Dynamic phase transitions in electromigration-induced step bunching

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Electromigration-induced step bunching in the presence of sublimation is studied theoretically in the attachment-limited regime. We predict a phase transition as a function of the relative strength of kinetic asymmetry and step drift. For weak asymmetry the number of steps between bunches grows logarithmically with bunch size, whereas for strong asymmetry at most a single step crosses between two bunches. In the latter phase the emission and absorption of steps is a collective process which sets in only above a critical bunch size and/or step interaction strength.

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I. INTRODUCTION

Much of the morphological structure and dynamics of crystal surfaces can be understood in terms of the behavior of steps that separate different exposed atomic layers.[1–3] Steps are long-lived structural defects subject to long-ranged interactions of entropic and elastic origin. When such a system of interacting steps is driven out of equilibrium by external forces, a rich variety of morphological patterns and dynamic phenomena emerge.

As was first discovered by Latyshev and co-workers,[4] steps on Si(111) surfaces can be manipulated by a direct heating current, which induces mass transport along the surface through the electromigration of adatoms.[5] A host of step patterns have since been found and studied experimentally in this system.[6,7] Of particular interest is the electromigration-induced formation of step bunches, and the long-standing puzzle posed by the multiple reversals of the current direction required to induce step bunching that occur as a function of substrate temperature.[4,8–10] A number of recent theoretical papers have addressed this issue on the level of linear stability analysis, and several mechanisms associated with the way in which atoms are exchanged at steps have been proposed.[11–14] However, in order to distinguish between alternative explanations and, thus, to uncover the true underlying microscopic processes, it seems mandatory to go beyond the linear regime and embark on a systematic investigation of the fully nonlinear step dynamics.

In this spirit, we report here on a dynamic phase transition that we have found in the most basic model of electromigration-induced step bunching.[5,15–17] In this model the steps are assumed to be straight, and the uniform step train is destabilized by an electromigration force in the downhill direction. The phase transition occurs as a function of a dimensionless parameter b, defined in (4) below, which gauges the relative importance of electromigration-induced kinetic asymmetry and step drift due to sublimation. This parameter can be tuned experimentally, e.g., by changing the electromigration force through the DC component of the heating current, or the sublimation rate through a change in temperature.

Step drift leads to the exchange of steps between bunches, which plays an important role in the evolution and coarsening of the bunch pattern.[16–18] The most striking visual signature of the phase transition is a qualitative change in the number of such *crossing steps*, and in the mechanism by which they are exchanged (Fig. 1). For $b<1$ (strong drift/weak asymmetry) the step density decreases smoothly in the outflow region of a bunch, and the number of crossing steps grows logarithmically with the bunch distance. In contrast, for $b>1$ (weak drift/strong asymmetry) there is at most a single free step between any two bunches, irrespective of their size. This feature should make the two regimes clearly distinguishable in experiments using reflection electron microscopy[4] or scanning probe microscopy.[8–10]

The dynamics in the regime $b>1$ are remarkably complex. The exchange of a step is a collective process involving both the expelling and the receiving bunch, which sets in only beyond a critical bunch size, and which is accompanied by breathing oscillations of the entire bunch. As a consequence, a stationary bunch shape amenable to a continuum description[19] of the type developed previously[20,21] for $b<1$ does not appear to exist.

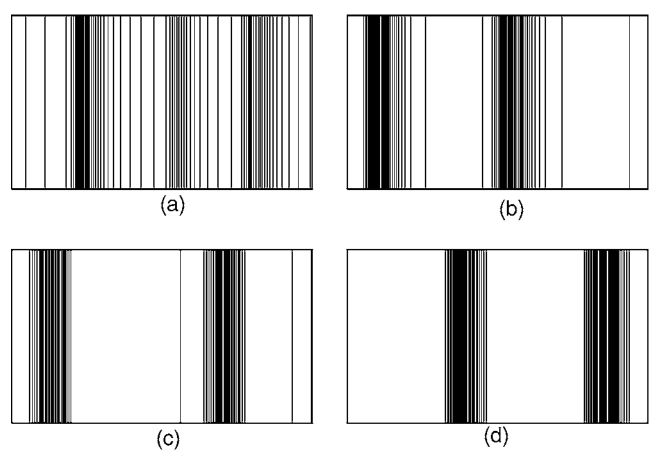

FIG. 1. Typical step configurations (top view of a vicinal surface) generated by numerical solution of (1). Graphs (a)–(d) correspond to $b=0.1, 0.5, 5, 20$, respectively, and in all cases $U/bl=0.6$. Each frame contains about 60 steps. Reduction of the number of crossing steps with increasing b is evident. In case (d) there are no crossing steps since the bunch sizes are within the dead zone region of Fig. 3.

