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著者	Sato Masahide, Deura Kaori
journal or publication title	Journal of Crystal Growth
volume	310
number	7-9
page range	1371-1375
year	2008-04-01
URL	http://hdl.handle.net/2297/9799

doi: 10.1016/j.jcrysgr.2007.12.019

Effect of Alternation of Kinetic Coefficients on Step Instabilities on Si(001) Vicinal face

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(Dated: November 21, 2007)

Abstract

With taking account of alternation of kinetic coefficients, we study the possibility of step instabilities on a Si(001) vicinal face. In sublimation, a step with large kinetic coefficient recedes faster than that with small kinetic coefficient, and step pairs are formed. The upper side step in the step pair is the step with large kinetic coefficient. An equidistant array of the pairs is unstable against bunching. Number of steps N_{\max} in bunches increases with time as $N_{\max} \sim t^\beta$. The exponent $\beta = 0.5$ when the bunch grows via successive collisions of step pairs, and $\beta \approx 1.2$ when the bunch grows via coalescence of bunches.

PACS numbers: 81.10.Aj, 05.70.Ln, 47.20.Hw, 68.35.Fx

Keywords: A1. Computer simulation, A1. Growth models, A1. Morphological stability A1. Crystal Morphology

I. INTRODUCTION

Dimerization of surface atoms occurs on a Si(001) surface [1]. On a Si(001) vicinal face, the dimers are parallel to the steps on T_A terrace and perpendicular to the steps on T_B terrace. The surface diffusion along the dimer rows is faster than that perpendicular to the dimer rows [2, 3]. The anisotropy of the surface diffusion changes alternately: on T_A , the surface diffusion parallel to the steps is faster than that perpendicular to the steps, and the relation is the opposite on T_B .

In addition to the type of terraces, the type of the steps changes alternately on the Si(001) vicinal face. The step in the lower side of T_A , which is called S_A , is smoother than that in the lower side of T_B , which is called S_B [2]. The difference in the smoothness changes properties of the two steps. For example, the step stiffness of S_A is larger than that of S_B [4–6], and the kinetic coefficient of S_A is probably smaller than that of S_B .

When the temperature is about 460°C, the vicinal face grows by the step-flow mode. The vicinal face is unstable and step bunching occurs [7, 8]. Frisch and co-workers [9] theoretically studied the step bunching by one-dimensional step flow model. They showed that the alternation of the surface diffusion is not important for the bunching. The bunching is caused by the alternation of the kinetic coefficients.

When a positive Ehrlich-Schwoebel effect [10–12] is present, the step bunching occurs in sublimation [13, 14]. With a strong ES effect [13], pairing of steps occurs. The bunches of step pairs are formed by coalescence of step pairs. With a weak ES effect [14], the fluctuation of step density occurs and the large bunches are formed. With the drift of adatoms [15], the step bunching occurs if the kinetic coefficients are finite [16–19]. In a conserved system, bunches grows by coalescence of bunches. With evaporation of adatoms, the collision of a single step and the bunches are repeated, and large bunches are formed [19].

The experiment [7, 8] was carried out only in growth, but the step bunching may also occur in sublimation. In this paper, we study the possibility of step instabilities in sublimation. In previous study [20, 21], with taking account of the drift of adatoms and the alternation of the diffusion coefficients, we studied the step bunching on the Si(001) vicinal face in sublimation. Since the step bunching does not occur without the drift, the alternation of the diffusion coefficients does not cause the step bunching [21]. Thus, in this paper, we study the step instabilities by alternation of kinetic coefficients. In Sec. II, we introduce

step flow model. We study the step bunching in Sec. III, and the step wandering in Sec. IV. In Sec. V, we summarize the results and carry out brief discussions.

