

Mean-field theory of habit change phenomenon during crystal growth

Lev N. Balykov*

*Department of Geology and Mineralogy, Graduate School of Science, Kyoto University, Sakyo, Kyoto 606-8502, Japan
and Institute for Physics of Microstructures, Russian Academy of Sciences, GSP-105, Nizhny Novgorod, 603600, Russia*

Masao Kitamura

Department of Geology and Mineralogy, Graduate School of Science, Kyoto University, Sakyo, Kyoto 606-8502, Japan

Igor L. Maksimov

Theoretical Physics Department, Nizhny Novgorod University, 23 Gagarin Avenue, Nizhny Novgorod 603000, Russia

(Received 16 December 2003; published 19 March 2004)

A description of a growth morphology diagram (morphodrom) of a two-dimensional crystal growing without impurities that shows a possibility of habit change is presented. By taking into account an additional channel of incorporation of growth units into kink sites by edge diffusion, a detailed analysis of the structures and growth rates of a two-dimensional island on a (001) surface of a Kossel crystal is performed. The change in habit from the squarelike shape near equilibrium to the diamondlike one at higher supersaturation conditions is predicted based on a consideration of growth rates.

DOI: 10.1103/PhysRevB.69.125411

PACS number(s): 81.10.Aj, 68.08.De, 68.43.Mn

I. INTRODUCTION

In general, how do crystals grow? What are their shape formation laws and conditions? What are the key parameters controlling the growth rate along a specific crystallographic direction? These are questions of particular importance not only for the crystal growth expert researcher but also for the occasional snowflakes observer. Among the most challenging tasks of the physics of complex systems is understanding the sequence of bifurcations, which is generic for a specific system driven far from equilibrium, and following how a macroscopic system possessing a great number of degrees of freedom reduces to a few key variables that play a decisive role in a spatio-temporal evolution system. This paper deals with one of the most long-standing problems of crystallophysics—the habit change during crystal growth.

The habit change of crystals during growth has been studied for many years. A huge amount of experimental data has been collected that demonstrate the change of habit during growth from one polyhedral shape to another (see, e.g., Ref. 1). Attempts to explain the habit change (HC) of crystals during growth have been made since the early 1950s (Ref. 2) based on a selective impurity effect³ or changes in the nature of growth surfaces such as surface roughening,⁴ changes due to dipolar solvents,⁵ etc., to name just a few. However, no quantitative theory was suggested until now for the habit change feature. A quite promising semiquantitative approach has been proposed recently⁶ relating the HC feature with the specific regimes of the vicinal surface/step evolution.

The problem of vicinal surface evolution during crystal growth has been addressed recently by many researchers. The roughening transition was shown^{7–11} to be a generic feature of the far-from-equilibrium quasi-one-dimensional array of growing steps. The effect of edge diffusion during vicinal surface growth was shown to be responsible for the meandering instabilities caused by either the step¹² or kink¹³ edge-barrier controlling mechanism. The kinetic

smoothing effect of the [110] step structure with regard to a series process via step site was quantitatively described in Ref. 14. A direct comparison of both the [110] and [100] (see Ref. 15) step structure growth rate permits a quantitative evaluation of the HC effect (a preliminary report on this subject was presented in the ICCG-13, Kyoto,¹⁶).

In the present paper, the self-consistent mean-field theory is formulated to describe the habit change during growth of a two-dimensional crystal without impurities. A detailed analysis of the structures and growth rates of the two-dimensional island is performed, taking into account an additional two-step channel of incorporation of growth units into kink sites by diffusion of adatoms on the edge of the step. The change in habit from the squarelike shape near equilibrium to the diamondlike one at higher supersaturations is predicted based on consideration of growth rates.

