Simultaneous Bunching and Debunching of Surface Steps: Theory and Relation to Experiments

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We study a model of two-dimensional step flow where the velocity of a step depends predominantly on the width of the terrace remaining behind it. While the uniform step train is unstable towards step bunching, the bunches themselves are unstable and tend to debunch. This leads to patterns where slow moving fairly straight bunches coexist with fast, strongly bent single steps, in qualitative agreement with experiments on electric current driven step motion on Si(111) surfaces. Analytical predictions of the shape and velocity of the single steps agree very well with Monte Carlo simulations.

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Several experimental groups [1–4] have shown that the motion of surface steps on Si(111) surfaces during evaporation by heating with direct electric currents depends crucially on the direction of the current relative to the step orientation. Current in one direction results in stable step flow, with the motion of more or less uniform and straight steps. Current in the opposite direction causes the steps to bunch together and form complex two-dimensional patterns. These exhibit very interesting dynamical properties, with the exchange of single steps between bunches. The microscopic mechanism responsible for these phenomena is very complicated and not at all understood [5]; there are three temperature regimes where the stable and unstable current directions change roles. Electromigration, which is the major cause for the deterioration of semiconductor electronic devices, has been suggested [1,5] as a candidate. In this work we propose a mesoscopic model [6] delineating crucial features of the physics on large scales, and achieve extraordinary qualitative agreement with experiment. We also make quantitative predictions that can be directly tested with new experiments.

A wide class of instabilities in step flow during both growth and evaporation can be understood in terms of a simple model [7–9] of step flow:

$$\frac{\partial X_n}{\partial t} = f_+(W_n) + f_-(W_{n-1}) + \gamma \frac{\partial^2 X_n}{\partial y^2}. \qquad (1)$$

Here $X_n(y,t)$ is the position of the nth step at time t, where y is the orthogonal coordinate along the step edge. The step index n increases in the direction of step motion. $W_n \equiv X_{n+1}(y,t) - X_n(y,t)$ is the width of the terrace in front of step n. The first two terms [7] on the right-hand side of (1) express the dependence of the velocity of a step on the widths of the terraces in front and behind it. They arise from an effective treatment of adatom attachment, detachment, and surface diffusion [10], and can be calculated explicitly using a microscopic theory such as the BCF theory [11]. The last term [9(d)] of (1) accounts for transverse step fluctuations; $1/\gamma q^2$ is the relaxation time of fluctuations along the step edge of wave number q, and γ is directly related to the step stiffness associated with step bending [12].

A straightforward linear stability analysis of (1) around the uniform step train configuration with terrace width W shows [7] that if

$$f'_-(W) > f'_+(W), \qquad (2)$$

the uniform step train is unstable towards step bunching. Here $f'_\pm$ are the derivatives of $f_\pm$. The asymmetry in the effective model may have several different microscopic physical origins. One possibility, discussed by Schwoebel and Shipsey [13], arises from the presence of different energy barriers associated with the exchange of adatoms between the step edge and the terraces in front or behind. Another possibility is an asymmetry in the diffusion of adatoms on terraces caused, e.g., by couplings to external electric fields or elastic strain fields.

To describe the long time behavior of an unstable system of steps we have to take into account two important physical effects that Eq. (1) does not treat. First, we prevent energetically costly step crossings or overhangs by imposing a restriction of a minimal distance, $D_{\min}$, between steps. Some researchers [14] have suggested simply stopping steps when the minimal distance is achieved. However, this misses a second basic physical effect: the contribution to the step velocity from terraces other than the nearest-neighbor ones considered in model (1). Consider for concreteness crystal growth (similar considerations apply to evaporation). When the steps are far apart, each step edge traps adatoms efficiently and multistep jumps are suppressed [15]. However, when the terrace widths approach the minimal distance, capture of adatoms by steps in the bunch becomes less efficient because this would make some terraces even narrower, leading to an energetically unfavorable configuration. Thus, surface diffusion over the entire bunch becomes more probable. Within our step flow model, this is equivalent to considering *effective multistep jumps* of adatoms, which permit continued evolution of the step bunching process. A reasonable way to take this physics into account is to

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modify Eq. (1) to

$$\frac{\partial X_n}{\partial t} = f_+(Z_n^{(f)}) + f_-(Z_n^{(b)}) + \gamma \frac{\partial^2 X_n}{\partial y^2}, \qquad (3)$$

for $W_n > D_{\min}$ and $\partial X_n/\partial t = 0$ otherwise. $Z_n^{(f)}$ ($Z_n^{(b)}$) is the width of the first terrace in front of (behind) the nth step that is larger than $D_{\min}$.

