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Macroscopic view of crystal-step transparency

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We study macroscopic aspects of crystal surface relaxation in $2+1$ dimensions by accounting for near-equilibrium kinetics of transparent steps at the nanoscale. For slowly varying step geometries, we show that step permeability can simply *renormalize* a parameter in a known relation between the large-scale surface flux and the step chemical potential. This leads to a nonlinear fourth-order partial differential equation for the surface height profile.

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Modeling epitaxial phenomena on crystalline materials requires the reconciliation of a wide range of scales, from the atomistic to the continuum. Below the roughening transition temperature, various features such as mounds and quantum dots on crystal surfaces evolve via the motion of atomic steps bounding nanoscale terraces [1,2]. Understanding how microscale kinetics influences surface morphological dynamics is important for predicting novel material properties. Basic kinetic processes are the diffusion of adsorbed atoms (adatoms) on terraces and the attachment-detachment of atoms at steps [3]. In this Brief Report, we incorporate step permeability (transparency), i.e., the direct exchange of adatoms between terraces [4], into a fully continuum theory in two independent space dimensions.

Signatures of step permeability have been detected in experimental observations of terrace width distributions in (Al, Ga)Sb lateral superlattice growth [5], decay of mounds on Si(100) [6], and electromigration-induced step bunching on Si(111) [7,8]. Step transparency is invoked in [9–12] in connection to long-range step dynamics and instabilities; a short review can be found in [13]. Recently, effective permeability rates were derived from the steady state of a kinetic model of epitaxial growth [14].

In this Brief Report we address the question: What is a fully macroscopic description of crystal surface morphological relaxation with step permeability in two dimensions? By "fully macroscopic" we mean a description based on partial differential equations (PDEs) for large-scale variables such as the continuous surface height profile, in the spirit of, e.g., [15–21]. Here, we show that *if the macroscopic limit is attained* (under plausible assumptions described below), step transparency is woven into the continuum theory via the additive renormalization of the kinetic attachment-detachment parameter in the PDE for the height. The two-dimensional (2D) structure of the relaxation PDE (without permeability) remains intact. Our derivation invokes systematic expansions in the step height; and yields an effective *tensor* macroscopic diffusivity that forms a natural extension to two dimensions of the scalar effective diffusivity given in [11] via circuit theory for straight steps. Our result extends to 2D morphologies a previous observation [22] limited to axisymmetry and self-similar slopes.

The starting point is the Burton-Cabrera-Frank (BCF) [3] model for the motion of steps near thermodynamic equilibrium. The kinetic processes are (i) isotropic and homogeneous diffusion of adatoms on terraces; (ii) attachment and detachment of atoms at step edges with an asymmetry, the Ehrlich-Schwoebel (ES) effect [23,24], in the up- and down-step directions; and (iii) step transparency with a finite, *positive* kinetic rate.

First, we describe the step geometry and related coordinates following [21]. The steps are modeled by non-self-intersecting and noncrossing smooth curves in the reference (basal) plane; these are numbered $i=1,2,\ldots$ where $i=1$ denotes the top step. Each step is described by the position vector $\mathbf{r}(\eta,\sigma,t)$; t is time, $\eta=\eta_i$ at the ith step, $\eta_i<\eta<\eta_{i+1}$ on the ith terrace, and σ indicates the position along each step. (For axisymmetric profiles, i.e., circular steps, η corresponds to the polar distance and σ is the angle.) The step train is monotonic, with steps descending with increasing η. The unit vectors normal and parallel to steps in the direction of increasing η and σ are $\mathbf{e}_\eta$ and $\mathbf{e}_\sigma$, respectively; $\mathbf{e}_\eta\cdot\mathbf{e}_\sigma=0$ (see Fig. 1). The requisite metric coefficients are $\xi_\eta:=|\partial_\eta\mathbf{r}|$ and $\xi_\sigma:=|\partial_\sigma\mathbf{r}|$.

