Solid-on-solid model with next-nearest-neighbor interaction for epitaxial growth

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We investigate a solid-on-solid model on two-dimensional substrates as a natural extension of the Wolf-Villain model for molecular-beam epitaxial growth [D.E. Wolf and J. Villain, Europhys. Lett. **13**, 389 (1990)]. In this extended Wolf-Villain model, freshly landed atoms relax into the local energy minima calculated within a next-nearest-neighbor approximation. We calculate the surface width, the correlation function, and the averaged step height and show the surface morphologies. In the transient regime, the model can be described by the nonlinear continuum equation proposed by Lai and Das Sarma [Z.W. Lai and S. Das Sarma, Phys. Rev. Lett. **66**, 2348 (1991)]. At relatively smaller lateral size $L_c \sim 35$ and earlier growth time $t_c \sim 1800$ than in the original model, it shows a crossover behavior to the Edwards-Wilkinson class, i.e., logarithmic kinetic roughening.

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I. INTRODUCTION

Recently, there has been much interest in epitaxially grown surfaces. In epitaxial growth such as molecular-beam epitaxy (MBE), incident particles with deposition flux R are deposited on substrates with temperature T and diffuse under chemical-bonding environment. To study the kinetic roughening of growing surfaces at low T (or high R), various growth models and continuum growth equations have been investigated numerically and analytically [1]. In particular, much attention has been paid to the universality class of growth models and continuum equations, which is mainly determined by the values of growth exponents governing the surface fluctuations. It has been expected that for an initially flat surface, the root-mean-square value of the surface fluctuation or the surface width W scales as

$$W(L,t) = L^{\alpha} f(t/L^{z}), \tag{1}$$

where L is the lateral size of the substrate, t is the growth time, α is the roughness exponent describing the saturated surface, z is the dynamic exponent, and the scaling function $f(x) \sim x^{\beta}$ (with the growth exponent $\beta = \alpha/z$) for $x \ll 1$ and $f(x) \to$ const for $x \gg 1$ [2]. Thus the surface width W grows as $W(t) \sim t^{\beta}$ for $1 \ll t \ll L^{z}$ and $W(L) \sim L^{\alpha}$ for $t \gg L^{z}$.

Various attempts using growth models may be classified into *real* MBE growth simulations and *kinetic* growth simulations. In a real MBE growth simulation, any surface atom can migrate by an Arrhenius-type hopping rate throughout the simulation. In a kinetic growth simulation, freshly landed atoms diffuse according to a given growth rule and become incorporated into the bulk. Considering some real MBE growth simulations such as the work by Wilby, Vvedensky, and Zangwill [3], one would expect that kinetic growth simulations give correct pictures of surface roughening in epitaxial growth, despite their ideality. In this work, we focus our attention on kinetic growth simulations.

Pioneering works initiating the study of growth models for MBE growth were carried out by Wolf and Villain [4] and Das Sarma and Tamborenea [5]. In the Wolf-Villain (WV) (or Das Sarma–Tamborenea) model, freshly landed atoms relax into sites with the largest coordination numbers (or neighboring kink sites). On one-dimensional (1D) substrates, Wolf and Villain and Das Sarma and Tamborenea obtained growth exponents β and α close to analytic values of the Herring-Mullins (HM) [6] linear equation. But numerical simulations on two-dimensional substrates [7,8] showed that in the transient regime, the WV model can be described by the nonlinear equation proposed by Lai and Das Sarma [9] and that the model may show a crossover to the Edwards-Wilkinson (EW) class [10]. The measurement of the surface diffusion current [11] also supported the EW behavior in the asymptotic regime. To clarify these crossovers, Šmilauer and Kotrla [12] carried out extensive simulations on 1D and 2D substrates. On 1D substrates, they observed a crossover from the HM to the Lai–Das Sarma (LD) behavior. But they did not draw a definite conclusion for the crossover to the EW class on 1D and 2D substrates.

If a more extensive simulation is not available, an indirect way of dealing with these crossovers may be to extend the original model without any important changes and to make it possible to observe the crossovers in smaller length and time scales than in the original model. In a previous work [13], we investigated a natural extension of the WV model where the binding energy calculation includes next-nearest-neighbor (NNN) interaction as well as nearest-neighbor (NN) interaction. As is well known from the theory of critical phenomena, further neighbor interactions would not lead to a change of the main results with NN interactions, especially for the universality class. Beyond expectation, we could observe the crossover behaviors on 1D substrates in much smaller length and time scales and in a clearer manner than in the original WV model.

