A continuum theory is used to predict scaling laws for the morphological relaxation of crystal surfaces in two independent space dimensions. Our goal is to unify previously disconnected experimental observations of decaying surface profiles. The continuum description is derived from the motion of interacting atomic steps. For isotropic diffusion of adatoms across each terrace, induced adatom fluxes transverse and parallel to step edges obey different laws, yielding a tensor mobility for the continuum surface flux. The partial differential equation for the height profile expresses an interplay of step energetics and kinetics, and aspect ratio of surface topography that plausibly unifies observations of decaying bidirectional surface corrugations.

Novel small devices rely on the stability of nanoscale surface features. The lifetimes of nanostructures decaying via surface diffusion scale as a large power of their size and increase with decreasing temperature. Below roughening, crystal surfaces evolve via the motion of atomic steps bounding nanoscale terraces.\textsuperscript{1,2} Experiments with decaying surface features\textsuperscript{3–6} are useful for testing step models. Particularly informative are observations of bidirectional corrugations relaxing below roughening.\textsuperscript{3–6} In lithography-based experiments,\textsuperscript{3} where initial wavelengths in two directions differ significantly and ratios are closer to unity and profiles evidently depend on two geometries; also, we relate scaling predictions of the PDE to relaxation experiments. We find that observed decay laws with time can arise from competition of step kinetics and surface topography. This effect is due to coupling of flux components via terrace diffusion and is distinct from the influence of step-edge diffusion; see, e.g., Ref. 18. The similar effect of anisotropic terrace diffusion on step meandering is studied in Ref. 19. By contrast to Ref. 19, our model has scalar microscopic parameters.

First, we describe the step flow model.\textsuperscript{1} A top terrace is surrounded by non-self-intersecting and noncrossing steps numbered \( i = 1, 2, \ldots; i = 1 \) denotes the top step. The projection of steps on the basal plane is described by the vector \( \mathbf{r}(\eta, \sigma, t) \); \( \eta \) is time, \( \eta = \eta_i \) at the \( i \)th step, \( \eta < \eta < \eta_{i+1} \) on the \( i \)th terrace, and \( \sigma \) is the position along each step (see Fig. 1). The unit vectors normal and parallel to steps in the direction of increasing \( \eta \) and \( \sigma \) are \( \mathbf{e}_\eta \) and \( \mathbf{e}_\sigma \); \( \mathbf{e}_\eta \cdot \mathbf{e}_\sigma = 0 \). The metric coefficients (to be used below) are \( \xi_\eta = |\partial_\eta \mathbf{r}| \) and \( \xi_\sigma = |\partial_\sigma \mathbf{r}| \).

Mass conservation for atoms is described by

\[

u_\eta = \frac{\partial \mathbf{r}}{\partial t} \bigg|_{\eta=\eta_i} = \frac{\Omega}{a} [J_\eta^{\eta}(\eta_i, \sigma, t) - J_\eta^{\eta}(\eta_i, \sigma, t)].
\]

\( u_\eta \) is the (normal) velocity of the \( i \)th step; \( \Omega \) is the atomic volume; \( a \) is the step height; \( J_\eta^{\eta} \) is the adatom flux (adatoms/length/time) normal to steps; \( J_\eta^{\eta} = -D_s \nabla C_\eta \) is the (vector) flux on the \( i \)th terrace; \( D_s \) is the terrace diffusivity, a scalar; and \( C_\eta(\mathbf{r}, t) \) is the adatom density [adatoms/(length)] on the \( i \)th terrace. In the quasistatic approximation, \( C_\eta \) solves the diffusion equation \( \nabla^2 C_\eta = 0 \), where no material is deposited from above. The boundary conditions for atom
attainment-dettachment at the \( i \)th and \((i+1)\)th steps are\(^{10}\)
\[
\dot{J}_i^\alpha(\eta_i, \sigma') = [k_i(\eta_i, \sigma') - C^i(\eta_i, \sigma')] 
\]
(2)
In Eq. (2), the \( t \) dependence is omitted, \( \dot{=} i \) (upper sign) or \( \dot{=} +1 \) (lower sign), \( k \) is the attachment-detachment rate, and \( C^i \) is the \( i \)th-step equilibrium atom density. Note that Eq. (2) is similar to those of other growth problems, but here, there is no morphological instability.

