2+1)$ (i.e., $d'=2$) dimensional layer-by-layer growth simulation results for the discrete limited mobility DT and WV models in Figs. 2–6. In Figs. 2–4 we present $(1+1)$-dimensional simulation results whereas Figs. 5 and 6 give $(2+1)$-dimensional simulational results for the two growth models. In each figure (to be described below) the panel (a) gives the simulated $W(t)$ as a function of growth time t for various values of the noise-reduction parameter m or the surface diffusion length l with the layer-by-layer oscillations manifestly obvious for larger values of m and l. (The original DT and WV models correspond to the simulation with $m=1$ and $l=1$, which has no layer-by-layer oscillations by construction.) In panel (b) of each figure we demonstrate our best computed scaling collapse of the $W-t$ plots [shown in panel (a)] for various values of m or l with a suitable scaling of time t to t/m^{μ} or $t/(l/L)^{\delta}$ as the case may be. For each scaling collapse in (b) we try various different values of the exponent μ or δ ro obtain the best statistical scaling in the simulated data. The system size used in each simulation is indicated in the corresponding figures and the captions. Here we should emphasize that the results shown in this paper represent only a typical fraction of our extensive DT and WV layer-by-layer growth simulations. The representative results presented here are of course in complete agreement with the full set of our simulation data, and our conclusion is based on a very large set of simulation results and not just on the results presented in this paper.

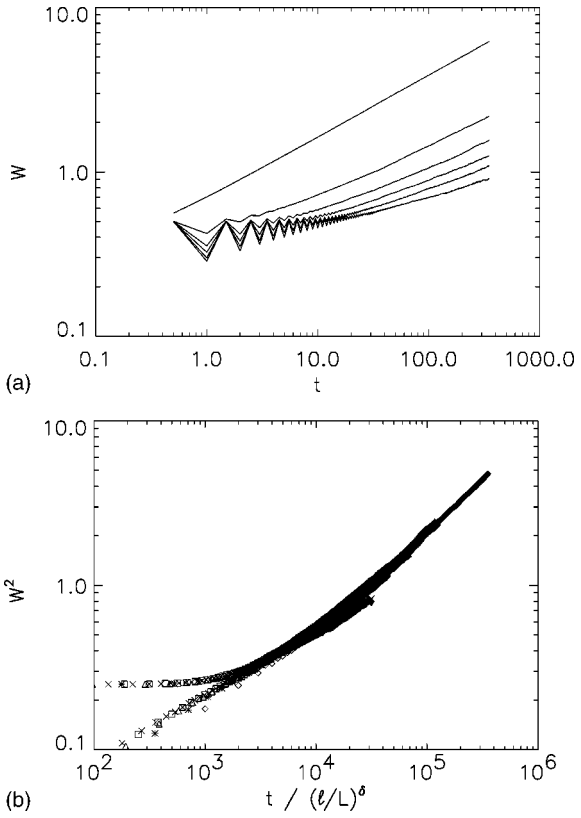

FIG. 2. (a) $W-t$ oscillations for $(1+1)$-DT ($L=1000$) with $m=1$ and $l=10$, 20, 30, 40, 50 (top to bottom); (b) scaling plot of systems in (a) using $\delta=1.5$. The interface width is measured in units of monolayers or lattice spacing and time is measured in units of number of deposited monolayers (i.e., average height of the surface).

In Fig. 2 we show our $(1+1)$-dimensional DT layer-by-layer growth simulation results for finite surface diffusion length l ($m=1, l>1$) for $l=10, 20, 30, 40, 50$. The layer-by-layer oscillations are visually obvious in Fig. 2(a)—the damping time t_c increases from $t_c \sim 10$ for $l=10$ to roughly