Next, we describe the step motion laws that express step transparency. For the ith terrace, this effect is described by the linear kinetic relations [4,13]

$$-J_{i,\perp}=k_{\mathrm{u}}(C_i-C_i^{\mathrm{eq}})+p(C_i-C_{i-1}),\qquad \eta=\eta_i,$$

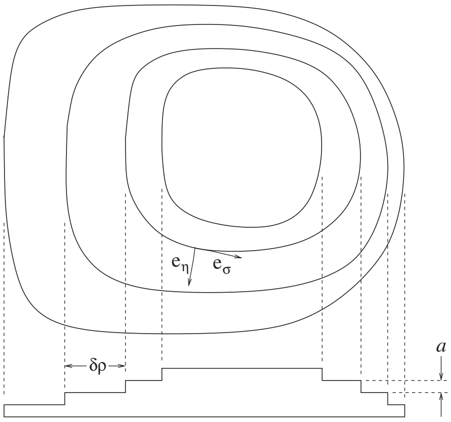

FIG. 1. Schematic of steps (top and side views). Local coordinates are (η,σ); $\eta=\eta_i$ at the ith edge.

$$J_{i,\perp} = k_{\mathrm{d}}(C_i - C_{i+1}^{\mathrm{eq}}) + p(C_i - C_{i+1}), \qquad \eta = \eta_{i+1}, \tag{1}$$

where $J_{i,\perp}(\eta,\sigma,t) \coloneqq \mathbf{e}_\eta \cdot \mathbf{J}_i$ is the transverse adatom flux component, $\mathbf{J}_i$ is the vector flux on the ith terrace, $C_i(\eta,\sigma,t)$ is the adatom concentration, k_{u} (k_{d}) is the kinetic rate for atom attachment-detachment at an up- (down-) step edge, p is the permeability rate, and $C_i^{\mathrm{eq}}(\sigma)$ is the equilibrium adatom density at the ith step edge [1]. The same σ is used in both Eqs. (1). Note that $\mathbf{J}_i = -D_{\mathrm{s}} \boldsymbol{\nabla} C_i$; D_{s} is the terrace diffusivity. This definition along with Eqs. (1) suffice for deriving a relation between the continuum limits of $\mathbf{J}_i$ and C_i^{eq}.

Emphasizing step permeability, we mention only briefly the remaining ingredients of step motion [1,3,21], since these do not involve p directly. The field C_i satisfies the diffusion equation, $D_s \nabla^2 C_i = \partial_t C_i \approx 0$ (in the quasisteady regime) for $\eta_i < \eta < \eta_{i+1}$; thus Eqs. (1) serve as boundary conditions. Furthermore, $C_i^{\mathrm{eq}} = C_{\mathrm{s}} \exp[\mu_i/(k_{\mathrm{B}}T)] \approx C_{\mathrm{s}}[1+\mu_i/(k_{\mathrm{B}}T)]$ where $\mu_i(\sigma)$ is the step chemical potential, C_{s} is the equilibrium density at a straight step edge, and $k_{\mathrm{B}}T$ is Boltzmann's energy [1]. This μ_i expresses step curvature and step-step interactions as detailed, e.g., in [21,22]. The step velocity u_i is dictated by mass conservation, viz.

$$u_i = (\Omega/a)(J_{i-1,\perp} - J_{i,\perp}), \qquad \eta = \eta_i, \tag{2}$$

where Ω is the atomic volume and a is the step height.

We now pass to the continuum limit of Eqs. (1), which yields a relation of the large-scale flux, $\mathbf{J}(\mathbf{r},t)$, and the macroscopic density $C^{\mathrm{eq}}(\mathbf{r},t)$, or the macroscopic chemical potential $\mu(\mathbf{r},t)$. We assume that, for all t of interest, the terrace width is much smaller than [21]: (i) the length λ over which the step density varies; and (ii) the step radius of curvature. Formally, we set $\delta\eta_i \coloneqq \eta_{i+1} - \eta_i \to 0$ by keeping fixed the step density $\mathcal{D}_i = \frac{a}{\delta\rho_i}$, $\delta\rho_i = (\xi_\eta)|_{\eta_i}\delta\eta_i$. Then, $\mathcal{D}_i \to |\boldsymbol{\nabla} h||_{\eta=\eta_i}$, where $h(\mathbf{r},t)$ is the continuum-scale height profile and $\boldsymbol{\nabla}$ operates in the (x,y) basal plane; $\boldsymbol{\nabla} = (\partial_x, \partial_y)$. In the same vein, $u_i \to [\partial_t h/|\boldsymbol{\nabla} h|]|_{\eta_i}$, the velocity of the level set for h [21]; $\boldsymbol{\nabla} h \neq 0$. In addition, we consider $D_{\mathrm{s}}/\nu = \mathcal{O}(a)$ [25] for $\nu = p, k_{\mathrm{u}}, k_{\mathrm{d}}$. This assumption (i) is consistent with the derivation of the continuum limit in previous works for $p=0$, e.g., [15–21]; and (ii) ensures that the (dimensionless) kinetic parameters $D_{\mathrm{s}}/(\nu a)$ are kept fixed, independent from the limit procedure. Imposing this scaling as a sufficient condition is further clarified below.

