

Quantitative theory of current-induced step bunching on Si(111)

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We use a one-dimensional step model to study quantitatively the growth of step bunches on Si(111) surfaces induced by a direct heating current. Parameters in the model are fixed from experimental measurements near 900 °C under the assumption that there is local mass transport through surface diffusion and that step motion is limited by the attachment rate of adatoms to step edges. The direct heating current is treated as an external driving force acting on each adatom. Numerical calculations show both qualitative and quantitative agreement with experiment. A force in the step down direction will destabilize the uniform step train towards step bunching. The average size of the step bunches grows with electromigration time t as t^β , with $\beta \approx 0.5$, in agreement with experiment and with an analytical treatment of the steady states. The model is extended to include the effect of direct hopping of adatoms between different terraces. Monte Carlo simulations of a solid-on-solid model, using physically motivated assumptions about the dynamics of surface diffusion and attachment at step edges, are carried out to study two-dimensional features that are left out of the present step model and to test its validity. These simulations give much better agreement with experiment than previous work. We find a step bending instability when the driving force is along the step edge direction. This instability causes the formation of step bunches and antisteps that is similar to that observed in experiment. [S0163-1829(98)03923-X]

I. INTRODUCTION

In 1989 Latyshev *et al.*¹ made the startling discovery that a direct heating current can induce step bunching on vicinal Si(111) surfaces. When the sample is resistively heated with direct current, steps can rearrange into closely spaced step bunches separated by wide terraces. Around 900°C, the step train is unstable towards step bunching when the current is in the step-down direction, but is stable when the current direction is reversed. Surprisingly, as the temperature is increased to 1190°C, the stable and unstable current directions are reversed, i.e., the step train is unstable with step-up current and stable with step-down current. There is another such reversal as the temperature is increased further.

Since then the phenomenon has received a great deal of attention. Theoretical work has mainly concentrated on two goals: understanding the microscopic physics underlying the instability towards step bunching and the reversal of the unstable current direction with temperature,²⁻⁴ and determining the mesoscopic evolution of the surface morphology as a result of the instability.⁵⁻¹¹ Recently Williams and co-workers¹²⁻¹⁵ carried out a series of measurements on Si(111) surfaces at 900 °C, to provide a quantitative understanding of the dynamics. By controlling the experimental system and comparing with theoretical models, they were able to extract detailed information about the mechanism and to determine quantitative values of relevant parameters. Although the details of the microscopic mechanisms leading to the change in the destabilizing current direction with varying temperature are still not fully understood,²⁻⁴ we show here that there exists a reliable mesoscopic theory that can provide quantitative agreement with a variety of experimental

results in the temperature regime (900 °C) studied by Williams *et al.*

In Secs. II and III, we briefly review some of the experimental and theoretical work that led to our present model. We focus on the case where the *step motion is limited by the attachment rate of adatoms to step edges* (in contrast to being limited by the diffusion rate on terraces). We also assume *local mass transport* by surface diffusion. These assumptions yield a minimal mesoscopic model that is consistent with all previous experimental results. In Sec. IV we give numerical results from this model using realistic parameter values and interpret and analyze some of the results in Sec. V. We briefly discuss in Sec. VI some effects of step permeability^{16,17} (direct adatom hops from one terrace to another), which might be important in other systems, e.g., Si(001). In Sec. VII we present some results of Monte Carlo simulations of a microscopic solid-on-solid model, using physically motivated assumptions about the dynamics of surface diffusion and attachment at step edges. These results are in qualitative agreement with experiment, in contrast to previous work⁹⁻¹¹ using conventional Metropolis dynamics. They also help in the understanding of additional two-dimensional (2D) features and instabilities that cannot be described by the simple 1D step model. Final remarks are given in Sec. VIII.

II. 1D STEP MODEL WITH EXTENDED VELOCITY FUNCTIONS

Vicinal surfaces, which are created by a miscut along a low-index plane below the roughening temperature, are most naturally and usefully described by a model of interacting

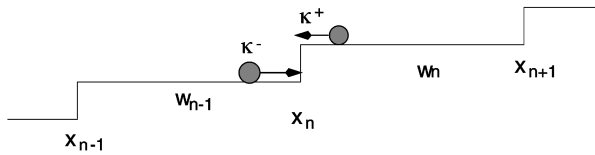


FIG. 1. Illustration of the labeling of steps and terraces and kinetic coefficients.

steps of the same sign. Because of the inherent anisotropy of the underlying crystal structure, these surfaces often exhibit quasi-one-dimensional features, thus making a 1D step model useful and accurate. Also the number of steps is often conserved as the surface evolves, permitting further simplifications in the analysis.

The change in the morphology of vicinal surfaces can be described in terms of the velocity of each step as long as no steps are created or destroyed. The classic Burton-Cabrera-Frank¹⁸ (BCF) treatment assumes that the mass transfer is governed by a set of adatom diffusion equations on each terrace, with steps acting as perfect sinks and sources for adatoms (or vacancies) so that local equilibrium is always maintained. However, the original BCF picture is valid only for simple materials where adatom diffusion is the rate limiting process. Extensions of the BCF model can be made to include a finite attachment/detachment term in the boundary conditions. This is needed for materials like silicon, where the atom exchange rate between steps and terraces is not fast enough to permit the adatom concentration near the step edge to achieve local equilibrium.

