Crossover behaviors in a molecular-beam epitaxial-growth model

C. S. Ryu and In-mook Kim
Department of Physics, Korea University, Seoul 136-701, Korea
(Received 18 October 1994)

We study a growth model for ideal molecular-beam epitaxial growth, in which landed atoms relax to local energy minima. In the calculation of the binding energy, we consider the next-nearest-neighbor interaction as well as the nearest-neighbor interaction. It is considered that this model, a natural extension of the Wolf-Villain model, is described by the most general continuum equation up to fourth order for a conservative growth. Numerical simulations on one-dimensional substrates show crossover behaviors of the growth exponents β and α; $\beta = \frac{1}{3}$ and $\alpha = 1$ change to $\beta = \frac{1}{4}$ and $\alpha = \frac{1}{2}$ (Edwards-Wilkinson class) via $\beta = \frac{3}{10}$ and $\alpha = \frac{3}{4}$. These results are supported by the calculations of the correlation function and the surface diffusion currents on tilted substrates. We also give an intuitive argument for these results.

PACS number(s): 05.40.+j, 61.50.Cj, 05.70.Ln, 68.55.Bd

I. INTRODUCTION

Recently, there has been much interest in kinetic growth phenomena. Various kinetic growth models and related continuum growth equations have been investigated numerically and analytically [1]. In particular, much attention has been paid to the universality class of various growth models, 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 the growth time, α the roughness exponent describing the saturated surface, z the dynamic exponent, and the scaling function $f(x) \sim x^{\beta}$ (with $\beta = \alpha/z$) 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 $t \gg L^{z}$.

More recently, several models are introduced for describing molecular-beam epitaxial (MBE) growth. *Ideal* MBE growth can be regarded as a conservative growth; there are no desorption and surface overhangs leading to bulk defects. Chemical-bonding environment is considered in the models for MBE growth, in which freshly landed atoms relax into local energy minima [3] or neighboring kink sites [4], instead of local height minima. It is noted that atoms landing at kink sites [with two nearest neighbors (NN's)] can move to empty trapping sites (with three NN's) in the Wolf-Villain (WV) model [3], while they stay at the kink sites in the Das Sarma-Tamborenea (DT) model [4]. On one-dimensional substrates, WV and DT obtained growth exponents β and α close to $\beta = 3/8$ and $\alpha = 3/2$ obtained from Herring-Mullins linear diffusion equation [5]

$$\frac{\partial h}{\partial t} = -\nu_1 \nabla^4 h + \eta, \tag{2}$$

where $h(\mathbf{x},t)$ is the height of the surface in $d = d' + 1$ dimension (d' is the substrate dimension) and η is an uncorrelated Gaussian noise.

But other recent works including the measurement of the surface current [6] and numerical simulations on two-dimensional substrates [6,7] suggested that the WV model is governed by the following nonlinear equation:

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

Equation (3) without $\nu \nabla^2 h$ term was solved to yield $\beta = 1/3$ and $\alpha = 1$ (in $d' = 1$) by Lai and Das Sarma (LD) [8]. In the presence of the $\nu \nabla^2 h$ term, the $\nu \nabla^2 h$ term becomes dominant in an asymptotic regime, leading to a crossover to Edwards-Wilkinson (EW) behavior ($\beta = 1/4$ and $\alpha = 1/2$ in $d' = 1$) [9]. In $d' = 1$, the crossover to EW behavior via LD behavior has been considered to be difficult to observe due to very small values of ν and λ_1. Very recently, Šmilauer and Kotrla [10] observed the crossover to the LD behavior. In spite of very large lateral size ($L \approx 40\,000$) and long time ($t \approx 10^8$) used in the simulations, it was not sufficient to draw a definite conclusion for the crossover to the EW class. Since it takes a very long time to arrive at the saturated regime, the crossover behavior of α has not been checked yet.