II. MODEL

We consider a system of straight, nontransparent steps[3] subject to electromigration and sublimation. We work in the attachment-detachment limited regime, where the kinetic length $d=D/k$, the ratio of surface diffusion coefficient D, and attachment rate k, is large compared to the step spacing.[3] The equations of motion for the step positions $x_i(t)$ then take the form (Ref. 15)

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

where the time scale has been normalized to the sublimation flux. Summing over i we see that the average step velocity v is equal to the mean terrace width l. In numerical solutions of (1) lengths are measured in units of l, i.e., we set $l=1$. The last term on the right-hand side represents stabilizing step-step interactions of strength U, where, for combined entropic or dipolar elastic repulsion (Ref. 1)

$$f_i = \left(\frac{l}{x_i - x_{i-1}}\right)^{\nu+1} - \left(\frac{l}{x_{i+1}-x_i}\right)^{\nu+1}, \tag{2}$$

with $\nu=2$. The parameter b governs the asymmetry between ascending and descending steps, relative to the mean step velocity, which induces step bunching when $b>0$. Linear stability analysis of (1) shows that the instability sets in at wavelengths corresponding to bunches containing more than M^* steps, with

$$M^* = 2\pi[\arccos(1 - bl/12U)]^{-1}. \tag{3}$$

In previous work more complicated variants of the step equations (1) have been studied numerically, and some of the features analyzed in this paper have been described on a qualitative level.[16,17] The advantage of using the attachment/detachment limited dynamics (1) lies in the linearity of the destabilizing terms, which allows to clearly expose the key role of the parameter b and the existence of a sharp phase transition.

In terms of physical quantities, the parameters b and U are given by (Ref. 15)

$$b = \frac{\Gamma F \tau_e}{2k_B T a^2}, \quad U = \frac{\Gamma \tau_e g}{2k_B T}\tan^3\theta, \tag{4}$$

where Γ is the step mobility for the Brownian motion of an isolated step,[1] a^2 is the atomic area, F is the electromigration force acting on an adatom, τ_e is the inverse desorption rate, $\theta=a/l$ is the miscut angle, and g is the step interaction parameter.

The model (1) is expected to apply[9] in two of the four temperature regimes[6,7] in which step bunching is observed on Si(111), around 900 °C (regime I) and around 1250 °C (regime III). The parameters given in Refs. 8 and 15 lead to the estimates $b\approx 14$ in regime I and $b\approx 0.3$ in regime III, which shows that both cases $b<1$ and $b>1$ are experimentally realizable.

Step equations of the form (1) can also be derived for step bunching induced by Ehrlich-Schwoebel (ES) barriers during sublimation[20] or by inverse ES barriers during growth.[20,22] In this sense (1) constitutes a rather generic model of step bunching kinetics. However, in step bunching induced by ES barriers the parameter b is restricted to the interval $0<b<1$, and hence the phenomena described in this paper do not occur.

III. STRUCTURE OF THE OUTFLOW REGION

In the presence of step drift, coarsening of step bunches is a very dynamic process during which steps continuously leave (flow out of) one bunch and join (flow into) its neighbor.[16,17] In Refs. 21 and 22 it was shown that the analysis of the outflow region provides key insights into the shape and dynamics of bunches for $b<1$. We shall see now that there are drastic differences between the outflow regions for the cases $b<1$ and $b>1$. We consider a bunch containing a large number $M\gg 1$ of steps, so that its shape can be considered quasistationary. We impose periodic boundary conditions $\Delta_i(t)=\Delta_{i+M}(t)$ for the terrace sizes $\Delta_i = x_{i+1}-x_i$. Stationarity implies then periodicity of each step trajectory (up to an overall shift with velocity $v=l$), with some period $\tau(b,U,M)$, during which each step i will once cross the plateau between two consecutive bunches. After time τ/M, each step i will substitute the position of step $i+1$ (up to a constant shift independent of i), so that