II. MODEL DESCRIPTION

To illustrate the concept, we consider a two-dimensional island on the (001) surface of the Kossel crystal. Our treatment will be based on the recently discovered kinetic smoothing effect that is due to the edge-diffusion-assisted incorporation of growth units into the step.¹⁴ It would be quite instructive briefly to present details of elementary structures of the [100] and [110] steps. In our model of the [110] step, which has been introduced in Ref. 14, we describe the step in terms of densities of “shifts” n_r with r jumps ($r \geq 1$) and elementary straight segments (ESS) n_0 [see Fig. 1(a)]: $\Gamma_s + n_0 = 1$, where $\Gamma_s \equiv \sum_{r=1}^{\infty} n_r$ is the total number of shifts of all heights. The symmetry of the [110] step suggests the following relation between n_0 and n_r : $n_r : n_0 = (1 - n_0)^2 n_0^{r-1}$ for any positive r , and allows for a description of the [110] step with only one variable, e.g., the total density of shifts Γ_s . Negative shifts and, therefore, overhangs are not included in the step structure. However, overhangs of height 1 are allowed for a transitional state

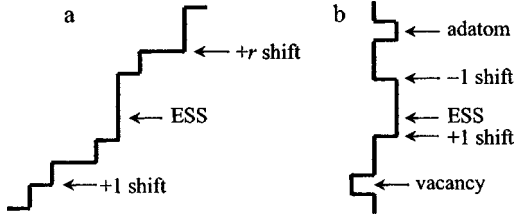


FIG. 1. (a) [110] step structure is described in terms of densities per unit site of shifts of single (+1) and multiple (+r) positive height and elementary straight segments (ESS). (b) [100] step structure is described in terms of densities per unit site of adatoms, vacancies, single positive (+1) and negative (-1) shifts, and ESS. Overhangs are excluded from the step structure for both [100] and [110] steps. The left-hand sides of the steps are one atomic layer higher than the right-hand sides.

during the incorporation of atoms into the shifts. Such a description of a [110] step implies that the average length of a straight segment is not very large and has been justified in Ref. 14. Therefore, we restrict our model to $\Gamma_s \geq \frac{1}{2}$ for the [110] step.

In the treatment of the [100] step, we go beyond our previous model developed in Ref. 15 and describe the [100] step in terms of densities per unit lattice n_a of adatoms, n_v of vacancies, Γ_s of shifts, and n_0 of ESS, where $\Gamma_s + n_0 = 1$, as we assume that the total length of the step is kept constant during growth [see Fig. 1(b)]. Hereafter, “shifts” of larger height and, consequently, overhangs are forbidden in our treatment of the [100] step. Since a very rough [100] step cannot be represented accurately in terms of shifts, adatoms, and vacancies, we limit the applicability of our model to $\Gamma_s \leq \frac{1}{2}$ for the [100] step.

A detailed description of elementary processes that change the structures of the [100] and [110] steps has been given in Refs. 15 and 14, respectively. The first and second neighbors’ bonds, ϕ_1 and ϕ_2 , and the activation barriers for adatom formation, $\Delta G_a^\ddagger$, vacancy formation, $\Delta G_v^\ddagger$, diffusion of adatom along the edge of the step, $\Delta G_s^\ddagger$, and adsorption from a terrace into a kink, $\Delta G_k^\ddagger$, are now taken into account.

III. [110] STEP

From the detailed balance argument, the net flux due to the direct process from terrace to shift along the [110] step J_{T-S}^{net} is given by

$$J_{T-S}^{\text{net}} = \frac{\Gamma_s^3}{\beta_1^2 \beta_2^2 \tau_{T-S}} \left(\alpha - \frac{\beta_2 n_0^2}{\Gamma_s^2} \right), \quad (1)$$

where $\tau_{T-S} = \nu^{-1} \exp(\Delta G_k^\ddagger / k_B T)$ is the relaxation time going from terrace to shift; $\beta_i = \exp(2\phi_i / k_B T)$, $i = 1, 2$; ν is the frequency factor; k_B is the Boltzmann constant; T is the temperature; $\alpha = 1 + \sigma$ is the saturation ratio; and σ is a supersaturation near the step.

