Instabilities at vicinal crystal surfaces: Competition between electromigration of adatoms and kinetic memory effect

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We studied the step dynamics during sublimation and growth when the adatoms on the crystal surface have a drift velocity $D_s F/kT$, where D_s is the surface diffusion coefficient and F is a force acting on the adatoms (F is related to the electric current heating the crystal). In the limit of fast surface diffusion and slow kinetics of atom attachment-detachment at the steps, we formulate a model that is free of the quasistatic approximation in the calculation of the adatom concentration on the terraces. The linear stability analysis of a step train results in an instability condition in the form $-(f\tau_s'/3\varepsilon)+(V/V_{cr})-1>0$, where τ_s' is the dimensionless lifetime of an adatom before desorption, f and ε are the dimensionless electromigration force and the force of step repulsion, respectively, and V and V_{cr} are the velocity of steps in the train and the critical velocity, respectively. As seen, instability is expected when either the velocity V is larger than V_{cr} (this instability is related to the "kinetic memory effect") or $-f\tau_s'/3\varepsilon>1$, i.e., when the electromigration force f is negative (but strong enough to dominate over the term V/V_{cr}), which means step-down direction of the drift. In the latter case, the initial stage of the step bunching process dramatically depends on the value of the ratio $-f/\varepsilon$. When $-f/\varepsilon \gtrsim 1$ the instability starts with a formation of small bunches (or pairs) of steps, whereas at $-f/\varepsilon \ll 1$, the instability starts with a formation of relatively large bunches containing $N_{initial}=2\pi/q_{max}=2\pi/(-f/3\varepsilon)^{1/2}$ steps. A numerical integration of the equations for the time evolution of the adatom concentrations and the equations of step motion reveals two different step bunching instabilities: (1) step density waves (small bunches that do not manifest any coarsening) induced by the kinetic memory effect and (2) step bunching with coarsening (eventually leading to a formation of very large bunches) when the dynamics is dominated by the electromigration. For the latter case, we obtained very instructive illustrations of the dynamical phase transition [V. Popkov and J. Krug, Phys. Rev. B **73**, 235430 (2006)] during sublimation and growth of a vicinal crystal surface.

I. INTRODUCTION

In a recent paper,[1] we advanced a new model for the step dynamics during sublimation and growth of a vicinal crystal surface. This model contains the same physics as the classical Burton–Cabrera–Frank (BCF) theory[2–4] but the mathematical treatment deviates from the BCF procedure. In contrast to their assumption that the adatom concentrations n_i on the terraces instantly reach their steady state for a given step configuration, we analyzed the non-steady-state problem.[1] This is relatively easy in the limiting case of fast surface diffusion and slow kinetics of atom attachment and detachment at the steps, since the fast diffusion provides for a constant value of the adatom concentration all over a given terrace. We derived equations for the time evolution of the adatom concentration on the terraces and equations for the step motion. In this way, we were able to accurately treat the case when the time to reach steady-state concentration of adatoms on the terraces is compatible with the time for non-negligible change in the step configuration. In such a situation, a new effect becomes important—the adatom concentration on a given terrace depends not only on the terrace size but on the "past of the terrace" as well. We call this a "kinetic memory effect". This effect provides a ground for a new type of instability of the regular step distribution. Step density compression waves appear at the vicinal surface with fast moving steps, as shown by both linear stability analysis and numerical integration of the equations for step motion.[1] In this way, we proved that as far as the stability of a vicinal surface is concerned, the quasistatic approximation fails (it does not predict any instability) when the velocity V of the steps exceeds its critical value V_{cr}. Therefore, it is of interest to reexamine the step bunching process, which is induced by the electric current heating the crystal, by going beyond the quasistatic approximation. Here, our aim is to see how the kinetic memory effect competes with another (well known) destabilizing factor—the electromigration of the adatoms.[5–11]

It is essential to mention earlier attempts to go beyond the quasistatic approximation in the BCF model and to clearly point out the differences from our treatment. The earlier attempts[12,13] were focused on finding the adatom concentration field $n_i(x)$ at the ith terrace on the basis of a modification of the classical Stefan problem. It is clear on physical ground that the concentration field is not symmetric (with respect to the middle of the terrace) because of the motion of the steps. This asymmetry could lead to step bunching instability when the velocity of steps is high enough. In a very recent paper, Dufay *et al.*[14] discussed the role of step flow advection effect and its competition with electromigration induced step bunching. Let us clearly say that our treatment does not account for the advection effect since we focus our attention on the limit of the fast surface diffusion and slow kinetics at the steps (the advection effect is negligible in this limit). The physics of the instability we reported[1] is related to the delay in establishing the adatom concentration that corresponds to the step distribution in the given moment. This retardation effect in achieving the steady-state concentration of adatoms generates step density waves during growth and sublimation of crystals.