By inspection of Eqs. (1), we observe that permeability couples the solution C_i of the diffusion equation with the densities $C_{i\pm1}$ of adjacent terraces, in contrast to the $p=0$ case [21]. This discrete coupling indicates, but does not warrant, that the macroscopic limit of C_i is required as a variable coupled with h. A similar formalism is invoked in [19], where the authors imply that permeability is connected to adatoms living on step edges but apparently give no explicit formula for the effect of p.

Accordingly, we choose to use as primary variables the values $C_i(\eta_i) \eqqcolon \mathcal{C}(i)$ and $J_{i,\perp}(\eta_i) \eqqcolon \mathcal{J}(i)$ [26]. These are viewed as the appropriate interpolations (or discretizations) of the continuous functions $C(\mathbf{r})$ and $J_\perp(\mathbf{r})$; h, C, and $J_\perp$ vary over the length scale $\lambda = \mathcal{O}(\xi_\eta)$, where $\lambda \gg a$. The specific choice of the point $(\eta = \eta_i)$ of evaluation on the ith terrace here becomes irrelevant in the continuum limit (as $\eta_{i+1} \to \eta_i$) in the absence of material deposition from above [21]. In principle, we expect to derive a system of equations for C and $J_\perp$.

By the definition of $\mathbf{J}_i$ we have $C_i(\eta_{i+1}) - \mathcal{C}(i) = -D_{\mathrm{s}}^{-1}(\xi_\eta|_{\eta_i}\delta\eta_i)\mathcal{J}(i) + o(\delta\eta_i)$ [27], which is viewed as a *microscale* Taylor expansion. The validity of this expansion follows from the nature of solutions to the diffusion equation (for C_i) on each terrace. Thus Eqs. (1) approximately yield

$$\begin{aligned} -\mathcal{J}(i) = k_{\mathrm{u}}[\mathcal{C}(i) - C_i^{\mathrm{eq}}] + p[\mathcal{C}(i) - \mathcal{C}(i-1) \\ + D_{\mathrm{s}}^{-1}(\xi_\eta|_{\eta_{i-1}}\delta\eta_{i-1})\mathcal{J}(i-1)], \end{aligned} \tag{3a}$$

$$\begin{aligned} \mathcal{J}(i) = k_{\mathrm{d}}[\mathcal{C}(i) - D_{\mathrm{s}}^{-1}(\xi_\eta|_{\eta_i}\delta\eta_i)\mathcal{J}(i) - C_{i+1}^{\mathrm{eq}}] \\ + p[\mathcal{C}(i) - D_{\mathrm{s}}^{-1}(\xi_\eta|_{\eta_i}\delta\eta_i)\mathcal{J}(i) - \mathcal{C}(i+1)], \end{aligned} \tag{3b}$$

where the neglected terms do not contribute in the macroscopic limit.

Next, we invoke the (macroscale) variables C and $J_\perp$ via the expansions $\mathcal{C}(i) - \mathcal{C}(i-1) = \delta\eta_{i-1}\partial_\eta C|_{\eta_i} + o(\delta\eta_{i-1})$, $\mathcal{C}(i+1) - \mathcal{C}(i) = \delta\eta_i\partial_\eta C|_{\eta_i} + o(\delta\eta_i)$, and $\mathcal{J}(i) - \mathcal{J}(i-1) = o(1)$. The expansions involving the macroscopic density C are valid if this is a reasonably smooth function of the surface height. Replace now the terrace widths $\xi_\eta|_{\eta_{i-1}}\delta\eta_{i-1}$ and $\xi_\eta|_{\eta_i}\delta\eta_i$ by $a/|\boldsymbol{\nabla} h|$. Thus

$$-\left(1 + \frac{pa}{D_{\mathrm{s}}}\frac{1}{|\boldsymbol{\nabla} h|}\right)J_\perp = k_{\mathrm{u}}(C - C^{\mathrm{eq}}) + \frac{pa}{|\boldsymbol{\nabla} h|}\partial_\perp C,$$

