

Non-conserved dynamics of steps on vicinal surfaces during electromigration-induced step bunching

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Abstract. We report new results on the non-conserved dynamics of parallel steps on vicinal surfaces in the case of sublimation with electromigration and step-step interactions. The derived equations are valid in the quasistatic approximation and in the limit $f^{-1} \gg l_D \gg l_{\pm} \gg l_i$, where f is the inverse electromigration length, l_D the diffusion length, l_{\pm} the kinetic lengths and l_i the terrace widths. The coupling between crystal sublimation and step-step interactions induces non-linear, non-conservative terms in the equations of motion. Depending on the initial conditions, this leads to interrupted coarsening, anticoarsening of step bunches or periodic switching between step trains of different numbers of bunches.

1 Introduction

For the theoretical study of homoepitaxial growth and sublimation of a crystal in contact with the gas phase it is important to have a model, which includes the kinetic processes and the different effects existing on the crystal surface. The classical model for the evolution of vicinal surfaces was introduced by Burton, Cabrera and Frank (BCF) [1]. It is based on the observation that the kink sites are those positions at the surface steps where the exchange between the adatom layer on the terraces and the solid phase takes place. On the mesoscopic scale the change of the crystal volume is a result of the movement of the steps. On this scale we can reduce a surface with straight steps to a one-dimensional step train. Such a surface may undergo *step bunching*, an instability where the steps move close to each other and form groups, called step bunches [2–5].

The theoretical description of step bunching instabilities within the framework of the BCF-model and its extensions has been the subject of much recent interest [6–14]. Here we focus specifically on the effect of non-conservative processes on the non-linear evolution of a step train. As we reported in [14] for the problem of sublimation in the presence of Ehrlich-Schwoebel (ES) barriers [15,16], non-conservative terms violating volume conservation in the co-moving frame arise generically from the interplay of sublimation and step-step interactions, and cause the interruption of the coarsening of the growing bunches or splitting of a large bunch into several smaller bunches. In the present paper we expand this analysis to include the experimentally relevant effect of surface electromi-

gration [2,5,17–27]. In 1989, Latyshev and collaborators discovered that by changing the direction of the direct heating current, a vicinal Si(111) surface switches between bunching and debunching [17]. Additionally, they observed several distinct temperature regimes. In the so called regimes I and III [5] the bunching instability occurs only if the heating current is applied in the down-step direction. On the other hand, for the same direction in regime II debunching occurs, and bunching requires an up-step current. Here, we consider the first temperature regime, where the temperature is low enough in order to neglect step transparency (the motion of adatoms across steps) [5,8,25].

Interrupted coarsening of electromigration-induced step bunches in the presence of sublimation was previously observed numerically by Sato and Uwaha [6], however a detailed analysis of the phenomenon was not carried out due to the complexity of their model. Other studies have approached the problem within the framework of weakly nonlinear amplitude equations, which can be systematically derived by an expansion around the instability threshold [5]. In this setting the non-conserved dynamics is described on large scales by the Benney equation, which displays either spatio-temporal chaos or an ordered array of bunches, but no coarsening [28,29]. This macroscopic behavior is consistent with the complex mesoscopic step dynamics revealed in the present work.

The paper is organized as follows. First, we sketch the derivation of the discrete step equations for the case of attachment-detachment limited kinetics and present the result of the linear stability analysis for very large wave lengths. Additionally, for comparison, we write down the corresponding equations for the case of growth. We then discuss the discrete equations and their continuum limits

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for two special cases, where the kinetic asymmetry between ascending and descending steps is caused solely by an ES-effect or by electromigration, respectively. Finally, we show the results of numerical simulations of the discrete step equations for the case with electromigration.