II. BASIC EQUATIONS

It is relatively easy to extend the treatment in Ref. 1 to account for the electromigration of the adatoms. The model is based on the following assumption: adatoms are subject of electromigration force (see Fig. 1) so that they more frequently jump in the direction of this force than in the opposite direction.[6,15] In addition, one assumes the surface diffusion to be much faster than the atom attachment-detachment kinetics at the steps. Since the electromigration force pushes the adatoms toward one of the steps that separate the terrace from its neighbors, the concentration close to this step is higher than the average concentration over the whole terrace. Therefore, more atoms attempt to attach to this step and become "crystal atoms." On the contrary, at the other step of the same terrace, the concentration of adatoms is lower and less atoms attempt to join the crystal. This simple physics should be incorporated into the equations for the time evolution of the terrace widths. In a linear approximation, the adatom concentration on a given terrace is $n_i(\xi,t)=n_i(t)[1+(F\xi/kT)]$, with $\xi=0$ being in the middle of the terrace. In this approximation, $n_i(t)$ is equal to the average value of the adatom concentration on the terrace.

The space nonuniformity of the adatom concentration has an impact on the rates of step motion. Really, now we have

$$v_i = -K\Omega\left[n_i\left(1 - \frac{Fl_i}{2kT}\right) - n_s^e(i)\right] - K\Omega\left[n_{i-1}\left(1 + \frac{Fl_{i-1}}{2kT}\right) - n_s^e(i)\right], \quad (1)$$

where K is the step kinetic coefficient (see Fig. 1) and Ω is the area of one atomic site at the crystal surface. On the basis of Eq. (1), one can write the expression for the time derivatives of the width of the terraces as

$$\frac{dl_i}{dt} = -\Omega\left\{K\left[n_{i+1}\left(1 - \frac{Fl_{i+1}}{2kT}\right) - n_s^e(i+1)\right] + K\left[n_i\left(1 + \frac{Fl_i}{2kT}\right) - n_s^e(i+1)\right] - K\left[n_{i-1}\left(1 + \frac{Fl_{i-1}}{2kT}\right) - n_s^e(i)\right] - K\left[n_i\left(1 - \frac{Fl_i}{2kT}\right) - n_s^e(i)\right]\right\}, \quad (2)$$

where n_i are functions of time.

As known, the equilibrium concentration of adatoms in the vicinity of the steps has the constant value n_s^e only when $l_i=l_{i-1}=l$, with l as the average terrace width determined by the miscut angle of the vicinal surface. In the general case, one has[16,17]

$$n_s^e(i) = n_s^e\left[1 + \tilde{A}\left(\frac{1}{l_{i-1}^3} - \frac{1}{l_i^3}\right)\right], \quad (3)$$

where $\tilde{A}=2\Omega A/kT$ and A is the strength of the entropic and stress-mediated repulsion between the steps.
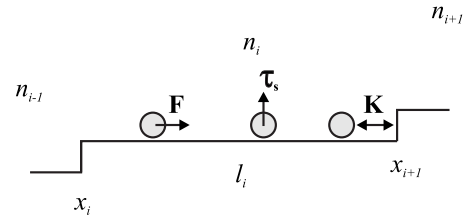

FIG. 1. Schematic view of a vicinal surface with straight steps. The adatoms have a drift velocity induced by an electric force F. The width of the ith terrace is $l_i=x_{i+1}-x_i$ and the concentration of adatoms on it is n_i. The desorption flux from the terrace is n_i/τ_s and the exchange between the crystal phase and the dilute layer of adatoms is characterized by the step kinetic coefficient K.

By introducing the dimensionless variables $\tau=Kt/l$, $\eta_i=l_i/l$, $c_i=n_i/n_s^e$, and $\varepsilon=\tilde{A}/l^3$, we obtain for the dimensionless width $\eta_i(\tau)$ of the ith terrace (details on the derivation of these equations can be found in Ref. 1),

$$\frac{d\eta_i}{d\tau} = -n_s^e\Omega\left\{c_{i+1} - c_{i-1} - f(\eta_{i+1}c_{i+1} - 2\eta_i c_i + \eta_{i-1}c_{i-1}) + 2\varepsilon\left(\frac{1}{\eta_{i+1}^3} - \frac{2}{\eta_i^3} - \frac{1}{\eta_{i-1}^3}\right)\right\}, \quad (4)$$

where $f=Fl/2kT$. The time evolution of the dimensionless concentrations of adatoms is given by

$$\frac{dc_i}{d\tau} = \frac{c_{st}}{\tau_s'} - \frac{c_i}{\tau_s'} - \frac{2}{\eta_i}c_i + \frac{2}{\eta_i} + \frac{1}{\eta_i}\varepsilon\left(\frac{1}{\eta_{i-1}^3} - \frac{1}{\eta_{i+1}^3}\right), \quad (5)$$

where $\tau_s'=\tau_s K/l$ and τ_s are the average time for desorption of an atom in a state of mobile adsorption on the crystal surface (see Fig. 1).