II. MODEL

We use a standard step flow model. x -axis is parallel to steps and y -direction is in the step-down direction. We neglect the anisotropy of the surface diffusion and the impingement. When the evaporation of adatoms is taken into account, the diffusion equation of adatom density is given by

$$\frac{\partial c(\mathbf{r}, t)}{\partial t} = -\nabla \cdot \mathbf{j}(\mathbf{r}, t) - \frac{1}{\tau} c(\mathbf{r}, t), \quad (1)$$

where $\mathbf{j}(\mathbf{r}, t)$ is the adatom current, and τ is the lifetime of adatoms. The adatom current is expressed as

$$\mathbf{j}(\mathbf{r}, t) = -D_s \left(\frac{\partial^2 c}{\partial y^2} \hat{\mathbf{e}}_y + \frac{\partial^2 c}{\partial x^2} \hat{\mathbf{e}}_x \right), \quad (2)$$

where D_s is the diffusion coefficient. The boundary conditions at the n th step are given by

$$\mp \hat{\mathbf{n}} \cdot \mathbf{j}|_{y_{n\pm}} = K_n (c|_{y_{n\pm}} - c_{\text{eq}}^{(n)}), \quad (3)$$

where $\hat{\mathbf{n}}$ is the unit vector normal to the step. y_{n+} (y_{n-}) is the lower(upper) side of the step. At the steps, the adatom current by the surface diffusion is equal to the number of solidified or melted adatoms, which is proportional to the difference of adatom density from the equilibrium value. The kinetic coefficient $K_n = K_A$ for S_A and $K_n = K_B$ for S_B . Since S_B is rougher than S_A , the kink density of S_B is higher than that of S_A . Solidification and melting at the step with high kink density is more frequent than those at the step with low kink density. Thus, K_B is larger than K_A .

The equilibrium adatom density $c_{\text{eq}}^{(n)}$ is expressed as

$$c_{\text{eq}}^{(n)} = c_{\text{eq}}^0 \left(1 + \frac{\Omega}{k_B T} \frac{\partial \xi_n}{\partial y_n} \right), \quad (4)$$

where c_{eq}^0 is the equilibrium adatom density of an isolated straight step, Ω is the atomic area and ξ_n is the repulsive interaction potential between steps. On the Si(001) vicinal face, the interaction potential is given by [22]

$$\xi_n = -A(\ln l_{n-1} + \ln l_n), \quad (5)$$

where $l_n = y_{n+1} - y_n$ is the width of the n th terrace.

By solving the diffusion equation eq. (1) with boundary conditions eqs. (3) in quasi-static approximation ($\partial c/\partial t = 0$), the adatom density is determined. The step velocity V is given by

$$V = \Omega \hat{\mathbf{n}} \cdot (\mathbf{j}|_{y_n^-} - \mathbf{j}|_{y_n^+}). \quad (6)$$

III. STEP BUNCHING

We assume that the steps are straight. From eq. (1) and eqs. (3), the adatom density $c_0^{(n)}(y)$ on the n th terrace is given by

$$c_0^{(n)}(y) = A_- e^{-y/x_s} + A_+ e^{y/x_s}, \quad (7)$$

where $y = 0$ is the position of the n th step and $y = l_n$ is that of the $(n + 1)$ th step. The coefficients, A_{\pm} are expressed as

$$A_{\pm} = \frac{(\lambda_{n+1} \mp 1)e^{\tilde{l}_n} c_n \pm (1 \pm \lambda_n)c_{n+1}}{2[(1 + \lambda_n \lambda_{n+1}) \sinh \tilde{l}_n + (\lambda_n + \lambda_{n+1}) \cosh \tilde{l}_n]}, \quad (8)$$

where the scaled step distance $\tilde{l}_n = l_n/x_s$ with $x_s = \sqrt{D_s \tau}$ and the parameter $\lambda_n = D_s/K_n x_s$. λ_n represents the effect of the kinetic coefficient. When $\lambda_n \ll 1$, the effect of the kinetic coefficient is neglected. The adatom density is in equilibrium at the steps. When $\lambda_n \gg 1$, The difference in the adatom density at the step and that in equilibrium is not neglected. From eq. (6), the step velocity V_n of the n th step is obtained as

$$V_n = \frac{\Omega D_s}{x_s} \frac{-(\lambda_{n-1} \sinh \tilde{l}_{n-1} + \cosh \tilde{l}_{n-1})c_{\text{eq}}^{(n)} + c_{\text{eq}}^{(n-1)}}{(1 + \lambda_n \lambda_{n-1}) \sinh \tilde{l}_{n-1} + (\lambda_n + \lambda_{n-1}) \cosh \tilde{l}_{n-1}} + \frac{\Omega D_s}{x_s} \frac{-(\lambda_{n+1} \sinh \tilde{l}_n + \cosh \tilde{l}_n)c_{\text{eq}}^{(n)} + c_{\text{eq}}^{(n+1)}}{(1 + \lambda_n \lambda_{n+1}) \sinh \tilde{l}_n + (\lambda_n + \lambda_{n+1}) \cosh \tilde{l}_n}. \quad (9)$$