In his important work on the instability induced by a direct heating current on Si(111), Stoyanov² proposed such a modified BCF model, including both a finite adatom attachment/detachment rate at step edges and an adatom drift velocity (or equivalently, an external driving force due to the electric field). Natori⁷ extended the work of Stoyanov to include step repulsions. The idea of incorporating step interactions in a generalized BCF model has been further developed by Sato and Uwaha.⁶

To describe the exchange of atoms or vacancies between steps and their neighboring terraces (attachment/detachment), we use a linear kinetics theory and write the net surface flux from step n to terrace n (see Fig. 1 for the labeling) as

$$j_n^+ = \frac{\kappa_+ c_{\text{eq}}}{kT} [\mu_n - \mu^t(x_n^+)], \quad (1a)$$

and the flux from step n to terrace $n-1$ as

$$-j_n^- = \frac{\kappa_- c_{\text{eq}}}{kT} [\mu_n - \mu^t(x_n^-)], \quad (1b)$$

where μ_n is the atom chemical potential at step n , defined as the increase in free energy per atom when atoms attach to the step. For a 1D step train with elastic and entropic step repulsions, this can be written as¹⁹

$$\mu_n = 2gh^3 a^2 \left(\frac{1}{w_{n-1}^3} - \frac{1}{w_n^3} \right), \quad (2)$$

where a^2 is the area of a single atomic cell on the surface, h the single step height, and $w_n \equiv x_{n+1} - x_n$ the width of terrace n . Here the parameter g is just the coefficient of the s^3 term in the well-known Gruber-Mullins²⁰ form for the projected free energy of vicinal surfaces with slope s . $\mu^t(x_n^\pm)$ is the adatom chemical potential on the terrace adjacent to step n , approaching x_n from the right-hand (+) and left-hand (-) side.²¹ c_{eq} is the equilibrium adatom concentration on terraces. We assume there is no asymmetry for the kinetic coefficient for adatom attachment from upper and lower terraces ($\kappa_+ = \kappa_-$).

To determine the adatom chemical potential on terraces $\mu^t(x)$, we need to know the mass transport mechanism on the surface. If it is much easier for adatoms to hop directly across a step edge from one terrace to another one in comparison to attachment at the step edge, then the adatom chemical potential becomes a constant on all terraces. We refer to this as *nonlocal mass transport* (case A). The other limit is when there is no significant hopping of adatoms over the step edge, as assumed in the BCF model; we call this the limit of *local mass transport* (case B). Experiments on Si(111) show that the relaxation rate of a step bunch with N steps scales with $N^{-\alpha}$ where $\alpha = 4.3 \pm 0.5$. As we have discussed in detail elsewhere,^{14,22} this is consistent with the *local mass transport limit* (case B), and we will assume this limit in most of this paper. In Sec. VI we will consider a more general scenario.

Assuming local mass transport, the step velocities $v_n(t)$ can be determined by solving the diffusion equation for adatoms on terraces with boundary conditions at step edges governed by linear kinetics. The equations can be written generally in an *extended velocity function form*:^{23,24}

$$v_n = f_+(w_n; \mu_n, \mu_{n+1}) + f_-(w_{n-1}; \mu_{n-1}, \mu_n). \quad (3)$$

We will not write down the general form for the velocity functions f_\pm for the electromigration problem since it is very complicated and not very instructive for our purpose. A simple limit that is consistent with experiment will be discussed below. More general expressions have been given by many authors.²⁵

In studies of surface dynamics, it is often convenient to consider idealized models where the kinetics is limited by a few slow processes on the surface, and the rates of other faster processes are taken to infinity. For BCF models, neglecting evaporation and deposition, there are two basic rates, the attachment/detachment rate κ , and the adatom diffusion rate D_s . Bartelt *et al.*²⁶ estimated the attachment/detachment rate from the step fluctuations of Si(111) at 900 °C under the assumption that attachment/detachment is the rate limiting process, while Pimpinelli *et al.*²⁷ estimated from the same data the diffusion rate under the assumption that adatom diffusion is the rate limiting process. It is useful to define a length scale $d \equiv D_s/\kappa$. When d is very small, the step dynamics is said to be *diffusion limited*, and when d is very large, the dynamics is *attachment/detachment limited*.

However, direct estimation of this ratio is difficult. For example, Table I lists several sets of parameters that give good agreement with experiments¹⁵ on the electromigration-driven relaxation of step bunches on Si(111), with d ranging from 10 to 10^5 Å. Physically d has to be finite and whether

TABLE I. Sets of parameters that give a good fit to the relaxation of step bunches. We use $\Gamma = 2c_{\text{eq}}a^4\kappa$ ($c_{\text{eq}}a^2 = 0.2$ ML) to compare with previous work. The table is taken from Ref. 15.

Parameter set	d (Å)	q (e)	Γ (Å ³ /s)	D_s (Å ² /s)
A	100 000	0.006	3×10^7	5.2×10^{11}
B	5000	0.006	4×10^7	3.4×10^{10}
C	100	0.03	3×10^8	5.2×10^9
D	10	0.2	2×10^9	3.5×10^8

a system is diffusion limited or attachment/detachment limited depends on the comparison of d with other length scales, e.g., the typical terrace width. Here we refer to the mathematical limit $d \rightarrow \infty$ as the *complete attachment/detachment limited* model, and we call the limit $d \rightarrow 0$ the *complete diffusion limited* model.

As suggested by Table I, the relaxation experiments can be explained using an effective charge q of the adatoms that reaches a finite and physically reasonable value as $d \rightarrow \infty$. In contrast, q must tend to the unphysical limit ∞ when $d \rightarrow 0$. Therefore the complete attachment/detachment limited model is well defined, and we will use this limit to illustrate the mechanism for electromigration. As we show below, Eq. (3) simplifies considerably in this limit. Moreover, a value of $d \geq 3000$ Å is predicted by extrapolating the diffusion rate from higher temperatures using a diffusion activation energy¹³ of 1.1 eV. However, numerical solutions of Eq. (3) using any of the parameter sets in Table I are consistent with the step bunching experiments, including the power law for coarsening.

As in the Stoyanov model,² we assume that there is a force F acting on each adatom because of the electric field. The adatom flux on terrace n under this driving force is

$$j_n = \frac{D_s c_{\text{eq}}}{kT} \left(-\frac{\partial \mu^t}{\partial x} + F \right). \quad (4)$$

With complete attachment/detachment limited kinetics, D_s tends to infinity relative to the attachment rate κ , or j_n . Therefore the adatom chemical potential $\mu^t(x)$ has a *constant gradient* F on each terrace. In general, $\mu^t(x)$ is affected by the motion of the neighboring steps, but usually the steps move very slowly so that they can be treated as effectively stationary as far as the diffusion of adatoms is concerned. Under this *quasistatic approximation*,²⁸ at any given time, the total surface flux into terrace n from the two neighboring steps ($j_n^+ - j_{n+1}^-$) equals the total amount of evaporation from this terrace, which is given by $c_{\text{eq}} w_n / \tau \equiv w_n / \tau_e$, where τ is the average lifetime of an adatom on the terrace before it evaporates. With these approximations, the step velocities in Eq. (3) can be written in the simple form^{23,24}

$$v_n = \frac{\kappa c_{\text{eq}} a^2}{2kT} (2\mu_n - \mu_{n+1} - \mu_{n-1}) + k_+ w_n + k_- w_{n-1}, \quad (5)$$

where

$$k_{\pm} = \pm \frac{\kappa c_{\text{eq}} a^2 F}{2kT} + \frac{1}{2\tau_e}. \quad (6)$$