Next, we close Eqs. (1) and (2) by relating \( C^i \) with the step positions. First, we introduce the \( i \)th-step chemical potential \( \mu(\eta_i, \sigma) \), the change in the step energy by adding or removing an atom at \( \eta_i, \sigma \) (Ref. 10): \( C^i = C_i e^{\mu_i/k_B T} - C_i[1 + \mu_i/k_B T] \), where \( |\mu_i| \ll k_B T \). \( C_i \) is the atom equilibrium density near a straight isolated step, and \( k_B T \) is Boltzmann’s energy.

Second, we provide a relation of \( \mu \) with the step positions. We use \( U(\eta, \sigma) \), the energy per length of the \( i \)th step; thus, the length \( \delta \eta_i = \xi_i \delta \sigma_i \) of the \( i \)th step has energy \( \delta W_i = U(\eta, \sigma) \delta \eta_i \). Addition or removal of atoms at \( \eta_i, \sigma \) changes \( \eta_i \) by \( \delta \eta \), assuming energy isotropy; so, the step moves along the local normal \( \langle \mathbf{e}_i \rangle \) by \( \delta \sigma = \xi_i \delta \eta \) and the step energy changes by \( \delta W_i = [\xi_i(\delta \eta_i)] \delta \eta_i \). By definition of \( \mu \),
\[
\mu_i = \Omega(\alpha) (\delta \eta_i) \xi_i^{-1} + \kappa U(\eta, \sigma) |\eta, \sigma|, \quad (3)
\]
where \( \kappa \) is the step curvature and \( U = \gamma + U_{\text{int}} \); \( \gamma \) is the step line tension, assumed a constant, and \( U_{\text{int}} \) accounts for interactions with other steps. For nearest-neighbor elastic-dipole or entropic repulsions, \( U_{\text{int}} \) is\(^{1,2,20}\)
\[
U_{\text{int}} = g \left[ \frac{\Phi(\eta_{i+1}, \sigma; \eta)}{(\eta_{i+1} - \eta)^2} + \frac{\Phi(\eta, \eta_{i-1}; \sigma)}{(\eta - \eta_{i-1})^2} \right], \quad (4)
\]
where \( g \) (energy/length) is positive and \( \Phi(\eta, \xi; \sigma) \) is geometry dependent, differentiable with \( \eta, \xi \) and satisfies \( \Phi(\eta_i, \eta_{i+1}) \delta \xi = \Phi(\eta_{i+1}, \eta_i) \delta \eta_{i+1} \)\(^{16}\). Supressing \( \eta_{i-1} \) and \( \eta_{i+1} \), Eqs. (3) and (4) yield \( \dot{\mu} = \dot{\mu}(\eta = \eta_i, \sigma) \).

Equations (1)–(4) describe step motion via adatom isotropic diffusion and atom attachment-detachment at steps. To enable predictions at length scales large compared to the terrace width \( \delta \eta_i \), we next derive a PDE for the continuum height profile \( h(\mathbf{r}, t) \). Thus, \( \delta \eta_i \) is much smaller than (i) the length over which the step density \( \frac{\partial}{\partial x} \), varies and (ii) the step radius of curvature, \( 1/\kappa \). We take \( \delta \eta_i = \eta_i + 1 \to 0 \) with fixed \( \frac{\delta \eta_i}{\delta \sigma_i} \). In this limit, \( \frac{\partial}{\delta \sigma_i} \to -\nabla h \), where \( \nabla h = (\partial h/\partial x, \partial h/\partial y, T) \), and \( T \to -\nabla h \).