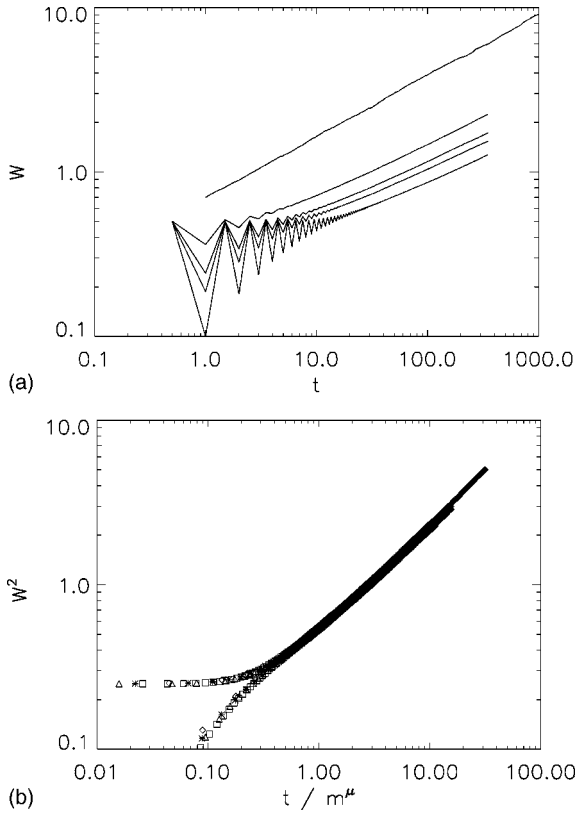

FIG. 3. (a) $W-t$ oscillations for $(1+1)$-DT ($L=1000$) with $l=1$ and $m=1$, 5, 8, 10, 15 (top to bottom); (b) scaling plot of systems in (a) using $\mu=1.5$. The units are as explained in the caption for Fig. 2.

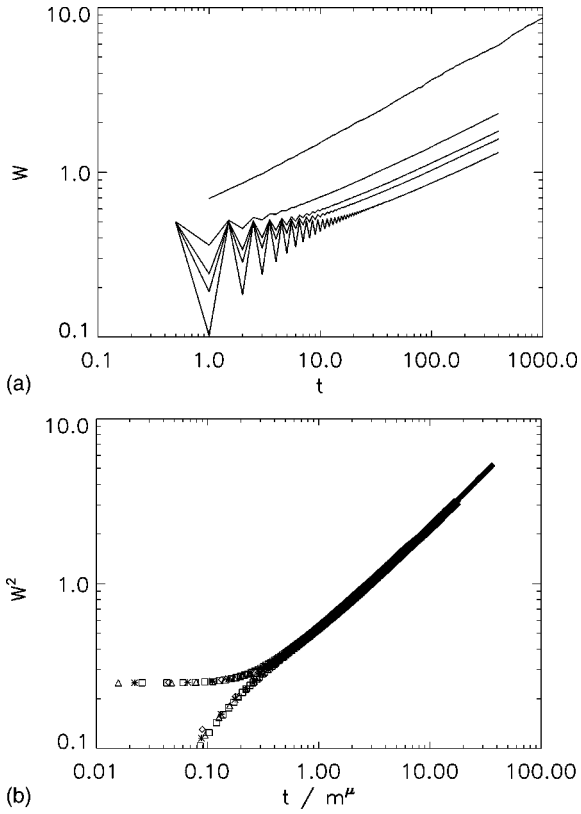

FIG. 4. (a) $W-t$ oscillations for $(1+1)$-WV ($L=1000$) with $l=1$ and $m=1$, 5, 8, 10, 15 (top to bottom); (b) scaling plot of systems in (a) using $\mu=1.5$. The units are as explained in the caption for Fig. 2.

$t_c \sim 50$ for $l=50$. In general, the magnitude of the oscillations decays exponentially with increasing time, and for $t>t_c$ we can only discern the power law increase of $W \sim t^{\beta}$, where β is the growth exponent in the model. In Fig. 2(b) we show our scaling collapse of the $W(t)$ data from Fig. 2(a), leading to the exponent value $\delta \approx 1.5$. Thus, $t_c \sim (l/L)^{1.5}$ in $d'=1$ DT model. We note that the scaling collapse in the kinetically rough growth regime (i.e., for $t>t_c$) is not excellent, but it is remarkable that the exponent δ that is meaningfully defined only in the layer-by-layer growth regime continues to provide an approximate reasonable description of the kinetically rough growth regime.