Besides the WV and the DT models, several models have been proposed to describe MBE growth properly. For ideal MBE growth, the most general continuum equation up to fourth order (see Ref. [11] for a brief review) can be written as

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

The $(\nabla h)^2$ term of the Kardar-Parisi-Zhang (KPZ) equation [12] is not allowed in the continuum equation for a conservative growth [13]. Since the $\lambda_2 \nabla \cdot (\nabla h)^3$ term giving $\beta = 3/10$ and $\alpha = 3/4$ (in $d' = 1$) [8] is more relevant

than the $\lambda_1\nabla^2(\nabla h)^2$ term in the renormalization group sense, for a model governed by Eq. (4), one can find rich crossover behaviors; the crossover scenarios are $3/8 \rightarrow 1/3 \rightarrow 3/10 \rightarrow 1/4$ for β and $3/2 \rightarrow 1 \rightarrow 3/4 \rightarrow 1/2$ for α.

In Sec. II, we introduce a $(1+1)$-dimensional growth model governed by Eq. (4), which is a natural extension of the WV model. In Sec. III, we show the crossover behaviors mentioned above. For this purpose, we calculate the surface width, the correlation function, and the surface diffusion currents. Section IV is devoted to a conclusion.

II. THE MODEL

In the model, freshly landed atoms relax to local energy minima, where the binding-energy calculation includes next-nearest-neighbor (NNN) interaction as well as NN interaction [14]. Considering a square lattice where the x and y axes correspond to the lateral and the growth directions, respectively, one can see that NNN interaction includes only diagonal interactions between the landed atom and the NNN's in the NN columns. We note that in the WV model, where an atom relaxes to the site with the largest coordination number, the binding energy is calculated within the NN approximation. The NNN approximation used in this *extended* WV model is expected to enhance the $\nu\nabla^2 h$ and $\lambda_1\nabla^2(\nabla h)^2$ terms, leading us to observe the crossover behaviors on much smaller length and time scales than in the original WV model.

Considering the NNN approximation on the square lattice, one can easily find that there are 16 different configurations at which the binding energy is calculated. Consideration of reflection symmetry reduces 16 configurations to 10 and the values of the binding energies can be expressed as $N_1E_1 + N_2E_2$, where the number of NN's is $N_1 = 1, 2$, and 3 and that of NNN's is $N_2 = N_1 - 1, N_1$, and $N_1 + 1$. Here E_1 is the binding energy between NN's and E_2 that between NNN's where $0 < E_2 < E_1$. The NNN interaction removes the degeneracies of the binding energies ($E_1, 2E_1$, and $3E_1$) calculated within the NN approximation. In 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. Figure 1 shows possible motions of atoms landing at the surface in the extended WV [Fig. 1(a)] and the WV [Fig. 1(b)] models.

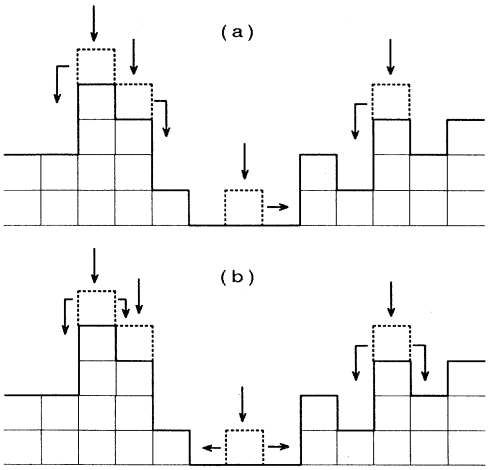

FIG. 1. Possible motions of atoms deposited at the surface are shown for the extended WV (a) and the WV (b) models.

III. NUMERICAL RESULTS

In this section, we present our numerical results in $d' = 1$. We calculated the surface width W up to the lateral size $L = 2000$ with $t = 10^5$ and took the statistical average over 300 to 1000 samples to obtain the growth exponents by linear regression analysis. In the simulation, we used the periodic boundary condition.

