Dynamic phase transitions in electromigration-induced step bunching

Vladislav Popkov and Joachim Krug
Institut für Theoretische Physik, Universität zu Köln, Germany.
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Electromigration-induced step bunching in the presence of sublimation or deposition 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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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, 2]. Since they entail a finite free energy cost per unit length, steps are long-lived structural defects which nevertheless, due to their one-dimensional nature, are highly sensitive to thermal fluctuations. These fluctuations induce long-ranged steric interactions between steps, which complement similar interactions mediated by bulk elasticity. When such a system of interacting steps is driven out of equilibrium by external forces, e.g. during growth or sublimation of the crystal surface, a rich variety of morphological patterns and dynamic phenomena emerge.

As was first shown by Latyshev and coworkers [3], step patterns on Si(111) surfaces can be efficiently manipulated by a direct heating current, which induces mass transport along the surface through the electromigration of adatoms. Subsequently a multitude of electromigration-generated step patterns have been found and studied experimentally [4, 5], including step bunches [6, 7, 8], step antibands [9], in-phase wandering steps [10] and step pairs [11], many of which still defy a comprehensive theoretical description.

In this Letter we report on the surprising discovery of a novel type of phase transition in the most basic model of electromigration-induced step bunching originally introduced by Stoyanov [12, 13, 14, 15]. 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 or growth. 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 [14, 15]. The most striking visual signature of the phase transition is a qualitative change in the number of such free 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 steps between bunches 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 [3, 7] or scanning tunneling microscopy [6, 8].

The dynamics in the regime $b > 1$ is 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 [16] of the type developed previously for $b < 1$ [17, 18] does not appear to exist.

Figure 1: Typical step configurations (top view of a vicinal surface) generated by numerical solution of (1). Graphs (a)-(d) correspond to $b = 0.1, 0.5, 1.0, 2.0$ respectively, and in all cases $U/bd = 0.6$. Each frame contains about 60 steps. Reduction of the number of free flowing steps with increasing $b$ is evident. In case (d) there are no free-flowing steps since the bunch sizes are within the dead zone region of Fig. 3.
Model. We consider a system of straight, non-transparent steps subject to electromigration and sublimation (including also a growth flux is straightforward). 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 $l$. The equations of motion for the step positions $x_i(t)$ then take the form

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

$$+ U (2f_i - f_{i-1} - f_{i+1})$$

(1)

where the time scale has been normalized to the sublimation (including also a growth flux is straightforward). 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,

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

(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

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

(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. The advantage of using the attachment/detachment limited dynamics 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

$$b = \frac{\Gamma F r_e}{2 k_B T a^2}, \quad U = \frac{\Gamma r_e g}{2 k_B T} \tan^3 \alpha$$

(4)

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

The model (1) is expected to apply in two of the four temperature regimes in which step bunching is observed on Si(111), around 900° C and around 1250° C. The parameters given in (4) lead to the estimates $b \approx 14$ in the low temperature regime and $b \approx 0.3$ in the high temperature regime, 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 or by inverse ES barriers during growth. 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.

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 neighbour. In 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 quasi-stationary. 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 $\tau/M$, each step $i$ will substitute the position of step $i + 1$ (up to a constant shift independent of $i$), so that

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

(5)

Deriving an equation for $\Delta_i(t)$ from (1) and substituting (4), we get a differential-difference equation for a single, $\tau$-periodic function $\Delta(t) = \Delta_i(t)$

$$\frac{d\Delta(t)}{dt} = -\frac{1 - b}{2} \Delta(t + \frac{\tau}{M}) + b\Delta(t) - \frac{1 + b}{2} \Delta(t - \frac{\tau}{M}) + U(...),$$

(6)

where for brevity the $U$-containing terms are only sketched. The (unknown) period $\tau$ determines the velocity of a bunch: after time $\tau$ the bunch shifts by $(−MI)$ in a frame co-moving with velocity $v$; in the laboratory frame its lateral velocity is then

$$V = l(1 - M/\tau).$$

(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 $b (\cosh(q) - 1) = \sinh(q) - qM/\tau$. To fix the unknown parameter $\tau$, we recall the Fourier analysis of
that for large $M$ a bunch.