The term c_{st}/τ_s' accounts for the deposition of atoms on the crystal surface since $c_{st}=R\tau_s/n_s^e$. When the deposition rate R has its equilibrium value R_e, one obtains $c_{st}=R_e\tau_s/n_s^e=1$. This means that $c_{st}<1$ corresponds to undersaturation and $c_{st}>1$ corresponds to supersaturation at the crystal surface.

The other terms in Eq. (5) also have a clear physical meaning—the adatom concentration decreases because of the desorption (the second term on the right hand side) and attachment of adatoms to the steps (the third term). The concentration c_i of adatoms increases because of the detachment of atoms from the steps [the next terms on the right hand side of Eq. (5)]. One should keep in mind, however, that now $c_i(\tau)$ is the average value of the dimensionless adatom concentration on the ith terrace.

It is essential to point out that our model completely neglects the transparency of the steps and Eq. (5) does not contain any terms describing the contribution of the exchange of adatoms between the neighboring terraces. This circumstance determines the temperature intervals of validity of our model. Really, the quasistatic treatment of the model with nontransparent steps[6,7] successfully reproduces the experimental data[5,8,10] on step bunching in the temperature intervals around 950 and 1250 °C (in both intervals bunching takes place at the step-down direction of the drift of the

adatoms). Therefore, the validity of our present results is restricted to these two temperature intervals. The impact of the step transparency will be considered elsewhere.

III. LINEAR STABILITY ANALYSIS

The simplest solution of Eqs. (4) and (5) is $\eta_i = 1$ and $c_i = c_0 = 1/1 + 1/2\tau'_s$, which is an equidistant step distribution and the corresponding constant concentration of adatoms in the absence of deposition rate (the impact of deposition will be discussed later). To study the stability of this solution, we follow the routine procedure and consider small deviations from the equidistant step train and constant adatom concentrations, i.e., $\eta_i = 1 + \Delta\eta_i(\tau)$ and $c_i = c_0 + \Delta c_i(\tau)$. By substituting these expressions into Eqs. (4) and (5), making use of series expansion, and keeping only the linear terms, we get

$$\frac{d\Delta\eta_i}{d\tau} = -n_s^e\Omega[\Delta c_{i+1} - \Delta c_{i-1} - (fc_0 + 6\varepsilon)(\Delta\eta_{i+1} - 2\Delta\eta_i + \Delta\eta_{i-1}) - f(\Delta c_{i+1} - 2\Delta c_i + \Delta c_{i-1})],$$ (6)

$$\frac{d\Delta c_i}{d\tau} = -\frac{\Delta c_i}{\tau'_s} - 2\Delta c_i + 2(c_0 - 1)\Delta\eta_i + 3\varepsilon(\Delta\eta_{i+1} - \Delta\eta_{i-1}).$$ (7)

Following the routine, we look for a solution of the type $\Delta\eta_j = e^{ijq}\eta_q(\tau)$ and $\Delta c_j = e^{ijq+i\phi}c_q(\tau)$, where q is a wave number and we already use i to denote the imaginary unit and j to denote the sequence number of the terrace. In addition, we allow for a phase shift ϕ of the wave describing the adatom concentrations Δc_j with respect to the wave, describing the terrace widths $\Delta\eta_j$. In this way, we arrive at a set of two differential equations for the time evolution of the amplitudes of the fluctuations in the terrace width distribution and adatom concentrations,

$$\frac{d\eta_q}{d\tau} = a_{11}\eta_q(\tau) + a_{12}c_q(\tau),$$

$$\frac{dc_q}{d\tau} = a_{21}\eta_q(\tau) + a_{22}c_q(\tau),$$ (8)

where the coefficients are given by the following expressions:

$$a_{11} = -2n_s^e\Omega(6\varepsilon + fc_0)(1 - \cos q),$$

$$a_{12} = -2e^{i\phi}n_s^e\Omega[i\sin q + f(1 - \cos q)],$$

$$a_{21} = 2e^{-i\varphi}[-(1 - c_0) + 3i\varepsilon\sin q],$$

$$a_{22} = -(2 + 1/\tau'_s).$$ (9)

This set of two linear differential equations has a solution of the type $e^{s\tau}$ and the real part of s is

$$s_r = 2n_s^e\Omega\left[-(6\varepsilon + fc_0)(1 - \cos q) + 2\frac{3\varepsilon\sin^2 q + f(1 - c_0)(1 - \cos q)}{(a_{11} - a_{22})} + 8\frac{(n_s^e\Omega)\sin^2 q[-(1 - c_0) + 3\varepsilon f(1 - \cos q)]^2}{(a_{11} - a_{22})^3}\right]$$ (10)

(details on the derivation of a similar expression when $f=0$ are given in Ref. 1).

