Local Electromigration Model for Crystal Surfaces

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The dynamics of crystal surfaces in the presence of electromigration is analyzed. From a phase field model with a migration force which depends on the local geometry, a step model with additional contributions is derived in the kinetic boundary conditions. These contributions trigger various surface instabilities, such as step meandering, bunching, and pairing on vicinal surfaces. Experiments are discussed.

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In the presence of an electric current, mobile atoms experience a diffusion bias, which is called electromigration. Surface electromigration is known as a major source of rupture of microstructures and is also an interesting tool for spontaneous nanostructure formation. In the past 15 years, kinetic instabilities at the micrometer or nanometer scales, such as step bunching [1], meandering [2], and pairing [3] on stepped surfaces under electromigration, have been the focus of a large body of literature. In the usual model for step motion under electromigration [4], mobile atoms between steps experience a constant drift force, while kinetic boundary conditions at steps are not affected by migration. It was suggested in Refs. [5,6] that migration could lead to additional terms in step kinetic boundary conditions, which may have some consequences on the dynamics of monolayer islands or vicinal surfaces. Nevertheless, a microscopic explanation was lacking.

From the work of Rous and Bly [7], it is known that, in the vicinity of steps, the backscattering of carriers strongly alters the migration force. In the present Letter, it is shown that the boundary conditions used previously in the literature should be modified to account for the variations of the migration force in the step region. To do so, a step model is derived from a phase field model with a nonconstant migration force. The analysis of this step model reveals that the new contributions may produce all the instabilities known so far on vicinal surfaces. Finally, the relevance of these results for experimental observations on various surfaces is discussed.

Let us start with a microscopic description in terms of a phase field model [8]. The surface is described by two fields ϕ and c, which, respectively, represent the normalized height of the surface and the local concentration of mobile atoms. More sophisticated models, which may account for arbitrary kinetics at the steps, are not considered here for the sake of simplicity [8]. The dynamics of the phase field ϕ is such that ϕ relaxes to a step and terrace structure. Terraces are wide regions where ϕ is constant, and steps have a typical width W, as shown in Fig. 1. The motion of the steps is driven by the departure from equilibrium in the step region, measured by the concentration variations: $(c - c_{\rm eq})$, where $c_{\rm eq}$ is the equilibrium concentration. The dynamics of ϕ is a relaxation with time scale τ_ϕ:

$$\tau_\phi \partial_t \phi = -\partial_\phi f + W^2 \nabla^2 \phi + \lambda (c - c_{\rm eq}) \partial_\phi g. \quad (1)$$

The free energy density f is a periodic function of ϕ with minima for the values of ϕ corresponding to the terraces. The coupling function g is also a function of ϕ, with $\partial_\phi g \geq 0$ in the step regions, $\partial_\phi g = 0$ on terraces, and $g_- - g_+ = 1$, where ± indicates the lower or the upper side of the step, respectively. The constant λ controls the strength of the coupling between mobile atoms and steps.

A global external force is present, related to the electric current in the bulk of the crystal. This current induces a local surface migration force $\mathbf{M}$ on mobile atoms. The

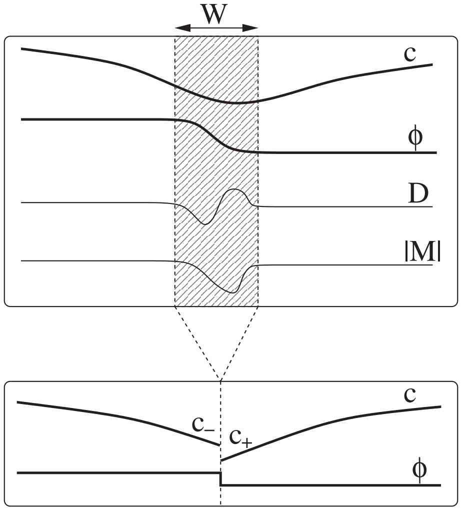

FIG. 1. Phase field model with a nonconstant migration force. The step region of width W is hatched. For the sake of simplicity, a 1D cross section is shown, but the model in the text is 2D. Notations: c is the mobile atom concentration, ϕ is the phase field, D is the local diffusion constant, and $|\mathbf{M}|$ is the local amplitude of the migration force. Using suitable asymptotics, one obtains a step model as shown in the lower part of the figure, where the concentration may be discontinuous at the step ($c_+ \neq c_-$), and ϕ is now a Heaviside function which indicates the step position.