The first phase of the two-step process is the exchange of atoms between a terrace and the step site neighboring a shift. The second phase is the exchange between the step and the shifts by the edge diffusion of adatoms. We classify the step

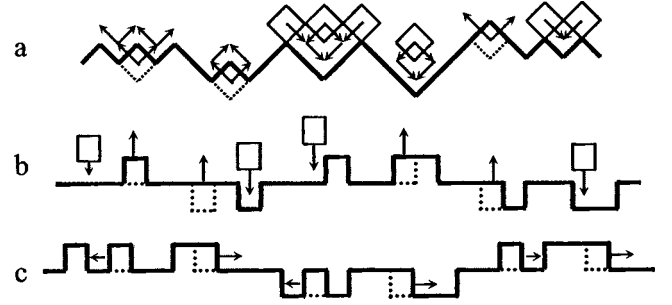


FIG. 2. (a) Elementary direct and series processes that explicitly change [110] step structure. There is an additional channel of incorporation by series process if the length of a straight segment adjacent to a shift is short. (b) Elementary direct processes that explicitly change the structure of the [100] step. (c) Elementary processes of motion of growth units along the edge of the step that explicitly change the structure of the [100] step. The upper side of the steps corresponds to the lower terrace.

sites into those on the short (of length 2) and long straight segments of the step, since the number of positions on the terrace sites, from which atoms can be incorporated into step sites, is different for these cases [see Fig. 2(a)]. By combining these two phases into a two-step process, we obtain an expression for the net flux from the terrace to shifts via step J_{T-St-S}^{net} in the following form:

$$J_{T-St-S}^{\text{net}} = \frac{n_0^2 \Gamma_s}{\beta_1^2 \beta_2^2} \left(\frac{2\Gamma_s}{\tau_{\text{short}}} + \frac{n_0}{\tau_{\text{long}}} \right) \left(\alpha - \frac{\Gamma_s^2}{\beta_2 n_0^2} \right), \quad (2)$$

where $\tau_{\text{short}} = \tau_{T-St} + 2\tau_{St-S} / \beta_1 \beta_2$, $\tau_{\text{long}} = \tau_{T-St} + \tau_{St-S} / \beta_1 \beta_2^2$, $\tau_{T-St} = \nu^{-1} \exp(\Delta G_a^\ddagger / k_B T)$ is the relaxation time going from the terrace to the step, and $\tau_{St-S} = \nu^{-1} \exp(\Delta G_s^\ddagger / k_B T)$ is the relaxation time going from the step site to a shift.

The general equation describing the nonequilibrium structure of the [110] step has the following form:

$$\frac{d\Gamma_s}{dt} = J_{T-St-S}^{\text{net}} - J_{T-S}^{\text{net}}, \quad (3)$$

where J_{T-S}^{net} and J_{T-St-S}^{net} are given by Eqs. (1) and (2), respectively. At the steady state, i.e., $d\Gamma_s / dt = 0$, by making use of the relation $\Gamma_s + n_0 = 1$, Eq. (3) thus becomes

$$\left\{ 2\gamma_s \Gamma_s + \gamma_l (1 - \Gamma_s) \right\} \left\{ \alpha (1 - \Gamma_s)^2 - \frac{\Gamma_s^2}{\beta_2} \right\} - \alpha \Gamma_s^2 + \beta_2 (1 - \Gamma_s)^2 = 0, \quad (4)$$

where $\gamma_l = \tau_{T-S} / \tau_{\text{long}}$ and $\gamma_s = \tau_{T-S} / \tau_{\text{short}}$. Figure 3(a) gives several examples of the density of shifts of the [110] step. Densities of shifts are calculated by exact solution of Eq. (4) for the simple case of $\Delta G_a^\ddagger = \frac{1}{2} \Delta G_k^\ddagger$. This relation has been discussed, for instance, in Ref. 17 and may correspond to a situation when a large growth unit is hydrated on the terrace sites adjacent to the solution. It is required to relinquish twice as many solvent molecules in the direct process as in each phase of the series process. Calculations are performed for the following strengths of nearest- and next-nearest-