We consider the vicinal face with the terrace width $l_A = l_B = l$ (Fig. 1(a)). On the vicinal face, the effect of the step repulsion vanishes. The equilibrium adatom density is given by $c_{\text{eq}}^{(n)} = c_{\text{eq}}^0$. The step velocity V_A of S_A and V_B of S_B are given by

$$V_A = \frac{-2\Omega D_s [\lambda_B \sinh \tilde{l} + \cosh \tilde{l} - 1] c_{\text{eq}}^0}{x_s [(\lambda_A \lambda_B + 1) \sinh \tilde{l} + (\lambda_A + \lambda_B) \cosh \tilde{l}]}, \quad (10)$$

$$V_B = \frac{-2\Omega D_s [\lambda_A \sinh \tilde{l} + \cosh \tilde{l} - 1] c_{\text{eq}}^0}{x_s [(\lambda_A \lambda_B + 1) \sinh \tilde{l} + (\lambda_A + \lambda_B) \cosh \tilde{l}]}. \quad (11)$$

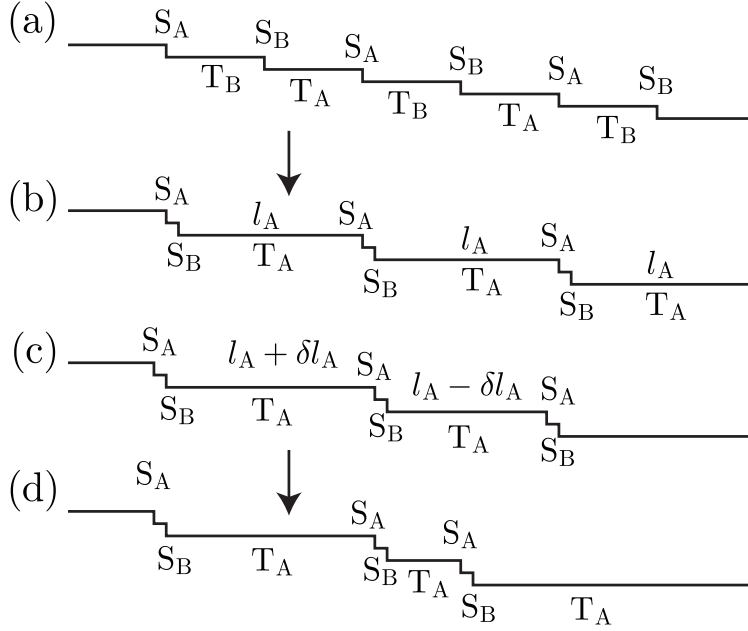


FIG. 1: Condition of surface in the stability analysis: (a) vicinal face with the step distance l , (b) an equidistant train of step pairs, (c) step pairs with alternation of width of T_A , and (d) unstable train of step pairs.

Since we assume K_B is larger than K_A , λ_A is larger than λ_B . Then, the vicinal face is unstable and S_B recedes faster than S_A . The step velocities are given by

$$V_A = \frac{\Omega D_s}{x_s} \frac{-(\lambda_B \sinh \tilde{l}_A + \cosh \tilde{l}_A)c_A^{(0)} + c_B^{(0)}}{(1 + \lambda_A \lambda_B) \sinh \tilde{l}_A + (\lambda_A + \lambda_B) \cosh \tilde{l}_A} + \frac{\Omega D_s}{x_s} \frac{-(\lambda_B \sinh \tilde{l}_B + \cosh \tilde{l}_B)c_A^{(0)} + c_B^{(0)}}{(1 + \lambda_A \lambda_B) \sinh \tilde{l}_B + (\lambda_A + \lambda_B) \cosh \tilde{l}_B}, \quad (12)$$

$$V_B = \frac{\Omega D_s}{x_s} \frac{-(\lambda_A \sinh \tilde{l}_A + \cosh \tilde{l}_A)c_B^{(0)} + c_A^{(0)}}{(1 + \lambda_A \lambda_B) \sinh \tilde{l}_A + (\lambda_A + \lambda_B) \cosh \tilde{l}_A} + \frac{\Omega D_s}{x_s} \frac{-(\lambda_B \sinh \tilde{l}_B + \cosh \tilde{l}_B)c_B^{(0)} + c_A^{(0)}}{(1 + \lambda_A \lambda_B) \sinh \tilde{l}_B + (\lambda_A + \lambda_B) \cosh \tilde{l}_B}. \quad (13)$$

where the width of T_A is l_A and that of T_B is l_B . Since the width l_A of T_A is larger than l_B of T_B , the equilibrium adatom density $c_A^{(0)}$ at S_A is larger than $c_B^{(0)}$ at S_B .