In Fig. 3 we show our $(1+1)$-dimensional DT model layer-by-layer growth oscillations [Fig. 3(a)] for the multiple hit noise-reduction technique ($l=1, m>1$) for different values of the noise-reduction factor m. In Fig. 3(b) we show the scaling collapse of the $W-t$ data in Fig. 3(a) for various m values. The scaling is excellent with an exponent $\mu=1.5$. Thus, $t_c \sim m^{1.5}$ in $d'=1$ DT model.

In Fig. 4 we depict our noise-reduced layer-by-layer growth oscillations [Fig. 4(a)] and the corresponding scaling collapse of the data for various values of m (with $l=1$) in the $(1+1)$-dimensional WV model simulations. Again, the scaling exponent μ is found to be $\mu=1.5$ for the best scaling collapse, indicating $t_c \sim m^{1.5}$ in both DT and WV noise-reduced models in $d'=1$. The finding of the apparent same exponent value $\mu=1.5$ in both DT and WV models in $(1+1)$-dimensional growth is consistent with the fact that the effective growth exponent β (obtained by plotting $\ln W$ against $\ln t$ in the simulated results) is almost identical in the two models: From the slope of the log-log plot in Fig. 2(b) we obtain $\beta \approx 0.338$ for the $d'=1$ DT model whereas from the slope of the log-log plot in Fig. 3(b) we obtain $\beta \approx 0.339$ for the $d'=1$ WV model. Thus within the effective time and length scales of our simulations the two models (DT and WV) have essentially the same effective dynamical universality class, which is consistent with the fact that they have the same effective exponent $\mu=1.5$ in $d'=1$. The fact that the asymptotic universality classes of the DT and WV models are different even in $d'=1$ dimension [31,38,41] does not seem to affect the effective values of μ we obtain in our simulations.

Before presenting our $(2+1)$-dimensional simulation results in Figs. 5–7 we first discuss the exponent values δ and μ, all of which have turned out to be approximately 1.5 in the $(1+1)$-dimensional DT and WV layer-by-layer epitaxial simulations. (We do not show here $l>1$, $m=1$ simulation results for the $d'=1$ WV model because they are very similar to those shown in Figs. 2–4 with the same exponent $\mu \approx 1.5$.) First we note that the exponent value is very close to (but somewhat above) the theoretically ''expected'' exponent $\mu=4/3=1.33$ predicted in Refs. [33,34] assuming that the continuum growth equation for these discrete growth models is that given in Eq. (5). We also note that the expected rela-

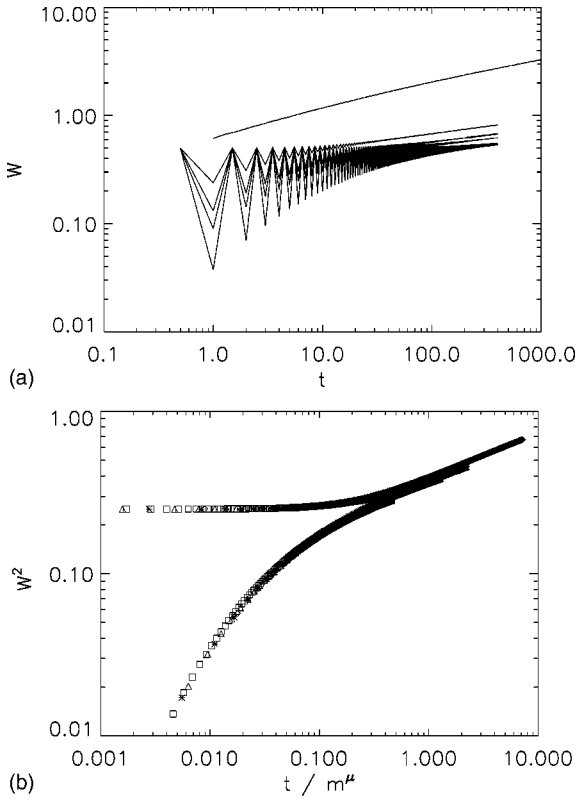

FIG. 5. (a) $W-t$ oscillations for $(2+1)$-DT ($L=10^3\times10^3$) with $l=1$ and $m=1$, 5, 8, 10, 15 (top to bottom); (b) scaling plot of systems in (a) using $\mu=2.5$. The units are as explained in the caption for Fig. 2.

