Drift-Induced Step Instabilities Due to the Gap in the Diffusion Coefficient

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Abstract

On a Si(111) vicinal face near the structural transition temperature, the 1×1 structure and the 7×7 structure coexist in a terrace: the 1×1 structure is in the lower side of the step edge and the 7×7 structure in the upper side. The diffusion coefficient of adatoms is different in the two structures. Taking account of the gap in the diffusion coefficient at the step, we study the possibility of step wandering induced by drift of adatoms. A linear stability analysis shows that the step wandering always occurs with step-down drift if the diffusion coefficient has a gap at the step. Formation of straight grooves by the step wandering is expected from a nonlinear analysis. The stability analysis also shows that step bunching occurs irrespective of the drift direction if the diffusion in the lower side of the step is faster. The step bunching disturbs the formation of grooves. If step-step repulsion is strong, however, the step bunching is suppressed and the straight grooves appear. Monte Carlo simulation confirms these predictions.

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I.   INTRODUCTION

On vicinal faces of Si(111) [1–3] and Si(001) [4], step wandering occurs at high temperatures when a specimen is heated by direct electric current. The current direction to cause the step wandering is step-down on the Si(111) vicinal face [1–3] and step-up on the Si(001) vicinal face [4].

The cause of the step wandering is drift of adatoms by the current [5–8]. The drift is in the same direction as the current [6, 7]. If the step is impermeable [9, 10], the step wandering occurs with step-down drift, as in the Si(111) vicinal face [1–3]. If there is alternation of the anisotropy in the diffusion coefficient on consecutive terraces [11], as in the Si(001) vicinal face [4] the step wandering occurs with step-up drift.

On Si(111) surfaces, the 1×1 structure is reconstructed and the 7×7 structure appears at low temperatures ($\leq 860°$C). In a vicinal face near the transition temperature, the 7×7 structure spreads from the upper side of the steps, and the two structures coexist in a terrace. Recently, Hibino and co-workers [12] observed step wandering near $860°$ C during growth. Due to the in-phase step wandering, grooves perpendicular to the steps appear on the vicinal face. Kato and co-workers [13] studied the step wandering theoretically. Focusing on the difference in diffusion coefficient of the two structures, they showed that the step wandering occurs in growth if the diffusion coefficient on the 1×1 structure is larger than that on the 7×7 structure.

With the two phases coexisting, the drift of adatoms may also cause the step wandering instability on the Si(111) vicinal face. In this paper, we study the possibility of morphological instabilities induced by the drift of adatoms with the gap in the diffusion coefficient on the upper and the lower terraces.

II.   MODEL

We consider a vicinal face where steps are running parallel to the x-direction bordering terraces of a width l on average. The y-direction is chosen toward the step-down direction. If impingement and evaporation of adatoms are neglected, the adatom density $c(\boldsymbol{r}, t)$ is determined by

$$\frac{\partial c(\boldsymbol{r}, t)}{\partial t} = \nabla \cdot \boldsymbol{j}(\boldsymbol{r}, t), \tag{1}$$

where $\boldsymbol{j}(\boldsymbol{r}, t)$ is the adatom current on the surface. With step-down drift, the adatom current is given by

$$\boldsymbol{j}(\boldsymbol{r}, t) = -D_{\mathrm{s}}(\boldsymbol{r}) \left(\boldsymbol{\nabla} c(\boldsymbol{r}, t) - \frac{F c(\boldsymbol{r}, t)}{k_{\mathrm{B}} T} \, \hat{\boldsymbol{e}}_{\boldsymbol{y}} \right), \tag{2}$$

where $D_{\mathrm{s}}(\boldsymbol{r})$ is the local diffusion coefficient, F the force to cause the drift and is positive for the step-down drift, and $\hat{\boldsymbol{e}}_{\boldsymbol{y}}$ the unit vector toward the step-down direction. We assume that the diffusion coefficient D_{s} takes two values in a terrace: $D_{\mathrm{s}} = D_1$ in the lower side of a step edge, $y_n < y < y_n + l_1^{(n)}$, and $D_{\mathrm{s}} = D_2$ in the upper side, $y_{n-1} + l_1^{(n-1)} < y < y_n$, where $y_n(x, t)$ is the position of the nth step and $l_1^{(n)}(x, t)$ is the terrace width of the lower side structure (1×1 in Si(111)).