It is seen directly from (1) that the last step now examine in more detail how steps are emitted from the oscillatory breathing of the bunch. They lie on the curve $\Delta(t)$ because of $\mathbf{8}$. Inset: $\Delta(t)$ for $b = 0.176$, $U/l = 0.108$. Oscillations are triggered by a step colliding with the front end of the bunch, but do not extend into the outflow region.

Number of steps between bunches. For $b < 1$, the existence of a solution $q$ of $\mathbf{8}$ 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 $N_f$ of free steps between two bunches of size $M$, we equate the total length $\sim \int \exp[qN_f] \sim qN_f \approx q^{-1}\ln M$. For small $b$, the solution of $\mathbf{8}$ can be approximated by $q \approx 3b$.

For $b \to 1$, $q$ diverges and $N_f$ vanishes. The absence of solutions of $\mathbf{8}$ means that the $U$-term in $\mathbf{1}$ 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 $\mathbf{1}$ and $\mathbf{2}$, 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. $\mathbf{1}$ we show numerically generated bunch configurations in the course of coarsening for $b < 1$ and $b > 1$, which confirm this conclusion.

Dynamics of emission and absorption of steps. We now examine in more detail how steps are emitted from a bunch. It is seen directly from $\mathbf{1}$ 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 $i$-th step has travelled sufficiently far. Bunches emit steps continuously, creating an outflow region governed entirely by the linear part of $\mathbf{1}$.

In contrast, for $b > 1$, the linear term $(1 - b)\Delta_i/2$ in $\mathbf{1}$ 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 $\mathbf{1}$]. 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 $i$-th. 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. $\mathbf{2}$).

When the emitted step finally 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 $b < 1$, however, the oscillations in the inflow region are damped and disappear towards the interior of the bunch. On the contrary, in the $b > 1$ phase...
the oscillations penetrate through the bunch, regain their large amplitude towards 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). The persistence of oscillations through the bunch interior implies correlations between the emission and absorption of steps, which should have important consequences for the coarsening dynamics; this question will be addressed elsewhere.

**Onset of step emission.** We have seen above that the emission of steps in the $b > 1$ regime is a nontrivial dynamical phenomenon 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 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 $\tau/M$ between emission of steps is infinite, and correspondingly bunches move with the mean step speed, $V = v$ [see 11]. The ratio $\tau/M$ decays monotonically to 1 with distance from the dead zone, reflecting the fact that $\lim_{M \to \infty} \tau/M = 1$ for any fixed $b$ [13].

Diagrams for different $b$ can be superimposed after rescaling $U_c$ and $M_c \equiv M_{c1,2}(U_c)$ according to the relations $U_c \approx 0.0105 \cdot b^\alpha$ and $M_c \approx 2.112 \cdot b^\beta$ with $\alpha \approx 2.87$, $\gamma \approx 0.935$ for all parameters investigated ($3 < b \leq 25$, $10^{-2} \leq U \leq 120$), with a relative error not exceeding 5%. Note that the relation $\gamma = (\alpha - 1)/2$ implies 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. 4. 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 $U \leq U_c/2$ late stage coarsening proceeds in two stages: without step exchange (for bunches sizes smaller than $M_{c2}$) and with step emission once the typical bunch size exceeds $M_{c2}$. Coarsening with or without step exchange has previously been associated with nonconserved ($b$ finite) and conserved ($b = \infty$, no sublimation) dynamics, respectively [15]; here we see that both types of behavior may coexist when $b > 1$.

**Conclusions.** We predict a new type of 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, which should be clearly visible in experiments on surfaces vicinal to Si(111). 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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* E-mail: popkov@thp.uni-koeln.de
1 E-mail: krug@thp.uni-koeln.de

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