By restricting the treatment to small wave numbers and the limit $\tau'_s \gg 1$, we obtain

$$s_r = B_2 q^2 - B_4 q^4,$$ (11)

where

$$B_2 = \frac{3n_s^e\Omega\varepsilon}{\tau'_s}\left[-1 - \frac{f\tau'_s}{3\varepsilon} + \frac{n_s^e\Omega}{6\varepsilon\tau'_s}\right],$$ (12)

$$B_4 = \frac{3n_s^e\Omega\varepsilon}{\tau'_s}\left[\frac{\tau'_s}{2} - \frac{f\tau'_s}{36\varepsilon} + \frac{n_s^e\Omega}{18\tau'_s\varepsilon}\right].$$ (13)

As shown above, Eqs. (11)–(13) can be reduced to the already known expressions (see Ref. 1) when the electromigration force F is zero and, therefore, $f=Fl/2kT=0$. In Ref. 1, we used the notation $V=n_s^e\Omega l/\tau_s$ for the velocity of steps in a perfectly regular train and $V_{cr} = 6K\varepsilon$ for the critical velocity.

Now, we are in a position to discuss the impact of the electromigration on the step dynamics. To have an instability of the step train (at small wave numbers, at least), we need a positive value of the constant B_2 [see Eq. (11)]. This could be achieved in two ways—either $n_s^e\Omega/6\varepsilon\tau'_s = V/V_{cr}$ is large enough to ensure a positive sign of the expression in the brackets of Eq. (12) or $f<0$ and the absolute value of $f\tau'_s/3\varepsilon$ dominates the term $n_s^e\Omega/6\varepsilon\tau'_s$ and is large enough to compensate -1. In the former case, we expect step density compression waves to propagate at the vicinal surface, as shown in Ref. 1. In the latter case, the step dynamics is expected to be dominated by the adatom electromigration effect. As for the value of B_4, it is definitely positive at $f<0$ [see Eq. (13)].

It is interesting to note that Eq. (12) defines a critical value of the electromigration force. Really, instability induced by electromigration takes place only when $f\tau'_s/3\varepsilon > 1$, which is equivalent to $F > F_c = 12\Omega A/K\tau_s l^3$. This expression for F_c was already published by Uwaha et al.[18]

In the presence of deposition rate, the linear stability criterion is slightly modified in the sense that the expression for B_2 now contains the quantity $c_{st} = R\tau_s/n_s^e$. Simple considerations result

$$B_2 = \frac{3n_s^e\Omega\varepsilon}{\tau'_s}\left[-1 - \frac{f\tau'_s}{3\varepsilon}\left(1 + \frac{c_{st}}{\tau'_s}\right) + \frac{n_s^e\Omega}{6\varepsilon\tau'_s}(1 - c_{st})^2\right].$$ (14)

Since the modifying factors $[1 + (c_{st}/\tau'_s)]$ and $(1 - c_{st})^2$ are always positive, the deposition rate does not qualitatively change the instability criterion. Step bunching with coarsen-

ing occurs at $f < 0$, i.e., step-down direction of the electric force when the step dynamics is dominated by the atom electromigration. A formation of small bunches that do not show coarsening takes place in both growth ($c_{st} > 1$) and sublimation ($c_{st} < 1$) when the dominating factor is the kinetic memory effect. The deposition rate is expected to have an impact only on the transition from one type of instability to the other one. As seen, the increase in the deposition rate (which means an increase in c_{st}) leads to large values of the last term in square brackets in Eq. (14). In other words, the increase in the deposition rate favors the formation of step density waves that are generated by the kinetic memory effect.

Equations (12) and (14) predict a stabilization effect of an electromigration force with step-up direction, i.e., $f > 0$. In other words, one can suppress the formation of step density waves, when the step velocity is higher than its critical value by manipulating the direction of the electric current heating the Si wafer.

Finally, for the wave number of the most unstable mode (the maximum value of s_r) during electromigration dominated sublimation, one obtains the following approximate expression:

$$q_{max} = \left[\frac{B_2}{2B_4}\right]^{1/2} \approx \left[\frac{-\dfrac{f}{3\varepsilon}}{1 - \dfrac{f}{18\varepsilon}}\right]^{1/2}. \qquad (15)$$

This expression has a simple limit—at $|f/18\varepsilon| \ll 1$, the most unstable mode has a wave number,

$$q_{max} = \left[-\frac{f}{3\varepsilon}\right]^{1/2}, \qquad (16)$$

which is also small; i.e., the instability starts with fluctuations having a large wavelength. The last expression provides a ground to conclude that the wavelength of the most unstable mode depends on the average terrace width l of the vicinal surface. Really, having in mind the definitions of f and ε, we find $q_{max} \sim l^2$ and, therefore, the corresponding wavelength of the most unstable mode

$$\lambda_{max} = j_{max}l = \frac{2\pi}{q_{max}}l \sim \frac{1}{l} \qquad (17)$$

is inversely proportional to the average terrace width.

According to Eq. (15), the increase in $|f/18\varepsilon|$ leads to an increase in the wave number q_{max} and the step bunching instability starts with the formation of very small bunches or pairs of steps, which later coalesce to form larger bunches.