Solidification and melting occur at step edges. In local equilibrium at a step, the adatom density is given by

$$c|_{y_n} = c_{\mathrm{eq}}^0 \left(1 + \frac{\Omega \tilde{\beta}}{k_{\mathrm{B}} T} \kappa + \frac{\Omega}{k_{\mathrm{B}} T} \frac{\partial U_n}{\partial y_n} \right), \tag{3}$$

where c_{eq}^0 is the equilibrium adatom density at the isolated step, Ω the atomic area, $\tilde{\beta}$ the step stiffness, κ the step curvature and U_n the step-step interaction potential.

From the continuity of the adatom current and the adatom density, the boundary conditions at the phase boundary are given by

$$\hat{\boldsymbol{n}}_{\mathrm{b}} \cdot \boldsymbol{j}|_{(y_n + l_1^{(n)})+} = \hat{\boldsymbol{n}}_{\mathrm{b}} \cdot \boldsymbol{j}|_{(y_n + l_1^{(n)})-}, \tag{4}$$

$$c|_{(y_n + l_1^{(n)})+} = c|_{(y_n + l_1^{(n)})-}, \tag{5}$$

where $\hat{\boldsymbol{n}}_{\mathrm{b}}$ is the normal vector of the boundary and $+(-)$ indicates the lower (upper) side of the step.

By solving the diffusion equation (1) in a static approximation with the boundary conditions, Eqs. (3)-(5), the adatom density is determined. The normal step velocity V_n is given by

$$V_n = \Omega \hat{\boldsymbol{n}}_{\mathrm{s}} \cdot (\boldsymbol{j}|_{y_n-} - \boldsymbol{j}|_{y_n+}), \tag{6}$$

where $\hat{\boldsymbol{n}}_{\mathrm{s}}$ is the normal vector of the step.

III. STABILITY ANALYSIS

When the steps and the boundaries of two phases are straight, the adatom density in the quasi-static approximation is given by $c(y) = A_0 + B_0 e^{f(y - y_n)}$, where $f = F/k_{\mathrm{B}} T$. In the

lower side of a step, the coefficients A_0 and B_0 are given by

$$A_0 = \frac{D_2 c_{\text{eq}}^0 (e^{fl^{(n)}} - 1)}{D_2 e^{fl_2^{(n)}}(e^{fl_1^{(n)}} - 1) + D_1(e^{fl_2^{(n)}} - 1)}, \tag{7}$$

$$B_0 = \frac{(D_1 - D_2)c_{\text{eq}}^0 (e^{fl_2^{(n)}} - 1)}{D_2 e^{fl_2^{(n)}}(e^{fl_1^{(n)}} - 1) + D_1(e^{fl_2^{(n)}} - 1)}, \tag{8}$$

where $l^{(n)} = y_{n+1} - y_n$ and $l_2^{(n)} = l^{(n)} - l_1^{(n)}$. In the upper side of the step, the coefficients are given by the same form with the replacement $D_1 \leftrightarrow D_2$ and $l_1^{(n)} \leftrightarrow l_2^{(n)}$. The adatom current j_0 is constant on the whole of the n-th terrace and is given by

$$j_0(l^{(n)}) = \frac{D_1 D_2 c_{\text{eq}}^0 f(e^{fl^{(n)}} - 1)}{D_2 e^{fl_2^{(n)}}(e^{fl_1^{(n)}} - 1) + D_1(e^{fl_2^{(n)}} - 1)}. \tag{9}$$

Since we have neglected the step repulsion, the current is a function of $l_1^{(n)}$ and $l_2^{(n)}$ and does not depend on the neighboring terrace widths.