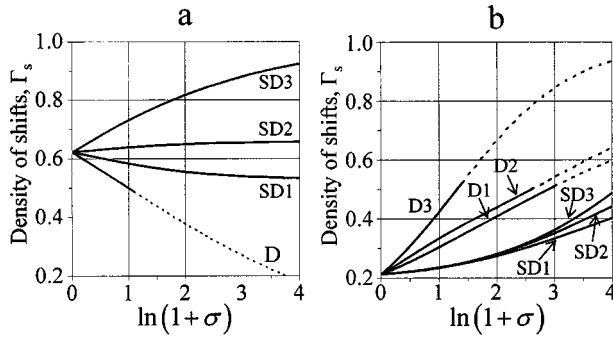


FIG. 3. Dependences of densities of shifts along [110] and [100] steps upon supersaturation (σ) in the absence ($\Delta G_s^\ddagger = \infty$, D lines) and presence ($\Delta G_s^\ddagger = \Delta G_a^\ddagger$, SD lines) of a series process. Lines numbered 1, 2, and 3 correspond to $\Delta G_a^\ddagger/k_B T = 0, 1$, and 10 , respectively. Calculations are performed for the following strengths of nearest- and next-nearest-neighbor bonds: $\phi_1/k_B T = 2$ and $\phi_2/k_B T = \frac{1}{2}$. (a) Density of shifts along the [110] step calculated for a simple case of $\Delta G_a^\ddagger = \frac{1}{2}\Delta G_k^\ddagger$ decreases with an increase of σ in the absence of a series process making the step rougher, independent of the height of the activation barrier for direct incorporation. Below $\Gamma_s \leq \frac{1}{2}$ the curve loses physical meaning. In the presence of a series process, the density of shifts may decrease or increase with an increase of σ . (b) Density of shifts along the [100] step calculated for a simple case of $\Delta G_a^\ddagger = \frac{1}{2}\Delta G_k^\ddagger = \frac{1}{3}\Delta G_v^\ddagger$ always increases with an increase of σ . The presence of a series process suppresses roughening up to high values of σ (SD lines). Above $\Gamma_s \geq \frac{1}{2}$, curves lose physical meaning.

neighbor bonds: $\phi_1/k_B T = 2$ and $\phi_2/k_B T = \frac{1}{2}$. The D line shows that in the absence of a two-step incorporation process ($\Delta G_s^\ddagger = \infty$), the [110] step roughens rapidly with an increase of supersaturation. On the other hand, the presence of a series process ($\Delta G_s^\ddagger = \Delta G_a^\ddagger$) has a great influence on the kinetics and structure of the [110] step that can become both *rough* and *smooth* during growth, as seen from the SD lines. Taking into account the attachment of adatoms and the formation of one-dimensional clusters on the straight segments of the [110] step could enhance the [110] step roughness. However, since the average length of a straight segment is small under the kinetic smoothening growth mechanism, this would only lead to a negligible increase of [110] step velocity, as we have already discussed in Ref. 14.

IV. [100] STEP

Combining elementary direct [see Fig. 2(b)] and edge diffusion [see Fig. 2(c)] processes that change the [100] step structure, we write a governing equation that describes the structure of the [100] step in terms of the density of shifts:

$$\frac{1}{2} \frac{d\Gamma_s}{dt} = \frac{\alpha n_0^2}{\beta_1^2 \beta_2^2 \tau_a} - \frac{n_a}{\beta_1 \beta_2^2 \tau_a} - \frac{\alpha n_v}{\beta_1^2 \beta_2^2 \tau_v} + \frac{n_0^2}{\beta_1^3 \beta_2^2 \tau_v} - \left(\frac{2n_a^2}{\tau_s} + \frac{\Gamma_s^2 n_a}{2\tau_s} - \frac{\Gamma_s^2 n_0^2}{\beta_1 \tau_s} + \frac{\Gamma_s n_0 n_a}{\tau_s} - \frac{\Gamma_s n_0^3}{\beta_1 \tau_s} \right), \quad (5)$$