Firstly, the familiar continuum mass conservation law for atoms comes from the step velocity law, Eq. (1). By using the continuous extension \( \mathbf{J}(\mathbf{r}) \) of \( J(\eta, \sigma) \), we have
\[
\frac{\partial h}{\partial \sigma_i} = -\frac{\Omega}{\xi_i \xi_\eta} [\partial_\sigma(\xi_\lambda J^\lambda) + \partial_\lambda(\xi_\lambda J^\sigma)] = -\Omega \nabla \cdot \mathbf{J}. \quad (5)
\]

Next, we apply Eq. (2) to relate \( \mathbf{J}(\mathbf{r}, t) \) to the continuum step chemical potential \( \mu(\mathbf{r}, t) = \mu(\eta, \sigma, t) \). The following procedure is more general than the analysis in Ref. 16. (i) We apply Eq. (2) with the upper sign for \( \sigma' = \sigma + \delta \sigma \) and with the lower sign for \( \sigma' = \sigma - \delta \sigma \). (ii) We expand the transverse current \( J^\sigma \), the density \( C_i \), and \( \mu \), each evaluated at \( (\eta_i, \sigma + \delta \sigma) \), at \( (\eta_i, \sigma) \) using \( \mathbf{J} = -\nabla C_i \). e.g., \( C_i(\eta_i, \sigma + \delta \sigma) \approx C_i(\eta_i, \sigma) + \delta \sigma \partial_\sigma C_i(\eta_i, \sigma) \), where \( Q_i(\eta_i, \sigma) \) and \( J^\sigma \) \( = \mathbf{J} \cdot \mathbf{e}_i \) is the longitudinal current. (iii) We subtract the \( \pm \) parts of Eq. (2) dropping terms that are negligible as \( \delta \eta_i \to 0 \). Thus, we find
\[
\left[ 1 + q \frac{\partial}{\partial \sigma_i} \right] J^\sigma + C_i D_i \frac{\partial \mu}{k_B T} \xi_\eta + \frac{\xi_\sigma}{\xi_\sigma} \left[ \frac{\sigma J^\sigma}{k_B T} + C_i D_i \frac{\partial \mu}{k_B T} \xi_\sigma \right] \frac{\partial \sigma}{\partial \sigma_i} = 0, \quad (6)
\]
where \( q = 2D_i / k_B T \). By setting \( \partial \sigma = 0 \) in Eq. (6), we obtain
\[
J^\sigma \to \mathbf{J}(\mathbf{r}, t) \cdot \mathbf{e}_\sigma = -\frac{D_i C_i}{k_B T} \frac{1}{1 + q \nabla |h|} \frac{\partial \phi_{\delta \eta_i}}{\xi_\sigma}, \quad (7a)
\]
where \( (a/\partial \phi) \) is q is fixed. Hence, Eq. (6) reduces to
\[
J^\sigma \to \mathbf{J}(\mathbf{r}, t) \cdot \mathbf{e}_\sigma = -\frac{D_i C_i}{k_B T} \frac{1}{1 + q \nabla |h|} \frac{\partial \phi_{\delta \eta_i}}{\xi_\sigma}. \quad (7b)
\]
By Eq. (7b), the longitudinal flux \( J^\sigma \) has the terrace diffusivity \( D_\sigma \), whereas the normal flux \( J^\sigma \), Eq. (7a), has the slope-dependent diffusivity \( D_\phi \), cf. Ref. 17. Note that \( D_\sigma = D_\phi \) for terrace diffusion limited (TDL) kinetics, \( |\nabla | h | \ll 1 \). The distinction between \( J^\sigma \) and \( J^\sigma \) results from coarse grainning in 2D, which combines atom attachment-detachment, terrace diffusion, and step topography. For ADL kinetics, \( |\nabla | h | \gg 1 \), \( J^\sigma \) depends on step variations of \( \mu \) since steps are sources and sinks of atoms by Eq. (2), whereas \( J^\sigma \) is sensitive to space variations of \( \mu \) along steps due to adatom diffusion between nonparallel steps. Equations (7a) and (7b) read \( \mathbf{J} = -\mathbf{C}_i \mathbf{M} \cdot \mathbf{\mu} \), where the mobility \( \mathbf{M} \) (length\(^2\)/energy/time) is a second-rank tensor. In the basal’s plane Cartesian system \((x, y)\), the matrix elements \( M_{ij} \) \((i, j = x, y)\) are
\[
M_{xx} = \frac{D_i}{k_B T} \frac{1}{|\nabla | h |} \left[ 1 + q \nabla |h| + \alpha^2 \right]. \quad (8)
\]