If the ratio of the width of the two structures $\gamma = l_2^{(n)}/l_1^{(n)}$ is fixed, Eq.(9) gives the current $j_0(l)$ as a function of the terrace width. When the steps are equidistant, the adatom current at the step positions from the upper terrace equals to that onto the lower terrace, $j_0|_{y_n-} = j_0|_{y_n+}$. The velocity of the steps vanishes and the steps do not move. If the step interaction is neglected, the stability of the equidistant steps for step pairing is determined by $j'(l)$ since it controls the balance of the incoming and outgoing current with a pairing fluctuation of the terrace width (the repulsive interaction tends to stabilize the system). It is unstable for step pairing if $j_0'(l) > 0$ and stable otherwise. From Eq. (9), $j_0'(l)$ is given by

$$j_0'(l) = \frac{(D_1 - D_2)D_1 D_2 c_{\text{eq}}^0 f^2 e^{fl_2} l_1 l}{l[D_2 e^{fl_2}(e^{fl_1} - 1) + D_1(e^{fl_2} - 1)]^2} \left(\frac{e^{fl} - 1}{l} - \frac{e^{fl_1} - 1}{l_1} \right). \tag{10}$$

When the surface diffusion in the lower side of the step is faster than that in the upper side ($D_1 > D_2$), the vicinal face is unstable for the step pairing. The stability is independent of the ratio γ of the widths and the drift direction.

To find the condition for the in-phase step wandering, we consider an equidistant train of straight steps tilted with an angle θ (Fig. 1). The adatom current on the terrace in the x-direction, $J_x^{(0)}$ is calculated as

$$\begin{aligned}
J_x^{(0)} &= \int_{y_n}^{y_{n+1}} dy(j_\parallel \cos\theta - j_\perp \sin\theta) \\
&= \frac{(D_1 - D_2 e^{f_\perp l_{1\perp}})(e^{f_\perp l_{1\perp}} - 1)(e^{f_\perp l_{2\perp}} - 1)(D_1 - D_2)c_{\text{eq}}^0 \tan\theta}{D_2 e^{fl_2}(e^{f_\perp l_{1\perp}} - 1) + D_1(e^{f_\perp l_{2\perp}} - 1)}
\end{aligned} \tag{11}$$

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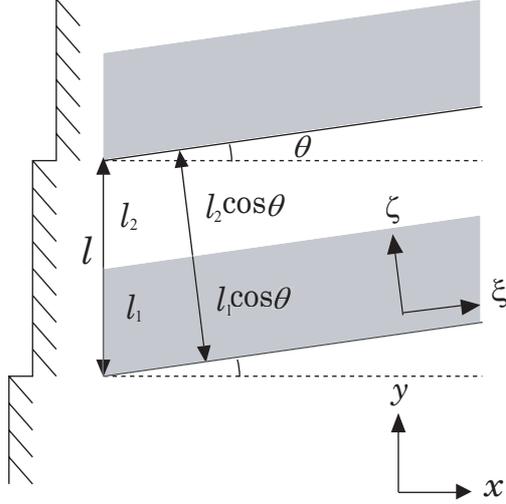
FIG. 1: A terrace bounded by tilted steps with an angle θ.

where $j_\parallel$ and $j_\perp$ indicate the adatom current in the ξ and ζ directions, $f_\perp = f\cos\theta$, $l_\perp = l\cos\theta$, $l_{1\perp} = l_1\cos\theta$ and $l_{2\perp} = l_2\cos\theta$. When the step distance is small enough, $f_\perp l_\perp \ll 1$, it becomes

$$J_x^{(0)} = \frac{(D_1 - D_2)^2 f l_1 l_2 c_{\text{eq}}^0}{(D_1 l_1 + D_2 l_2)} \frac{\eta_x}{1 + (\eta_x)^2}. \tag{12}$$

where we assume $y_n(x,t) = nl + \eta(x,t)$ and $\eta_x = \partial\eta/\partial x$. In addition to Eq. (12), there is current $J_x^{(1)}$ caused by the change of the chemical potential along the step:

$$J_x^{(1)} = -\cos^2\theta(D_1 l_{1\perp} + D_2 l_{2\perp})c_{\text{eq}}^0 \frac{\partial}{\partial x}\left(\frac{\mu}{k_{\text{B}}T}\right), \tag{13}$$

where $\mu = \Omega\tilde{\beta}\kappa$.