As seen in Fig. 2, the slope ($L = 2000$) in the log-log plots of W vs t, β changes from $\beta = 0.329 \pm 0.001$ to $\beta = 0.255 \pm 0.001$ via $\beta = 0.298 \pm 0.001$. As seen in Fig. 3, the slope in the log-log plot of W vs L, α changes from $\alpha = 1.02 \pm 0.01$ to $\alpha = 0.52 \pm 0.02$ via $\alpha = 0.76 \pm 0.02$, where L ranges from 15 to 340. Numerically obtained values of β and α are in very good agreement with analytic values in the crossover scenarios proposed from Eq. (4). The calculation of β and α suggests that the extended WV model is described by Eq. (4).

As seen in the figures, the log-log plots of W vs t and W vs L do not show the region with $\beta \approx 3/8$ and that with $\alpha \approx 3/2$ in small length and time scales, that is, the extended WV model shows the LD behavior at early times. It is considered that λ_1 is large enough to suppress the crossover regime from $\beta = 3/8$ and $\alpha = 3/2$ to $\beta = 1/3$ and $\alpha = 1$. The effect of increasing λ_1 on β was discussed in the nonlinear curvature model by Kim and Das Sarma [15]. It is also noted that the WV model in $d' = 2$ shows LD behavior at early times [6,7].

The roughness exponent $\alpha > 1$ implying unstable

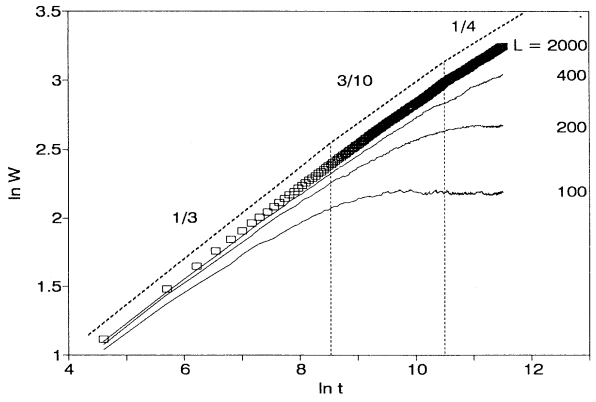

FIG. 2. The log-log plots of W vs t for $L = 100$, 200, 400, and 2000. The slopes of the dotted lines are shown in the figure.

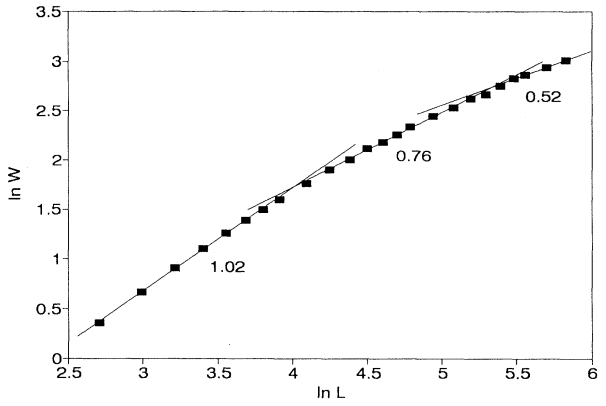

FIG. 3. The log-log plot of W vs L. The slopes of the solid lines are shown in the figure.

growth of the surface has been investigated in recent works including groove instability [16] and anomalous scaling behavior [17]. In the extended WV model (regardless of values of E_2), $\alpha > 1$ is not allowed and $\alpha = 1$ changes to $\alpha = 3/4$ at relatively small lateral size ($L \sim 50$). Noticing that an arbitrary small NNN interaction leads to the absence of $\alpha > 1$ (the hierarchy of binding energies does not depend on E_2), we consider that the extended WV model with the NNN interaction is more physically reliable than the WV model with only the NN interaction from the viewpoint of describing a real growth process.