$$\begin{aligned} \left(1 + \frac{(p+k_{\mathrm{d}})a}{D_{\mathrm{s}}}\frac{1}{|\boldsymbol{\nabla} h|}\right)J_\perp \\ = k_{\mathrm{d}}(C - C^{\mathrm{eq}}) - \frac{k_{\mathrm{d}}a}{|\boldsymbol{\nabla} h|}\partial_\perp C^{\mathrm{eq}} - \frac{pa}{|\boldsymbol{\nabla} h|}\partial_\perp C, \end{aligned} \tag{4}$$

for all $\mathbf{r}$, where $\partial_\perp = \xi_\eta^{-1}\partial_\eta$ and $C^{\mathrm{eq}}(\mathbf{r})$ is the continuum limit of C_i^{eq}. Solving for $J_\perp$ we obtain the formula

$$J_\perp = -D_{\mathrm{s}}\frac{(2p/k)\partial_\perp C + \partial_\perp C^{\mathrm{eq}}}{1 + 2p/k + q|\boldsymbol{\nabla} h}, \tag{5}$$

where $k \coloneqq 2(k_{\mathrm{u}}^{-1} + k_{\mathrm{d}}^{-1})^{-1}$, essentially the harmonic average of k_{u} and k_{d}, and $q \coloneqq 2D_{\mathrm{s}}/(ka)$ [21].

It remains to find a relation between C and C^{eq}. With recourse to Eqs. (5) we have

$$\begin{aligned} \left(1 + \frac{(k+2p)a}{2D_{\mathrm{s}}}\frac{1}{|\boldsymbol{\nabla} h|}\right)(C - C^{\mathrm{eq}}) \\ = -\frac{a}{|\boldsymbol{\nabla} h|}\frac{k_{\mathrm{d}}}{k_{\mathrm{u}}+k_{\mathrm{d}}}\left(1 + \frac{pa}{D_{\mathrm{s}}}\frac{1}{|\boldsymbol{\nabla} h|}\right)\partial_\perp C^{\mathrm{eq}} \\ + \frac{a}{|\boldsymbol{\nabla} h|^2}\frac{k_{\mathrm{d}}}{k_{\mathrm{u}}+k_{\mathrm{d}}}\frac{pa}{D_{\mathrm{s}}}\partial_\perp C = 0. \end{aligned} \tag{6}$$

The dominant balance of terms in this equation yields a formula for $J_\perp$: as $a/\lambda \to 0$, consistent with $\delta\eta_i \to 0$, the right-hand side vanishes since $pa/D_{\mathrm{s}} = \mathcal{O}(1)$. Hence we establish that $C = C^{\mathrm{eq}}$. Thus Eq. (6) entails

$$J_\perp = -\frac{D_\mathrm{s}\partial_\perp C^\mathrm{eq}}{1+q_\mathrm{eff}|\nabla h|} = -\frac{D_\mathrm{s}C_\mathrm{s}}{k_\mathrm{B}T}\frac{\partial_\perp \mu}{1+q_\mathrm{eff}|\nabla h|}, \tag{7}$$

where $q_\mathrm{eff} := 2D_\mathrm{s}/(k_\mathrm{eff}a)$ and $k_\mathrm{eff} := k+2p$. We stress the manifestly additive renormalization of k to $k+2p$.

The adatom flux component $J_\parallel$ parallel to step edges can be derived through differentiation of Eq. (3a) with respect to σ. The derivation here is not different from that for the $p=0$ case studied in [28]. We find

$$\begin{aligned} a\partial_\parallel \mathcal{J}(i) = {}& \frac{k_\mathrm{u}a}{D_\mathrm{s}}[\mathcal{J}(i) + D_\mathrm{s}C_i^\mathrm{eq}] \\ & + \frac{pa}{D_\mathrm{s}}\{\mathcal{J}(i) - \mathcal{J}(i-1) + D_\mathrm{s}^{-1}\partial_\parallel[(\delta\eta_{i-1}\xi_\eta)\mathcal{J}(i)]\}, \end{aligned} \tag{8}$$

where $\partial_\parallel = \xi_\sigma^{-1}\partial_\sigma$. Letting $a/\lambda \to 0$, or $\delta\eta_i \to 0$ with fixed $\xi_\eta\delta\eta_i/a$, yields the continuum-scale flux component

$$J_\parallel = -D_\mathrm{s}\partial_\parallel C^\mathrm{eq} = -\frac{D_\mathrm{s}C_\mathrm{s}}{k_\mathrm{B}T}\partial_\parallel \mu, \tag{9}$$

which is identified with the longitudinal flux of the $p=0$ case [21,28]. Note that this $J_\parallel$ is not affected explicitly by p since permeability enters the flux directly only through boundary conditions (1) for the transverse component $J_\perp$. At the risk of redundancy, we emphasize that, from a mathematical standpoint, setting $q, q_\mathrm{eff}, D_\mathrm{s}/(pa), D_\mathrm{s}/(k_{\mathrm{u(d)}}a) = \mathcal{O}(1)$ is a crucial *condition* for the derivation of the above limits.