The evolution of the step position is determined by the adatom current in the x-direction as

$$
\begin{aligned}
\frac{\partial\eta}{\partial t} &= -\Omega\frac{\partial(J_x^{(0)} + J_x^{(1)})}{\partial x} \\
&= -\frac{\partial}{\partial x}\left[\frac{\alpha_2\eta_x}{1 + \eta_x^2} + \frac{\alpha_4}{1 + \eta_x^2}\frac{\partial}{\partial x}\left(\frac{\eta_{xx}}{(1 + \eta_x^2)^{3/2}}\right)\right],
\end{aligned} \tag{14}
$$

where the coefficients α_2 and α_4 are

$$\alpha_2 = \Omega\frac{(D_1 - D_2)^2 f l_1 l_2 c_{\text{eq}}^0}{(D_1 l_2 + D_2 l_1)}, \tag{15}$$

$$\alpha_4 = \Omega(D_1 l_1 + D_2 l_2)c_{\text{eq}}^0\frac{\Omega\tilde{\beta}}{k_{\text{B}}T}. \tag{16}$$

If the step position is of the form $\eta(x) = \eta_0 e^{iqx + \omega_q t}$, the linear amplification rate is

$$\omega_q = \alpha_2 q^2 - \alpha_4 q^4. \tag{17}$$

The coefficient α_4 is always positive and suppresses the step fluctuation. With step-up drift ($f < 0$) the coefficient α_2 is negative and suppresses the step fluctuation, while α_2 becomes positive and the step wandering occurs with step-down drift ($f > 0$).

Equation (14) is the same type of equation describing the step wandering in other conserved systems [13–16]. The solution of the equation shows a regular periodic pattern whose amplitude increases in a power law of time as $t^{1/2}$ [14]. As a result periodic grooves will be formed.

IV. MONTE CARLO SIMULATION

We perform Monte Carlo simulation for solid-on-solid steps of a square lattice model. The boundary condition is helical in the y-direction and periodic in the x-direction. We assume that γ is fixed to 1 so that the phase boundary is at $(y_n + y_{n+1})/2$ when the steps move. In the lower side of a step (supposedly the 1×1 region), an adatom on the site (i, j) moves to $(i \pm 1, j)$ with the probability $p_d = D_1/4$ and to $(i, j \pm 1)$ with the probability $p_d = D_1(1 \pm fa/2)/4$. In the upper side of a step (the 7×7 region), the parameter D_1 is replaced by D_2. The diffusion across the boundary of the two regions takes D_2. In our simulations, we assume that $(D_1, D_2) = (1, \alpha)$ or $(\alpha, 1)$ with $\alpha < 1$.

Solidification and melting occur at the lower edge of the step positions [17]. The probabilities for solidification p_+ and melting p_- are given by

$$p_\pm = \left[1 + \exp\left(\frac{\Delta E_s + \Delta U \mp \phi}{k_B T} \right) \right]^{-1}, \tag{18}$$

where ΔE_s is the increment of the step energy, ϕ the potential gain by solidification and ΔU is the change of the step-step interaction potential. We assume the repulsive interaction potential U_n of the nth step takes the form

$$U_n = \sum_{m=n\pm 1} \frac{A}{[y_n(x_i) - y_m(x_i)]^2}, \tag{19}$$

where $y_n(x_i)$ is the position of the nth step at $x = x_i$.