We now analyze the stability of the bunches. We find, quite surprisingly, that under some conditions the bunches are *unstable towards debunching*. Consider first the dynamics of a single bunch of N straight steps separated by the minimal distance. Denote the width of the wide terrace behind step 1 by $Z^{(b)}$ and the width of the one in front of step N by $Z^{(f)}$. Initially, only step N can move. After it has moved a small distance, it "sees" in (3) the narrow terrace directly behind it (which hardly contributes to its velocity) and the wide terrace in front. Therefore, the velocity of step N is approximately $f_+(Z^{(f)})$.

Once step N has moved, step $N-1$ can move forward; its initial velocity from (3) is approximately $f_-(Z^{(b)})$. However, after it moves slightly, its motion is controlled by the relatively narrow terraces directly behind and in front, so step $N-1$ slows down significantly. Successive steps behave in the same way until step 1 moves and slows down within the minimal distance from step 2. Now, according to (3), step $N-1$ can again move more quickly and the entire process repeats itself. In effect, each step moves with velocity $f_-(Z^{(b)})$ for an infinitesimal time interval δt, and then waits for the rest of the steps to move for a time interval $(N-2)\delta t$. We therefore conclude that at least for a while, steps $1,\ldots,N-1$ stay in one bunch that moves with an average velocity $f_-(Z^{(b)})/(N-1)$. Thus if

$$f_+(Z^{(f)}) > f_-(Z^{(b)})/(N-1), \qquad (4)$$

step N will move faster than the $N-1$ steps behind it, and will *escape* from the bunch. In this case the bunch is unstable towards debunching, and releases a step that moves into the terrace in front of it, until it reaches the bunch ahead. This instability of the bunch leads to an *exchange of single steps* between neighboring bunches. Although this argument for the debunching instability used a specific initial configuration of straight steps, we expect it to hold in many experimentally relevant situations where steps can bend. The bunches should then remain straighter than the single steps moving on the terraces, since the effective stiffness of a bunch of steps is much greater than that of a single step.

In the limit relevant to the sublimation experiments of Si(111) in the presence of an electric current, the diffusion length (the average distance an adatom diffuses before it desorbs) is large compared to terrace widths [1,16]. Therefore $f_+(W) = k_+W$, and $f_-(W) = k_-W$. The uniform step train is unstable towards step bunching when $k_- > k_+$, with $k_- > 0$. (The net step velocity of the uniform step train is non-negative by convention.) If $k_+ > 0$ as well [17], Eq. (4) will hold for small enough $Z^{(b)}$ or large enough N. We then expect to see the unique signature of the instability mechanism discussed in this work: *simultaneous bunching and debunching of steps.*

To test these ideas, we carried out Monte Carlo simulations of the following two-dimensional coarse-grained model of step flow. M steps, each consisting of L segments, reside on a square lattice with periodic boundary conditions in both directions. The position of the yth segment of the nth step is denoted by $X_n(y)$. Distances are measured in units of the lattice spacing and time in Monte Carlo cycles. Each Monte Carlo cycle consists of a "step flow" sweep followed by a "line tension" sweep. In a step flow sweep, we first calculate the maximal possible distance of motion for each individual step segment in one unit of time: $D_n(y) \equiv k_+Z_n^{(f)}(y,t) + k_-Z_n^{(b)}(y,t)$. To use this in our lattice model, we define an integer distance I_n, such that $I_n(y) = [D_n(y)] + 1$ if a random number $0 < x < 1$ is smaller than $D_n(y) - [D_n(y)]$. Otherwise, $I_n(y) = [D_n(y)]$. Here $[D_n]$ is the integer part of D_n. Next we move all the step segments taking into account the minimal distance restriction by setting $X_n(y,t+1) = X_n(y,t) + \min[I_n(y), W_n(y,t) - D_{\min}]$.

The energetics of step bending is taken into account in the line tension sweep. Here we choose a step segment at random and attempt to move it forward or backward with probability 1/2. If the move violates the minimal distance restriction, it is rejected. If the restriction is not violated, we reject the attempted move with probability $1 - \exp(-\beta\Delta E)$ if it raises the line tension energy by an amount ΔE, and accept it otherwise. The line tension energy is $E = \tilde{\gamma}/2\sum_{y,n}[X_n(y+1) - X_n(y)]^2$, and β is an inverse temperature parameter. In each sweep this process is repeated ML times.