IV. LONG TIME BEHAVIOR OF VICINAL SURFACES

We study the nonlinear dynamics of the steps by numerical integration of Eqs. (4) and (5). Our aim is to look for a confirmation of the results that are obtained by the linear stability analysis. On the other hand, it is interesting to explore the capability of this model (free of the quasistatic approximation) to manifest the dynamic phase transition that was predicted by Popkov and Krug.[19]
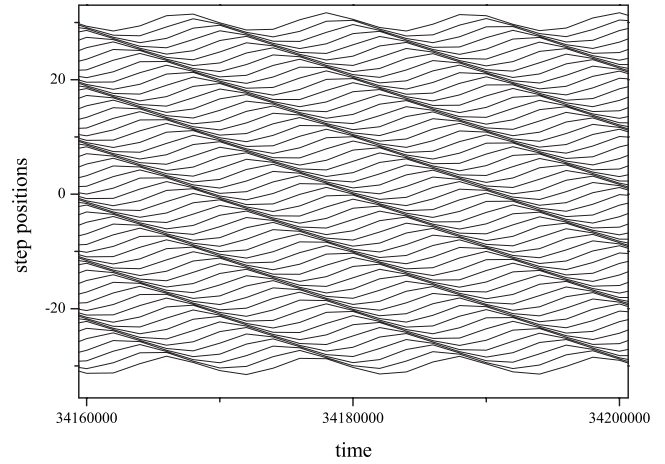

FIG. 2. Step trajectories (sublimation) in a frame moving with a velocity averaged over all of the steps in the system (containing 60 steps). Compression waves propagate when the model parameters are $\tau'_s = 100$, $\varepsilon = 10^{-5}$, $n^e_s\Omega = 0.1$, and $f = -10^{-4}$.

The set of Eqs. (4) and (5) involves $2N$ variables because we use circling boundary conditions $\eta_{N+1} = \eta_1$ and $c_{N+1} = c_1$. To obtain an instructive picture of the sublimation dynamics, we calculate the step trajectories in a frame moving with the average velocity of the step train (containing N steps). The calculated trajectories clearly show (Fig. 2) the propagation of step density waves (due to a kinetic memory effect) and the formation of bunches of steps (Fig. 3) due to the electromigration of the adatoms. Some comments on these two instabilities will be made on the basis of Eq. (12). By substituting the values of the model parameters $\tau'_s = 100$, $\varepsilon = 10^{-5}$, $n^e_s\Omega = 0.1$, and $f = -10^{-4}$ (which we used to obtain the trajectories shown in Fig. 2) into Eq. (12), we see that $-f\tau'_s/3\varepsilon = 333.3$ and $n^e_s\Omega/6\varepsilon\tau'_s = 16.6$.

As seen above, the term containing the electromigration force has a much stronger contribution to the amplitude of fluctuations [see Eqs. (11) and (12)]. On this basis, one intu-
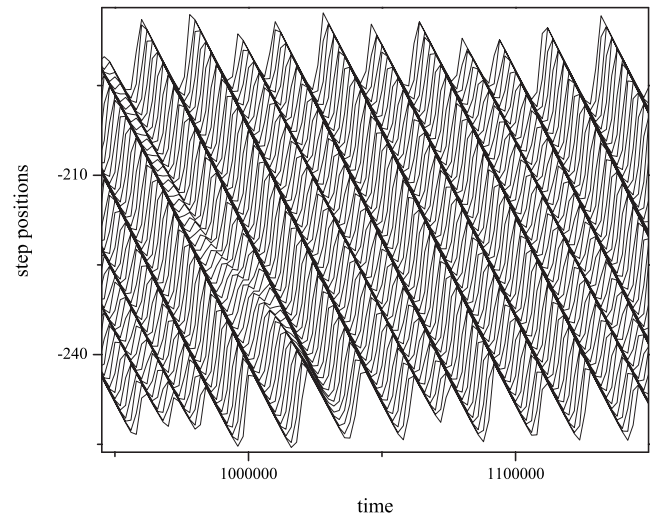

FIG. 3. Step trajectories (sublimation) manifest coarsening—step bunches grow in size with the sublimation time. The model parameters are $\tau'_s = 100$, $\varepsilon = 10^{-5}$, $n^e_s\Omega = 0.1$, and $f = -10^{-3}$.
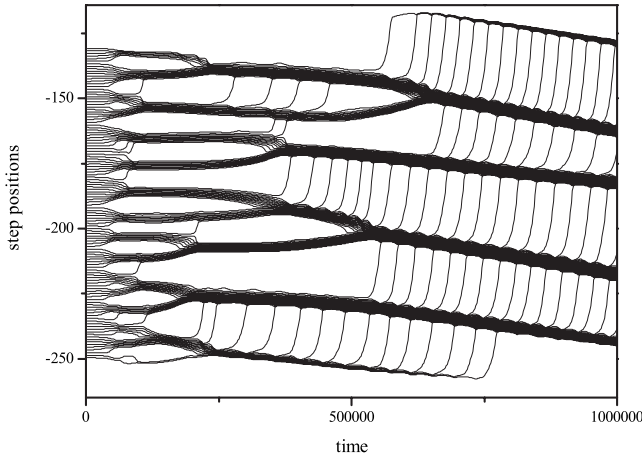
FIG. 4. The step trajectories (during sublimation) obtained at $n_s^e\Omega=0.1$, $f=-2.0\times10^{-3}$, $\varepsilon=10^{-3}$, and $\tau_s'=2000$ manifest three interesting features of the step dynamics: (1) long wavelength instability, (2) existence of a dynamic regime with all steps involved in bunches (Ref. [19]), and (3) existence of critical size above which the bunch starts to emit single steps (Ref. [19]).