We first carry out the simulation with the diffusion coefficients $(D_1, D_2) = (1, 0.1)$, the stiffness $\tilde{\beta}/k_{\mathrm{B}}T = 1.64$ and the equilibrium density $c_{\mathrm{eq}}^0 = 0.18$. The system size is 512×512 and the initial step distance is $l = 16$ (the step number is $N = 32$). Initially, the steps are straight and equidistant, and there are a few adatoms on the terraces.
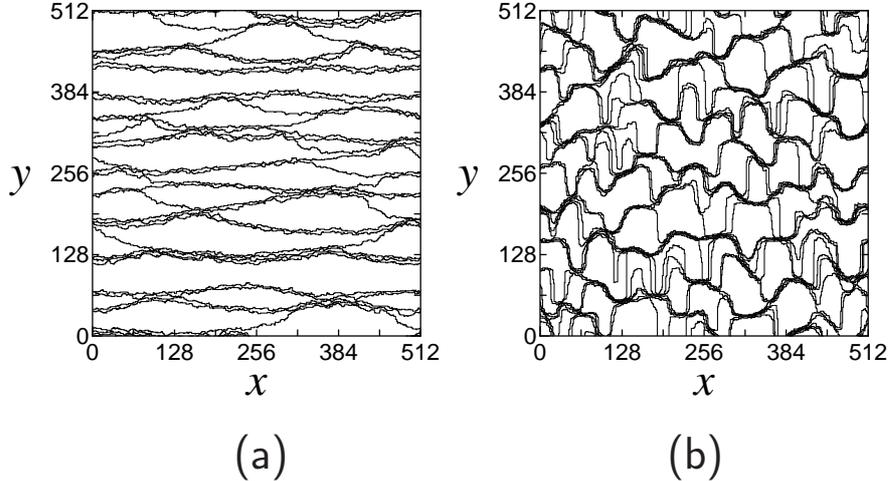

FIG. 2: Snapshots of a destabilized vicinal face for $D_1 > D_2$: with (a) step-up drift and (b) step-down drift .
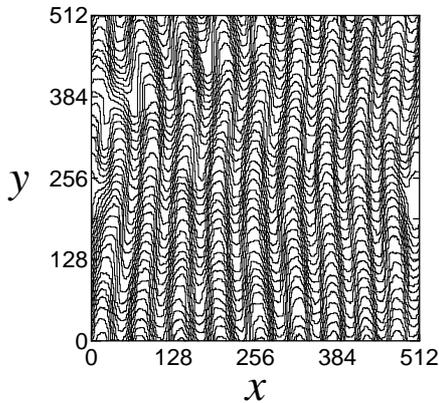
FIG. 3: Snapshot of step wandering induced by step-down drift with a strong step-step repulsion for $D_1 > D_2$. Except for the repulsive potential, the parameters are the same as in Fig. 2(b).

When we neglect the step-step repulsive interaction, the vicinal face is unstable for the step bunching (Fig. 2(a)),which agrees with the analysis in Sec. III. The step bunching occurs irrespective of the drift direction, but the form of the bunches changes with the drift direction. The bunches are straight with step-up drift (Fig. 2(a)) and wander with step-

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down drift (Fig. 2(b)). With a strong repulsive interaction, $A = 300$, the step bunching is suppressed and the in-phase step wandering occurs. Grooves parallel to the drift are produced (Fig. 3), which agrees with the solution [14, 15] of Eq. (14).
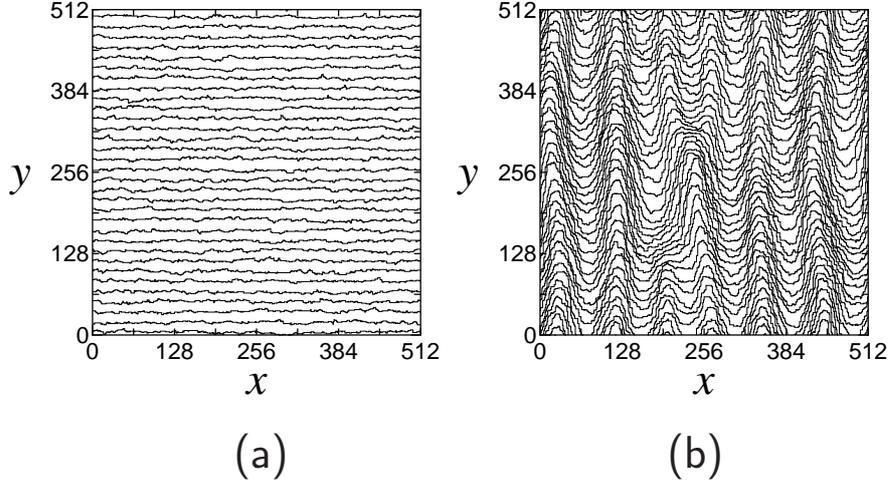

FIG. 4: Snapshots of a vicinal face for $D_1 < D_2$: with (a) step-up drift and (b) step-down drift. The repulsive interaction is absent.