In this work we investigate this extended WV model on 2D substrates of real situations in experiments, which shows the LD behavior in a transient regime and the

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EW behavior in an asymptotic regime, i.e., logarithmic kinetic roughening. In Sec. II we briefly summarize the previously obtained results that are relevant to this work. In Sec. III we calculate the surface width, the correlation function, and the averaged step height and show the surface morphologies. Section IV is devoted to a brief summary.

II. SOLID-ON-SOLID MODELS FOR EPITAXIAL GROWTH

Ideal MBE growth can be regarded as a conservative growth; there are no desorption and surface overhangs leading to bulk defects. For a conservative growth, the most general continuum equation up to fourth order (see Ref. [14] for a brief review) can be written as

$$\frac{\partial h}{\partial t} = -\nabla \cdot \mathbf{j} + \eta \quad (2)$$
$$= \nu \nabla^2 h - \nu_1 \nabla^4 h + \lambda_1 \nabla^2 (\nabla h)^2 + \lambda_2 \nabla \cdot (\nabla h)^3 + \eta,$$

where $h(\mathbf{x}, t)$ is the height of the surface in $d = d' + 1$ dimension (d' is the substrate dimension), $\mathbf{j}(\mathbf{x}, t)$ the surface current, and η an uncorrelated Gaussian noise. The $(\nabla h)^2$ term of the Kardar-Parisi-Zhang equation [15] is not allowed in the continuum equation for a conservative growth.

The $\nu \nabla^2 h$ term in Eq. (2), which was introduced by Edwards and Wilkinson [10], produces $\alpha = (3-d)/2$ and $\beta = (3-d)/4$ and is known to describe a growing surface in the presence of gravitation. The $-\nu_1 \nabla^4 h$ term introduced by Herring and Mullins [6] gives $\alpha = (5-d)/2$ and $\beta = (5-d)/8$. α and β consistent with the analytic values were obtained in larger curvature models [16] in which freshly deposited atoms relax into sites with larger curvatures. The $\lambda_1 \nabla^2 (\nabla h)^2$ term solved by Lai and Das Sarma [9] yields $\alpha = (5-d)/3$ and $\beta = (5-d)/(7+d)$. [In this paper, the $\lambda_1 \nabla^2 (\nabla h)^2$ term is referred to as LD behavior.] As a model showing the LD behavior, they introduced a model where an atom moves to a neighboring kink site and breaks the bond to find another kink with smaller step height. Recently, another model belonging to the same universality class was proposed. In the model, a modification of the restricted solid-on-solid (RSOS) model [17], atoms can move to the nearest sites satisfying the RSOS condition instead of being rejected [18].

The characteristics of another nonlinear $\lambda_2 \nabla \cdot (\nabla h)^3$ term introduced by Lai and Das Sarma have not been as clear as the others. A dimensional analysis yielded critical exponents $\alpha = (5-d)/4$ and $\beta = (5-d)/(6+2d)$ [9]. In the work by Das Sarma and Ghaisas [7,19] and in the extended WV model on 1D substrates [13], the values of α and β mentioned above were obtained in transient regimes before crossovers to the EW class. On the other hand, one can regard the $\lambda_2 \nabla \cdot (\nabla h)^3$ term as a higher-order correction of the EW term, considering a Hamiltonian in a generalized Langevin equation $H \sim \int d^{d'}x \sqrt{1 + (\nabla h)^2}$ describing surface tension. More recently, a renormalization group (RG) analysis [20] and a direct integration [21] showed that the $\lambda_2 \nabla \cdot (\nabla h)^3$ term alone yields α and β consistent with the EW class. In view of the results of the RG analysis and the direct integration, we consider that the previous numerical results consistent with the dimensional analysis are simply artifacts owing to a very slow crossover to the EW class. In fact, we have *not* observed such a behavior in the extended WV model on 2D substrates of real situation in experiments.