2 Model

We consider an ascending one-dimensional step train with step edges located at positions x_i (Fig. 1). The starting point for the derivation of the equations of motion for the steps is the balance equation

$$\frac{\partial n_i}{\partial t} = D_s \left[\frac{\partial^2 n_i}{\partial x^2} - f \frac{\partial n_i}{\partial x} \right] - \frac{n_i}{\tau} + F \stackrel{!}{=} 0 \quad (1)$$

for the concentration of adatoms $n_i(x, t)$ on the i th terrace of width $l_i = x_{i+1} - x_i$. Here D_s is the surface diffusion constant, and τ is the average life time of an adatom before desorption. Together these two quantities define the diffusion length $l_D = \sqrt{D_s \tau}$, which sets the scale of spatial variation for the adatom concentration. The terrace is losing adatoms due to desorption at rate $1/\tau$ and gaining adatoms because of deposition with constant flux F . The applied direct heating force F_{el} causes a drift of adatoms for which we can use the Einstein relation and define a further length scale, the so called electromigration length $f^{-1} = k_B T / F_{el}$ [6,18]. As is common in the field, we assume that the adatom concentration adjusts instantaneously to the slowly moving steps, an assumption that is known as the quasistatic approximation and amounts to setting $\partial_t n_i(x, t) = 0$ in (1). An approach that goes beyond this approximation was recently presented by Rangelov and Stoyanov [24,25].

The general solution $n_i(x)$ of the ordinary differential equation (1) can be specified using mass conservation at the steps as boundary conditions. A terrace of width l is bounded by two steps with positions $x = \pm l/2$, at which the flux continuity conditions

$$\begin{aligned} \frac{f_-}{\Omega} &= D_s \left[\frac{\partial n}{\partial x} - f n \right] = +k_- [n - n_{eq}], \quad x = -\frac{l}{2}, \\ \frac{f_+}{\Omega} &= D_s \left[\frac{\partial n}{\partial x} - f n \right] = -k_+ [n - n_{eq}], \quad x = +\frac{l}{2}, \end{aligned} \quad (2)$$

must hold, where Ω is the cross section of an atomic site at the step. The labels $+/-$ refer to quantities corresponding to the lower/upper terrace of a step. The fluxes f_{\pm} depend on both the difference of the adatom concentration $n(x)$ compared to its equilibrium value n_{eq} and on the attachment/detachment to the steps with kinetic coefficients k_{\pm} . If the condition $k_+ > k_-$ is fulfilled we speak about a standard ES effect [15]. It induces an asymmetry in the concentration profiles $n_i(x)$ quantified by the asymmetry parameter

$$b_{ES} \equiv \frac{k_+ - k_-}{k_+ + k_-} = \frac{l_- - l_+}{l_- + l_+}, \quad (3)$$

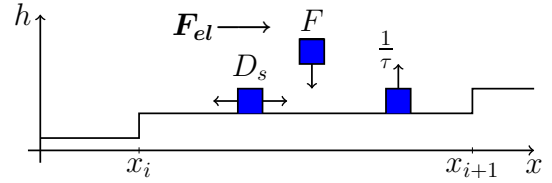


Fig. 1. (Color online) Sketch of the elementary processes in the Burton-Cabrera-Frank model.

where $l_{\pm} = D_s/k_{\pm}$ are called kinetic lengths.

Apart from the attachment kinetics, a second effect incorporated into the boundary conditions (2) is the step-step repulsion. The equilibrium concentration n_{eq} is determined by the chemical potential $\Delta\mu_i$ at the i th step through the relation $n_{eq} \approx n_{eq}^0 (1 + \Delta\mu_i/k_B T)$, and $\Delta\mu_i$ depends on the widths of the two neighboring terraces l_i and l_{i-1} according to [2,30]

$$\frac{\Delta\mu_i}{k_B T} = -g \left(\frac{l_i^3}{l_i^3} - \frac{l_i^3}{l_{i-1}^3} \right) = : g\nu_i, \quad (4)$$

where l is the mean terrace spacing and g is a *dimensionless* measure for the strength of repulsion between the steps [2,31].