We also carry out the simulation with $D_1 < D_2$ (Fig. 4). The diffusion coefficients are $(D_1, D_2) = (0.1, 1)$ and the step-step repulsion is neglected ($A = 0$). Other parameters are the same as Fig. 2. With step-up drift (Fig. 4(a)), neither step wandering nor step bunching occurs. With step-down drift, the step wandering occurs, but no indication of bunching is seen (Fig. 4(b)). As time passes, periodicity selection proceeds, resulting in straight grooves as those of Fig. 3.

V. SUMMARY AND DISCUSSION

In this paper, we studied the drift-induced morphological instabilities on a vicinal face with two phases. As is seen from Eq. (15), the step wandering occurs with step-down drift unless $D_1 = D_2$. If $D_1 = D_2$, our model reduces to a model of a simple vicinal face. We have already found [10] that, with step-down drift, wandering instability occurs in such a simple vicinal face if the kinetic coefficient of the step is finite. Although in the present paper we have assumed an infinite kinetic coefficient (local equilibrium at the steps), it must be finite in reality. Therefore we may say wandering instability is expected irrespective of

the diffusion ratio. In all cases straight grooves parallel to the drift is produced due to the in-phase step wandering in Monte Carlo simulation if step bunching is suppressed.

In the Si(111) vicinal face near the transition temperature, the diffusion coefficient in the lower side of a step is larger than that in the upper side [18]. With the assumption that the ratio of the widths of the two structures is constant, the step bunching occurs irrespective of the current direction and the step wandering occurs with step-down current. In a vicinal face of large inclination, however, the step bunching is suppressed due to the strong step-step repulsion, and only grooves induced by the step wandering may be observed with step-down current.

In our model, we assumed that the boundary of the two structures moves in concert with the steps and the ratio of the widths of two structures is constant. The drift direction to cause the instabilities does not depend on the ratio of the widths. In reality the ratio of the widths changes with temperature [19] and the motion of the boundary does not automatically follow the steps. To study the morphological development of this system in detail, we need to extend our model to include the freedom of the motion of the boundary [20].

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Figure Captions

Figure 1: A terrace bounded by tilted steps with an angle θ.

Figure 2: Snapshots of destabilized vicinal face for $D_1 > D_2$: with (a) step-up drift and (b) step-down drift.

Figure 3: Snapshot of step wandering induced by step-down drift with a strong step-step repulsion for $D_1 > D_2$. Except for the repulsive potential, the parameters are the same as in Fig. 2(b).