The main features of this work are that the $\lambda_2 \nabla \cdot (\nabla h)^3$ term is present in the continuum equation [Eq. (4)] and that the extended WV model shows asymptotic behavior (the EW behavior) at much smaller length and time scales than the WV model. We support the first by calculating the correlation function $G(r,t) \equiv \langle [h(r_0,t) - h(r_0+r,t)]^2 \rangle$, where $\langle\,\rangle$ denotes a spatial average, and the second by measuring the surface diffusion currents.

As seen in Fig. 4, the scaling plot of $G(r,t)/r^{2\alpha}$ as a function of $r/t^{1/z}$ with $\alpha = 3/4$ and $z = 5/2$ shows a very good data collapse, which yields the consistent results with α and β calculated from the surface width W. This indicates that the result of $\beta = 3/10$ and $\alpha = 3/4$ at intermediate times is not an artifact owing to a very slow crossover from the LD behavior to the EW behavior. As generally accepted, in the case of $\alpha < 1$, the correlation function gives the same results as the surface width.

Next, we calculate the surface diffusion currents $j(m)$ on tilted substrates with slope m. The measurement of the surface current was proposed to determine the asymptotic behavior of a growth model before its influencing the scaling properties of the surface [6]. When the surface current is in the downhill direction, it is concluded that the asymptotic behavior is governed by the EW class. As seen in Fig. 5, the extended WV model has *large* surface currents in a downhill direction. This conforms the crossover to the EW class in the asymptotic regime. But difficulties for obtaining asymptotic behaviors of $j(m)$ prevented us from estimating the value of ν, the coefficient of the EW term. We supported our main results for the surface width by calculating the correlation function and the surface current. The detailed results of the correlation function and the surface current will be published elsewhere.

Finally, we give a simple and intuitive argument for our main results by comparing with other work. Recently, Kim, Park, and Kim [18] proposed a model considered to be governed by the equation

$$\frac{\partial h}{\partial t} = -\nu_1 \nabla^4 h + \lambda_1 \nabla^2 (\nabla h)^2 + \eta. \tag{5}$$

In the model, a modification of the restricted solid-on-solid model [19] (MRSOS), an atom can move to the nearest site satisfying the restricted solid-on-solid condition, instead of being rejected. It is very plausible to consider that the extended WV model is described by Eq. (4), if discrepancies between possible moves of atoms in the extended WV model and those in the MRSOS model lead to the $\nu \nabla^2 h$ and $\lambda_2 \nabla \cdot (\nabla h)^3$ terms. Examples are illustrated in Fig. 6.

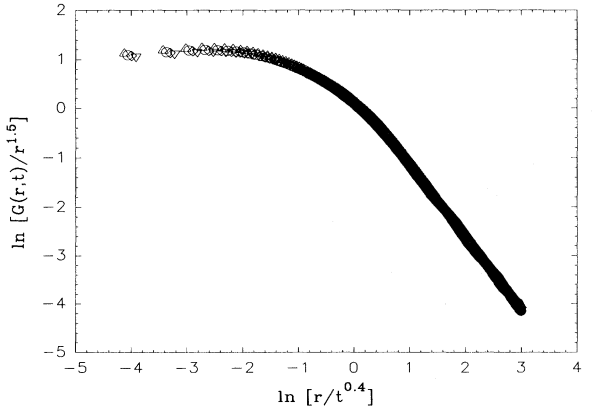

FIG. 4. Scaling plots of $G(r,t)$ for $t = 18\,000$ ($\triangle$), $22\,000$ ($\bigcirc$), and $26\,000$ ($\diamond$) with $L = 5000$ and for $t = 30\,000$ ($\triangledown$) with $L = 2000$. Statistical averages were taken over 200 samples.