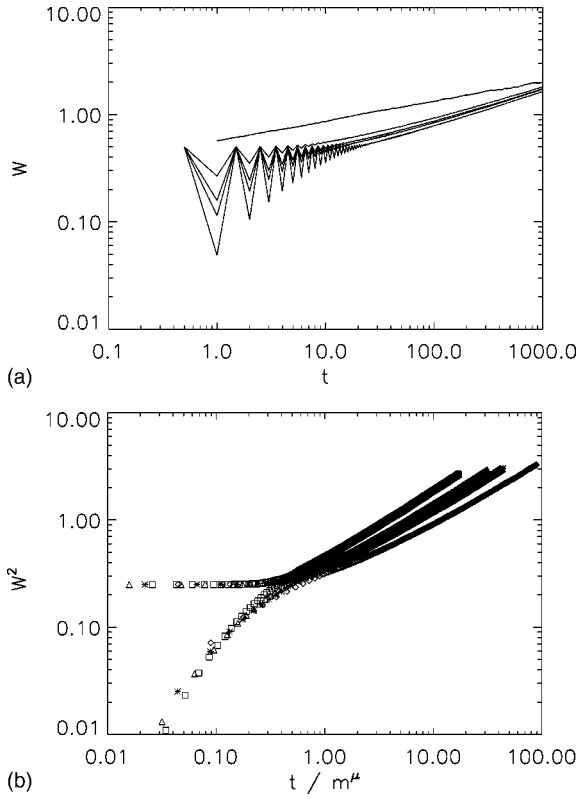

FIG. 6. (a) $W-t$ oscillations for $(2+1)$-WV ($L=100\times100$) with $l=1$ and $m=1$, 5, 8, 10, 15 (top to bottom); (b) scaling plot of systems in (a) using $\mu=1.5$. The units are as explained in the caption for Fig. 2.

tionship between μ and δ, namely $\delta=\mu d'$ that becomes $\delta=\mu$ in $d'=1$, is obeyed by our simulation results. We also add that the measured exponent $\mu\simeq1.5$ is consistent with other findings in the literature [25,26,30]. The cause for our calculated $\mu\,(\simeq1.5)$ to be somewhat (by roughly 10%) higher than the theoretical value ($\mu=4/3$) is not very clear at this stage. We do not, however, believe this discrepancy to be particularly significant because of a number of reasons: (i) our scaling collapse are in fact not inconsistent with an exponent of 1.33 although an exponent value of 1.5 is definitely a statistically better fit for our scaling collapse; (ii) it is quite conceivable that there are some systematic finite size and finite time corrections to scaling (which are known to be very important in DT and WV models); (iii) finally, at least in the WV model that is definitely known [31,38,41] to asymptotically belong to the EW universality class, it is possible that our simulated exponent $\mu\simeq1.5$ is showing some effects of the asymptotic universality class since the theoretically expected [33,34] μ for the linear EW equation [our Eq. (7)] is $\mu=2$ in $d'=1$ dimensions.

Our $d'=2$ dimensional noise-reduced ($m>1,l=1$) results for the DT, the WV, and the F model are shown in Figs. 5–7, respectively. These $(2+1)$-dimensional layer-by-layer growth results (as well as the results shown in Fig. 2) in limited mobility models are completely new and do not exist anywhere in the literature. We carried out our $(2+1)$ layer-by-layer growth simulations using only the noise-reduction technique since our $(1+1)$-dimensional results (compare Figs. 2 and 3) indicate that the finite diffusion length and the multiple hit noise-reduction techniques are essentially equivalent and the $(2+1)$-dimensional simulations with $l>1$ are particularly cumbersome to carry out. Our $d'=2$ layer-by-layer epitaxy simulations seem to have produced a few surprising results as discussed below.