Figure 4: Snapshots of vicinal face for $D_1 < D_2$: with (a) step-up drift and (b) step-down drift. The repulsive interaction is absent.
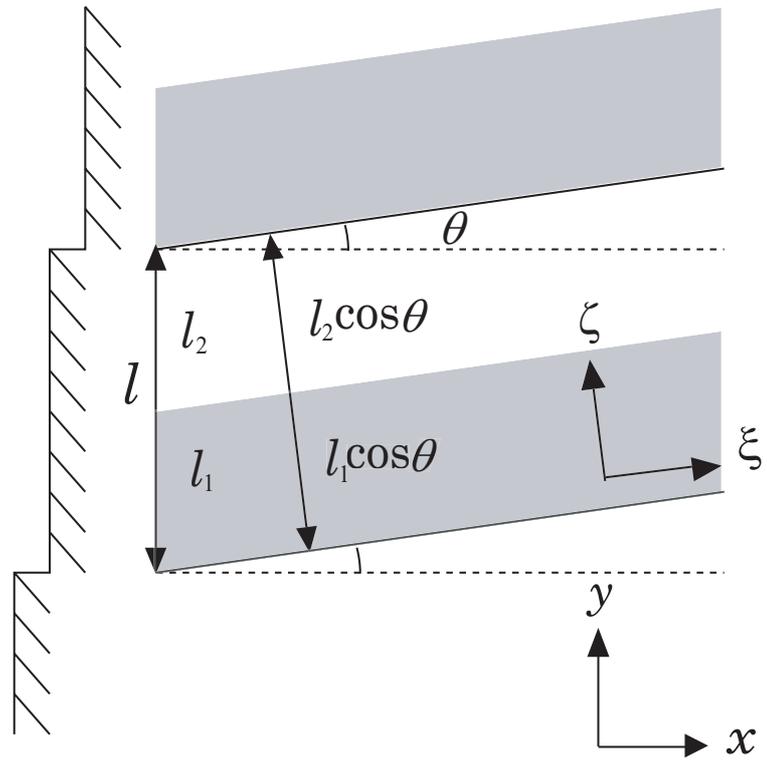
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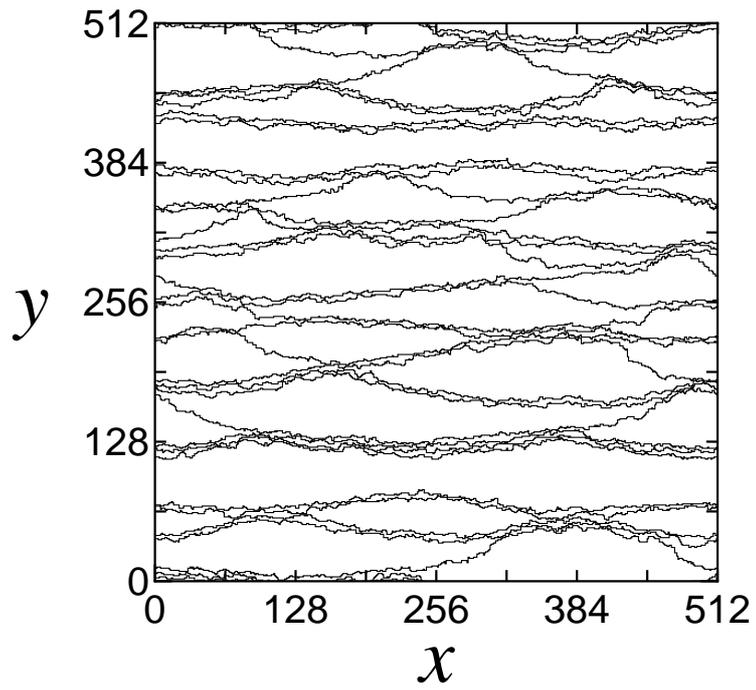
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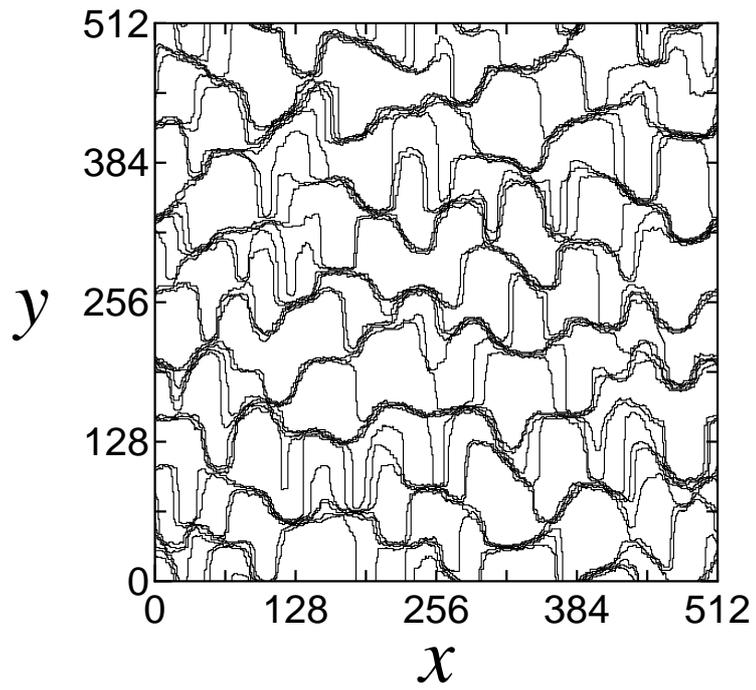