We started the simulation with $M = 30$ uniformly spaced straight steps of length $L = 1000$, and performed repeated step flow and line tension sweeps. The initial terrace width was $W = 50$. In Fig. 1 we show a typical configuration after 160 000 cycles with $\beta\tilde{\gamma} = 0.2$, $k_+ = 0.001$, and $k_- = 0.004$. We find fairly straight bunches (the thick lines) coexisting with single strongly bent steps that reside on the terraces. These patterns differ dramatically from the ones we found for the Frank instability [9(d)]. We followed the dynamics of the system and found that, indeed, the bunches move slowly and exchange fast moving single steps between them. We also note that the single steps often arrange themselves into surprisingly uniform *crossing arrays*, where successive steps separate from a bunch into the terrace in front of it, forming large angles with respect to the bunch in the middle of the terrace, and then join the upper bunch. The distance d between steps in a crossing array, in the direction parallel to the bunch behind, and their velocities (in

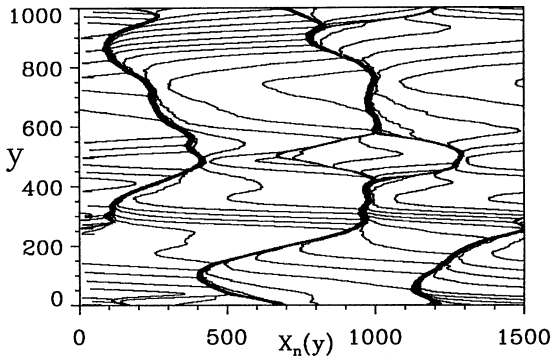

FIG. 1. A snapshot of a system of 30 steps after 160 000 Monte Carlo sweeps of the kinetic model. The simulation parameters for all data reported are $\beta\bar{\gamma} = 0.2$, $k_+ = 0.001$, and $k_- = 0.004$. Steps move from left to right and are marked by solid lines. Heavy solid lines correspond to step bunches.

the same direction) are constant within an array (to a good approximation), but vary from array to array.

We now calculate analytically the shape and velocity of the steps in the crossing arrays as a function of d. Since these steps escape from the bunches, we start by linearizing (1) around an infinite uniform bunch of straight steps, separated by narrow terraces of width $\bar{W}$, with $d \gg \bar{W} \approx D_{\min}$, and tilted with an angle α with respect to the y axis. The infinite uniform bunch configuration is $X_n^0 = n\bar{W} + y\tan\alpha + (k_+ + k_-)\bar{W}t$.

To describe properties of the crossing arrays, we now look for particular solutions with the property that the distance d between two neighboring steps in the direction parallel to the bunch is *independent* of the step index. A family of such solutions for $\delta X_n \equiv X_n - X_n^0$ that also satisfies the boundary condition $\delta X_n(y \to \infty) = 0$ is

$$\delta X_n(y,t) = e^{-[q(y-n\Delta)-\omega(q)t]}, \tag{5}$$

where both q and ω are, in principle, complex numbers, and $q^r > 0$. Here $\Delta \equiv d\cos\alpha$ and superscripts r and i stand for the real and imaginary parts, respectively. A large value of q^r corresponds to a sharp angle that the steps in the crossing array form with respect to the y axis. The linear dispersion relation corresponding to solutions (5) is

$$\omega = k_+(e^{q\Delta} - 1) + k_-(1 - e^{-q\Delta}) + \gamma q^2. \tag{6}$$

We anticipate that the value of q^i yielding the maximal growth rate ω^r will dominate in actual patterns. In our case it is the $q^i = 0$ mode, where both ω and q are real.

It is difficult to measure q^r accurately in the simulations, but it is quite easy to measure $V \equiv \omega^r/q^r$. V is the step velocity parallel to the bunch projected on the y axis. We find, within the numerical accuracy of our simulations, that V takes a well-defined, *unique* value for each value of Δ. In order to explain the selection of definite $q^r(\Delta)$ and $V(\Delta)$, we invoke the ansatz of *marginal stability* [18]. This selection mechanism, although not justifiable rigorously, works well in many cases [9,18]. Marginal stability predicts that the selected q^r satisfies the equation $\omega^r/q^r = \partial\omega^r/\partial q^r$. This leads to the following relation between q and Δ:

$$k_+G(q\Delta) - k_-G(-q\Delta) + \gamma q^2 = 0, \tag{7}$$

where $G(x) \equiv (x - 1)\exp(x) + 1$ and we assumed $q^i = 0$. Further analysis [19] shows that a step pairing mode causes the crossing arrays to be *unstable* for $q\Delta < (q\Delta)_m \equiv 1/2\ln(k_-/k_+)$, the value of $q\Delta$ at the maximum of the $q(\Delta)$ curve given by (7). Equation (7) is physically meaningful for $\Delta \geq \Delta_m \approx 2\Delta_c$, where $\Delta_c \equiv \sqrt{2\gamma/(k_- - k_+)}$; it predicts that q goes to zero for large Δ as Δ^{-1}. We can also obtain the behavior of V from the relation

$$V = \Delta(k_+e^{q\Delta} + k_-e^{-q\Delta}) + 2\gamma q. \tag{8}$$

For large values of Δ, $V \approx a\Delta + b/\Delta$, where a and b are known functions of k_+, k_-, and γ.

To compare with simulations we measured Δ and V for several crossing arrays such as the ones in Fig. 1. These results are shown in Fig. 2 as full circles. Correlation functions associated with step fluctuations [12] suggest the value $\gamma = 0.0665$. We then solved Eq. (7) numerically, and found the function $q(\Delta)$. Using this in Eq. (8) determines $V(\Delta)$. This result is plotted as a solid line in Fig. 2. The agreement between the simulations and the theory for our model is evidently very good.

But is the model itself physically realistic? Both our effective treatment of adatom diffusion through the use of velocity functions $f_\pm(Z(y))$ and the linearization of the curvature in the line tension term in Eq. (4) are quantitatively accurate only for relatively straight steps or bunches oriented close to the y axis. It is quite appropriate to question the validity of our model's description of the sharply angled steps in the middle of the terraces. We now argue that our model is indeed relevant for experimental systems.

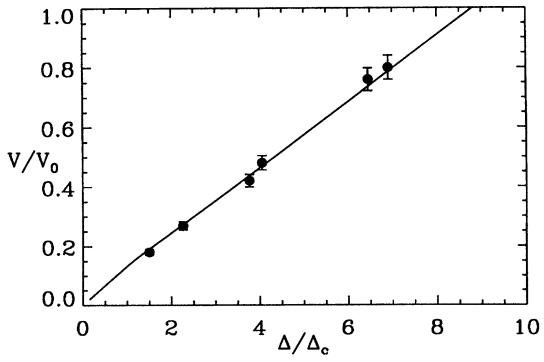

FIG. 2. The velocity V of crossing arrays in the direction parallel to the bunch behind, projected on the y axis, in units of the initial step velocity $V_0 = (k_+ + k_-)W$, as a function of Δ/Δ_c (see text for definition). The solid line is the theoretical prediction based on the marginal stability ansatz. The full circles are results from simulations of the kinetic model.

As discussed above, the essential physics of both the bunching and debunching instabilities is seen in the one-dimensional limit. Thus we do not require the delicate and inherently two-dimensional coupling of the diffusion field and the line tension terms needed for Mullins-Sekerka type instabilities. Since our model is accurate near the relatively straight step bunches, it should properly describe the way a crossing array initially separates from a bunch, and the way it joins another bunch. Note that the marginal stability ansatz we have used to derive the basic relations (6) and (7) relies only on a *linearization* around a solution of straight steps.

Moreover, while our description of the steeply angled steps in the middle of a terrace is certainly inadequate, it does keep those steps relatively straight. Using the full curvature in the line tension term should not change this behavior significantly. We expect that any reasonable approximation scheme which matches the nontrivial behavior of the crossing arrays near the bunch behind to that of the bunch in front would produce qualitatively similar behavior between the bunches.

Thus we expect our results to be directly relevant to experiments. Indeed, there is a striking resemblance between the patterns obtained in the experiments and the ones we get in our simulations. Both the thick fairly straight bunches and the crossing arrays predicted by our model (see Fig. 1) are observed in experiments (see Fig. 1 of [3] and Fig. 1(c) of [4]). Moreover, Latyshev, Krasilnikov, and Aseev [4] observed the escape of single steps from bunches as described above. This strongly suggests that an asymmetry of the type discussed in this work, coupled with effective multistep jumps of adatoms, is responsible for the interesting step behavior observed in the experiments. It should be possible to measure both $q(\Delta)$ and $V(\Delta)$ from the experimental data. One can then evaluate the effective parameters of the model, k_+, k_-, and γ, and check the consistency of the predictions in (7) and (8). To the best of our knowledge, this is the first proposal for a direct experimental determination of these important parameters from step patterns. We hope this work will spur new experiments as well as more detailed theories that will uncover the microscopic origin of the basic asymmetry.

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