$$\Delta_{i\pm 1}(t) = \Delta_i\left(t \pm \frac{\tau}{M}\right). \tag{5}$$

Deriving an equation for $\Delta_i(t)$ from (1) and substituting (5), we get a differential-difference equation[21] for a single, τ-periodic function $\Delta(t)=\Delta_i(t)$

$$\frac{d\Delta(t)}{dt} = \frac{1-b}{2}\Delta\left(t+\frac{\tau}{M}\right) + b\Delta(t) - \frac{1+b}{2}\Delta\left(t - \frac{\tau}{M}\right) + U(\cdots), \tag{6}$$

where for brevity the U-containing terms are only sketched. The (unknown) period τ determines the velocity of a bunch: after time τ the bunch shifts by $(-Ml)$ in a frame comoving with velocity v; in the laboratory frame its lateral velocity is then (Ref. 21)

$$V = l(1 - M/\tau). \tag{7}$$

Big bunches are separated by wide plateaux, and for the steps crossing a plateau (in case there are many) the U term in (6) should become negligible. In this outflow region one can solve the remaining linear part by the ansatz $\Delta(t) \sim \exp(qMt/\tau)$ obtaining the transcendental equation

$$b[\cosh(q) - 1] = \sinh(q) - qM/\tau. \tag{8}$$

To fix the unknown parameter τ, we recall the Fourier analysis[21] of (1), which shows, irrespective of the value of b, that for large M generically $\tau(M)\approx M+O(1)$. Thus

$$b[\cosh(q) - 1] = \sinh(q) - q \tag{9}$$

which has a real positive solution for each $b<1$ but no solutions for $b>1$. In the following section we explore the consequences of this fact.

IV. NUMBER OF CROSSING STEPS

For $b<1$, the existence of a solution q of (9) implies a smooth decrease of the step density in the outflow region, with the terrace widths increasing exponentially, as $\Delta_k/\Delta_{k-1}\approx\exp(q)$. To estimate the number N_c of free steps between two bunches of size M, we equate the total length $\sim l\exp[qN_c]$ occupied by N_c terraces to the typical distance Ml between the bunches, and obtain

$$N_c \approx q^{-1}\ln M. \tag{10}$$

For small b, the solution of (9) can be approximated by $q \approx 3b$.

For $b\to 1$, q diverges and N_c vanishes. The absence of solutions of (9) means that the U term in (1) can never be neglected and that correspondingly there can be at most one step crossing the plateau between two bunches, at any stage of evolution. One can check, using (1) and (2), that any configuration with more than one step between two bunches is unstable for $b>1$, so that all steps except at most one will be pushed back to the bunch they originated from. In Fig. 1 we show numerically generated bunch configurations in the course of coarsening for $b<1$ and $b>1$, which confirm this conclusion.

V. DYNAMICS OF EMISSION AND ABSORPTION OF STEPS

We now examine in more detail how steps are emitted from a bunch. It is seen directly[18] from (1) that the last step (with label i, say) of the bunch at position x_i, which is trailing a wide terrace of width $\Delta_i=x_{i+1}-x_i\gg l$, will be driven to escape from the bunch by the linear term $(1-b)\Delta_i/2$, provided $b<1$. This term indeed gives the main contribution to the dynamics of the last step of the bunch, as we see from numerical analysis. The emitted step does not perturb the remaining steps; the $(i-1)$-th step which has become the last, is free to escape once the ith step has traveled sufficiently far. Bunches emit steps continuously, creating an outflow region governed entirely[21,22] by the linear part of (1).

In contrast, for $b>1$, the linear term $(1-b)\Delta_i/2$ in (1) gives a negative contribution to the step velocity, and the only way to move the last step i away from the bunch is by step-step interactions [the U term in (1)]. Since the next step $i-1$ cannot be emitted before step i has landed at the next bunch, the configuration of steps at the end of the bunch has to be changed by the emission process—if it were unchanged, the next step $i-1$ would be emitted immediately after the ith. This gives rise to oscillations of the bunch profile at the end, which spread to the whole bunch, and whose amplitude grows with increasing b. Such oscillations at the outflow end of the bunch are completely absent in the $b<1$ phase (Fig. 2).