itively expects the step bunching instability induced by electromigration to dominate the step dynamics. Contrary to these expectations, the nonlinear dynamics manifests step density waves, which are characteristic for the kinetic memory effect (no coarsening is seen in Fig. 2). It is essential to point out that Fig. 2 manifests the failure of the quasistatic approximation when the step velocity is high enough ($n_s^e\Omega/6\varepsilon\tau_s'=V/V_{cr}=16.6$). Really, the theory of electromigration affected step dynamics[7] makes use of this approximation and predicts bunching of steps at the value $f=-10^{-4}$, which is used in Fig. 2. This prediction is in clear contradiction with the trajectories (shown in Fig. 2) manifesting the existence of step density waves but not of bunches and coarsening.

To study the transition from step density wave instability to step bunching (with coarsening) instability, we gradually increased the absolute value of the parameter f and integrated Eqs. (4) and (5) at $f=-2\times10^{-4}$ and $f=-3\times10^{-4}$. The results manifest the formation of step density waves, which display very slow coarsening (at $f=-3.0\times10^{-4}$, coarsening is a bit faster). A further increase in the absolute value of the parameter f (see Fig. 3) leads to coarsening with a substantial rate.

It is interesting to check the results of the linear stability analysis for the most unstable mode in the electromigration dominated bunching of steps. Equations (15)–(17) and the discussion after them predict the bunching process to start either with a formation of small bunches (or even pairs) or with a formation of much larger bunches depending on the value of the parameter $|f/18\varepsilon|$. A good illustration of long wavelength instability is provided by Fig. 4, which shows the step trajectories when $\tau_s'=2000$, $\varepsilon=10^{-3}$, $n_s^e\Omega=0.1$, and $f=-2\times10^{-3}$. By substituting these values into the ratio $|f/18\varepsilon|$, one obtains $1/9$, which is much smaller than unity so that the wave number of the most unstable mode is given by Eq. (16). Thus, one obtains $q_{max}=[-f/3\varepsilon]^{1/2}=\sqrt{2/3}=0.816$. The corresponding wavelength measured in number

of terraces is $j_{max}=2\pi/q_{max}\approx7.7$, which is in good agreement with the results shown in Fig. 4.

Figure 4 shows the trajectories when the average step velocity is much smaller than its critical value ($V/V_{cr}<0.01$). Under this condition, the kinetic memory effect is negligible and the quasistatic assumption is a good approximation. That is why Fig. 4 successfully reproduces the results for the most unstable mode already obtained [see the Eqs. (33) and (60) in Ref. [20] and Eq. (4) in Ref. [21]] in the treatment based on the quasistatic approximation.

Finally, it is interesting to explore the capability of this model to manifest dynamic phase transition that was predicted by Popkov and Krug[19] on the basis of the quasistatic approximation. The dynamic phase transition is, in fact, a transition between two regimes of step dynamics, which are qualitatively different. In the most clear situation, one of the regimes is characterized by the absence of single steps at the crystal surface, i.e., all steps are in bunches (such a situation is shown in Fig. 4 for a relatively short time of evaporation). The second regime can be described as a coexistence of bunches and single steps crossing the terraces [an essential requirement is to have several steps simultaneously crossing each terrace (see Fig. 4)]. Popkov and Krug[19] found the transition to take place at the critical value $b=1$ of the asymmetry parameter b, which is defined by the following expression:[19]

$$\frac{dx_i}{dt}=\frac{1-b}{2}(x_{i+1}-x_i)+\frac{1+b}{2}(x_i-x_{i-1})+U, \qquad (18)$$

where x_i is the position of the ith step, U accounts for the step-step repulsion, and the time scale is normalized to the sublimation rate. To express b through the parameters of our model, we rewrite Eq. (1) to introduce dimensionless parameters and make use of the steady-state solution $c_i=1/1+\eta_i/2\tau_s'$ of Eq. (5) at $c_{st}=0$. Thus, for the limit $\tau_s'\gg1$, one obtains the following expression for the step rate during sublimation (the deposition rate is zero):

$$\frac{dx_i}{dt}=-K\Omega n_s^e\{[c_i(1-f\eta_i)-1]+[c_{i-1}(1+f\eta_{i-1})-1]\}$$

$$=K\Omega n_s^e\left\{\frac{\eta_i}{2\tau_s'}(1+2f\tau_s')+\frac{\eta_{i-1}}{2\tau_s'}(1-2f\tau_s')\right\}. \qquad (19)$$

After appropriate normalization of the time scale, Eq. (19) can be transformed into Eq. (18) with $b=-2f\tau_s'$.