3 Step equations of motion

Using equations (1), (2) and (4) we find the concentrations $n_i(x)$ for all terraces. The velocity of the i th step is then given by the superposition of the fluxes coming from the two neighboring terraces as $dx_i/dt = f_- + f_+$. Since the non-conservative terms of primary interest here arise from sublimation, we discuss separately the limiting cases of pure sublimation ($F = 0$, $\frac{1}{\tau} > 0$) and pure growth ($F > 0$, $\frac{1}{\tau} = 0$); of course, in a typical experimental setup both processes may proceed simultaneously. For the case of pure sublimation and in the limit $f^{-1} \gg l_D$, we obtain the nonlinear system

$$\begin{aligned} R^{-1} \frac{dx_i}{dt} &= \frac{\left[\left(\frac{l_+}{l_D^2} + \frac{f}{2} \right) s_i + \frac{1}{l_D} c_i \right] \gamma_i - \frac{1}{l_D} e^{-\frac{f l_i}{2}} \gamma_{i+1}}{\left[\frac{f(l_- - l_+)}{2} + 1 \right] s_i + \frac{l_- + l_+}{l_D} c_i} \\ &+ \frac{\left[\left(\frac{l_-}{l_D^2} - \frac{f}{2} \right) s_{i-1} + \frac{1}{l_D} c_{i-1} \right] \gamma_i - \frac{1}{l_D} e^{\frac{f l_{i-1}}{2}} \gamma_{i-1}}{\left[\frac{f(l_- - l_+)}{2} + 1 \right] s_{i-1} + \frac{l_- + l_+}{l_D} c_{i-1}}, \end{aligned} \quad (5)$$

where $R = n_{eq}^0 \Omega D_s$, $s_i = \sinh(l_i/l_D)$, $c_i = \cosh(l_i/l_D)$ and $\gamma_i = 1 + g\nu_i$. The result (5) contains all four length scales and illustrates the complicated functional dependence for a simple one-dimensional step train.

To simplify these expressions we use the approximation of attachment-detachment limited kinetics, $l_D \gg l_{\pm} \gg l$ [31,32]. After some calculations along the lines of [14]

we arrive at

$$R_e^{-1} \frac{dx_i}{dt} \approx \gamma_i \left[\frac{(1 - b_{ES})}{2} l_i + \frac{(1 + b_{ES})}{2} l_{i-1} \right] + U (2\nu_i - \nu_{i+1} - \nu_{i-1}) - \frac{b_{el}}{2} [(\gamma_i + \gamma_{i+1}) l_i - (\gamma_i + \gamma_{i-1}) l_{i-1}]. \quad (6)$$

Here a second asymmetry parameter

$$b_{el} \equiv -\frac{fl_D^2}{l_- + l_+} \quad (7)$$

incorporating the strength of electromigration has been introduced, $R_e = (n_{eq}^0 \Omega D_s) / l_D^2 = n_{eq}^0 \Omega / \tau$ is the constant rate with which the surface changes volume in a unit time (in the absence of non-linear, non-conservative terms, see below) and $U = (gl_D^2) / (l_- + l_+)$.

4 Linear stability

Equations similar to (6) can be derived when the surface is subject to a growth flux but sublimation is absent ($F > 0$, $\frac{1}{\tau} = 0$). In that case the factor γ_i in front of the square bracket on the right hand side of (6), which depends nonlinearly on the step coordinates, is replaced by the constant

$$\gamma = -\frac{F\tau}{n_{eq}^0}. \quad (8)$$

Analogous to the problem considered in [14], this implies qualitatively different instability conditions for growth and sublimation. Performing a standard linear stability analysis, in the limit of large wavelength perturbations we find the instability conditions

$$b^{sub} \equiv 2b_{el} + b_{ES} > 6g \quad \text{for sublimation} \quad (9)$$

$$b^{gr} \equiv 2b_{el} - \frac{F\tau}{n_{eq}^0} b_{ES} > 0 \quad \text{for growth.} \quad (10)$$

In the case of growth step bunching merely requires the compound asymmetry parameter b^{gr} to be positive, whereas for sublimation the corresponding quantity b^{sub} needs to exceed a positive threshold value $6g$. This is an important consequence of the qualitatively different contributions to the balance equation (1) that arise from desorption and deposition, respectively. Note that in a general situation the instability conditions (9), (10) can be combined into the form $b = (1 - F\tau/n_{eq}^0) b_{ES} + 2b_{el} > 6g$, which was already obtained in [12].