Due to the repulsive interaction between the steps, the step with double height are not formed. An equidistant train of step pairs whose upper side step is S_A are formed (Fig. 1(b)). From the condition $V_A = V_B$, the difference $\Delta c = c_A^{(0)} - c_B^{(0)}$ in the equilibrium adatom densities is determined. When the step distance is much smaller than the surface diffusion

length, from eqs. (12) and (13), Δc is approximately expressed as

$$\Delta c = \frac{(\lambda_A - \lambda_B)l}{4x_s} c_{\text{eq}}^0, \quad (14)$$

where we assumed that $\lambda_A l \ll 1$ and $\lambda_B l \ll 1$. From eqs. (12)-(14), the velocity of the step pair is given by

$$V_{\text{pair}} = -\frac{\Omega D_s l}{x_s^2} c_{\text{eq}}^0. \quad (15)$$

When the repulsion is weak, the step distance l_B is much smaller than l_A . An equidistant train of step pairs, which is separated by large T_A appears. From eqs. (4) and (5), the distance in a pair, l_B is expressed as

$$l_B = \frac{8\Omega A x_s}{k_B T (\lambda_A - \lambda_B) l} \quad (16)$$

We give a small perturbation to the width l_A of large T_A and study the stability of the equidistant train of step pairs. We assume that large T_A with the width $l_A + \delta l_A$ and small T_A with the width $l_A - \delta l_A$ appear alternately (Fig. 1(c)). We consider the step pair is very tight and l_B is much smaller than l_A . We neglect the change of the equilibrium adatom density, and assume that the step pairs are stable. At a step pair with large upper T_A terrace, the change of adatom current δj_- from the upper large T_A and that δj_+ to the lower small T_A are given by

$$\delta j_- \approx -\frac{D_s}{x_s} \frac{\lambda_B c_{\text{eq}}}{\lambda_A + \lambda_B} \frac{\delta l_A}{x_s}, \quad (17)$$

$$\delta j_+ \approx \frac{D_s}{x_s} \frac{\lambda_A c_{\text{eq}}}{\lambda_A + \lambda_B} \frac{(-\delta l_A)}{x_s}. \quad (18)$$

Then, the change of the velocity δV_{pair} of step pair with large upper terrace is given by

$$\delta V_{\text{pair}} = \Omega(\delta j_- - \delta j_+) = \frac{\Omega D_s}{x_s} \frac{\lambda_A - \lambda_B}{\lambda_A + \lambda_B} \frac{\delta l_A}{x_s} c_{\text{eq}}^0, \quad (19)$$

where we assume that the step distance in a pair does not change by the fluctuation. Since δV_{pair} is positive, the step pair recedes slower. For the step pair with small upper terrace, the change of velocity of step pair is $-\delta V_{\text{pair}}$. The step pair recedes faster. Thus, the equidistant array is unstable against the fluctuation (Fig. 1(d)).

To derive eq. (19), we assumed that the step pairs are stable, but this assumption may be not correct. It is not clear whether large bunches are formed. To see the motion of unstable array of step pairs, we carry out numerical simulations of eq. (9). Figures 2 and