FIG. 1. Schematic of steps on the basal plane. Local coordinates relative to a top terrace are \((\eta, \sigma)\). The arrow shows longitudinal flux directed to a valley. Dots denote many steps.
We set

\[ M_{xy} = M_{yx} = -\frac{D_s}{k_BT} q|\nabla h| (\partial h)^2, \]

where \( \alpha = \frac{\partial h}{\partial h} \) is the (aspect) ratio of dominant (maximum-amplitude) wavelengths in \( x \) and \( y \); we take \( \lambda_x \leq \lambda_y \), and thus, \( \alpha \approx 1 \).

Next, we obtain a PDE for the height profile \( h(r,t) \). First, we derive a relation between \( \mu \) and \( \nabla h \) via Eqs. (3) and (4) by expanding in \( (\eta_i - \zeta) \) the function \( \Phi(\eta_i, \zeta; \sigma) \) of Eq. (4), where \( \zeta = \eta_1 + \eta_2 \). After some algebra, the limit \( \delta \eta_i \to 0 \) of \( \mu_i = \tilde{\mu} = \eta_1 - \eta_2 \) yields

\[ \mu = \tilde{\mu} |\nabla h| = \frac{\nabla}{|\nabla h|} \cdot \left( \frac{\nabla h}{|\nabla h|} \right), \tag{11} \]

where \( \kappa = -\nabla \cdot \frac{\nabla h}{|\nabla h|} \) is the step-edge curvature, \( g_1 = \frac{\alpha}{\sigma} \), and \( g_3 = \frac{\alpha}{\alpha^2} \Phi(\eta_1, \eta_2, \eta_3) \) have dimensions of energy per area. This \( \mu \) also results from the variational derivative of the surface energy \( E = \int dxdy \left[ g_1 |\nabla h| + (g_1/3)|\nabla h|^3 \right] \). By Eqs. (5), (7a), (7b), and (11),

\[ \partial_t h = B \nabla \cdot \left[ -\mu \left( \nabla \cdot \left( \frac{\nabla h}{|\nabla h|} \right) + g_3 \nabla \cdot \left( |\nabla h| |\nabla h| \right) \right] \right], \tag{12} \]

where \( \mu = \frac{k_BT}{\beta} \) and \( B = \frac{D_{c_{1,2}} \alpha^2}{k_BT} \) are [length]^4/time. By Eqs. (8)–(10) for \( \mu \), Eq. (12) describes an interplay of step energetics and kinetics, and aspect ratio \( \alpha \). This dependence on \( \alpha \) is absent in previous studies of morphological evolution below roughening. It is tempting to compare ingredients of Eq. (12) to similar treatments of steps, e.g., Ref. 19 for a step meander without deposition. The last term of Eq. (14) in Ref. 19 pertains to the flux along the step edge, which depends on the step-edge slope; in the small-slope limit, this term appears to agree with Eq. (7b). We emphasize that the (isotropic) physics for each terrace in our model is different from that of Ref. 19 where anisotropic terrace coexists with step-edge diffusion.

We now show that Eq. (12) reduces to known macroscopic laws for everywhere parallel steps. We set \( J^y = 0 \) by which the effective mobility becomes \( M = \frac{D_s}{k_BT} (1+q|\nabla h|)^{-1} \), a scalar. For straight steps (in 1D), \( \eta = x \), we additionally have \( \kappa = 0 \) and the PDE becomes

\[ \partial_t h = -B_3 \left[ (1+q|\partial h|)^{-1} \right] \partial_{xx}(|\partial h| |\partial h|), \]

where \( B_3 = \frac{D_{c,1,2} \alpha^2}{k_BT} \), which is constant, e.g., with Ref. 9. The reduced PDE is applied to smooth regions of periodic corrugations. For concentric circular, descending steps of radius \( r \) (in 2D), we have \( \kappa = 1/r \) and Eq. (12) becomes

\[ \partial_t h = B r^{-1} \left[ \left( 1+q|\partial h| \right)^{-1} \right] \partial_{rr}(|\partial h| |\partial h|), \]

where \( m = \frac{1}{r} |\partial h| \), which is suitable for smooth regions of axisymmetric mounds.