where $\tau_j = \nu^{-1} \delta_j$, $\delta_j = \exp(\Delta G_j^\ddagger/k_B T)$, and $j = a, v, s, k$. The number of shifts along the step can only be changed by the creation and the annihilation of an adatom and a vacancy. If only direct processes (terms outside brackets) are considered and the densities of adatoms and vacancies are assumed equal and related to the density of shifts as $n_a = n_v = \frac{1}{4} \Gamma_s^2$, Eq. (5) becomes equivalent to that suggested by van der Eerden.¹⁸ However, it has been demonstrated both analytically¹⁵ and numerically¹⁹ that adatoms and vacancies are not just “two consecutive shifts of opposite signs” but essential elements of a step having their own kinetic behavior. Therefore, two more equations should be added to describe the kinetics of adatoms and vacancies on the [100] step:

$$\frac{dn_v}{dt} = \frac{n_0^2}{\beta_1^3 \beta_2^2 \tau_v} - \frac{\alpha n_v}{\beta_1^2 \beta_2^2 \tau_v} + \frac{\alpha \Gamma_s^2 n_0}{2\beta_1^2 \beta_2^2 \tau_k} - \frac{2n_v n_0}{\beta_1^2 \beta_2^2 \tau_k}, \quad (6)$$

$$\frac{dn_a}{dt} = \frac{\alpha n_0^2}{\beta_1^2 \beta_2^2 \tau_a} - \frac{n_a}{\beta_1 \beta_2^2 \tau_a} + \frac{\Gamma_s^2 n_0}{2\beta_1^2 \beta_2^2 \tau_k} - \frac{2\alpha n_a n_0}{\beta_1^2 \beta_2^2 \tau_k} - \left(\frac{4n_a^2}{\tau_s} + \frac{\Gamma_s^2 n_a}{2\tau_s} - \frac{3\Gamma_s^2 n_0^2}{2\beta_1 \tau_s} + \frac{\Gamma_s n_0 n_a}{\tau_s} - \frac{\Gamma_s n_0^3}{\beta_1 \tau_s} \right). \quad (7)$$

Terms in parentheses in Eqs. (5) and (7) account for the motion of growth units along the edge of the step. When deriving Eqs. (6) and (7), we treat the configuration “ $\Gamma + \Gamma - \Gamma + \Gamma -$ ” as two neighboring adatoms, not a vacancy neighbored by two shifts; “ $\Gamma - \Gamma + \Gamma - \Gamma +$ ” as an adatom surrounded by two shifts, not two neighboring vacancies; and “ $\Gamma + \Gamma - \Gamma + n_0$ ” and “ $n_0 \Gamma - \Gamma + \Gamma -$ ” as an adatom neighboring a kink site, not a vacancy surrounded by a shift and an ESS. This procedure is justified when the probability for vacancy annihilation is much lower than that for an adatom diffusion or annihilation. This seems to be the case if the activation barrier for a vacancy annihilation is higher than that for an adatom annihilation or motion along the edge of the step, e.g., during growth from the solution. Consequently, processes involving edge diffusion only change the number of adatoms, shifts, and ESS in our model.

The governing system of Eqs. (5)–(7) describes the time evolution of the [100] step structure. We should emphasize that this set of equations, having an appearance quite similar to the analogous “reaction-type” kinetic balance equations (see, e.g., Ref. 20), nevertheless was derived from the “first-principles” consideration by performing the correct summation of elementary probabilities within a mean-field approach.²¹ Moreover, the system (5)–(7) provides a quite correct asymptotic description of both near-equilibrium [100] step growth kinetics²² and the thermodynamic-equilibrium state²³ (for more details, see Ref. 15).