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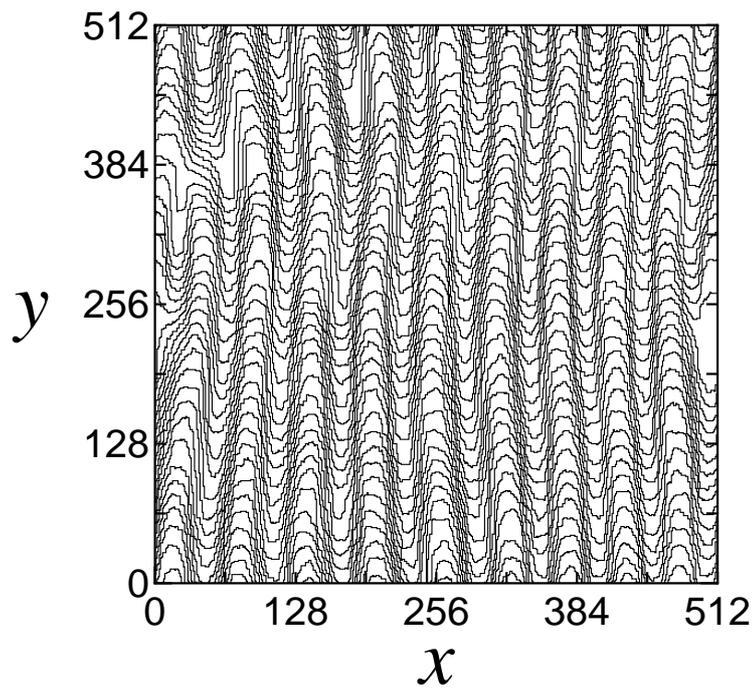
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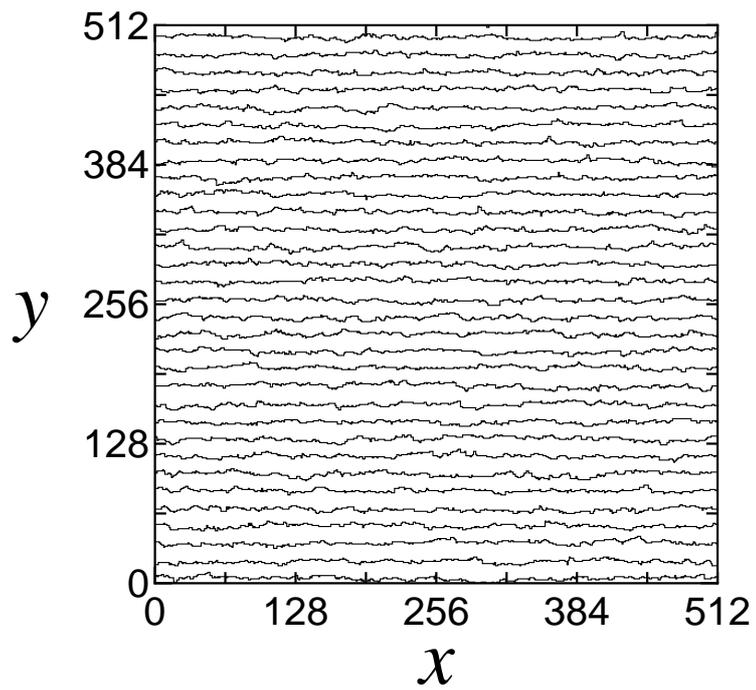
Fig.4(a) M. Sato et al., ICCG14-0416

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Fig.4(b) M. Sato et al., ICCG14-0416