III. EXPERIMENTAL DETERMINATION OF THE PARAMETERS

Williams *et al.* devised a series of experiments to measure the various parameters and also test the assumptions about the mass transport limits discussed in the previous section. As mentioned earlier, on Si(111) at 900 °C, the relaxation of step bunches is consistent with the local mass transport limit. The step interaction parameter g can be measured^{29,30} from the distribution of terrace widths and step positions at equilibrium, and has an estimated value around 0.015 eV/Å². Assuming attachment/detachment limited kinetics for mass transport, the kinetic coefficient κ can be measured independently from the thermal fluctuations of the steps and the relaxation of step bunches. Rather than using κ , we define $\Gamma = 2c_{\text{eq}}a^4\kappa$ to compare with earlier work. Γ gives the step mobility for the Brownian motion of an isolated step²⁶ and is measured to be around 5×10^7 Å³/s. This value also gives a good fit to the relaxation of step bunches, thus providing additional evidence supporting the local mass transport assumption aside from the scaling behavior mentioned before.

The force on adatoms due to the direct heating current can be measured from the relaxation of the step bunches that occurs after reversing the current to the stable direction.¹⁵ The force acting on each adatom can be conveniently described in terms of an effective charge q , with $F = qE$, where the experimental value of $E = 7$ V/cm. Table I lists four sets of parameters that give good fits to the decay of step bunches with a direct current in the stabilizing direction at 900 °C. As mentioned before, as d becomes very large, the values of q and Γ reach limiting values, which we use in the complete attachment/detachment limited model. Other relevant parameters include the average terrace width $w_0 = 1100$ Å, and the evaporation time for one monolayer $\tau_e = 1250$ s.¹³

IV. NUMERICAL RESULTS AND COMPARISON WITH EXPERIMENTS

When the adatom drift velocity is in the step down direction ($F < 0$), one can show from a linear stability analysis using the parameters in Table I that a uniform step train is unstable towards step bunching. The evolution of the step bunches is determined by numerically integrating Eq. (3), starting from a step train with small random deviations from uniform spacing. The system continues to coarsen by forming larger and larger step bunches. Figure 2 shows the average bunch size N_{ave} for a system of 2049 steps as a function of time, using Eq. (5) for the complete attachment/detachment limited model. A bunch is defined by a number of adjacent steps with no terraces between them larger than $w_0/2$. The average bunch size is defined by $\Sigma_n n \rho_n / \Sigma_n \rho_n$, where ρ_n is the number (or density) of bunches consisting of n steps. It can be fitted by a t^β growth rate with $\beta \approx 0.50$. Results using the more complicated velocity functions in Eq. (3) obtained from solutions of a generalized BCF equation and parameter set A are almost indistinguishable on this scale.

This compares very well with the STM results of Yang, Fu, and Williams.¹³ They show at both 945 °C and 1245 °C that the growth of the facet sizes Z between two step bunches satisfies t^β where $\beta \approx 0.5$. It is a good approximation to re-

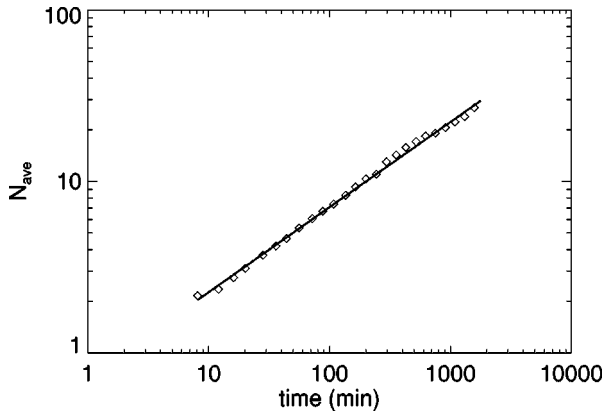


FIG. 2. Evolution of the average bunch size using parameter set A. The solid line is a fit to t^β with $\beta=0.50$.

late the average number of steps in a bunch N_{ave} to Z/w_0 . They observed that at 945 °C, after about 120 min of annealing time, the average terrace width between step bunches grows to about 3500 Å. In the numerical simulations, the average terrace width grows to about 6800 Å in the same time. We consider this quite satisfactory agreement, given the uncertainties in the values of the experimental parameters we used. Thus, not only does the step model give the correct power-law growth rate, it also gives good quantitative agreement with experimental results at 945 °C.

Simulations using other sets of parameters in Table I produce slightly different results, but all agree with the experimental data within the errors in measured parameters. Moreover, they all have approximately a $t^{1/2}$ coarsening rate during the time simulated and experimentally observed. Therefore we cannot determine a unique set of microscopic parameters accurately from the coarsening rate alone.

Dobbs and Krug¹¹ also obtained a $t^{1/2}$ coarsening rate from simulations of a 2D solid-on-solid model using Metropolis dynamics. However, they obtained the $t^{1/2}$ behavior only when there is significant *lateral* fluctuations of step bunches as can sometimes occur in the later stages of coarsening, while initially the growth law they observed went as $t^{1/4}$. Experimentally there is no such transition, and we obtain this kind of coarsening from a 1D model with straight steps. Moreover, as discussed below in Sec. VII, there are other unphysical surface features that arise from the use of Metropolis dynamics to describe Si(111) and related systems, and we suggest there an alternative dynamical scheme that gives good qualitative agreement with experiment.