At the steady state, i.e., when all the elements of the step are in kinetic balance, the system of Eqs. (5)–(7) becomes algebraic as the following condition should hold: $dn_0/dt = dn_a/dt = dn_v/dt = 0$. Figure 3(b) gives several examples of the density of shifts on the [100] step. Densities calculated by numerical solution of Eqs. (5)–(7) at the steady state for a simple case of $\Delta G_a^\ddagger = \frac{1}{2}\Delta G_k^\ddagger = \frac{1}{3}\Delta G_v^\ddagger$, which corresponds to

the case used for [110] step analysis, are shown. Calculations are performed for the following strengths of nearest- and next-nearest-neighbor bonds: $\phi_1/k_B T = 2$ and $\phi_2/k_B T = \frac{1}{2}$. D lines show that in the absence of edge diffusion ($\Delta G_s^\dagger = \infty$), the [100] step roughens rapidly with an increase of supersaturation. In the presence of edge diffusion ($\Delta G_s^\dagger = \Delta G_a^\dagger$), roughening of the [100] step during growth can be suppressed up to high supersaturations as seen from SD lines.

V. GROWTH RATES

Analysis of governing equations shows that [110] step structure kinetics can be reversed from roughening to smoothing during growth. Thus the [110] step will remain well-defined up to high supersaturations and, therefore, will not necessarily tend to disappear from the shape of a two-dimensional island or crystal. In fact, if the [100] step starts to roughen during growth while the [110] step remains smooth, the two-dimensional island or crystal may experience a change of habit from a squarelike shape to a diamondlike one. In order to determine the relative presence of [100] and [110] steps in the shape of a two-dimensional island or crystal, we need to consider the rates of advance of these steps.

Traditionally, when estimating the growth rate of the [100] step, only growth at kink sites is considered. What is meant by a kink or “half-crystal” site is a position on a step in which the addition of an atom or the removal of an adatom changes the number of atoms in the bulk crystal by 1 but does not change the step structure. Obviously, the kink is then a “single-shift-ESS” combination. Calculating the growth rate only at these sites may be justified for the ± 1 -shift model if the number of adatoms and vacancies is much smaller than that of kink sites. This, however, would lead to a great underestimation of the growth rate for the $+n$ -shift model of the [110] step. Therefore, in our analysis we shall calculate the growth rate of both the [100] and [110] steps, taking into account all processes that involve breaking or recovering two nearest-neighbor bonds. Thus, for example, the direct incorporation process of the creation of an adatom that can further move along the edge of the step is excluded from the growth rate. The following approximate expression is then used to estimate the growth rates V for both the [100] and [110] steps:

$$V = \frac{\nu \sigma}{\beta_1^2 \beta_2^2 \delta_k} \Gamma_{\text{eff}}, \quad (8)$$

where

$$\Gamma_{\text{eff}} \cong \Gamma_s (1 - \Gamma_s) \left\{ (1 + \Gamma_s) \frac{\delta_k}{\delta_s} + 1 \right\} \quad (9)$$

is the effective density of kinks along a step that is determined by the step structure as well as the growth mechanism.

The rates are calculated either for the whole range of supersaturations under study or up to the limits of the respective step model applicability, i.e., $\Gamma_s \leq \frac{1}{2}$ for the [100] step and $\Gamma_s \geq \frac{1}{2}$ for the [110] step. As is seen from Fig. 4, the