Compared with the solid-on-solid models mentioned above, the WV model has a simpler growth rule. Unexpectedly, it shows complex crossover behaviors that are not fully convincing; previous studies mentioned in the Introduction suggest that the WV model is governed by

$$\frac{\partial h}{\partial t} = \nu \nabla^2 h - \nu_1 \nabla^4 h + \lambda_1 \nabla^2 (\nabla h)^2 + \eta. \quad (3)$$

In a previous work [13] we investigated the extended WV model on 1D substrates, which was found to be governed by Eq. (3). We showed the crossover behaviors of α and β from the calculation of the surface width; the extended WV model shows the LD behavior ($\alpha = 1$ and $z = 3$) in a transient regime and crosses over to the EW behavior ($\alpha = 1/2$ and $z = 2$) in an asymptotic regime. We also confirmed the EW behavior in the asymptotic regime from the measurement of the surface diffusion current. We note that the crossover from the HM ($\alpha = 3/2$ and $z = 4$) to the LD behavior is suppressed in the extended WV model in $d' = 1$ as in the WV model in $d' = 2$. In the extended WV model, an arbitrary small NNN interaction leads to the absence of $\alpha > 1$ implying unstable growth of the surface. Thus we consider that the extended WV model with a NNN interaction is more physically reliable than the WV model with only a NN interaction from the viewpoint of describing real growth processes.

III. NUMERICAL RESULTS ON 2D SUBSTRATES

In this section we present the numerical results on 2D substrates of size $L \times L$ ($d' = 2$). In our numerical simulation, an atom is added to the top of a randomly chosen column. If the binding energy is the largest at the chosen site, the atom stays; otherwise it moves to the empty site of the NN column offering the strongest binding. In the simulation, we used the periodic boundary condition.

In the extended WV model, the values of the binding energies E can be expressed as $N_1 E_1 + N_2 E_2$, where the number of NN's, the coordination number, is $N_1 = 1, \ldots, 5$ and that of NNN's is $N_2 = N_1 - 1, \ldots, N_1 + 7$. (See Fig. 1.) Here E_1 is the binding energy between NN's and E_2 is that between NNN's. The hierarchy of binding energies is independent of the value of E_2 in $d' = 1$, while it is not in $d' = 2$. If $E_1 > 7E_2$, larger N_1 corresponds to larger E. Otherwise a site with smaller N_1 can have larger E. To fulfill a natural extension of the WV model, we set $E_1 = 8$ and $E_2 = 1$. At the end of this section, we also deal with the opposite case of $E_1 < 7E_2$ with the values of $E_1 = 2$ and $E_2 = 1$.

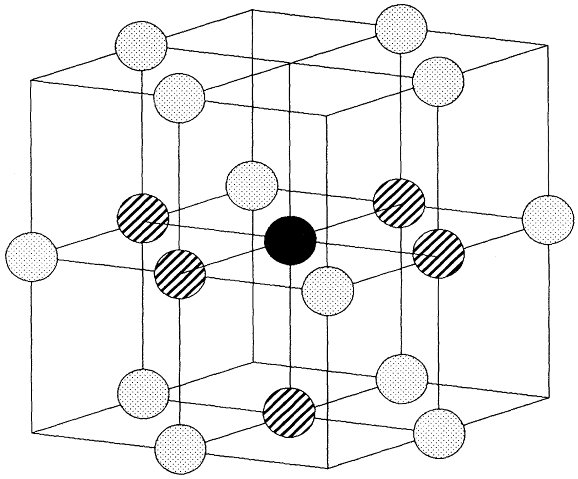

FIG. 1. Nearest neighbors (hatched spheres) and next nearest neighbors (dotted spheres) of a given site (the solid sphere) on a two-dimensional substrate.

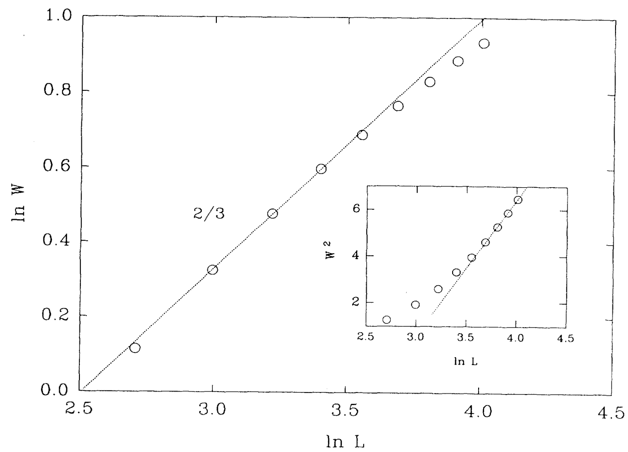

FIG. 3. The log-log plot of W vs L. The slope of the guide dotted line is 2/3. The inset shows $W^2 \sim \ln L$ in the asymptotic regime.