As another application of the step model, we also simulate the step bunching occurring under *growth conditions*. It is well known that a Schwoebel barrier³¹ has very different effects on growth and evaporation. For example, if there is an additional barrier for an adatom to attach to a step edge from the upper terrace, a 1D step train will be stabilized under growth and destabilized under evaporation. However, even in the absence of a Schwoebel asymmetry (that is, even when $\kappa_+ = \kappa_-$) as assumed here, simulations of the present step model under growth conditions show a *decrease* in the bunching rate with increasing deposition rate. Figure 3 shows the dependence of the average bunch size as a function of time for different growth conditions. It is useful to define $\bar{R} = R\tau/c_{eq}$ as the ratio between deposition and de-

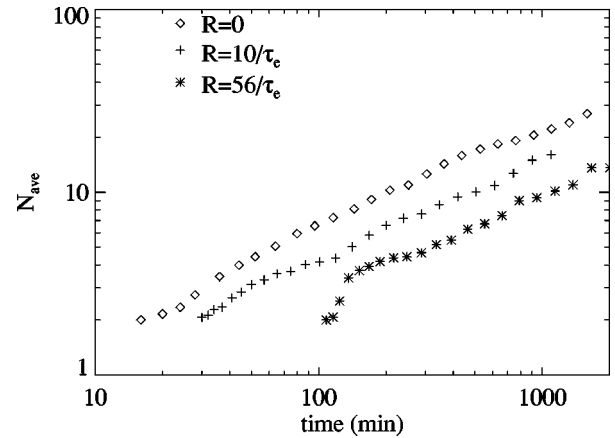


FIG. 3. Effects of growth conditions on the step bunching.

sorption rates. As \bar{R} increases, the bunching rate decreases, in good agreement with the experimental results of Yang, Fu, and Williams.¹³ This decrease in the coarsening rate with increasing deposition was also noted by Tersoff *et al.*³² in their study of stress-induced step bunching. As Kandel and Weeks⁵ argued, when the step train is traveling in a certain direction (e.g., due to deposition or evaporation), a step at the front end of a step bunch can leave the bunch and join with the step bunch in front of it, causing an exchange of steps between step bunches. As the growth rate increases, the velocities of these crossing steps get larger and larger, so more debunching occurs, thus reducing the coarsening rate. A detailed study of the effect of debunching requires a 2D model and is beyond the scope of this paper.

V. ANALYSIS: QUASI-STEADY STATE AND COARSENING RATE

In this section we will try to understand analytically some of the numerical results. Although it is straightforward to use the solution of the diffusion equation to determine the velocity functions in Eq. (3) when simulating the step bunching numerically, it is more convenient and instructive to consider the simple linear velocity function model of Eq. (5). Initially we also neglect any deposition or evaporation.

When the surface flux j is a constant everywhere, the surface is in a steady state. As we will show later, such a steady state is possible when F is in the step down direction. Figure 4 is snapshot of the profile of the surface during one particular simulation. Assuming that the step bunch between A and B is in a steady state with a surface atom flux j^* , we have from Eqs. (1a), (1b), and (14),

$$(N_s - 1)j^* = \frac{\kappa c_{eq}}{2kT} (\mu_A - \mu_B + F w_{AB}), \quad (7)$$

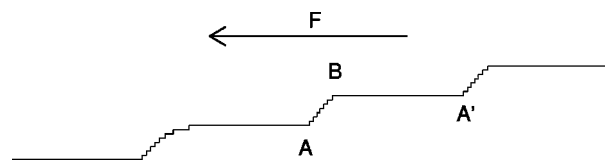


FIG. 4. Typical profile of step configurations when step bunches are induced by a force acting on adatoms in the step down direction.

where N_s is the number of steps in the bunch and w_{AB} is the distance between A and B . On the flat terrace between B and A' the flux is given by

$$j^* = \frac{\kappa c_{\text{eq}}}{2kT} (\mu_B - \mu_{A'} + F w_{BA'}). \quad (8)$$

For a *periodic array* of step bunches, $\mu_A = \mu_{A'}$, and the steady-state flux is then

$$j_{\text{ss}}^* = \frac{1}{N_s} \frac{\kappa c_{\text{eq}}}{2kT} F w_{AA'} = \frac{\kappa c_{\text{eq}} F w_0}{2kT}. \quad (9)$$

Therefore the steady-state adatom current is independent of the size of the step bunches. This result is valid for a periodic array of step bunches and is important in determining the time scaling exponent, as will be shown at the end of this section.

The steady-state profile of the step bunch can be calculated numerically. Here we go to the continuum limit, which is a good approximation when the number of steps in the bunch is large. If $z(x)$ denotes the height of the surface, then the slope is $z_x(x) \equiv \partial z / \partial x$. Using the continuum version of the Gruber-Mullins free energy functional^{20,33}

$$H = L_y \int g |z_x(x)|^3 dx, \quad (10)$$

which is appropriate for vicinal surfaces below the terrace roughening temperature, we can write the adatom chemical potential as³³

$$\mu(x) = a^2 h \frac{\delta H}{\delta z} = -6 g a^2 h |z_x| z_{xx}. \quad (11)$$

Here z_{xx} is the second derivative of $z(x)$ with respect to x . As in the step model discussed earlier, we can drop the linear step energy term if no new steps are created. Note that in the continuum description slopes of different signs correspond to positive and negative steps. Here we consider step profiles with positive slopes everywhere and thus can set $|z_x| = z_x$. In the attachment/detachment limit, the surface flux is given by

$$j(x) = \frac{\kappa c_{\text{eq}}}{2kT} \frac{h}{z_x} \left[F - \frac{\partial}{\partial x} \mu(x) \right]. \quad (12)$$

The above equation has the physical property that the adatom mobility (as the response to an external field) is inversely proportional to the slope z_x . To calculate the step bunch profile $z(x)$, we can neglect the first term in Eq. (12) if the terrace width inside a bunch is much smaller than the distance between bunches. For an isolated step bunch with $j(x) = j^*$, using Eq. (11), we have

$$\frac{3gh^2 \kappa c_{\text{eq}}}{kT} \frac{\partial}{\partial x} (z_x z_{xx}) = j^* z_x. \quad (13)$$

This can be reduced to

$$z_x^3 = \frac{j^* kT}{2\kappa gh^2 c_{\text{eq}}} [(z - z_0)^2 - (H/2)^2], \quad (14)$$

where z_0 and H are integration constants. The step bunch profile $z(x)$ can be easily calculated numerically by integrat-

ing Eq. (14), or analytically in terms of hypergeometric functions. Equation (14) was first derived by Nozières³³ in his study of surface dynamics below the roughening transition. The maximum slope of the step bunch with $j^* < 0$ is given by $z_x^{\text{max}} = [-j^*(H/2)^2 kT / (2\kappa gh^2 c_{\text{eq}})]^{1/3}$. For a true steady state with a periodic array of step bunches, $j^* = j_{\text{ss}}^*$ [Eq. (9)]. We expect j^* to fluctuate around this value for a system in a quasi-steady state. H is approximately the height of the step bunch for large bunches. We have $z_x^{\text{max}} \sim N_s^{2/3}$ since j_{ss}^* is independent of N_s . This can be experimentally tested by measuring the average slope of the step bunch as a function of the average bunch size. Note that the continuum limit breaks down near the edges of a step bunch where sharp changes in the local slope occur.