5 Conservative and nonconservative dynamics

Beyond the linear stability properties, a fundamental difference between the scenarios of pure growth and sublimation is that the surface dynamics is *conservative* during growth but not during sublimation [8,14]. Here conservative dynamics implies that the rate of volume

change of the crystal, obtained by summing the equations of motion over all steps x_i , is independent of the surface configuration [33]. Indeed, replacing the γ_i in front of the square brackets on the right hand side of (6) by the constant (8) and summing over i , one readily obtains $\sum_i \dot{x}_i = -F\Omega L$, where L is the total length of the crystal.

It is instructive to compare the structure of the non-conservative contributions induced during sublimation by the configuration-dependent factors γ_i in (6) for the two step bunching instabilities driven by electromigration and by an ES-effect, respectively. First we neglect the ES-effect, setting $b_{ES} = 0$, which simplifies (6) into the form

$$R_e^{-1} \frac{dx_i}{dt} = \frac{\gamma_i}{2} (l_i + l_{i-1}) + U (2\nu_i - \nu_{i+1} - \nu_{i-1}) - b_{el} (l_i - l_{i-1}) + \frac{gb_{el}}{2} \times (\nu_i l_{i-1} - \nu_i l_i + \nu_{i-1} l_{i-1} - \nu_{i+1} l_i). \quad (11)$$

The second group of terms on the RHS of equation (11) with prefactor U arises from equilibrium step-step interactions and stabilizes the regular step train, while the third group of terms describes the effect of electromigration, which is stabilising or destabilising depending on the sign of b_{el} , i.e., the direction of F_{el} . The last group of terms arises from the interplay of electromigration and step-step interactions. Since the terms in this group cancel pairwise under summation over i , their contribution is conservative and the only non-conservative contributions in equation (11) are the first terms multiplied by γ_i .

For comparison, setting $b_{el} = 0$ equation (6) reduces to the equations derived in [14],

$$R_e^{-1} \frac{dx_i}{dt} = \frac{\gamma_i}{2} (l_i + l_{i-1}) + U (2\nu_i - \nu_{i+1} - \nu_{i-1}) - \frac{b_{ES}}{2} (l_i - l_{i-1}) + \frac{gb_{ES}}{2} (\nu_i l_{i-1} - \nu_i l_i). \quad (12)$$

The difference between the two cases is that the terms proportional to gb_{ES} on the RHS of equation (12) do not cancel under summation with respect to i , and thus are non-conservative. As will be shown in the next section, this gives rise to distinct contributions in the continuum limit.

6 Continuum equations

In previous work a systematic method for deriving continuum equations of motion from the discrete step dynamics was developed [9,10] which was applied to the model (12) in [14]. Briefly, the method can be seen as a kind of Lagrange transformation [34] which replaces the ‘Lagrangian’ dynamics of particle-like steps by the ‘Eulerian’ evolution of the step density $m(x, t)$. The latter in turn defines a continuous height profile $h(x, t)$ through $m(x, t) = \frac{\partial h}{\partial x}$.

Here we wish to compare the two instability mechanisms described by equations (11) and (12), respectively, on the continuum level. Following the procedure outlined

in [14] for both models, we find that the continuum evolution equation takes the general form

$$\frac{\partial h}{\partial t} + \frac{\partial}{\partial x} \left[-\frac{3gm^2}{2} - \frac{m'}{6m^3} + \frac{3U(m^2)''}{2m} - J_b \right] + 1 = -\frac{3g(m^2)'}{2} \left[\frac{m'}{6m^3} \right]' - \Phi_b, \quad (13)$$

where primes denote spatial derivatives. Here time t is rescaled by R_e , length x by the average step distance l , and height h is measured in units of the monoatomic step height. The terms inside the square brackets on the LHS are conservative, and the non-conservative contributions are collected on the RHS of equation (13). The two models (11) and (12) differ in the form of the contribution J_b to the conserved surface flux, and of the non-conservative term Φ_b . Labeling the contributions due the ES-effect by ES and those due to electromigration by el , respectively, the conservative terms are given by

$$J_b^{ES} = \frac{b_{ES}}{2m}, \quad J_b^{el} = \frac{b_{el}}{m} - 3gb_{el}m', \quad (14)$$

and the non-conserved contributions are

$$\Phi_b^{ES} = \frac{3gb_{ES}(m^2)'}{2} \left[\frac{1}{2m} \right]', \quad \Phi_b^{el} \equiv 0. \quad (15)$$