From the results in $d' = 1$, we expect that in a transient regime the model shows the LD behavior ($\beta = 1/5$ and $\alpha = 2/3$) and that in an asymptotic regime it shows the EW behavior (logarithmic kinetic roughening). First, we calculate the surface width with the statistical average on 150 – 600 samples. As seen in Fig. 2, we obtained $\beta = 0.2 \pm 0.001$ at small time scales, which is in excellent agreement with 1/5. As shown in the inset, W^2 behaves as $\ln t$ for $t > 2000$. Figure 3 shows the log-log plot of W vs L where $\alpha = 0.675 \pm 0.006$, close to 2/3, at small length scales and $W^2 \sim \ln L$ for $L > 40$. Therefore, the extended WV model shows the LD behavior in the transient regime and a crossover to the EW class in the asymptotic regime. We estimate the crossover time $t_c \sim 1800$ and the crossover length $L_c \sim 35$. The crossover time t_c in the extended WV model is much smaller than that ($\sim 20\,000$) in the original WV model [11]. Since it takes a very long time to arrive at a saturated regime, the crossover length L_c has not been estimated for the WV model in other works. We have successfully obtained the same results as in the WV model but in much smaller time and length scales and in a clearer manner.

Second, we calculate the correlation function to confirm our results obtained from the surface width. The height-difference correlation function is defined as $G(\mathbf{r},t) = \langle [h(\mathbf{x}+\mathbf{r},t) - h(\mathbf{x},t)]^2 \rangle$, where $\langle\,\rangle$ denotes a spatial average. In an isotropic growth, we can write $G(\mathbf{r},t) = G(r,t)$, where $G(r,t)$ scales as

$$G(r,t) = r^{2\alpha} g(r/t^{1/z}), \quad (4)$$

with the scaling function $g(x) \to$ const for $x \ll 1$ and $g(x) \sim x^{-2\alpha}$ for $x \gg 1$. We obtained the LD behavior in the transient regime by scaling $G(r,t)$ with the exponents of the LD term. Figure 4 shows a very good data collapse of the scaling plots of $G(r,t)/r^{2\alpha}$ vs $r/t^{1/z}$ with $\alpha = 2/3$ and $z = 10/3$, which confirms the LD behavior in the

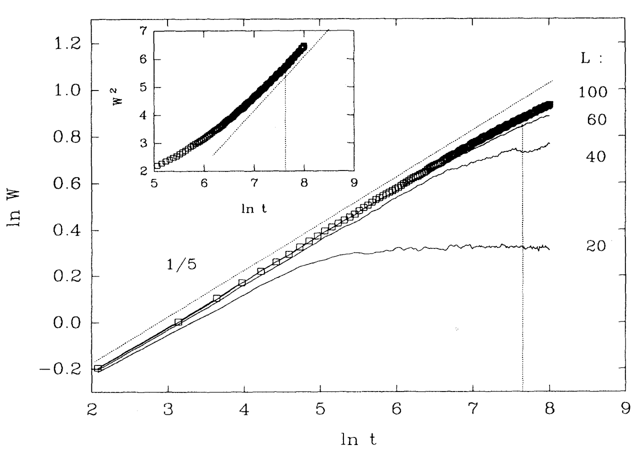

FIG. 2. The log-log plots of W vs t for L = 20, 40, 60, and 100. The slope of the guide dotted line is 1/5. The inset shows $W^2 \sim \ln t$ in the asymptotic regime where $L = 100$.

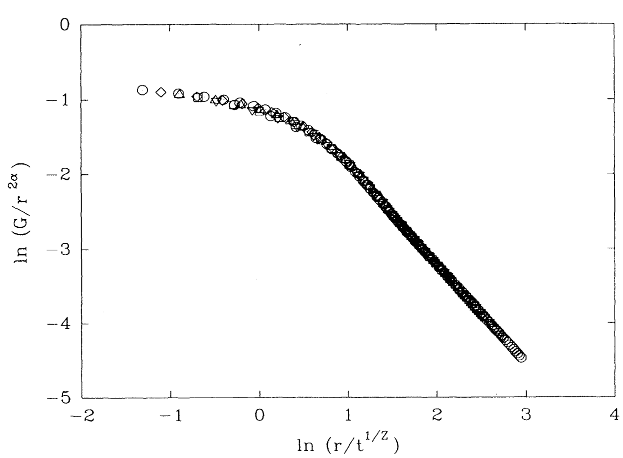

FIG. 4. Scaling plots of $G(r,t)$ for t = 25, 50, 100, 200, 400, and 800 with $L = 100$. We have $\alpha = 2/3$ and $z = 10/3$. Statistical averages were taken over 150 samples.