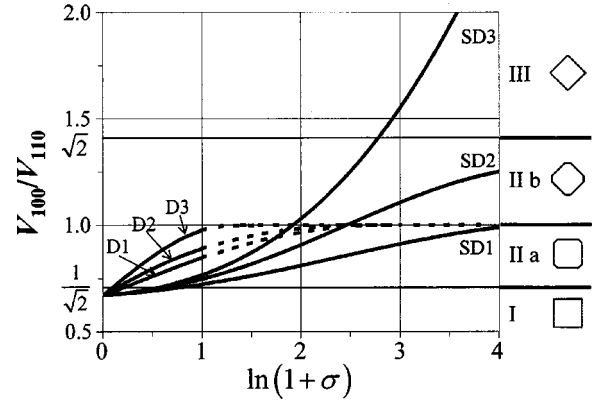


FIG. 4. Ratio of growth velocities (V) of [100] and [110] steps as a function of supersaturation calculated for a simple case of $\Delta G_a^\dagger = \frac{1}{2} \Delta G_k^\dagger = \frac{1}{3} \Delta G_v^\dagger$. Horizontal lines at $V_{100}/V_{110} = 1/\sqrt{2}$ and $V_{100}/V_{110} = \sqrt{2}$ define regions of square, mixed, and diamond habits. In the absence of a series process ($\Delta G_s^\dagger = \infty$, D lines), no habit change occurs. Only in the presence of a series process ($\Delta G_s^\dagger = \Delta G_a^\dagger$, SD lines) does a diamond shape appear at high activation barriers. Lines numbered 1, 2, and 3 correspond to $\Delta G_a^\dagger/k_B T = 0, 1$, and 10, respectively. Calculations are performed for the following strengths of nearest- and next-nearest-neighbor bonds: $\phi_1/k_B T = 2$ and $\phi_2/k_B T = \frac{1}{2}$.

[100] step never grows faster than the [110] step for the case of direct processes only. And the fact that the velocities of the [100] and [110] steps are equal indicates a circular shape of a two-dimensional island that corresponds to a conventional kinetic roughening under an adhesive growth mechanism. In contrast to that, in the case of both direct and series processes there is a crossover in growth rates. This implies a change of habit of a two-dimensional island from a squarelike shape bound by [100] steps to a diamondlike one bound by [110] steps. The position of the crossover point for a selected strength of the nearest- and second-nearest-neighbor bonds depends strongly on the height of the activation barriers. With an increase of the activation barriers, the [110] step will smooth faster with an increase of supersaturation. At very high activation barriers, the structure of the [110] step will be determined only by the presence of edge diffusion and will not depend on the height of the barrier. On the other hand, the structure of the [100] step is mainly determined by the presence of edge diffusion for any height of the activation barriers. This means that higher activation barriers will cause a habit change to occur at lower supersaturations. There also exists an asymptotic threshold supersaturation, below which a habit change does not occur.

VI. MORPHODROM

On the basis of the developed theory, one may quantitatively describe three basic growth habits of the two-dimensional island. As is well known, the shape of a crystal is defined by the most slowly growing surface (this is true for both two- and three-dimensional crystal as well as for the two-dimensional island). The island shape will be predominantly squarelike when the rate of the [100] step advance is slower than that of the [110] step by a factor of $\sqrt{2}$ or

more: $V_{100}/V_{110} < 1/\sqrt{2}$ (see region I in Fig. 4). When the [100] step advances at about the same rate as does the [110] step ($V_{100}/V_{110} \approx 1$), the two-dimensional island will have a “mixed” habit (regions IIa and IIb in Fig. 4). If, on the contrary, the [100] step advances faster than the [110] step by a factor of $\sqrt{2}$ or more, $V_{100}/V_{110} > \sqrt{2}$, the two-dimensional island will have a predominantly diamondlike shape, as is shown in Fig. 4 (region III). Thus, Fig. 4 clearly demonstrates that when the activation barriers for elementary processes are high in the presence of edge diffusion, there is a change in habit of a two-dimensional island during growth without impurities. This is in contrast with the conclusions of Beinfait²⁴ and Lian,²⁵ who attributed habit change to the impurities effect. It should be noted that adding the effect of impurities to our model intensifies the differences between [100] and [110] step kinetics, allowing habit change to occur at lower supersaturations. This case has been considered and details are to be published elsewhere.

VII. SUMMARY

We have studied the structures and growth rates of two-dimensional islands and crystals by considering the [100]

and [110] steps on the (001) surface of the Kossel crystal. By employing physically transparent key variables, we have succeeded in formulating simple self-consistent mean-field-type equations governing system evolution. On the basis of the developed approach, three basic growth habits are described of the two-dimensional island. This explains the change in habit of a two-dimensional island without impurities from the shape bounded by [100] steps at equilibrium to one bounded by [110] steps at higher supersaturations. The demonstrated habit change of two-dimensional islands and crystals is a direct effect of introducing edge diffusion into the models for [100] and [110] step kinetics.