In Fig. 5 we present our $d'=2$ layer-by-layer growth simulations in the DT model using the noise-reduction technique. The results are depicted in the same manner as in the $d'=1$ case shown in Fig. 3—in particular, Fig. 5(a) shows the actual layer-by-layer growth oscillations for various values of m whereas Fig. 5(b) shows the scaling collapse. It is obvious from comparing Figs. 3 and 5 that the layer-by-layer growth regime is substantially stronger in $d'=2$ case compared with the $d'=1$ case, which is consistent with a much larger value of the damping exponent $\mu\simeq2.5$ (compared with 1.5 in $d'=1$) in Fig. 5(b) in the $d'=2$ system. Our calculated damping exponent $\mu\simeq2.5$ for the $d'=2$ dimensional DT model is substabtially (by 25%) higher than the corresponding theoretical prediction [33,34] of $\mu=2$ for the LDV equation [Eq. (5)], which is generally thought to be the continuum description for MBE growth. This large discrepancy between our simulated damping exponent ($\mu\simeq2.5$) and the theoretical damping exponent ($\mu=2$) corresponding to Eq. (5) in $d'=2$ dimensions may be a real effect, arising from the recently discovered fact [42] that the DT model in $(2+1)$ dimensions has actually a very small (but nonzero) EW ∇^2h term in its continuum description in contrast to the

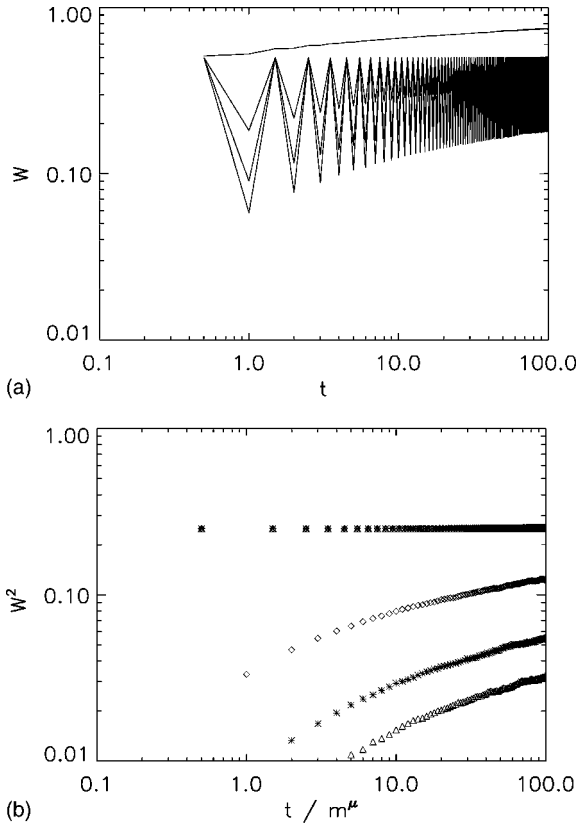

FIG. 7. (a) $W-t$ oscillations for $(2+1)$ Family ($L=100\times100$) with $l=1$ and $m=1$, 5, 8, 10 (top to bottom), (b) scaling plot of systems in (a) using $\mu=0.0$. The units are as explained in the caption for Fig. 2.

$(1+1)$-dimensional DT model, where the EW $\nabla^2 h$ term strictly vanishes [i.e., $\nu_2=0$ in Eq. (7)] by virtue of a topological symmetry [6] in the DT model. Thus the DT model in $(2+1)$ dimensions may actually asymptotically belong (but with a very small value of ν_2) to the EW universality class which according to Eq. (6) has an infinite value of the damping exponent μ. It is, therefore, possible that the value $\mu\simeq2.5$ in Fig. 5(b) may be indicative of a small correction in the $(2+1)$-dimensional DT model arising from the EW term in the continuum equation. More work is needed to conclusively settle this issue. This would also explain why the scaling collapse for the damped oscillations in Fig. 5(b), particularly for the data in the kinetically rough ($t>t_c$) growth regime, is not as good as the corresponding $(1+1)$-dimensional results shown in Fig. 3—the asymptotic corrections to the LDV equation [Eq. (5)] arising from a small $\nabla^2 h$ term (which is present in $d'=2$ DT simulation results of Fig. 5.