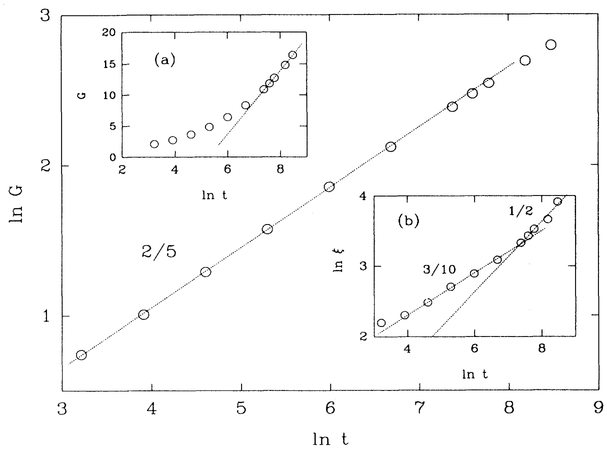

FIG. 5. The log-log plot of G vs t with $L = 100$, where $t = 25, 50, 100, 200, 400, 800, 1600, 2000, 2400, 3600$, and 4800. The slope of the guide dotted line is 2/5. Inset (a) shows $G(r,t) \sim \ln t$ in the asymptotic regime. Inset (b) shows the log-log plot of the correlation length ξ vs t. The slopes of the guide dotted lines correspond to $1/z$.

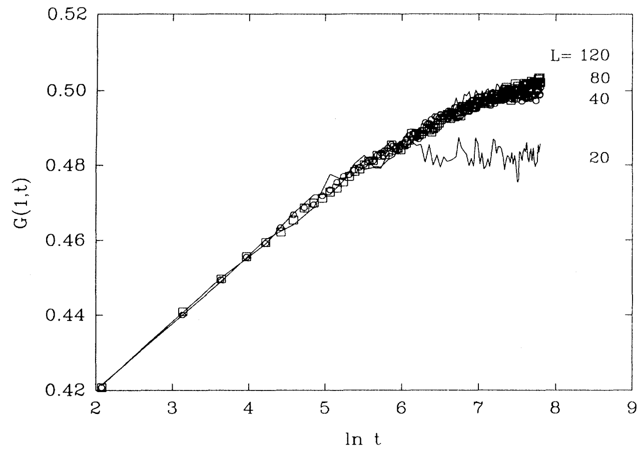

FIG. 6. Semilogarithmic plots of $G(1,t)$ vs t for $L = 20$ (——), 40 (——), 80 (○), and 120 (□). In this time scale, the data for $L = 40$, 80, and 120 are overcrowded. Statistical averages were taken over 150 – 300 samples.

transient regime. We also obtained the log-log plot of G vs t from the saturated values of $G(r,t)$, as shown in Fig. 5. We obtained $\beta = 0.201 \pm 0.001$ in the transient regime and $G \sim \ln t$ for $t > 1600$, as shown in inset (a), which is consistent with the results obtained from the surface width. Inset (b) shows the log-log plot of the correlation length $\xi \sim t^{1/z}$ against t. We obtained ξ for several values of t as the value of r at which $G(r,t)$ begins to saturate. As seen in inset (b), the dynamic exponent z changes from 10/3 to 2. This confirms that the logarithmic behaviors such as $W^2 \sim \ln t$ and $G \sim \ln t$ in the asymptotic regime are not artifacts owing to the saturation by finite-size effects.

Third, we calculate the averaged square of the step height $G(1,t) = \langle (\nabla h)^2 \rangle$. In $d' = 1$, both the extended WV and the WV models show initial power-law increases of $G(1,t)$ [12,22,23]. In $d' = 2$, $G(1,t)$ grows as $\ln t$ at early times, as shown in Fig. 6, in the extended WV model while it increases as a power law in the WV model [12]. The absence of an initial power-law increase of $G(1,t)$ in $d' = 2$ implies that the value of α measured from the surface width is the same as that from the correlation function. As shown in Fig. 6, $G(1,t)$ saturates to a constant value. However, our data are not sufficient to draw a definite conclusion whether the constant saturated value is independent of L [22] or it has a correction of order $1/L$ [24].