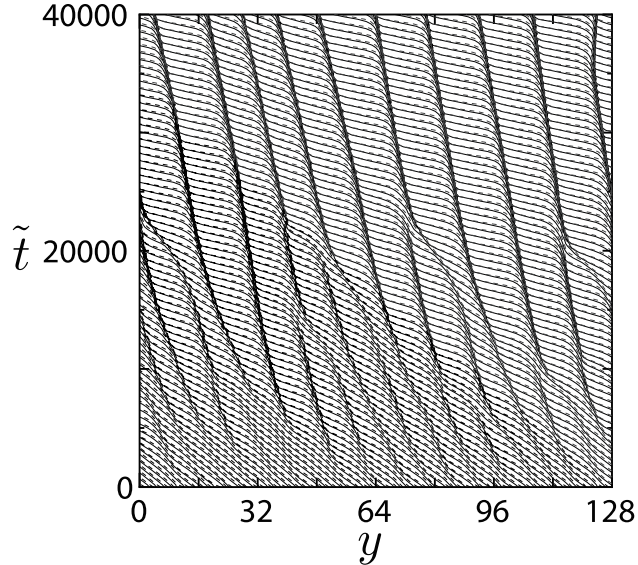


FIG. 2: Time evolution of step positions. The number of steps is 128 and the system width is 128 with the periodic boundary condition.

3 show the time evolution of steps. The dotted lines represent the motions of S_B and the solid lines represent those of S_A . y -axis represents the dimensionless time, $\tilde{t} = \Omega D_s c_{\text{eq}}^0 t / x_s^2$. The dimensionless parameters are $l/x_s = 2^{-8}$, $\Omega D_s c_{\text{eq}}^0 / x_s^2 = 2^{-16}$, $\lambda_A = 10$, $\lambda_B = 1$, and $\Omega A / c_{\text{eq}}^0 k_B T x_s = 2^{-8} \times 10^{-2}$. The number of steps is 128. The system width is 128 with the periodic boundary condition. Initially, the steps are equidistant with a small random fluctuation.

In an early stage, the step pairs whose upper side step is S_A are formed, which is expected from eqs. (10) and (11). An equidistant array of the step pairs is unstable against the fluctuation of the width of T_B , and the step bunching occurs.

When $\tilde{t} \leq 2000$, both the separation of step pairs and the collision between small bunches repeatedly occur. When $2000 \leq \tilde{t} \leq 4000$, the collision to bunches does not occur. The separation of step pairs repeatedly occurs, and the bunch size seems to be saturated. When $\tilde{t} \geq 4000$, the collision starts again and the bunch size grows rapidly (Figure 3).

The change of the frequency of the collision affects the time evolution of bunch size. Figure 4 shows the time evolution of the size N_{max} of the largest bunch, which is averaged over 100 runs. The size of bunches grows with time as t^β . When $2 \times 10^4 \leq \tilde{t} \leq 7 \times 10^4$ and $2 \times 10^5 \leq \tilde{t}$, the collision between bunches seldom occurs and the exponent $\beta \approx 0.5$. In an early stage ($\tilde{t} \leq 20000$) and middle stage ($70000 \leq \tilde{t} \leq 200000$), the collision between step

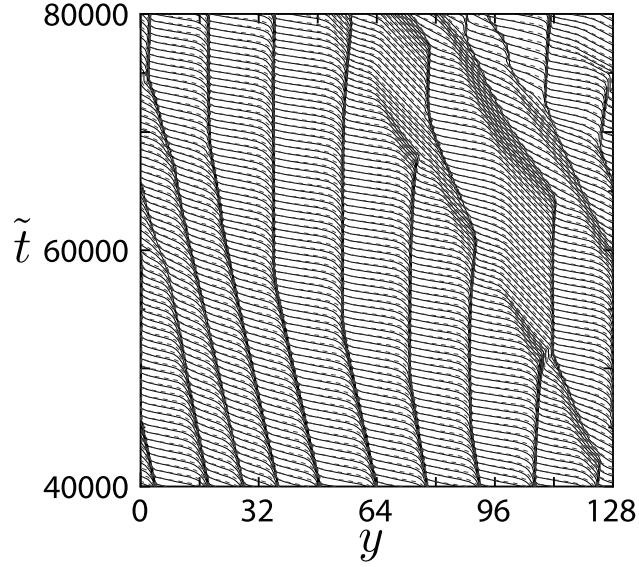


FIG. 3: Time evolution of step positions in a later stage. The parameters are the same as those in Fig. 2.