ACKNOWLEDGMENTS

L.N.B. would like to thank the Japan Society for Promotion of Science (JSPS) for financial support. I.L.M. appreciates the support and hospitality of Satellite Venture Business Laboratory at the University of Tokushima during his short-term stay at the SVBL. This work was partly supported by Grants-in-Aid for Science Research from the Ministry of Education, Culture, Science and Technology in Japan, and for the 21st Century COE Program (Kyoto University, G3).

*Email address: balykov@kueps.kyoto-u.ac.jp

¹I. Markov, *Crystal Growth for Beginners* (World Scientific, Singapore, 1995).

²H. E. Buckley, *Crystal Growth* (Wiley, New York, 1951).

³P. Bennema, in *Handbook of Crystal Growth*, edited by D. T. J. Hurle (North-Holland, Amsterdam, 1993), Vol. 1a, p. 477.

⁴I. Sunagawa, in *Morphology of Crystals*, edited by I. Sunagawa (Terra, Tokyo, 1987), p. 509.

⁵T. Kuroda and R. J. Lacmann, *J. Cryst. Growth* **56**, 189 (1982).

⁶M. Kitamura and K. Nishioka, *Proc. Jpn. Acad., Ser. B: Phys. Biol. Sci.* **74**, 100 (1998); **76**, 108 (2000).

⁷R. Savit and R. Ziff, *Phys. Rev. Lett.* **55**, 2515 (1985).

⁸J. Kertesz and D. E. Wolf, *Phys. Rev. Lett.* **62**, 2571 (1989).

⁹M. Uwaha and Y. Saito, *Phys. Rev. Lett.* **68**, 224 (1992).

¹⁰U. Alon, M. R. Evans, H. Hinrichsen, and D. Mukamel, *Phys. Rev. Lett.* **76**, 2746 (1996).

¹¹J. M. Lopez and H. J. Jensen, *Phys. Rev. Lett.* **81**, 1734 (1998).

¹²O. Pierre-Louis, C. Misbah, Y. Saito, J. Krug, and P. Politi, *Phys. Rev. Lett.* **80**, 4221 (1998).

¹³O. Pierre-Louis, M. R. D’Orsogna, and T. L. Einstein, *Phys. Rev. Lett.* **82**, 3661 (1999).

¹⁴L. N. Balykov, M. Kitamura, I. L. Maksimov, and K. Nishioka,

Europhys. Lett. **52**, 692 (2000).

¹⁵L. N. Balykov, M. Kitamura, I. L. Maksimov, and K. Nishioka, *Philos. Mag. Lett.* **78**, 411 (1998).

¹⁶L. N. Balykov, M. Kitamura, and I. L. Maksimov, *J. Cryst. Growth* **237–239**, 39 (2002).

¹⁷G. H. Gilmer and P. Bennema, *J. Appl. Phys.* **43**, 1353 (1972).

¹⁸J. P. van der Eerden, in *Handbook of Crystal Growth*, edited by D. T. J. Hurle (North-Holland, Amsterdam, 1993), Vol. 1a, p. 406.

¹⁹H. M. Cuppen, H. Meekes, W. J. P. van Enckevort, P. Bennema, and E. Vlieg, *Surf. Sci.* **525**, 1 (2003).

²⁰H. T. Dobbs, D. D. Vvedensky, A. Zangwill, J. Johansson, N. Carlsson, and W. Seifert, *Phys. Rev. Lett.* **79**, 897 (1999).

²¹M. Ohara and R. C. Reid, *Modeling Crystal Growth Rates from Solution* (Prentice-Hall, Englewood Cliffs, NJ, 1973), p. 229.

²²V. V. Voronkov, *Sov. Phys. Crystallogr.* **15**, 13 (1970).

²³W. K. Burton, N. Cabrera, and F. C. Frank, *Philos. Trans. R. Soc. London, Ser. A* **243**, 299 (1951).

²⁴M. Bienfait, R. Boistelle, and R. Kern, in *Adsorption et Croissance Cristalline* (CNRS, Paris, 1965), p. 515.

²⁵L. I. Lian, K. Tsukamoto, and I. Sunagawa, *J. Cryst. Growth* **99**, 150 (1990).