Our numerical integration of Eqs. (4) and (5) reproduces the results of Popkov and Krug.[19] Figure 4 manifests the existence of a regime of step dynamics, where all of the steps are in bunches (there are no steps crossing the terraces) at $b=-2f\tau_s'=8$. Figure 4, which is obtained at values $n_s^e\Omega=0.1$, $f=-2.0\times10^{-3}$, $\varepsilon=10^{-3}$, and $\tau_s'=2000$, manifests another interesting feature of the step dynamics, which was predicted by Popkov and Krug[19]—when the bunch exceeds some critical size it starts to emit single steps. It is essential to say that a somewhat softer definition of the "bunched phase" was used in Ref. [19]—steps crossing the terraces are not completely excluded. In the original paper[19] the requirement is to have no more than one step crossing the terrace at
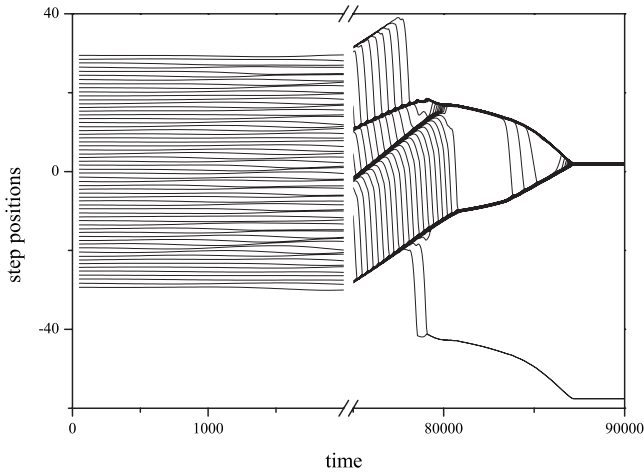
FIG. 5. Step trajectories during crystal growth (the model parameters are $\tau'_s = 100$, $\varepsilon = 10^{-3}$, $n^e_s \Omega = 0.01$, $c_{st} = 50$, and $f = -0.04$) clearly manifest a dynamic phase transition (Ref. [19]). This transition is accompanied by a remarkable compression of the bunches.

a time. This softer definition requires smaller asymmetry parameter ($b=1$) to obtain bunched phase (our Fig. 4 is obtained at $b=8$).

V. INSTABILITIES DURING GROWTH

During growth, one could reach higher rates of step motion in comparison to the crystal sublimation. Therefore, one could expect the quasistatic approximation to more frequently fail. It is of interest to study the nonlinear dynamics of steps in the presence of deposition rate. Figure 5 shows the trajectories of the steps, which are obtained by integrating Eqs. (4) and (5) for $\tau'_s = 100$, $\varepsilon = 10^{-3}$, $n^e_s \Omega = 0.01$, $c_{st} = 50$, and $f = -0.04$. Let us immediately point out that this value of the electromigration parameter f is unrealistically high (about 2 orders of magnitude higher than the estimations given in Refs. 8 and 10). That is why one cannot expect the results shown in Fig. 5 to reproduce real experimental observations. We include this figure in the paper just to show the behavior of our model in the limit of very strong electromigration of the adatoms. In this sense, Fig. 5 contains interesting results. Step trajectories show the initial formation of pairs (short wavelength instability), which later coalesce to form larger bunches (hierarchical bunching). Single steps detach from the bunches and quickly cross the terraces to join the bunch ahead. This familiar picture of coexisting bunches and single steps is suddenly transformed into a single bunch containing all 60 steps in the system studied by numerical integration of the equations of step dynamics. The sharp transformation is a very instructive manifestation of the Popkov–Krug dynamical phase transition. The more familiar (and frequently observed) scenario is based on a long wavelength instability with bunches that do not exchange single steps until they reach their critical size (see, for instance, Figs. 4 and 6).

When the electromigration of the adatoms is not so strong the time evolution of the train of steps during the crystal growth (see Fig. 6) is different from that shown in Fig. 5.
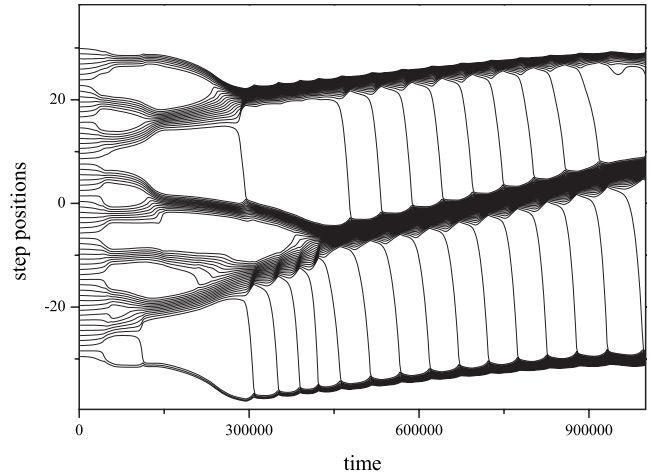

FIG. 6. Step trajectories during crystal growth (model parameters are $\tau'_s = 10000$, $\varepsilon = 10^{-3}$, $n^e_s \Omega = 0.1$, $c_{st} = 5$, and $f = -0.002$).