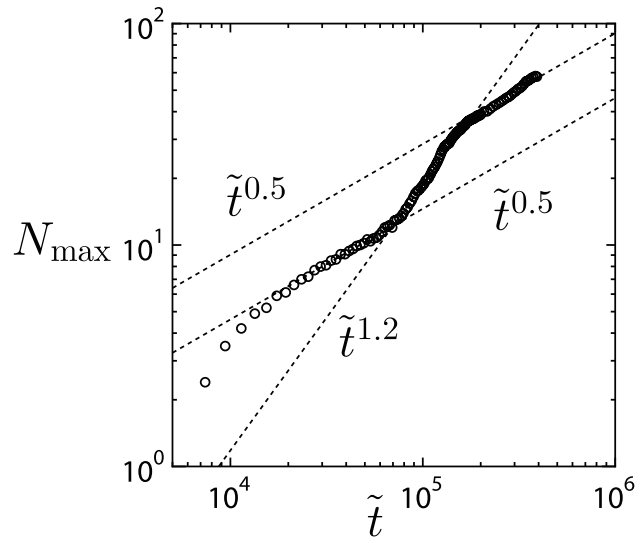


FIG. 4: Time evolution of bunch size. N_{\max} represents the number of steps in the largest bunch.

bunches occurs frequently. In the early stage, the exponent β is not clearly defined, but in the middle stage, the exponent β is approximately given by $\beta \approx 1.2$.

IV. STEP WANDERING

When we studied the step bunching, we assumed that the steps are straight. However, another type of step instability, the step wandering may occur. We consider an equidistant array of step pairs and study the wandering instability. We assume that both S_A and S_B are fluctuated as $\zeta(t, x) = \delta\zeta(t) \cos qx$.

When the step pairs are tight, the adatom current on T_A is larger than that on T_B . The stability of the step pairs is determined by the modification of adatom density on large T_A . By the step fluctuation, the adatom density on T_A is given by $c(x, y) = c_0(y) + c_1(y) \cos qx$, where $c_0(y)$ is the adatom density for straight steps and the second term is the modulation of adatom density induced by the step fluctuation. From eq. (1), the diffusion equation for $c_1(y)$ is given by

$$\frac{d^2 c_1(y)}{dy^2} = \left(\frac{1}{x_s^2} + q^2 \right) c_1(y). \quad (20)$$

When the amplitude of the step fluctuation is small, the boundary conditions on T_A are given by

$$D_s \left. \frac{dc_1}{dy} \right|_0 = K_B (c_1|_0 - c_B^{(1)}), \quad (21)$$

$$-D_s \left. \frac{dc_1}{dy} \right|_{l_A} = K_A (c_1|_{l_A} - c_A^{(1)}), \quad (22)$$

where $c_B^{(1)}$ and $c_A^{(1)}$ are defined as

$$c_B^{(1)} = - \left(\left. \frac{dc_0}{dy} \right|_0 - \frac{\lambda_B}{x_s} c_0|_0 \right) \delta\zeta, \quad (23)$$

$$c_A^{(1)} = - \left(\left. \frac{dc_0}{dy} \right|_{l_A} + \frac{\lambda_A}{x_s} c_0|_{l_A} \right) \delta\zeta. \quad (24)$$

By solving the diffusion equation (20) with boundary conditions (21) and (22), the adatom density is determined, and the time evolution of $\delta\zeta$ is given by

$$\begin{aligned} \frac{d\delta\zeta}{dt} &= -\frac{\Omega D_s}{2x_s^2} (c_0|_{l_A} - c_0|_0) \delta\zeta - \frac{\Omega D_s}{2x_s} \left(\left. \frac{dc_1}{dy} \right|_{l_A} - \left. \frac{dc_1}{dy} \right|_0 \right) \\ &= -\frac{\Omega D_s}{2x_s^2} \frac{(c_B^0 - c_A^0) \sinh(l_A/x_s) + (\lambda_A c_B^{(0)} - \lambda_B c_A^{(0)}) [\cosh(l_A/x_s) - 1]}{(1 + \lambda_A \lambda_B) \sinh(l_A/x_s) + (\lambda_A + \lambda_B) \cosh(l_A/x_s)} \\ &\quad - \frac{\Omega D_s}{2x_s} \Lambda_q \frac{(c_B^{(1)} - c_A^{(1)}) (\cosh \Lambda_q l - 1) + (\lambda_A c_B^{(1)} + \lambda_B c_A^{(1)}) \sinh \Lambda_q l}{(1 + \lambda_A \lambda_B) \sinh(\Lambda_q l_A) + (\lambda_A + \lambda_B) \cosh(\Lambda_q l_A)}, \end{aligned} \quad (25)$$

where $\Lambda_q = \sqrt{q^2 + x_s^{-2}}$. When the step distance is much smaller than the surface diffusion length, eq. (25) is approximated as

$$\frac{d\delta\zeta}{dt} = \omega_q \delta\zeta, \quad (26)$$

The amplification rate ω_q is given by

$$\omega_q = -\frac{1}{2} \frac{D_s^2 \Delta c}{(\lambda_A + \lambda_B) x_s} q^2. \quad (27)$$

Since ω_q is negative, the amplitude of the fluctuation rapidly decreases. The wandering of step pairs does not occur.