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force $\mathbf{M}$ depends on the surface local geometry via the phase field ϕ. The resulting mass flux, oriented in the direction $\mathbf{n}_e = \mathbf{M}/|\mathbf{M}|$, reads:

$$j_e = Dc\frac{\mathbf{M}}{k_B T} = \frac{Dc}{\xi}\mathbf{n}_e, \tag{2}$$

where D is the local ϕ-dependent diffusion constant, and $\xi = k_B T/|\mathbf{M}|$ is a ϕ-dependent length scale which characterizes the amplitude of the force. Far from the steps, the diffusion constant and migration length are constant: $D \to D_0$ and $\xi \to \xi_0$. Local mass conservation reads:

$$\partial_t c = \nabla\left[D\left(\nabla c - \frac{c}{\xi}\mathbf{n}_e\right)\right] + F - \frac{c}{\tau} - \partial_t h, \tag{3}$$

where h is the solid concentration, i.e., the number of solid atoms per unit area above a plane of arbitrary height parallel to the terraces. The variations of h are analogous to that of ϕ, and the jump of h across a step is constant $h_- - h_+ = 1/\Omega$, where Ω is the atomic area. Following Refs. [8,9], we use a thermodynamically consistent formulation, with $g = h\Omega$.

Writing Eq. (3), we have discarded the tensorial coupling between the geometrical anisotropy of the step and the migration direction (i.e., the orientation of $\mathbf{n}_e$), which could change the local migration direction in the vicinity of a step. We shall rather focus on another effect which is, in my opinion, the most important one: the variation of the migration force amplitude in the step region. The phase field model presented here also neglects the dependence of the migration force on the adatom density. Indeed, this effect should occur at high densities only [10], while the adatom densities which are relevant for the experiments mentioned below are small.

We now derive a step model from the phase field model (1) and (3). When step kinetics is fast, one may perform the thin interface asymptotics reported in Refs. [8,9]. We shall assume that steps are slowly driven out of equilibrium, so that the step width W is much smaller than the smallest cutoff length ℓ_c related to the diffusion field on terraces, defining a small parameter $\epsilon \sim W/\ell_c$. The length ℓ_c may, for example, be the typical distance to other steps, the migration length ξ, or the desorption length $(D\tau)^{1/2}$. We also consider a finite coupling constant $\lambda \sim 1$ and a small departure from equilibrium at the steps $(c - c_{\mathrm{eq}}) \sim \epsilon$. The latter condition indicates that step kinetics is fast enough to keep the concentration in the step region close to c_{eq}. Finally, the relaxation of ϕ must be fast, so that the dynamics does not affect the step and terrace structure. We therefore need $\tau_\phi \sim \epsilon^2$.

Different expansions of c and ϕ are performed in the step region (i) and far from the steps (o). Defining an inner variable $\eta = z/\epsilon$, where z is a coordinate along the normal to the step, and the arclength s along the step, we have

$$c^{(i)}(\eta, s) = \sum_{n=0}^{\infty} \epsilon^n c_n^{(i)}(\eta, s); \quad c^{(o)}(\mathbf{r}) = \sum_{n=0}^{\infty} \epsilon^n c_n^{(o)}(\mathbf{r}), \tag{4}$$

where $\mathbf{r} = (x, y)$. A similar expansion is used for ϕ. Finally, the (i) and (o) expansions of c and ϕ must be matched at the border between the two regions.

To 0th order in the expansion, $c_0^{(i)} = c_0^{(o)} = c_{\mathrm{eq}}$. Moreover, ϕ_0 is an arbitrary immobile step and terrace structure (with steps far from each other).