We next apply separation of variables to Eq. (12) for smooth regions of bi-periodic profiles, aiming to unify decay laws in relaxation experiments. Consistent with step simulations in 1D and kinetic Monte Carlo simulations in 2D, both for initial sinusoidal profiles, we set \( h(0,t) = A(t)H(r) \) and find \( A(t) \). This variable separation, which we call a “scaling ansatz,” is satisfied only approximately: additive terms in \( \mu \) and \( M \) scale differently with \( A \). In \( \mu \), Eq. (11), the step line tension \( (g_1 \text{ term}) \) scales with \( \alpha^0 \) and the step interaction \( (g_3 \text{ term}) \) scales with \( \alpha^2 \). In \( M \), Eqs. (8)–(10), the kinetic term \( \beta = (1+q|\nabla h|)^{-1} \) must be compared to the aspect ratio squared, \( \alpha^2 \); \( \langle |\nabla h| \rangle = r \) is a typical slope.

Here, we do not address the evaluation of \( H(r) \), which solves a nonlinear PDE. Because boundary conditions for \( H \) at facet edges require feedback from step simulations, a numerical scheme for \( H \) within continuum is not currently feasible. By Refs. 9 and 13, the scaling ansatz seems reasonable for long \( t \) and initial sinusoidal profiles.

We next focus on ADL kinetics, \( \beta \ll 1 \), distinguishing four cases. In the first case, (i) step interactions dominate, \( |g_3 \nabla \cdot (|\nabla h| \nabla h)| \gg |g_1| \), or \( \beta > \alpha^2 \) by dimensional analysis for sinusoidal profiles, where \( \nu = \frac{h_{01}}{h_{02}} \) and \( h_{01} \) is the peak-to-valley height variation, and (ii) \( \beta < \alpha^2 \) so that longitudinal fluxes are considerable. Thus, \( \mu \) scales with \( A^2 \), and the matrix elements of \( \Lambda \) are \( \Lambda_{xx} = \frac{(\alpha h)^2}{|\nabla h|^2} \), \( \Lambda_{yy} = \frac{(\alpha h)^2}{|\nabla h|^2} \), and \( \Lambda_{xy} = \frac{\alpha h (\alpha h)}{|\nabla h|^2} \), which scale with \( A^0 \) as in TDL kinetics. We find \( dA/dt = -cB_3 A^2 \), hence

\[ A(t) = A_0 (1 + cB_3 A_0 t)^{-1}, \quad A_0 := A(0). \tag{13} \]

The constant \( c \) depends on \( H \) and is thus influenced by facets. Equation (13) suggests an inverse linear decay with time if the (y) adatom flux in the direction of the longer wavelength \( \lambda_y \) is significant.

In the second case, (i) step interactions remain dominant and (ii) \( \beta > \alpha^2 \), so that transverse fluxes prevail. Thus, we obtain \( dA/dt = -C h^3 A, \) where

\[ A(t) = A_0 e^{-C R^3}, \tag{14} \]

where \( C \) is influenced by \( H \). The remaining cases for ADL kinetics follow similarly. The results are summarized in Table I. The square-root decay with time when line tension dominates and \( \beta > \alpha^2 \) is in agreement with Ref. 13.

Our predictions, based on Eq. (12) with ADL kinetics, can be extended to TDL kinetics. The mobility \( M \) then reduces to \( \frac{D_0}{k_BT} \). Thus, we obtain Eq. (13) or Eq. (14), regardless of \( \alpha \), for step-interaction- or line-tension-dominated \( \mu \).