Strictly speaking, the above analysis only holds for steady-state profiles with complete attachment/detachment limited kinetics, but we expect it to be a good approximation for the quasi-steady-state profiles that arise as the step bunches slowly coarsen with time. Indeed simulations and experiment agree that the step bunches coarsen with time as t^β with $\beta \approx 0.5$. This $t^{1/2}$ power law can be justified by a scaling argument. We assume that as $t \rightarrow \infty$, there is only one characteristic length for the system, which scales as t^β . We can thus write all the variables on the surface in terms of the scaled length x/t^β at time t . We have noted that the steady-state flux is independent of the size of the step bunches [Eq. (9)]. For a system in a quasi steady state, we thus assume that the flux can be written as a function of the scaled length only and there is no extra time dependence, i.e.,

$$j(x, t) = J(x/t^\beta). \quad (15)$$

In contrast, the surface profile should maintain a constant average slope and thus must have the following scaling form:

$$z(x, t) = t^\beta Z(x/t^\beta). \quad (16)$$

Substituting these into the equation expressing microscopic mass conservation:

$$\frac{\partial}{\partial t} z(x, t) \sim - \frac{\partial}{\partial x} j(x, t), \quad (17)$$

we have $\beta = 1/2$ by comparing the leading exponents on both sides of this equation. This prediction is in good agreement with both the experimental and the numerical work, as shown in Fig. 2.

VI. EFFECTS OF STEP PERMEABILITY

In the previous sections we focused our study of the step model on vicinal Si(111) surfaces around 900 °C. Our basic approach can be applied more generally, though the limits we used above are not necessarily satisfied. However as long as the mass transport is local, other differences from our present model, e.g., a finite diffusion length or asymmetric step edge attachment rates (Schwoebel barriers), can be studied using the extended BCF model and the velocity function approach of Eq. (3) in a more or less straightforward way.

We will not detail this work here, but instead turn to the conceptually interesting case of *step permeability*, which can make the extended BCF picture no longer valid. This is motivated in part by recent work by Tanaka *et al.*¹⁷ and

Stoyanov.³⁴ In the classic BCF picture with local mass transport, the only way to achieve adatom transport from one terrace to another is through attachment to and subsequent detachment from the step edge separating them. However, if there is *direct hopping* of adatoms across a step edge without incorporating into the step edge first, this causes a coupling of diffusion fields on adjacent terraces that must be taken into account.

In a BCF-like picture, the adatom chemical potentials can have discontinuities at step edges either because steps are perfect sinks, or because there is a strong diffusion barrier near the step edge. In their analyses of island flattening on Si(001), Tanaka *et al.*¹⁷ introduced an adatom hopping term between adjacent terraces over a step edge proportional to the difference between the chemical potentials on the two terraces. Although this interterrace hopping could be fast compared with attachment of adatoms to the step edge, it could still be slow compared with diffusion on flat terraces, thus allowing discontinuities in the adatom concentration field at step edge positions. Here we take this limit, assuming that the adatom diffusion rate on flat terraces is always much faster than *both* the attachment and interterrace hopping rates.

The effect of step permeability on the relaxation of step bunches due to step repulsions can be studied straightforwardly. In the absence of an external driving force, the adatom chemical potential on each terrace is a constant denoted by μ_n^t (see Fig. 1 for the labeling of terraces). We can write the net surface flux at the right-hand side of step n as

$$j_n^+ \sim \kappa (\mu_n - \mu_n^t) + p (\mu_{n-1}^t - \mu_n^t), \quad (18a)$$

and the flux at the left-hand side of step n as

$$j_n^- \sim \kappa (\mu_{n-1}^t - \mu_n) + p (\mu_{n-1}^t - \mu_n^t). \quad (18b)$$

Assuming again the quasistatic limit where $j_n^+ = j_{n+1}^-$, we can solve μ_n^t for any given set of μ_n from a system of linear equations. The analytic solution is given in the Appendix. It is easy to see that the $p = \infty$ and $p = 0$ limits correspond to case *A* and case *B* dynamics, respectively.

As mentioned in Sec. II, experiments¹⁴ on the relaxation rate of step bunches on Si(111) near 900 °C show a size scaling exponent $\alpha = 4.3 \pm 0.5$, consistent with the $p = 0$ limit ($\alpha = 4$). In comparison, if we assume $p = 2\kappa$, we obtain $\alpha = 3.6$ for the bunch sizes used in experiments and larger p will give even smaller α . Therefore we conclude $p < 2\kappa$ and can dismiss the importance of step permeability for Si(111) at 900 °C. However, p can be large for other systems, or even perhaps for Si(111) at different temperatures. Tanaka *et al.*¹⁷ estimated $p = 36\kappa$ on Si(001) at 950 °C. Here we discuss some interesting effects step permeability has on electromigration, which could be a way to detect any significant permeability if it exists.