The noise-reduced $d'=2$ WV model simulations, shown in Fig. 6, are even more surprising. The layer-by-layer growth oscillations in the initial transient time (up to $t\sim10$ or so) are apparent in Fig. 6(a) for finite values of m although the oscillations are already weaker than the corresponding DT results shown in Fig. 5. This is not expected based on the $d'=1$ results, where Fig. 3 (DT) and Fig. 4 (WV) are essentially identical within our simulation sizes and times. Even more surprising is the scaling collapse in the $d'=2$ WV model shown in Fig. 6(b) which is clearly not a good scaling behavior—in fact, beyond the layer-by-layer regime (i.e., for $t>t_c$) there is essentially no scaling behavior in the $d'=2$ WV results. The scaling behavior of the WV model (Fig. 6) in the $(2+1)$-dimensional WV model is clearly very different from (and worse than) the corresponding DT results. The best damping exponent value for μ we obtain from Fig. 6(b) is $\mu\approx1.5$, which is the same as the corresponding WV value in $d'=1$ as depicted in Fig. 4. We should emphasize that this estimate ($\mu\approx1.5$) for the damping exponent in the $(2+1)$ dimensional WV model should be taken at best as a crude estimate for an effective exponent since there is no scaling behavior in the WV data shown in Fig. 6. Given the very similar $d'=1$ behavior in the DT and WV models (as shown in Figs. 3 and 4) it is very surprising that the $d'=2$ behavior in the two models (including the effective damping exponent values $\mu\simeq2.5$ and 1.5, respectively, for the DT and the WV model) is so completely different.

What is the explanation for this striking difference in the $d'=2$ layer-by-layer epitaxial growth behavior in the DT (Fig. 5) and the WV (Fig. 6) model (particularly in view of their essentially identical behavior in $d'=1$ as seen in Figs. 3 and 4)? The explanation actually lies in the recently discovered fact [42,43] that, while the $d'=1$ dimensional WV model obeys [38,41,42] the continuum growth equations given in Eqs. (5) and (7) with ν_4 and $\lambda_{22}\neq0$ and ν_2 very small but having a nonvanishing positive value, the $d'=2$ dimensional WV growth model is *actually unstable* in the sense that the WV morphology in the $(2+1)$-dimensional growth forms a regular mounded structure with the mound edges having approximately constant slopes. Such an epitaxial mounding instability [43] in the $(2+1)$-dimensional WV model becomes particularly manifest under the noise-reduction technique as discussed in details in Ref. [43]. This unstable mounded morphological growth [43] in the noise-reduced $d'=2$ WV model leads to the peculiar behavior seen for late times in Fig. 6, where the epitaxial mounding instability prevents the usual layer-by-layer growth regime from behaving in the ''usual'' manner depicted in Figs. 2–5.

The effective low value of the WV damping exponent $\mu\leqslant1.5$ (rather than the ''expected'' larger value $\mu\sim2.5$ based on the $d'=1$ dimensional WV result) is interestingly consistent with the earlier finding [44] in the literature on the dependence of the onset time t_{ES} of the Ehrlich-Schwoebel (ES) instability (which produces a mounding instability similar to that seen [42,43] in the noise-reduced $d'=2$ WV simulations) on the diffusion length. In particular, it has been found [44] that $t_{ES}\sim l_{ES}^{-2}$, where l_{ES} is the characteristic length controlling the strength of the ES instability. If we now interpret our exponent relationship defined by Eq. (2) to be valid for the ES instability [and only the magnitudes of the exponents enter Eq. (2)], then we conclude that for $d'=2$, $\mu=|\delta|/d'=1$ fpr $|\delta|=2$, where l_{ES} now replaces the diffusion length as the characteristic length. Our $d'=2$ WV layer-by-layer growth simulations shown in Fig. 5 are actually consistent with a value of the exponent $\mu=1$. Further work is needed to conclusively establish this speculative

connection between our $d'=2$ noise-reduced WV simulations and the ES instability.