As was discussed above, the terms in equation (11) proportional to gb_{el} give rise to a conservative contribution, whereas the terms in (12) proportional to gb_{ES} contribute to the non-conservative part of the continuum equation.

In earlier work based on the continuum approach [9,10] the non-conservative contributions were generally neglected because of the smallness of g [14], and it was therefore concluded that step bunching phenomena induced by electromigration and by the ES-effect belong to the same universality class [9,35]. However, it has subsequently become clear that small non-conservative terms may qualitatively change the nonlinear dynamics of surface steps [14], and the fact that these terms are of different form for the two instability mechanisms implies that their equivalence needs to be reexamined. In the following we therefore explore the nonlinear behavior of the electromigration model (11) using numerical simulations.

7 Nonlinear step dynamics

Numerical simulations of equation (11) were carried out using an odeint-type procedure [36] for systems of M steps with periodic boundary conditions. We consider the following ranges for the four independent parameters of the model: $b_{el} \in [0, 0.5]$, $U \in [0, 0.5]$, $g \in [0, 0.1]$ and $M < 100$. Another degree of freedom is provided by the choice of the initial condition. In general, we start the simulations with two types of initial step train configurations: either a randomly disturbed equidistant step train, or an initial shock of closely spaced steps and a single large terrace. Step trajectories are shown in the co-moving coordinate system $\tilde{x}_i(t) = x_i(t) - lt$, and we normalize both the height of the (monoatomic) steps and the average terrace width l to unity. The time t is rescaled by R_e and we measure the integration time in time units (t.u.). For the description of the bunch geometry we use two measures: the maximal slope $m_{\max} \equiv \max_i\{m_i\}$ and the minimal curvature $\kappa_{\min} \equiv \min_i\{\kappa_i\}$, where $m_i = 1/l_i$ and $\kappa_i = -8(l_{i+1} - l_i)/(l_{i+1} + l_i)^3$ respectively. A step is defined to belong to a bunch, if its distance to the next closest step of the bunch is smaller than $l = 1$.

An important consequence of the non-conservative character of the dynamics is the phenomenon of *anti-coarsening*, where an initial large step bunch splits into smaller bunches [14]. In Figure 2 we show an example of this behavior for parameter values $b_{el} = 0.4$, $g = 0.05$, $U = 0.05$, and $M = 80$. Figure 2a displays the movement of the first step in the train. After 11 180 t.u. the large bunch splits for the first time into a large and a very small bunch, which is reflected in a clear change in the velocity of the step; smaller bunches move faster. Focusing on the splitting region one finds that the initial small bunch disappears again after 4 t.u., an event which is repeated at 11 258 t.u. with a small bunch life time of 19 t.u. In Figure 2b we show the trajectories of all steps in the train in the time window 11 280–11 470 t.u. At 11 333 t.u. the small bunch appears for the third time and at 11 415 t.u. a third bunch arises for the first time. The switching between different numbers of bunches continues until the step train relaxes into four bunches, as seen in the height profiles in Figure 2c. Figure 2d shows the corresponding time evolution of the minimal curvature κ_{\min} and the maximal slope m_{\max} , along with the number of bunches. Both κ_{\min} and m_{\max} show clear changes in the region of the splitting; however, whereas the maximal slope remains essentially the same after the splitting event, there is a significant decrease in the minimal curvature.