The dynamics is obtained to first order. The concentration of atoms far from the steps obeys Eq. (3), with $D \to D_0$, and without the exchange term $\partial_t h$.

Global mass conservation at the step reads:

$$\frac{V_n}{D_0}(\Omega^{-1} + c_- - c_+) = \mathbf{n}\cdot\nabla c_+ - \mathbf{n}\cdot\nabla c_- + \frac{c_- - c_+}{\xi_s}, \tag{5}$$

with $1/\xi_s = (\mathbf{n}\cdot\mathbf{n}_e)/\xi_0$. Equation (5) is obtained to first order without the terms $(c_- - c_+)/\xi_s$ and $(c_- - c_+)V_n/D$, which are higher order contributions. Two additional equations are obtained to first order. They are interpreted as the step kinetic boundary conditions:

$$D_0\mathbf{n}\cdot\nabla c_\pm - \frac{D_0 c_\pm}{\xi_s} = \pm\nu_\pm\left(c_\pm - \tilde{c}_{\mathrm{eq}} - \beta\frac{V_n}{\Omega} \pm Q_\pm\frac{c_{\mathrm{eq}}}{\xi_s}\right), \tag{6}$$

where $\tilde{c}_{\mathrm{eq}} = c_{\mathrm{eq}}(1 + \Gamma\kappa)$. These expressions involve some coefficients, the definitions of which follow [11]:

$$\begin{aligned}
\Gamma c_{\mathrm{eq}} &= 2^{1/2}\frac{W}{\lambda}\int d\phi (f - f_{\mathrm{min}})^{1/2}, \\
\beta &= 2^{1/2}\frac{\tau_\phi}{W}\int d\phi (f - f_{\mathrm{min}})^{1/2} \\
&\quad - \int dz (g - g_-)(g_+ - g)D^{-1}, \\
\nu_\pm^{-1} &= \pm\int dz (g_\mp - g)(D^{-1} - D_0^{-1}), \\
Q_\pm &= \pm\int dz (g_\mp - g)(1 - \xi_0/\xi),
\end{aligned} \tag{7}$$

where f_{min} is the value of f at its minima. Integrals are taken through one step in the normal direction z. The constants $g_\pm$ are the values of g on both sides of the steps.

If we ignore the terms proportional to $Q_\pm$, Eq. (6) is equivalent to the general linear kinetic boundary condition of steps when step transparency and attachment-detachment on both sides of the steps are taken into account [8]. Using linearized nonequilibrium thermodynamics, Eq. (6) could have been stated directly, with the new terms proportional to $Q_\pm$ [12]. Hence, Eq. (6) is more general than the specific limit of the specific phase field model derived above. The relevance of our derivation from a phase field model comes from the explicit link between the terms of Eq. (6) and microscopic quantities. Indeed, from Eq. (7), Q_+ or Q_- account for the variations of $|\mathbf{M}|$ on the lower or the upper side of the step, respectively. Moreover, $Q_\pm > 0$ or $Q_\pm < 0$, respectively, account for a decrease or an increase of $|\mathbf{M}|$ in the step region.

Instead of analyzing the consequences of the full boundary conditions (6), we shall now focus on a simplified limit.

We define:

$$Q = Q_+ + Q_- = \int dz(1 - \xi_0/\xi). \tag{8}$$

In order to grasp some of the main consequences of the new contributions, we shall focus on a simplified limit where $Q_+ = Q_- = Q/2$. This may be obtained in the phase field model with an antisymmetric g and a symmetric variation of ξ. This assumption amounts to considering that the main effect of the force variation in the step region is an average increase or decrease of the amplitude of the force, respectively, leading to a negative or positive Q. We also focus on a specific limit for step kinetics. We define:

$$d_0 = D_0(\nu_+^{-1} + \nu_-^{-1}) = \int dz(D_0/D - 1). \tag{9}$$

Comparing the last term in the left-hand side with the last term in the right-hand side of Eq. (6), we infer that $Q_\pm$ will be relevant when it is of the same order of magnitude as $D_0/\nu_\pm$. We therefore take the limit where $\nu_\pm$ is large, which implies d_0 small. We also take D to be symmetric across the step so that $\nu_+ = \nu_-$.