Finally, in Fig. 7 we show our $(2+1)$-dimensional Family model (FM) simulations under noise-reduced ($m>1$, $l=1$) conditions. The FM, which by construction is designed to follow exactly the linear second order Edwards-Wilkinson equation [Eq. (7)] with $\nu_4=\lambda_{22}=0$ in Eq. (5) and $\nu_2\neq 0$, has very simple growth rules (not shown in Fig. 1): Each randomly deposited atom seeks to find the site of local height minina as the incorporation site. The FM is essentially the discretized version of the EW equation [Eq. (7)], and as such has the EW dynamical exponents: $\beta=0$ and $\mu=\infty$ in $d'=2$. The growth exponent β [defines the presaturation kinetic roughening of W as $W\sim t^{\beta}$ [1–6]] being zero (i.e.. $W\sim\ln t$) in the FM, growth is already very smooth because the roughening is only logarithmic in time. In the presence of noise reduction, therefore, the FM [Fig. 7(a)] shows persistent layer-by-layer growth oscillations in $d'=2$ with only logarithmic damping of the oscillation induced by kinetic surface roughening. Since the damping exponent μ in the noise-reduced $d'=2$ FM is infinity [Eq. (6)], we cannot obtain any scaling collapse of the layer-by-layer growth simulation data of Fig. 7(a) which is obvious in the ''scaling plot'' shown in Fig. 7(b). A very large value of the exponent μ (≈ 100, for example) will of course produce trivial (and meaningless) data collapse, but we have checked that no finite reasonably small (up to $\mu=10$) value of the damping exponent μ produces scaling in Fig. 7(b). Thus, our $d'=2$ FM results are consistent with the theoretical prediction [33,34] of μ being infinity in the $d'=2$ EW equation. We note that we have also carried out $d'=1$ dimensional noise-reduced layer-by-layer growth simulations (not presented in this paper) in the FM, obtaining excellent scaling collapse with the theoretically predicted value of $\mu=2$. Our results for the $d'=1$ FM layer-by-layer growth are consistent with those reported in Ref. [45].

We mention that very recently a numerical simulation of layer by layer growth in the full Arrhenius diffusion model has appeared in the literature [46]. In general, the parameter $R\equiv D/F$ in this full diffusion model [46], where D is the surface diffusivity and F is the deposition flux, corresponds qualitatively to our parameters l/L or m.

IV. CONCLUSION

We have presented numerical results for extensive computer simulations of various noise-reduced limited mobility DT [10], WV [11], and FM [40] growth models in $(1+1)$ and $(2+1)$ dimensions in order to study the damping of the layer-by-layer growth epitxy invariably induced by the shot noise inherent in the deposition beam fluctuations. We have used both multiple hit noise-reduction technique and the long surface diffusion length method to obtain the layer-by-layer growth in our simulations, and have shown that these two different techniques for obtaining layer-by-layer growth are essentially equivalent. Our simulation results in general exhibit (with two exceptions noted below) very good scaling connecting the layer-by-layer growth regime with the kinetically rough growth regime. Our calculated damping exponents agree well with theoretical predictions where applicable. The two exceptions noted above are the $(2+1)$-dimensional WV and the FM, where scaling fails for different reasons. The $(2+1)$-dimensional noise-reduced WV model is known [43] to manifest unstable growth with spectacular epitaxial mounding, which inhibits layer-by-layer growth leading to the failure of scaling collapse. The $(2+1)$-dimensional FM on the other hand exhibits very strong and persistent layer-by-layer growth oscillations (with little kinetic roughening) whose damping is expected on theoretical grounds to be extremely weak leading to an infinite value of the damping exponent, which is equivalent to saying that there is essentially no scaling since in the presence of noise-reduction layer-by-layer growth regime lasts forever.

ACKNOWLEDGMENTS

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