In Figure 3 we summarize results obtained from simulations starting from a randomly disturbed equidistant step array of 40 steps. Figure 3a shows the phase diagram of the system in the g - b_{el} plane at fixed $U = 0.2$. Below the line $b_{el} = 3g$ the system is linearly stable and $m_{\max} = m(x) \equiv 1$. Above this line we see three qualitatively different types of long-time behavior: steady solutions with one single bunch, with two bunches, and time-dependent solutions that switch periodically between one and two bunches. In Figure 3b we plot the time evolution of the maximal slope m_{\max} at five points along the line $b_{el} = 0.35$. For $g = 0$ we see the usual coarsening behavior, in which the number of bunches decreases in a step-wise fashion until a single bunch configuration is reached and the system relaxes to a stationary periodic state with a clearly bounded maximal slope. The remaining temporal periodicity of m_{\max} is due to the permanent step exchange between the front and back end of the bunch (see also inset in Fig. 2a). Increasing g the maximal slope decreases (while still maintaining the single bunch configuration), until at $g = 0.05$ the regime of periodic switching is reached, leading to a complex periodic pattern in m_{\max} .

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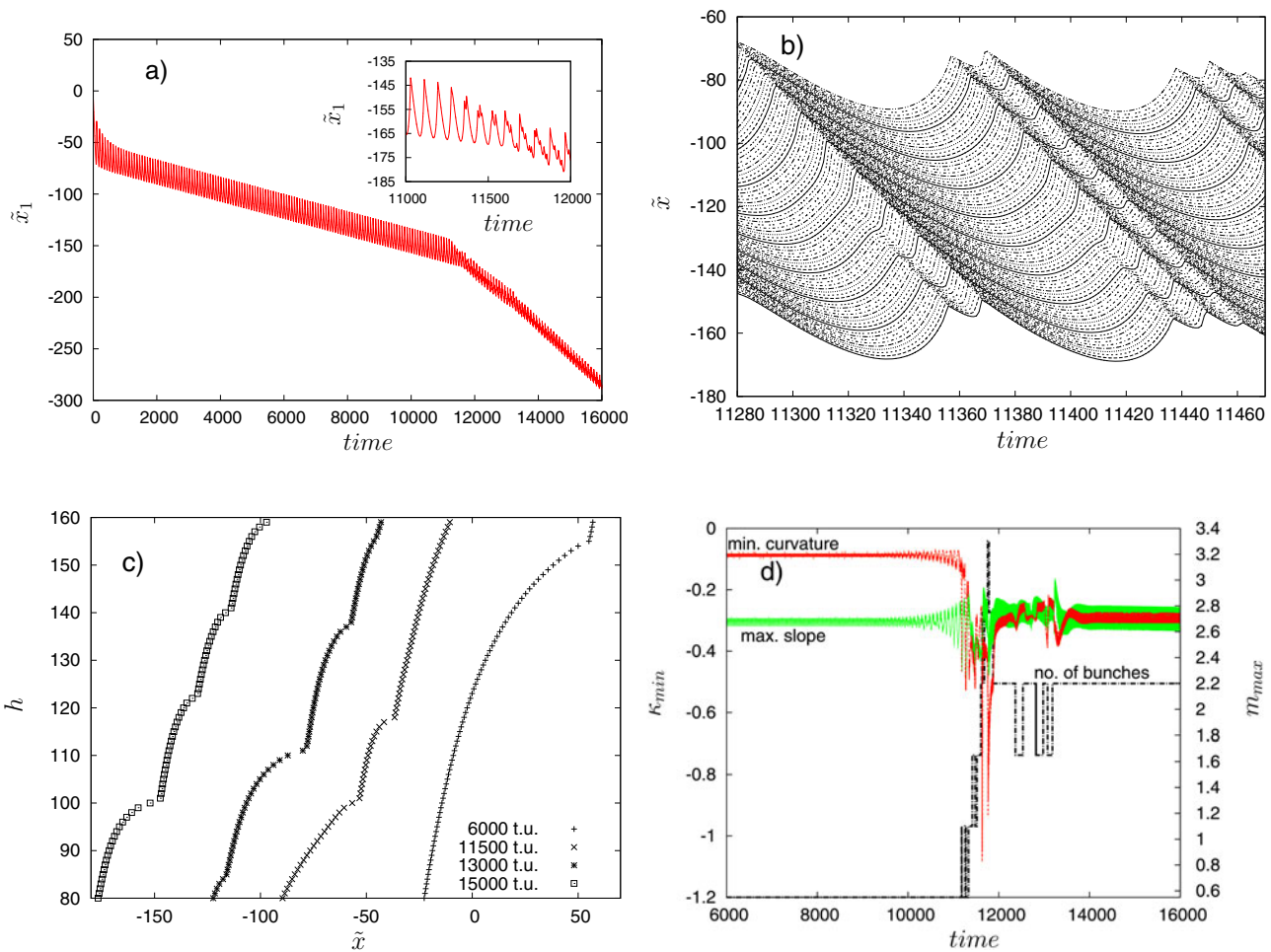