The adatom concentration field has a constant gradient on each terrace when there is a driving force. We need to be more precise in our description of the microscopic origin of step permeability to obtain a complete theoretical description. Here we consider the case where the step permeability is proportional to the difference between the *local* adatom chemical potentials immediately to the left- and right-hand sides of the step. Equations (18a) and (18b) then become

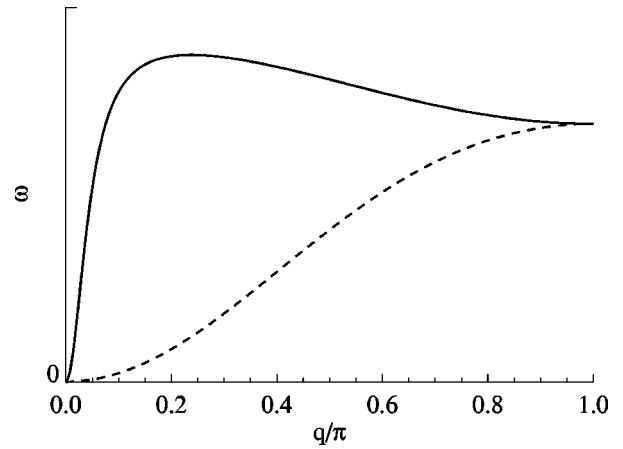


FIG. 5. Linear instability of a uniform step train. The local hopping of adatoms over step edges (step permeability), coupled with the step repulsions, moves the maximum instability away from the step pairing mode ($q = \pi$) to longer wavelengths. The solid line is for $p/\kappa = 100$ and the dashed line is for $p = 0$. Other parameters are taken from set *A* in Table I, although we do not attempt to describe Si(111) realistically here.

$$j_n^+ \sim \kappa [\mu_n - \mu^t(x_n^+)] + p [\mu^t(x_n^-) - \mu_n^t(x_n^+)] \quad (19a)$$

and

$$j_n^- \sim \kappa [\mu^t(x_n^-) - \mu_n] + p [\mu^t(x_n^-) - \mu_n^t(x_n^+)]. \quad (19b)$$

$\mu^t(x)$ can be determined in much the same way as before by assuming there is a gradient, F , in $\mu(x)$ on each individual terrace. In a uniform step train, the external force creates a local chemical potential gradient, and introduces a ‘leak’ of surface flux from the permeability term. We now show that the ‘leak’ will create a *long wavelength* bunching instability, in contrast to the pairing instability familiar from the BCF picture.

In a linear stability analysis, the step positions are written as

$$x_n(t) = \sum_{\phi} e^{in\phi + \omega(\phi)t} u_{\phi}(0) + nw_0, \quad (20)$$

where

$$u_{\phi}(0) = \frac{1}{N} \sum_n e^{-in\phi} [x_n(0) - nw_0], \quad (21)$$

for small perturbations from uniform configurations. Figure 5 plots the amplification exponent ω of a uniform step train as a function of a dimensionless wave number ($\phi = \pi$ corresponds to the pairing mode). The solid line is for $p/\kappa = 100$ and the dashed line is for $p = 0$. The maximum linear instability has shifted to much longer wavelengths. Note that very strong repulsive interactions could also produce such a shift.⁶ However, for systems with step permeability, there is a very rapid (almost linear) growth in the average size of step bunches in the initial stage, which then crosses over to the $t^{1/2}$ behavior. In contrast, for the purely repulsive system, the growth rate is approximately $t^{1/2}$ at all times. These characteristics could be used to detect step permeability if it is very large.

VII. MONTE CARLO SIMULATIONS OF THE 2D SOLID-ON-SOLID MODEL: MODIFIED ARRHENIUS DYNAMICS

For Si(111) at 900 °C the steps are mostly straight and a 1D model is adequate for most purposes. However, at higher temperatures, there exists noticeable bending of steps. For example, at around 1100 °C when evaporation is significant, 2D arrays of *crossing steps* form between step bunches. Kandel and Weeks⁵ proposed a (quasi) 2D step model where the velocity of each step depends only on the *local* neighboring terrace widths in the direction perpendicular to the average step edge direction. This model reproduced many features of the crossing arrays quite accurately.¹² Further developments along these lines have been reported in Refs. 23 and 24.

A full 2D step model taking account of 2D adatom diffusion on terraces with boundary conditions on the moving curved steps is very difficult to study. Also it is necessary to go beyond the BCF framework, which excludes the creation of new steps to explain the antistep bunches reported by Latyshev *et al.*³⁵ Here we study a generalized 2D solid-on-solid (SOS) model that takes explicit account of a *step edge barrier* in the kinetics of adatom attachment/detachment at step edges. We believe this is probably the simplest 2D microscopic model that can provide a physically reasonable description of both adatom diffusion and step motion in Si(111) and related systems. However, there is almost no hope of simulating the long time behavior of such a microscopic model using realistic parameter values. Thus, in contrast to the 1D step model we studied above, here we concentrate only on qualitative properties. Specifically, we consider a very large external driving force along with a very small average terrace width. These extreme choices will permit significant step motion in the computer time available to us.

The SOS model is defined on a square lattice with total energy

$$\mathcal{H} = \sum_{\langle ij \rangle} \epsilon |h_i - h_j|, \quad (22)$$

where h_i is the column height and $\langle ij \rangle$ denotes nearest-neighbor pairs on a square lattice. Surface diffusion is simulated by exchange of atoms on top of a nearest-neighbor pair of columns ($h_i \rightarrow h_i - 1$ and $h_j \rightarrow h_j + 1$, where i, j are nearest-neighbor sites). A driving force is simulated by asymmetric attempt frequencies in directions along and opposite to the force direction. For example, when the force is in the x direction, we assume the attempt frequencies in the $+x$ and $-x$ directions satisfy the following relation:

$$p_{+x}/p_{-x} = \exp(2Fa/k_B T). \quad (23)$$

Our next task is to describe how the probability Γ_{ij} for an adatom hopping from i site to j depends on surface configurations. Often in statistical physics, *Metropolis dynamics* is used, where the hopping probability depends on the energy difference between the final and initial state ΔE_{ij} through the relation

$$\Gamma_{ij} = \min[1, \exp(-\Delta E_{ij}/kT)]. \quad (24)$$

This dynamics often has the virtue of fast equilibration in the absence of a driving force since there are no barriers for

movements with no change in energy, but it does not usually provide a physically realistic description of actual dynamical processes.

Krug and Dobbs⁹⁻¹¹ have studied in detail the effects of an external driving force combined with Metropolis dynamics in a SOS model. They used these simulations along with a continuum model to “describe the *universal* features” of the electromigration problem. However, the resulting surface structures have several artificial features that do not resemble experiments on Si(111) surfaces. For example, in their simulations, a surface instability develops regardless of the current direction and then there are no extended flat regions of the surface with $\nabla h = 0$. Experiments on Si(111) generally reveal flat terraces and individual steps coalescing into bunches when the current is in the unstable direction, and reversing the current direction will *stabilize* the uniform step train.