V. SUMMARY

In this paper, we studied the effect of the alternation of kinetic coefficients on the step instabilities on the Si(001) vicinal face. In sublimation, S_B recedes faster than S_A , and the step pairs whose upper side is S_A are formed. In our model, if we assume that K_A is larger than K_B , the upper side step in step pairs is S_B . However, the results do not change: the step pairs are unstable against the bunching and stable for the wandering.

An equidistant array of the step pairs are unstable against the fluctuation of the large terrace, and the step bunching occurs. The number N_{\max} of step in the largest bunch increases with time as $N_{\max} \sim t^\beta$. The exponent $\beta \approx 0.5$ when the bunches grow only via collision of step pairs and $\beta \approx 1.2$ when the collision of step bunches frequently occurs. In many systems [19, 21], the exponent $\beta \leq 1.0$. The exponent $\beta \approx 1.2$ is very large. We have not haven the easy explanation why such a large exponent is obtained. Now we are studying how the exponent is determined.

Though we neglected the step stiffness in eq. (25), the step pairs are stable for the step wandering. If we take account of the step stiffness, the step pairs is more stable for the step wandering. When the step pairs are formed, the kinetic coefficient at the lower side step, K_B is larger than that the upper side step, K_A . The step pairs is regarded as the single step with the positive ES effect. The wandering of single step with the positive ES effect does not occur in sublimation [23]. Thus, in the present case, the wandering of step pairs does not occur.

When the step instabilities are caused by the drift of adatoms, the step bunching and the step wandering can occur simultaneously [24–26]. The recombination by the step wandering

affects the growth rate of bunch size. In the present case, however, the step wandering does not occur. The effect of the wandering of step pairs on the step bunching is neglected, and we can use the one-dimensional model to study the growth law of bunch size.

On the Si(001) vicinal face, the step bunching in growth has been observed at low temperature [7, 8]. The results of theoretical study [9] agree with the experiments [7, 8]. However, the step motion in sublimation at the low temperature has not been observed. At low temperature the surface diffusion length is so long that the condition is probably similar to our simulation. Thus, the confirmation of the present results are expected.

Acknowledgments

This work was supported by Grant-in-Aid for Scientific Research from Japan Society for the Promotion of Science.

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Figure Captions:

Figure 1:

Condition of surface in the stability analysis: (a) vicinal face with the step distance l , (b) an equidistant train of step pairs, (c) step pairs with alternation of width of T_A , and (d) unstable train of step pairs.

Figure 2:

Time evolution of step positions. The number of steps is 128 and the system width is 128 with the periodic boundary condition.

Figure 3:

Time evolution of step positions in a later stage. The parameters are the same as those in Fig. 2.

Figure 4:

Time evolution of bunch size. N_{\max} represents the number of steps in the largest bunch.