With these assumptions—symmetric $1/\xi$ and D and fast kinetics—the boundary conditions (6) read:

$$c_\pm = \tilde{c}_{\text{eq}} \mp Q^* c_{\text{eq}}/2\xi_s + \beta V_n/\Omega, \tag{10}$$

where

$$Q^* = Q + d_0 = \int dz\left(\frac{D_0}{D} - \frac{\xi_0}{\xi}\right). \tag{11}$$

Equations (10) account for transparent steps when β is large and nontransparent steps with fast attachment-detachment kinetics when β is small. The expression (11) indicates in a remarkably compact formulation that a single quantity Q^* accounts at the same time for the variations of the migration force and for the variations of the diffusion constant in the step region. Therefore, in the present limit, one cannot distinguish between the relative contributions of diffusion and migration force variations from the analysis of step dynamics.

We shall now see how all known elementary instabilities of vicinal surfaces are obtained from the model with kinetic boundary conditions (10). Let us analyze the stability of an array of initially straight and parallel steps separated by the same distance ℓ. We assume that the direction of migration is orthogonal to the average step direction, defined as the x axis. We also consider the conserved regime where $F = 0$ and $1/\tau \to 0$. The mth step is perturbed by a small deviation

$$\zeta_m(x, t) = \zeta_{\omega\phi} \exp[i\omega t + im\phi + iqx], \tag{12}$$

where ϕ and q are, respectively, the phase shift from step to step and the wave vector along the step. An instability is indicated by a positive $\Re e[i\omega]$. Substituting Eq. (12) in the model equations, one finds the general expression of $i\omega$ as a function of ϕ and q.

In the case of in phase meandering ($\phi = 0$), one finds in the long wavelength and weak electromigration limit $[(q\ell)^2 \sim \ell/\xi_0 \ll 1]$:

$$i\omega \approx \Omega c_{\text{eq}} D\left[\frac{Q^*}{\xi_0} q^2 - \Gamma \ell q^4\right]. \tag{13}$$

Therefore, the train of steps is unstable when $Q^*/\xi > 0$, and the most unstable wavelength is $\lambda_m = 2\pi(2\xi_0\Gamma\ell/Q^*)^{1/2}$.

In order to analyze step bunching, we shall introduce a repulsion between steps. If this interaction is of elastic or entropic origin, its free energy per unit step length is $\mathcal{A}/\ell^2$ [13]. The local equilibrium concentration is then

$$\tilde{c}_{\text{eq}} = c_{\text{eq}}(1 + \Gamma\kappa + 2A(\ell_+^{-3} - \ell_-^{-3})), \tag{14}$$

where we have considered the interaction between neighboring steps only, and $A = \Omega\mathcal{A}/k_B T$. For the sake of simplicity, we analyze the stability of the mode $q = 0$, which accounts for the bunching of straight steps. In the limit of weak electromigration, the linear stability analysis leads to:

$$\frac{i\omega}{\Omega D c_{\text{eq}}} = \frac{(2Q^*/\xi_0\ell^2)[1 - \cos(\phi)] - (12A/\ell^5)[1 - \cos(\phi)]^2}{1 + 2[1 - \cos(\phi)]\Omega D\beta/\ell}. \tag{15}$$

Once again, an instability appears if $Q^*/\xi > 0$. The stability analysis thus indicates that a vicinal surface is simultaneously destabilized with respect to bunching and meandering for a downhill flux when migration or diffusion is weaker in the step region and for an uphill migration when migration or diffusion is enhanced in the vicinity of the steps.