When the emitted step collides with the receiving bunch it provokes perturbations in the inflow region of the bunch, which are visible both for $b<1$ and $b>1$. In the case $b<1$ the oscillations in the inflow region are damped and disappear toward the bunch interior. In the $b>1$ phase, however, the oscillations penetrate through the bunch, regain their amplitude toward the bunch tail, and culminate in the emission of the last step of the bunch, provided that the initial impact was sufficiently strong (Fig. 2). This implies correlations between the emission and absorption of steps, which should have important consequences for the coarsening dynamics.

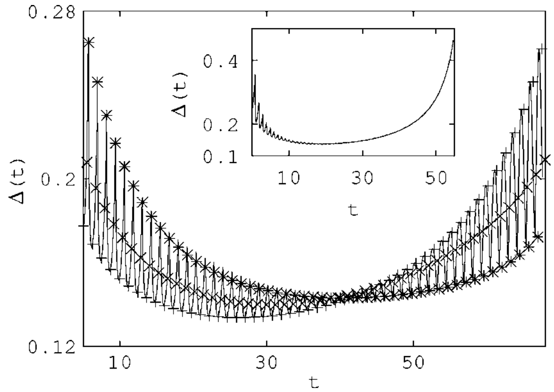

FIG. 2. Full line: Distance $\Delta(t)$ between a pair of neighboring steps moving through the bunch interior (initial and end regions excluded), for $b=10$, $U/l=6$, $M=64$. Symbols show the sizes of 42 consecutive terraces (out of 64), at times $t=0(+)$, $t=0.34\cdot(\tau/M)$ $(\times)$, $t\approx 0.55\cdot(\tau/M)(*)$, illustrating the oscillatory breathing of the bunch. They lie on the curve $\Delta(t)$ because of (5). Inset: $\Delta(t)$ for $b=0.176$, $U/l=0.108$. For $b<1$ the oscillations do not extend into the outflow region.

VI. ONSET OF STEP EMISSION

We have seen above that the emission of steps for $b>1$ is facilitated by a large step-step repulsion U, and suppressed by the kinetic asymmetry b. For small U (or large b) the oscillatory breathing of the bunch may not be able to trigger

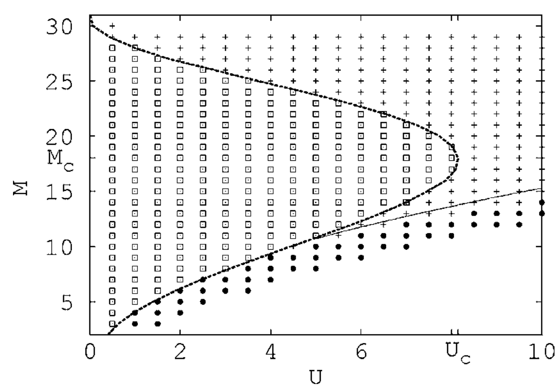

FIG. 3. Phase diagram characterizing quasistationary step bunches of size M as function of step-step repulsion U for $b=10$, $l=1$. Three phases can be defined: (a) Bunches smaller than the size M^* deduced from linear stability analysis [Eq. (3)] dissolve (filled circles). (b) Dead zone: for $M_{c1}(U)<M<M_{c2}(U)$ bunches are stable but do not emit steps (squares). (c) For $M>M_{c2}(U)$ and $M^*<M<M_{c1}(U)$ bunches are stable and emit steps (crosses). The lines $M_{c1}(U), M_{c2}(U)$ terminate at a critical interaction strength $U_c(b)$ beyond which stable bunches always emit steps.

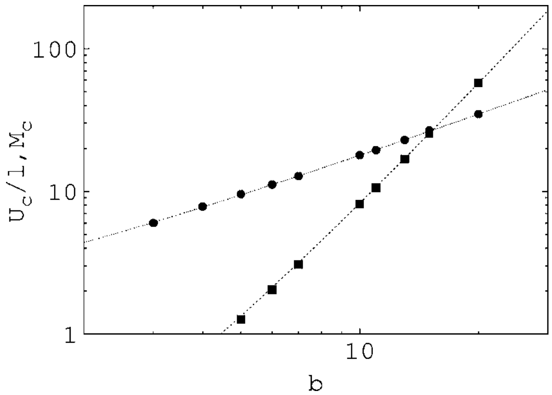

FIG. 4. Values of critical interaction strength U_c (squares) and bunch size M_c (circles) for different b, determined from numerical simulations. Straight lines illustrate the power laws (11).