Of course it is possible that at much later times some limiting features of both the experiments and simulations could be insensitive to the choice of dynamics, and hence universal. For example, most driven surfaces eventually become “rough”; because of transverse step fluctuations³⁶ this probably holds true in principle for the experiments at sufficiently large length and time scales even when the current is in the nominally “stable” direction. We show here that with a more physically motivated choice of dynamics, the SOS model can provide a qualitatively accurate description of the length and time scales probed by present experiments, as well as of any longer time “universal” features, if such exist.

Diffusion on surfaces is usually an activated process with an energy barrier. A different dynamical scheme, *Arrhenius dynamics*, takes this physics into account in an extreme way by assuming that the energy barrier is simply the binding energy of the atom, independent of the final configuration. However, Krug *et al.*¹⁰ found that there is *no* morphological change for this dynamics under an external driving force. They showed in general that instabilities in a continuum model are associated with the dependence of the adatom mobility on the local slope, while instabilities in a microscopic model require a dependence of the hopping probability on the final configuration. Here we obtain such a configuration dependence by modifying the original Arrhenius dynamics, which provides a reasonable description of activated processes such as surface diffusion, to include an *extra barrier* that arises from the presence of steps.

This energy barrier is motivated by the physics of rebonding and surface reconstruction that can occur near steps. The surface atoms near steps on Si(111) surfaces usually rearrange themselves and rebond in characteristic ways to lower the step energy.³⁷ To incorporate an *additional* adatom into the step usually involves the collective motion of many atoms as this rebonding is modified. This process has a higher activation energy than the simple pairwise additive bond picture in the usual SOS model would suggest. Also, in many cases the repeatable step unit, the kink, has a complex structure, and requires the incorporation of two adatoms to bring about its movement. To take account of this physics in our simulations in a simple way, we assign an additional barrier for any movement that lowers the energy, since all attachment events are associated with a decrease in energy. So that

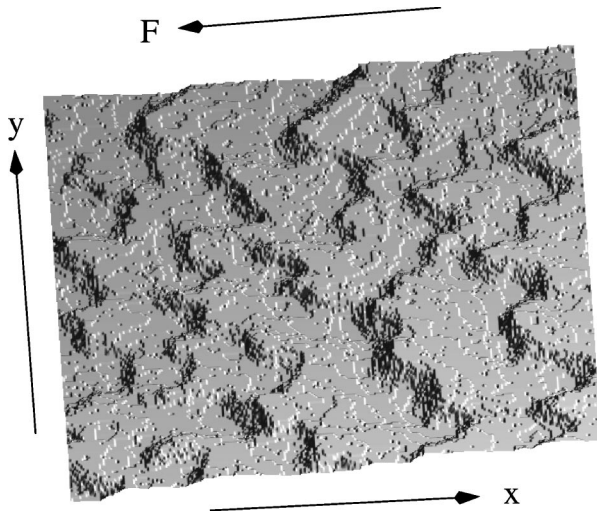


FIG. 6. Snapshot of a simulation using a solid-on-solid model with a diffusion bias perpendicular to the step edge direction, after about 2.1×10^6 Monte Carlo steps. Parameters used here are $kT = 0.8\epsilon$, $p_x = 0.3$, $p_{-x} = 0.7$, $p_y = p_{-y} = 0.5$, and the size of the system is 512×512 . The dark lines are normal (up) steps and white lines are antisteps (down steps).

detailed balance holds in equilibrium, the same barrier must also be added to a movement that increases the energy. We call this scheme *modified Arrhenius dynamics* and thus assume

$$\Gamma_{ij} = \begin{cases} \exp(-2\epsilon n_i), & \Delta E_{ij} = 0 \\ b \exp(-2\epsilon n_i), & \Delta E_{ij} \neq 0, \end{cases} \quad (25)$$

where $b < 1$ and n_i is the number of horizontal bonds the surface atom at site i has.

This way of introducing an attachment barrier was suggested by Bartelt *et al.*³⁸ in their study of step fluctuations. We view modified Arrhenius dynamics as a convenient but not necessarily unique microscopic scheme that produces the ‘‘right’’ boundary conditions (giving in particular a finite value for the kinetic coefficient κ) in the mesoscopic step models discussed in previous sections. Thus the dynamical behavior of mesoscopic and macroscopic scale features in the simulations should be physically meaningful.

We start the simulations with a uniform step train with steps orientated along the vertical (y) direction. The height of the surface increases along the positive x direction. Periodic boundary conditions are used along the y direction. In the x direction we require $h(x + L_x, y) = h(x, y) + N_0$, where N_0 is the initial number of steps in the system. For a system of size $L_x \times L_y$, the initial average terrace width $w_0 = L_x / N_0$. With a diffusion bias in the average step down direction ($p_{+x} < p_{-x}, p_{+y} = p_{-y}$), the system is unstable towards step bunching. The step bunches continue to coarsen, consistent with the results of previous sections. Figure 6 is a snapshot of a typical configuration after some bunching has occurred. The dark regions are step bunches, and single height crossing steps are visible between them.

The qualitative features of the simulations are very similar to the experiments, and also to the predictions of the step model. Vicinal surfaces are stable during the time simulated when the driving force is in the step up direction, and un-

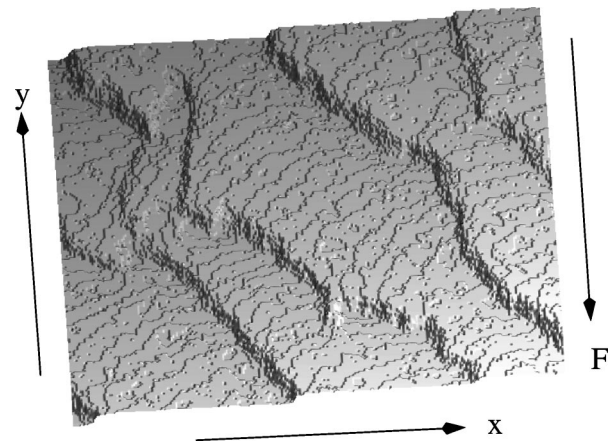


FIG. 7. The same parameters as in Fig. 6, except that the driving force direction is parallel to the average step edge direction ($p_y = 0.3$, $p_{-y} = 0.7$, $p_x = p_{-x} = 0.5$). The initial steps are along the vertical (y) direction.

stable towards step bunching when the force is in the step down direction. Crossing steps form when there is significant evaporation. Preliminary results show that the coarsening rate is consistent with the $t^{1/2}$ power law, but so far the system size and simulation time are too small to determine the exponent accurately.