Fig. 2. (Color online) An example for the splitting of a large bunch in a system of 80 steps with parameters $b_{el} = 0.4$, $g = 0.05$, $U = 0.05$. (a) Time evolution of one of the steps. The inset shows a blowup around the onset of splitting. The period of oscillations prior to the breakup reflects the time required for the step to once traverse the (single) bunch. (b) Plot of all step trajectories between 11 280 t.u. and 11 470 t.u. (c) Comparison of the profiles after 6000 t.u., 11 500 t.u., 13 000 t.u. and 15 000 t.u. (d) Time evolution of the globally maximal slope, the globally minimal curvature and the number of bunches.

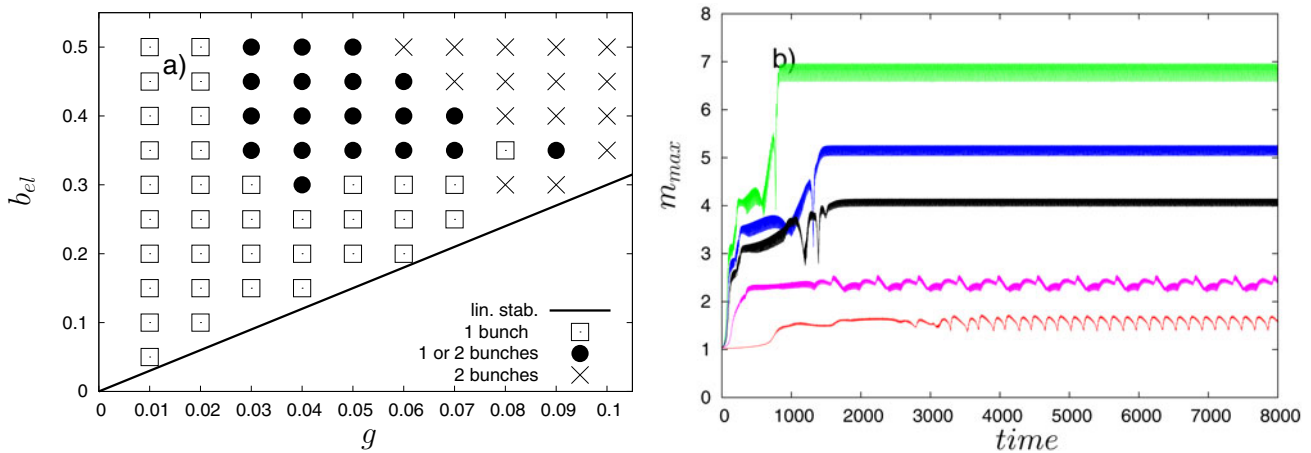


Fig. 3. (Color online) System with $U = 0.2$, $M = 40$ and fluctuating initial condition. (a) Stability/instability diagram, showing the number of bunches in the final state, for different combinations of b_{el} and g — \square : 1 bunch, \bullet : 1 or 2 bunches, \times : 2 bunches, below the line $b_{el} = 3g$: stability. (b) Time evolution of the maximal slope m_{max} with $b_{el} = 0.35$ and (from top to bottom) $g = 0.00, 0.01, 0.02, 0.05,$ and 0.09 .