Let us now discuss some of the experimental results of the literature. On Si(111), atoms drift along the current direction [14]. Three high temperatures regimes, denoted I, II, and III, were found [1,15]. In regimes I and III, bunching is observed for a downhill current during both growth and sublimation [15]. In regime II, bunching is observed during growth for a downhill current and during sublimation for an uphill current [15]. From the step model without the Q contribution, it was concluded that regimes I and III correspond to opaque steps (β, ν_+, and ν_- small), and regime II corresponds to fast step kinetics (ν_+ and ν_- large) [15,16].

In the case of opaque steps, the Q terms are probably a small contribution, unless an unexpected dramatic increase or change of sign of migration is observed in the step region. In the case of fast kinetics, the Q terms become important. From the study of step pairing [3], it was concluded that β is large, which means that steps are transparent. In Ref. [3], the occurrence of pairing was related to a small negative kinetic length $d_0 \approx -0.13$ Å. In the present analysis, this result generalizes to $Q^* = Q + d_0 \approx -0.13$ Å.

We therefore obtain an alternative explanation for the pairing based on $Q < 0$, i.e., on a stronger migration in the step regions, and $|Q| \gg |d_0|$. Quantitatively, we find $Q \approx -0.13$ Å. This may, for example, account for an increase of 1% of the migration force in a step region of width 13 Å.

Step meandering is also observed during sublimation with downhill migration in regime II [2]. If $Q < 0$, as suggested above, then Q rather favors the stabilization of the meander for a downhill flux. Meandering therefore requires an extended study, which would also account for the sublimation rate and which is beyond the scope of the present Letter [17].

Another system of interest is Si(100), which undergoes a dimer-row reconstruction, rotated $\pi/2$ from one layer to the next one. We here assume that dimer rows are perpendicular $\perp$ or parallel $\|$ to steps and that the current is perpendicular to the average step orientation. The difference of the diffusion constant ($D_\perp$, $D_\|$) perpendicularly to the step leads to pairing [4] and subsequent bunching of pairs [18] for both directions of the electric current. The crucial quantity is the migration mass flux $j \sim c_{\mathrm{eq}} D/\xi$. Therefore, the phenomena that are attributed to a variation of D can also be the result of a variation of ξ. More precisely, stronger migration along dimer rows reproduces pairing and bunching of pairs for both directions of the current, as observed in experiments [19]. Furthermore, following Ref. [20], one could consider pairs as effective steps with an effective value of Q, coming from the internal variations of D and ξ. This would help to analyze the pair bunching dynamics.

We also expect the effect presented above to be relevant for the case of metals. Indeed, from microscopic models on metallic surfaces [7], strong variations of the migration force in the vicinity of the steps were found. Rous and Bly [7] indicate a decrease up to ~50% in a region ~20 Å for Na. Using Eq. (8) with an approximate integration of the results of Ref. [7], we find $Q \approx 10$ Å. This contribution is 2 orders of magnitude larger than that found above for Si(111) and should therefore have drastic consequences on the dynamics of metal surfaces under electromigration. Experiments on Au surfaces [21] indicate that, in the presence of an electric current, the Ehrlich-Schwoebel (ES) effect [22] (a decrease of interlayer mass transport related to a low value of ν_-) disappears, and a significant increase of interlayer mass transport is observed. The disappearance of the ES effect is probably caused by the increase of the temperature due to the joule heating effect. Indeed, it was shown in Ref. [23] that the ES effect disappears at high temperature on Au. Therefore, kinetics is fast, and the tendency to bunching indicated in Ref. [21] could be caused by the nonconstant migration scenario presented above.

Electromigration also leads to step bunching for some other metal surfaces [24] and on GaN(0001) during epitaxial growth [25]. But quantitative microscopic information is lacking for a precise discussion of the nonconstant migration force scenario in these systems.

In conclusion, variations of the migration force in the vicinity of the steps may lead to step bunching, meandering, or pairing during electromigration. This mechanism defines a novel scenario for surface instabilities, which may compete or combine with the other destabilizing mechanisms analyzed in the literature. Finally, the relevance of these results for some semiconductor and metal surfaces was discussed.

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