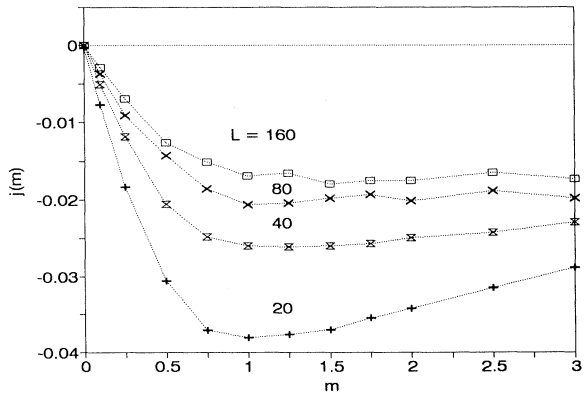

FIG. 5. Surface currents measured on tilted substrates with slope m. Averages were taken over 10^6 or 5×10^6 ($L = 160$) layers in saturated regimes. Dotted lines are guides to the eye.

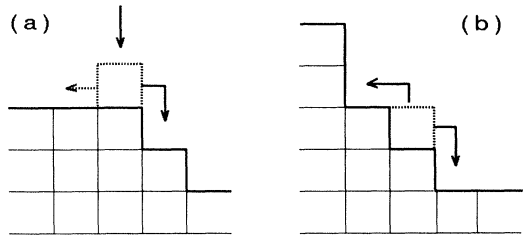

FIG. 6. (a) Possible motions of an atom landing at the ith column in the extended WV (solid line) and the MRSOS (dotted line) models. (b) Possible motions of an atom landing at the ith column where neighboring sites give the same binding energy.

An atom landing at the ith column moves to the $(i-1)$th column in the MRSOS model while it moves to the $(i+1)$th one in the extended WV model, as seen in Fig. 6(a). This downward hopping gives rise to the $\nu\nabla^2 h$ term. In addition to the $\nu\nabla^2 h$ term, the $\lambda_2\nabla\cdot(\nabla h)^3$ term should be included in the continuum equation to describe the extended WV model. Figure 6(b) shows the NN columns with the same binding energy $2E_1 + 2E_2$; an atom deposited at the ith column can move upward or downward with equal probabilities in the extended WV model. In the MRSOS model, an atom landing at the ith column moves to the $(i+1)$th column (downward hopping). The presence of the $\nu\nabla^2 h$ term would also make the atom move downward. Since the $(i-1)$th column offers the largest value $(7\lambda_2)$ of the $\lambda_2\nabla\cdot(\nabla h)^3$ term [the values are 0 at the ith column and λ_2 at the $(i+1)$th one], the presence of the $\lambda_2\nabla\cdot(\nabla h)^3$ term can make an upward hopping. The competition of the $\lambda_2\nabla\cdot(\nabla h)^3$ term with the other ones enables the atom to move upward or downward with equal probabilities as in the extended WV model. Although the above argument is crude and not sufficient, this shows intuitively that the $\nu\nabla^2 h$ and the $\lambda_2\nabla\cdot(\nabla h)^3$ terms are present in the continuum equation for the extended WV model [20].

IV. CONCLUSION

In conclusion, we have investigated the extended WV model, a natural extension of the WV model for MBE growth. The essence of this extension is that in the binding-energy calculation, not only the NN interaction but also the NNN interaction is considered in the extended WV model while only the NN interaction is considered in the WV model. We consider that the extended WV model is described by the most general equation [Eq. (4)] for a conservative growth. Thus the asymptotic behavior of the extended WV model is governed by the EW behavior. To our knowledge, the extended WV model is the first one to show the full crossover behaviors expected by Eq. (4). We expect that the WV model also shows the same crossover behaviors as those in the extended WV model but in a slower manner [21]. In this sense, the extended WV model is more tractable than the original WV model.

ACKNOWLEDGMENTS

We are grateful to B. Kahng, J. M. Kim, and H. Park for valuable discussions and K. P. Heo for numerical calculation. This work was supported in part by Korea Research Foundation, KOSEF, and the Ministry of Education (Project No. BSRI-94-2409), Korea.