By Eqs. (7) and (9), the vector-valued adatom flux is $\mathbf{J} = -C_\mathrm{s}\mathbf{M}(\nabla h)\cdot\nabla\mu$ where $\mathbf{M}$ is essentially the surface mobility (second-rank) tensor derived in [21,28]; the permeability rate p enters $\mathbf{M}$ through q_eff. A PDE for h is subsequently obtained via (i) the mass conservation statement $\partial_t h + \Omega\nabla\cdot\mathbf{J} = 0$, which stems from the step velocity law (2); and (ii) a formula for the continuum-scale step chemical potential, $\mu(\mathbf{r},t)$, in terms of the step curvature and step-step interactions. Details on μ can be found in [21]. The resulting PDE for the surface height h reads [21,28]

$$\partial_t h = B\nabla\cdot\left\{\boldsymbol{\Lambda}\cdot\nabla\left[\nabla\cdot\left(\frac{\nabla h}{|\nabla h|}\right) + \frac{g_3}{g_1}\nabla\cdot(|\nabla h|\nabla h)\right]\right\}, \tag{10}$$

where $\boldsymbol{\Lambda} = -\frac{k_\mathrm{B}T}{D_\mathrm{s}}\mathbf{M}$ and $B = \frac{D_\mathrm{s}C_\mathrm{s}g_1\Omega^2}{k_\mathrm{B}T}$; g_1 expresses the step line tension and g_3 is the step-step interaction strength for elastic dipole or entropic step interactions.

The additive renormalization of the kinetic coefficient k_eff in Eq. (7) for two dimensions is in agreement with an electric-circuit analog for one-dimensional (1D) steps; see, e.g., [1,10,11,15]. Our calculation extends systematically this simple physical analogy from one dimension to two dimensions, bringing about the distinction between transverse and longitudinal adatom fluxes and revealing the role played mathematically by certain kinetic conditions, e.g., the $\mathcal{O}(a^{-1})$ scaling of p/D_s. The effective diffusion constant $D_\mathrm{s}(1+q_\mathrm{eff}|\nabla h|)^{-1}$ emerging from Eq. (7) enters only the transverse flux component; cf. Eq. (9).

Our results have possible limitations. Negative values of p, which, for example, are predicted for electromigration-induced step pairing on Si(111) [8], might not be taken into account within the fully continuum approach. An indication of this inadequacy is given by the plausible, for $p \lesssim -k/2$, large negative values of q_eff, which generate nonphysical singular behavior at slopes $|\nabla h| = -1/q_\mathrm{eff}$. On the other hand, the limit of large p by which $C_i = C_{i+1}$ at $\eta = \eta_i$ would amount to $q_\mathrm{eff}|\nabla h| \to 0$, i.e., diffusion-limited kinetics [22]. This case should lie within the continuum approach.

In conclusion, we derived a macroscopic, anisotropic equation for the surface flux that incorporates step permeability in two dimensions. This description *emerges* mathematically from the limit $a/\lambda \to 0$ (a: step height; λ: macroscopic size) with *fixed* step density and kinetic parameters such as pa/D_s. The main result is that step transparency simply modifies an effective kinetic attachment-detachment rate, k_eff in the q_eff of Eq. (7) for the transverse adatom flux. The step geometry is assumed to be sufficiently slowly varying to allow for the continuum limit; and kinetic lengths defined in this theory are taken to be comparable to the length of a few terraces. Despite the latter formal restriction, which ensures independence of the kinetic parameters from the limit procedure, the results hold for a wide range of values of these parameters. For example, attachment-detachment limited kinetics by which $q_\mathrm{eff}|\nabla h| \gg 1$ is included. The renormalization of k_eff derived here appears to be meaningful for $p \geqslant 0$. The comparison of continuum predictions with discrete step simulations for finite, even negative, p deserves future attention.

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