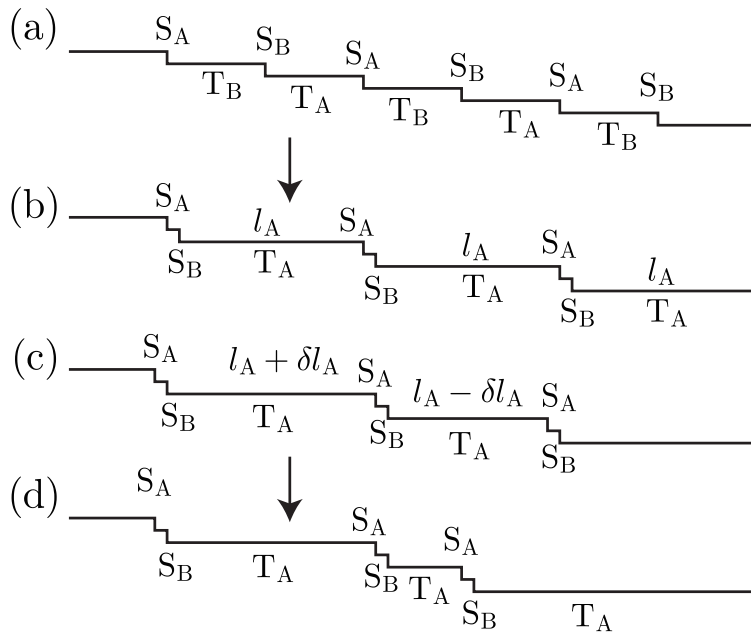


Figure 1: M. Sato and K. Deura

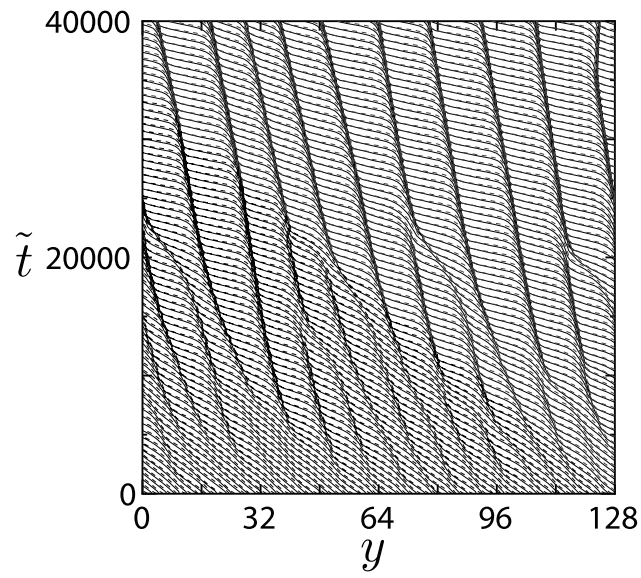


Figure 2: M. Sato and K. Deura

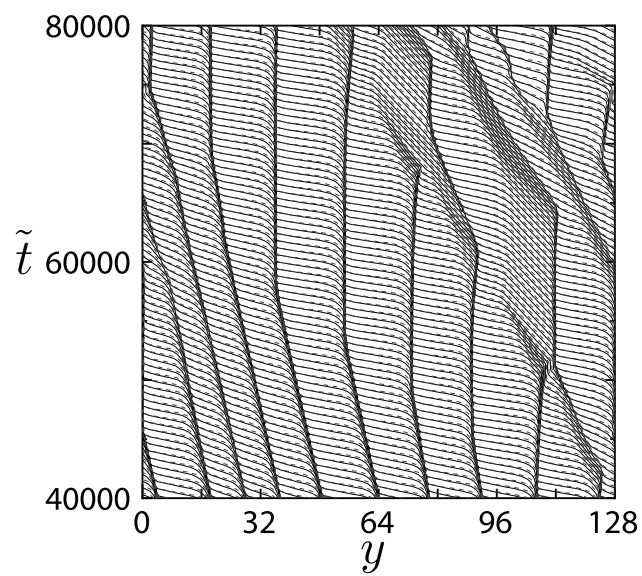


Figure 3: M. Sato and K. Deura

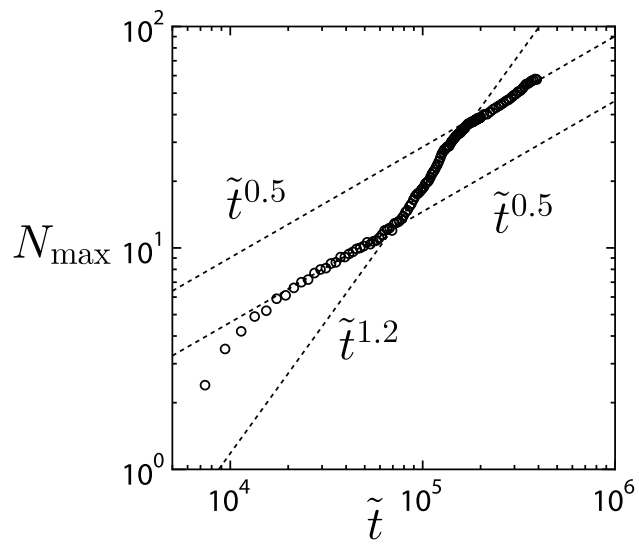


Figure 4: M. Sato and K. Deura