Finally, we show the surface morphologies in both the transient regime and the asymptotic regime. Figure 7 shows the surface morphologies at $t = 200$ (10^5), where the extended WV model shows the LD (EW) behavior. As seen in the figure, the surface shows a short-wavelength fluctuation in the transient regime [Fig. 7(a)] and a long-wavelength fluctuation in the asymptotic regime [Fig. 7(b)].

Here we discuss a point relevant to our results. We have chosen $E_1 = 8$ and $E_2 = 1$ throughout the simulation. As mentioned at the beginning of this section, if $E_1 < 7E_2$, a deposited atom can move to the site with smaller coordination number but larger binding energy. Thus atoms can move in different ways to the original WV model. To see this effect on the surface roughness we calculated the surface width with $E_1 = 2$ and $E_2 = 1$

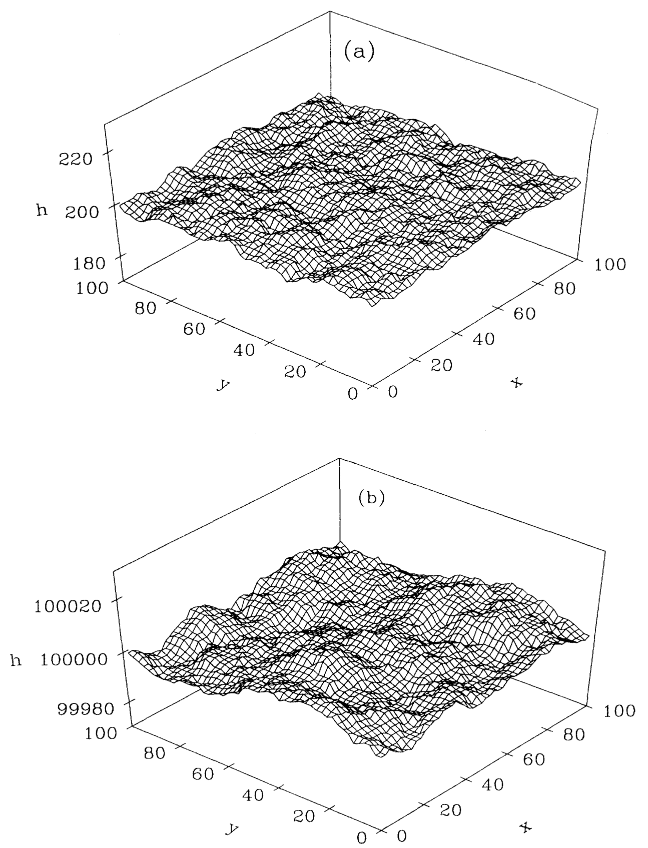

FIG. 7. Surface morphologies at (a) $t = 200$ and (b) $t = 10^5$.

but obtained the same results for α and β as with $E_1 = 8$ and $E_2 = 1$. From this result, we consider that growth models, such as the WV and the extended WV models where deposited particles move to local energy minima, are governed by the EW class in asymptotic regimes, regardless of the details of growth rules.

IV. SUMMARY

We have carried out a kinetic growth simulation for molecular-beam epitaxy with a solid-on-solid model with next-nearest-neighbor interactions, which is a natural extension of the Wolf-Villain (WV) model. On two-dimensional substrates, we have calculated the surface width, the correlation function, and the averaged step height. We have also shown the surface morphologies in both the transient and the asymptotic regimes. The extended WV model shows the Lai–Das Sarma [the $\lambda_1\nabla^2(\nabla h)^2$ term] behavior in the transient regime and the Edwards-Wilkinson behavior, i.e., a logarithmic kinetic roughening in the asymptotic regime. We have observed the same crossover behavior as in the original WV model but in much smaller length and time scales and in a clearer manner.

On one-dimensional substrates, the behavior of another nonlinear $\lambda_2\nabla\cdot(\nabla h)^3$ term was reported from the calculations of the surface width and the correlation function [13]. We consider that this result on one-dimensional substrates is an artifact owing to a very slow crossover to the Edwards-Wilkinson class, in view of the present results on two-dimensional substrates, which are consistent with the recent works of Das Sarma and Kotlyar [20] and Kim and Das Sarma [21].

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