In the 2D step model studied in Refs. 5 and 24, the steps are all ascending (or descending) at a given y position. Although there is significant step bending, steps cannot form overhangs since step positions $x_n(y)$ are defined as single-valued functions of y . In the SOS model, there is no such restriction. Indeed, we can see from Fig. 6 that some crossing steps have bent so much that they have created *antisteps* at certain y positions, i.e., steps of opposite sign to the initial ones at particular fixed y positions. In our simulations, the temperature is still well below the roughening temperature of the flat surface, but it is not energetically forbidden to create new steps or overhangs, in contrast to the step models previously studied.

It is interesting to compare these results with the experiment by Latyshev *et al.*³⁵ They observed *antistep bunch formation* taking place after step bunch formation. The first stage of the antistep bunch formation occurs through the bending of the single height crossing steps between the step bunches, creating a region of bunched steps of the opposite sign. Indeed, we have directly observed step bunches created from this kind of step bending in our model with modified Arrhenius dynamics when we applied the external force in a direction *parallel* to the initial (and average) step edge direction. In Fig. 7, the initial (and average) step edge direction is in the vertical (y) direction. The bias is in the downwards ($-y$) direction. The dark regions are step bunches formed by steps bending in the opposite direction to those individual steps (black lines) on the terraces. As in the previous case, following the bias ($-y$) direction, there are regions of steps going up, and regions of antistep bunches going down. We derive elsewhere³⁹ from a 2D BCF-like model a new linear instability when the diffusion bias is parallel to the step edge direction that we believe underlies the patterns seen here.

These features obtained from simulations of the new SOS model are quite different from the ripple structure reported

by Dobbs and Krug¹¹ using Metropolis dynamics, where there are no distinct steps and facets after the surface develops large structures. Here the steps and terraces are easily discernible. Because of our more realistic treatment of the physics of surface diffusion and attachment at steps and the favorable comparison with experiment, we believe that *modified Arrhenius dynamics* provides a better description for current-induced step bunching on Si(111).

VIII. CONCLUSION

In summary, the evolution of the structure of Si(111) surfaces during electromigration at 900° C can be understood quantitatively using a one-dimensional step model, with parameter values and the mass transport mechanism determined from experiment. Specifically, the $t^{1/2}$ power-law growth rate for step bunch sizes is reproduced. We concentrate on the case where mass transport is limited by the rate of adatom attachment to a step edge, but the method can be easily generalized, as illustrated by our discussion of direct adatom hopping between different terraces.

The 1D step model has averaged over the individual movement of adatoms and atomic scale fluctuations of the steps, thus permitting simulations of the long time behavior using realistic parameter values. However, at higher temperatures, when 2D effects such as step bending can be seen, even the quasi-2D step models considered to date^{5,24} may not be sufficient. Moreover an exact BCF-like treatment of full 2D diffusion problem seems prohibitively difficult. To examine these issues, we carried out Monte Carlo simulations of a 2D solid-on-solid model, using physically motivated assumptions about the dynamics of surface diffusion and attachment at step edges. In particular we used *modified Arrhenius dynamics* with an extra barrier for attachment of adatoms at step edges and find good qualitative agreement with experiment. A new step bending instability is seen when there is a force acting on adatoms along the step edge direction that may be related to experiments by Latyshev *et al.*³⁵ In general we believe that this approach of combining information from experiment, microscopic simulations, and mesoscopic step models may prove useful in a number of different problems in surface science.

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APPENDIX

By requiring $j_n^+ = j_n = j_{n+1}^-$ and imposing periodic boundary conditions in Eqs. (18a) and (18b), we arrive at the following system of linear equations for the terrace chemical potentials for a system of N steps:

$$\begin{bmatrix} c_0 & c_1 & 0 & \cdots & c_1 \\ c_1 & c_0 & c_1 & \cdots & 0 \\ 0 & c_1 & c_0 & \cdots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ c_1 & 0 & 0 & \cdots & c_0 \end{bmatrix} \begin{bmatrix} \mu_1^t \\ \mu_2^t \\ \mu_3^t \\ \vdots \\ \mu_N^t \end{bmatrix} = \kappa \begin{bmatrix} \mu_1 + \mu_2 \\ \mu_2 + \mu_3 \\ \mu_3 + \mu_4 \\ \vdots \\ \mu_N + \mu_1 \end{bmatrix}, \quad (\text{A1})$$

where

$$c_0 = 2(\kappa + p) \quad c_1 = -p, \quad (\text{A2})$$

and $\mu_i^t = \mu^t[(x_i + x_{i+1})/2]$. μ_i^t can be solved for analytically since the matrix on the left-hand side of the equation is a circulant⁴⁰ matrix. The result can be expressed as

$$\mu_n^t = \sum_m K_m (\mu_{n+m} + \mu_{n+m+1}), \quad (\text{A3})$$

where in the limit $N \rightarrow \infty$

$$K_m = \frac{\kappa}{2(\kappa + p)} \frac{1}{\sqrt{1 - a^2}} \left(\frac{1 - \sqrt{1 - a^2}}{a} \right)^m, \quad (\text{A4})$$

and

$$a = \frac{p}{\kappa + p}. \quad (\text{A5})$$

K_m describes the correlation between the adatom chemical potential at a given terrace with the adatom chemical potential m steps away. K_m decays exponentially as m increases. It is convenient to define N_c as the number of steps over which K_m decreases by half, i.e.,

$$K_{N_c} = K_0/2. \quad (\text{A6})$$

When $p \gg \kappa$, we have

$$N_c \approx \ln(2) \sqrt{\frac{p}{2\kappa}}. \quad (\text{A7})$$

Since N_c is the range of correlation between the chemical potential values on different terraces, the mass transport is effectively nonlocal over a number of steps much smaller than N_c , and is local over a number of steps much larger than N_c .

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