Here, (in Fig. 6), we have more realistic values of the electromigration force. The further decrease in the electromigration force leads to a step dynamics dominated by the kinetic memory effect and the formation of step density waves, as shown in Fig. 7.

VI. CONCLUSIONS

Going beyond the quasistatic approximation in the treatment of step dynamics, we see the vicinal surface to manifest two different step bunching instabilities. The formation of small bunches of constant size (absence of any coarsening) occurs when the electromigration is relatively weak and the step dynamics is dominated by the kinetic memory effect (see Figs. 2 and 7). This is, in fact, the instability (the formation of step density waves) reported in our previous paper.[1] Another type of instability (step bunches manifest coarsening so that the average bunch size increases with the
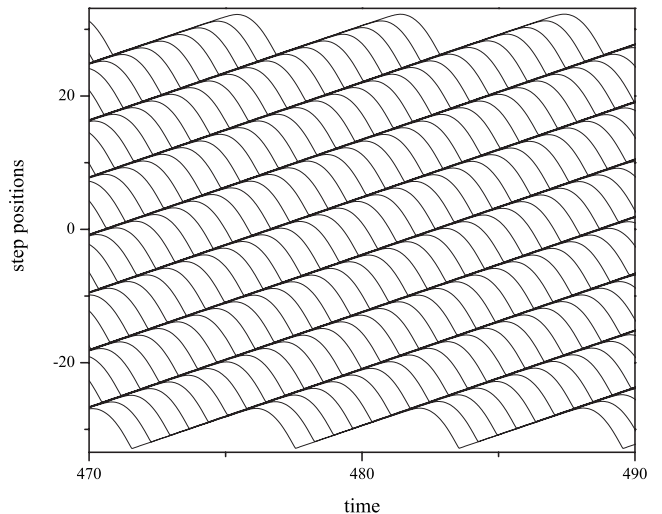

FIG. 7. Step trajectories during crystal growth. The model parameters have the values $\tau'_s = 10$, $c_{st} = 500$, $\varepsilon = 10^{-3}$, $n^e_s \Omega = 0.1$, and $f = -10^{-5}$.

sublimation time) occurs when the electromigration of adatoms is the dominating factor and the kinetic memory effect has a secondary role in the step dynamics. It is interesting to note that these two factors play identical roles in the linear stability theory where the instability condition reads $-(F/F)_c + (V/V_{cr}) - 1 > 0$, with $F_c = 12\Omega A/K\tau_s l^3$ [see Eqs. (11) and (12)]. As far as the nonlinear dynamics of steps is concerned, these two factors have a rather different impact. The electromigration of adatoms ($F \gg F_c$) leads to the formation of large bunches which manifest clear coarsening, whereas the deviation from the equilibrium ($V \gg V_{cr}$) produces a regular array of small bunches of constant size (there is no coarsening at all).

The step bunching due to the factor F/F_c is similar to the already studied[22] instability induced by electromigration in the limit of slow step kinetics and fast surface diffusion (the model in Ref. 22 makes use of the quasistatic approximation). Analyzing this instability, we reproduce already published (by other authors) results for this limit. For example, our Eq. (16) coincides with Eqs. (33) and (60) in Ref. 20 and Eq. (4) in Ref. 21 These results of the linear stability analysis for the most unstable mode are supported by Figs. 4 and 5, which show the step trajectories that are obtained by numerical integration of Eqs. (4) and (5). Finally, our non-steady-state treatment of the step dynamics at the crystal surface manifests the dynamic phase transition that was predicted by Popkov and Krug for the model based on the quasistatic approximation.[19]

In conclusion, one should say that the quasistatic approximation is quite relevant when the velocity of the steps does not exceed its critical value. As far as the experiments[8,10,11] aimed at revealing the fundamentals of the step bunching instability are concerned a clear coarsening, which leads to a gradual increase in the number of steps in the bunch has been reported; i.e., the step kinetics is dominated by the electromigration. The experimental results for the shape of the bunches and the scaling relation between the width of the bunch and the number of steps in it are in good agreement with the theoretical results[7] that are obtained in the framework of the quasistatic approximation. To reach the experimental conditions where the step dynamics is not dominated by the electromigration of the adatoms, one should decrease the electric current through the crystal and keep the temperature high enough by some additional heating (for instance, irradiative heating of the wafer). It is clear, however, that in the molecular beam epitaxy growth of Si, for instance, the rate of crystal growth (and therefore the rate of step motion) is a key technological parameter. To obtain the required thickness of a layer in an acceptable deposition time, one could need a step velocity much higher than the critical one. Then, the step density waves will appear at the growing crystal surface.

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