Next, we compare our predictions with observations on Si(001) (Refs. 3 and 5) and Ag(110). In Si(001), with \( \ell = 2D_0/k \approx 1000 \text{ nm}^2 \) and terrace width \( \Delta w \approx 10 \text{ nm} \), \( q(|\nabla h|) = \frac{n}{3\ell^2} \approx 100 \) which suggests ADL kinetics. We find
Further, the assumed dipole interactions can dominate with $\beta = \frac{2\kappa}{\ell} \leq 0.01$, with the aspect ratio squared, $\alpha^2 \approx \left(\frac{\delta_{22}}{\hat{g}_{11}}\right)^2$, and the relative strength of step interactions, $\frac{\delta_{22}}{\hat{g}_{11}}$, with $(\hat{g}_{11})^2$. In Ref. 3, $\alpha \approx 10^{-3}$ and thus $\beta \gg \alpha^2$. Also, $\nu = 1/30$ and $\frac{\delta_{22}}{\hat{g}_{11}} > 1$.21 and thus $\frac{\delta_{22}}{\hat{g}_{11}} > (\hat{g}_{11})^2$. Equation (14) follows, in agreement with Ref. 3. In Ref. 5, $\alpha \approx 10^{-1}$, $\nu = 1/15$, and $\frac{\delta_{22}}{\hat{g}_{11}} = 100$.21 So, $\beta < \alpha^2$ and $\frac{\delta_{22}}{\hat{g}_{11}} > (\hat{g}_{11})^2$. Equation (13) follows, in agreement with Ref. 5.

Note that Si(001) may have properties not entirely consistent with Eq. (12), e.g., terrace diffusion is anisotropic and step interactions can deviate from (4).22 Although the terrace anisotropy modifies $M$, it does not change the scalings with time.22 Further, the assumed dipole interactions can dominate on Si(001) when terraces are sufficiently narrow, e.g., when double steps form.22

We now discuss observations on Ag(110) (Ref. 6) where step interactions are mainly entropic.24 By $\beta < 10^{-36}$ and $\alpha \approx 1/15$,25 we have $\beta < \alpha^2$. We estimate $\frac{\delta_{22}}{\hat{g}_{11}}$ by $\frac{\delta_{22}}{\hat{g}_{11}} = \frac{k_B T}{\hbar^2} \ln(\cosh(\frac{\delta_{22}}{2\hbar^2}))$ and $\frac{\delta_{22}}{\hat{g}_{11}} = \frac{\pi \kappa \delta_{22}}{2\hbar^2} \left[ \sinh\left(\frac{\delta_{22}}{2\hbar^2}\right) \right]^2$, where $\epsilon_0$ is the kink formation energy, 0.04 eV $\leq \epsilon_0 \leq 0.1$ eV,26 $\alpha = 1.4$ Å, $\alpha_0 = 4$ Å, and $T = 210$ K; thus, $\frac{\delta_{22}}{\hat{g}_{11}} \approx 1$. With $\nu = 2/25$,27 $\frac{\delta_{22}}{\hat{g}_{11}} = O(\frac{\nu}{\alpha^2})$; thus, our criterion for step energetics appears inaccurate for scaling. A possible reason is the terrace anisotropy of Ag(110), which may modify the numerical values of requisite parameters. Further study of the dynamics with reliable boundary conditions at facets is indicated.

Our work forms a basis for a general approach to morphological evolution below roughening. Extensions in 2D include the Ehrlich-Schwoebel (ES) barrier,28 long-range step interactions, step-edge diffusion, anisotropy of step stiffness, and material deposition. Inclusion of the ES barrier28 with rates $k_u$ and $k_d$ amounts effectively to $k = 2(1/k_u + 1/k_d)^{-1}$ in Eq. (12).16 Step-edge diffusion contributes to longitudinal fluxes but may not be important for Si(001).2

Connecting predictions of Eq. (12) to actual experimental situations has yet to be explored. Our scaling ansatz should be tested for realistic initial profiles. Despite couplings caused by nonlinearities,14 our scaling should be valid for a range of prevailing wavelengths.5,6,14 Other predictions of our approach include crossovers from exponential to inverse linear profile decay via aspect-ratio changes of the surface shape. Our work should stimulate further studies and relaxation experiments on crystal surfaces below roughening.

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