[1] *Dynamics of Fractal Surfaces*, edited by F. Family and T. Vicsek (World Scientific, Singapore, 1991); J. Krug and H. Spohn, in *Solids Far From Equilibrium: Growth, Morphology and Defects*, edited by C. Godreche (Cambridge University Press, New York, 1991).
[2] F. Family and T. Vicsek, J. Phys. A **18**, L75 (1985).
[3] D. E. Wolf and J. Villain, Europhys. Lett. **13**, 389 (1990).
[4] S. Das Sarma and P. I. Tamborenea, Phys. Rev. Lett. **66**, 325 (1991).
[5] C. Herring, in *The Physics of Powder Metallurgy*, edited by W. E. Kingston (McGraw-Hill, New York, 1951); W. W. Mullins, J. Appl. Phys. **28**, 333 (1959).
[6] J. Krug, M. Plischke, and M. Siegert, Phys. Rev. Lett. **70**, 3271 (1993).
[7] S. Das Sarma and S. V. Ghaisas, Phys. Rev. Lett. **69**, 3762 (1992); M. Kotrla, A. C. Levi, and P. Šmilauer, Europhys. Lett. **20**, 25 (1992).
[8] Z. W. Lai and S. Das Sarma, Phys. Rev. Lett. **66**, 2348 (1991).
[9] S. F. Edwards and D. R. Wilkinson, Proc. R. Soc. London, Ser. A **381**, 17 (1982).
[10] P. Šmilauer and M. Kotrla, Phys. Rev. B **49**, 5769 (1994).
[11] S. Das Sarma, Fractals **1**, 784 (1993).
[12] M. Kardar, G. Parisi, and Y. C. Zhang, Phys. Rev. Lett. **56**, 889 (1986).
[13] Growth models, allowing bulk defects, for epitaxial growth were considered by H. Yan, Phys. Rev. Lett. **68**, 3048 (1992) and D. A. Kessler, H. Levine, and L. M. Sander, *ibid.* **69**, 100 (1992). Asymptotic behaviors of these models are governed by KPZ dynamics.
[14] Our model employs the NNN interaction and NN diffusion in a horizontal direction. It is not clear whether or not this extension to NNN interaction gives rise to an effective increase of a diffusion length.
[15] J. M. Kim and S. Das Sarma, Phys. Rev. Lett. **72**, 2903 (1994).
[16] J. G. Amar, P. -M. Lam, and F. Family, Phys. Rev. E **47**, 3242 (1993).
[17] M. Plischke, J. D. Shore, M. Schroeder, M. Siegert, and D. E. Wolf, Phys. Rev. Lett. **71**, 2509 (1993); M. Schroeder, M. Siegert, D. E. Wolf, J. D. Shore, and M. Plischke, Europhys. Lett. **24**, 563 (1993); S. Das Sarma,

S. V. Ghaisas, and J. M. Kim, Phys. Rev. E **49**, 122 (1994).
[18] Y. Kim, D. K. Park, and J. M. Kim, J. Phys. A **27**, L533 (1994).
[19] J. M. Kim and J. M. Kosterlitz, Phys. Rev. Lett. **62**, 2289 (1989).
[20] Using the nonlinear curvature model [15], one can give a similar argument for Fig. 6.
[21] One would expect that an inclusion of NNN interaction does not change the main results with NN interaction. In this sense, our results suggest that the WV model is described by Eq. (4) rather than Eq. (3). The difference of the two equations is the presence of the $\lambda_2 \nabla \cdot (\nabla h)^3$ term, the behavior of which shows up in transient regime. To our knowledge, the characteristics of the $\lambda_2 \nabla \cdot (\nabla h)^3$ term is not so well-known as the other terms. It needs further numerical and analytical studies to confirm the suggestion to the WV model.