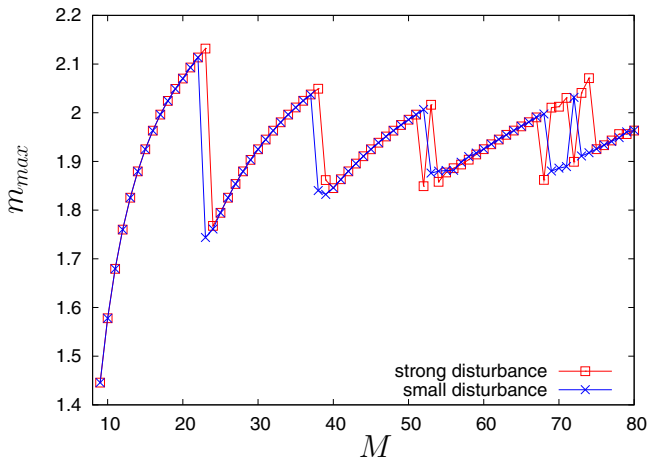


Fig. 4. (Color online) Dependence of the globally maximal slope m_{\max} on the number of steps M for $g = U = 0.04$, $b_{el} = 0.2$ and for large and small noise amplitude in the random initial condition.

Finally, in Figure 4 we plot the behavior of the maximal slope as a function of the number of steps for two different amplitudes of the initial disturbance, $g = U = 0.04$, and $b_{el} = 0.2$. Here m_{\max} is the global maximal slope, measured as its largest value for the last 500 t.u. of the simulation. We see that m_{\max} generally increases with M , but this behavior is interrupted by downward jumps every time the number of bunches that can fit into the system increases by one. This shows that the existence of stationary solutions with multiple bunches can be seen as a consequence of the fact that, in the presence of non-conservative processes, the maximal slope is bounded from above [14]. Near the transition between different numbers of bunches the system behavior depends very sensitively on the amplitude of fluctuations in the initial configuration, an effect that is particularly pronounced around $M = 70$.

8 Conclusion

In this work we have extended the non-conservative step bunching model presented in [14] to include the effect of electromigration. The model applies to the first of the experimentally observed temperature regimes on the Si(111) surface, where step transparency can be neglected. The general step equations of motion incorporating sublimation, the Ehrlich-Schwoebel effect, electromigration and step-step interactions were derived from the classical BCF model in the quasistatic approximation. For the case of attachment-detachment limited kinetics we compared the equations for growth and sublimation. In previous publications [9–11,31] non-conservative contributions were neglected, because of the experimentally small prefactor g [2,14]. Those terms were now taken into account and some important consequences were identified. First, on the level of linear stability analysis, they shift the instability condition on the dimensionless asymmetry parameter b by $6g$, as was first pointed out in [12]. This shift is present in

the case of sublimation, but not in the case of growth [14]. Moreover, in the case of sublimation the structure of the non-conservative terms differs depending on the underlying mechanism inducing the asymmetry between ascending and descending steps. This leads to different continuum equations for step bunching caused by an ES-effect or by electromigration, respectively.

Nevertheless, the numerical integration of the discrete step equations for the case with sublimation and electromigration reproduces qualitatively the results of [14]. The non-linear, non-conservative terms supply a richness of dynamical behaviors in this simple one-dimensional step model. There are steady solutions which contain more than one bunch, periodic switching between step trains of different numbers of bunches, and a sensitive dependence on the initial condition. This shows that the notion of universality between different types of step bunching mechanisms, which was originally formulated on the basis of conservative continuum equations [9,35], can be applied also in the presence of non-conservative dynamics.

In previous work on the conservative version of (6) a dynamical phase transition was identified which separates two qualitatively different regimes of step bunching distinguished by the presence or absence of crossing steps between bunches [11]. In our units this transition occurs at $b_{el} = 1/2$, and experimental evidence for its existence in the Si(111) system has recently been reported [26]. In order to clearly bring out the effects due to the non-conservative nature of the dynamics, in the present study we have restricted ourselves to the parameter range $b_{el} \in [0, 0.5]$, but the influence of non-conservative terms on the phase transition reported in [11] is clearly an interesting topic for future work.

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