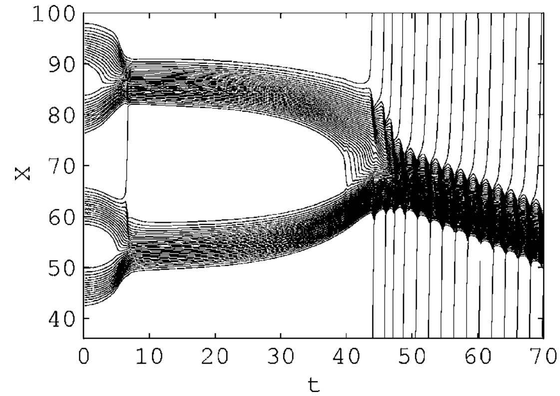

FIG. 5. Space-time plot of individual step trajectories during coarsening, in a frame comoving with the step velocity v, for $b=20$, $U/l=12$. Step emission starts when the bunch size exceeds a critical value. The isolated emission event around $t\approx 7$ occurs because the bunch shapes are not yet stationary. The initial configuration is a slightly perturbed train of bunches consisting of 16 steps each.

the emission of steps when bunches are small. The typical behavior of bunches as a function of size M and step interaction U at a fixed value of $b>1$ is summarized in the phase diagram in Fig. 3. For any given $b>1$, there exists a critical value $U_c(b)$ such that for $U<U_c$ bunches emit steps only for sizes $M^*<M<M_{c1}$ and $M>M_{c2}$, whereas for $U>U_c$ stable bunches always emit steps. Inside the dead zone $M_{c1}<M<M_{c2}$ the time interval τ/M between emission of steps is infinite, and correspondingly bunches move with the mean step speed, $V=v=l$ [compare to (7)]. The ratio τ/M decays monotonically to 1 with distance from the dead zone, reflecting the fact[21] that $\lim_{M\to\infty} \tau/M=1$ for any fixed b.

Diagrams for different b can be superimposed after rescaling U_c and $M_c \equiv M_{c1,2}(U_c)$ according to the relations

$$U_c \approx 0.0062 \cdot (b+1)^{\alpha}, \quad M_c \approx 1 + 1.69 \cdot b^{\gamma}, \qquad (11)$$

with $\alpha\approx 3$, $\gamma\approx 1$ for all parameters investigated ($3<b\leqslant 20$, $10^{-2}\leqslant U\leqslant 60$), see Fig. 4. Note that the relation $\gamma=(\alpha-1)/2$ implies an invariance of the linear instability curve (3) at large U under rescaling.

Different step kinetics for bunches of different sizes implies a change in coarsening dynamics, highlighted in Fig. 5. For $b\gg 1$, depending on the value of U different coarsening scenarios are possible. For $U>U_c$ steps are exchanged throughout the coarsening process, while for $U\lesssim U_c/2$ late stage coarsening proceeds in two stages: without step exchange (for bunch sizes smaller than M_{c2}) and with step emission once the typical bunch size exceeds M_{c2}. Coarsening with or without a step exchange has previously been associated with nonconserved (b finite) and conserved ($b=\infty$, no sublimation) dynamics, respectively;[17] here we see that both types of behavior may coexist when $b>1$.

VII. CONCLUSIONS

We predict a dynamic phase transition in electromigration-induced step bunching within the regime of nontransparent steps and attachment-detachment limited kinetics. The transition is characterized by a dramatic change in the number and behavior of the free steps that are exchanged between bunches. Qualitative changes in the behavior of crossing steps have repeatedly been reported in electromigration experiments on Si(111). For example, Homma and Aizawa[9] observed single crossing steps consistent with our $b>1$ scenario in temperature regime III, while Gibbons *et al.* found a dramatic reduction in the number of crossing steps on approaching equilibrium conditions.[10] We therefore expect that a systematic investigation of crossing steps will considerably deepen our understanding of step bunching in this system. Theoretical challenges for the future include the development of a continuum description for $b>1$, and the investigation of the correlated coarsening dynamics in this regime.

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