HOMOGENIZATION OF COMPOSITE VICINAL SURFACES: EVOLUTION LAWS IN 1+1 DIMENSIONS

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Abstract. We apply classical homogenization to derive macroscopic relaxation laws for crystal surfaces with *distinct inhomogeneities* at the microscale. The proposed method relies on a multiscale expansion in one spatial coordinate. This approach transcends the coarse graining applied previously via Taylor expansions. Our work offers an extension of the static homogenization formulated in [Margetis, *Phys. Rev. E*, 79 (2009), 052601] in order to account for surface evolution. The starting point is the Burton-Cabrera-Frank (BCF) model for the motion of line defects (steps) separating nanoscale terraces. We enrich this model with sequences of distinct material parameters, i.e., disparate diffusivities of adsorbed atoms (adatoms) across terraces, kinetic sticking rates at step edges, and step energies for elastic-dipole interactions. Multiscale expansions for the adatom concentration and flux are used, with a slow diffusive time scale consistent with the quasi-steady regime for terrace diffusion. This procedure is complemented by a weak formulation for the step chemical potential in terms of a step free energy. The evolution laws incorporate appropriate averages of the microscale parameters.

Key words. Burton-Cabrera-Frank (BCF) model, line defect, homogenization, multiscale expansion, composite, epitaxial relaxation, step chemical potential, variational principle

AMS subject classifications. 76M50, 74A50, 35Q99

1. Introduction.

Crystal surface structures are critical ingredients of thin film epitaxy as well as surface chemistry and catalysis. Aspects of surface evolution remain an area of active interest [8, 14, 23]. A crucial issue is to elucidate how the microscale dynamics of constituent atomic defects influence the surface morphological evolution at large scales. This concern broadly motivates the present article.

Vicinal crystal surfaces are characterized by nanoscale flat regions (terraces) oriented in the high-symmetry direction and separated by line defects (steps) which are typically one atomic layer high. The steps considered here are monotonic (of the same 'sign'). Their number is fixed by the miscut angle set in laboratory experiments [14].

A standard approach to deriving macroscopic limits of stepped surfaces essentially relies on Taylor expansions for the step positions and step density; see, e.g., [5, 21, 22, 28, 29, 32, 34, 42]. This coarse graining is appropriate when material parameters remain unchanged across terraces. However, this approach is in principle inadequate if the surface has strong inhomogeneities, namely, distinct diffusivities and kinetic rates across terraces at the nanoscale. We call such a stepped surface 'composite'.

In this article, we address the question: what is the macroscale description of surface relaxation consistent with the microstructure inhomogeneities of a composite stepped surface? To provide an answer, we invoke singular perturbations, i.e., classical homogenization and multiscale expansions, in one spatial coordinate. This approach is deemed more general than the previous coarse graining. Our main results comprise evolution laws that contain microscale averages of material parameters.

It is tempting to claim that homogenization is not needed since the requisite averaging may stem from an electric circuit analog for the stepped surface [28, 31]. In this view, for instance, sequences of terrace diffusivities correspond to in-series electric conductances per unit length; hence, the average diffusivity would be the

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appropriate, effective circuit parameter. We believe that this view is incomplete for at least two reasons. First, it is static, leaving out the issue of time scales and evolution. Second, in this picture effective circuit parameters are determined by averaging at the (microscale) level of a few steps and terraces. This kind of averaging is conceptually different from the averaging required by homogenization in the macroscopic limit. One can in fact argue that the electric-circuit analog is a byproduct of, rather than a substitute for, homogenization.

The present work forms an extension of a recent, brief report on Fick's law for surface diffusion [19]. Here, more details on the derivation of Fick's law are provided, the roles of time scales and the mesoscale are quantified and discussed extensively, and the continuum laws comprise more variables, e.g., the step chemical potential.

Our analysis leaves open questions. The main focus is the *consistency* of macroscopic laws with a microscale model of a composite surface. Accordingly, the continuum limit is *assumed* to exist. Our derivation of Fick's law relies on formal arguments in terms of the classical solution for a microscale diffusion equation. In contrast, our treatment of the step chemical potential is based on a weak formulation [21]. It is expected that a weak formulation (not based on classical, closed-form solutions) can be applied for all evolution laws. This aspect is not fully developed here. We restrict attention to one spatial coordinate; the theory of a composite stepped surface in 2+1 dimensions is not addressed. We believe that the extension of continuum laws to 2+1 dimensions would bear no surprising results, yet it would require a homogenization technology different from the one applied here. It is hoped that our work will serve as an invitation to further investigations.

We assume that the reader is familiar with the basic concepts of epitaxial relaxation. For reviews on related topics, see, e.g., [8,14,16,23].

1.1. Physical motivation. There are at least two categories of applications that physically motivate our study. First, semiconductor surfaces may naturally exhibit structural phases that depend on the temperature and crystal misorientation angle. This 'surface reconstruction' amounts to material parameters that can vary appreciably across adjacent terraces and has received considerable attention. In particular, the Si(001) system[1] manifests a reconstruction in which dimer rows (chains of bonded atoms) alternate from perpendicular to parallel to step edges across terraces [4,17,25,36,40].

Another category of phenomena involves surface compounds created by small amounts of solutes added on a crystal. Such additions can cause dramatic morphological changes, affecting the crystal shape and stability [6,11,15,33,37,39,41]. This observation implies that artificial, composite surfaces may have interesting, unconventional properties, and therefore need to be explored systematically.

A feature common in both of the above cases is the existence of microstructure inhomogeneities. However, it is reasonable to expect that the surface appears homogeneous at a large enough length scale.

1.2. Averaging and mesoscale. Our homogenization approach is adopted from the basic theory of composites, e.g., [24,27]. The central theme is to identify separate scales from the physical setting and governing equations for steps, and then average out microscopic details in order to pass to the full continuum limit. The averaging procedure is intimately connected to the structure of the governing laws at the microscale. By requiring that classical solutions, e.g., the mass concentration

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[1] The index (001) here indicates the surface orientation, i.e., the respective normal vector.

and flux, of step flow remain bounded in the limit of a large number of steps, one can obtain solvability (non-growth) conditions giving rise to an effective macroscopic description.

In this framework, the notion of the *mesoscale* arises naturally. Its use facilitates the interpretation of averages [24]. Intuitively, one may think of the mesoscale as any surface region small enough to capture the underlying inhomogeneities yet sufficiently large to allow for reliable averaging. The variation of material parameters and variables (e.g., step velocity) across steps and terraces is evident at the mesoscale; but the corresponding averages of interest vary slowly across such mesoscale regions so that the full continuum limit makes sense.

The above idea permeates the study of diffusion in layered media [12]. The homogenization scheme in [12] is static from the outset; and includes the layer boundaries through the values of a fast, continuous spatial variable. The mass (adatom) concentration and flux are assumed to be continuous across these boundaries [12]. In contrast, our setting encompasses relaxation dynamics. First, the step edges (terrace boundaries) are moving; hence, the step velocity law plays a pivotal role. Second, densities and fluxes are in principle discontinuous across step edges because of the assumed attachment-detachment kinetics at steps. Third, the microscale Fick's law for diffusion is complemented with a step chemical potential, which expresses thermodynamically the change in the free energy for each step.

1.3. Kinetic processes. It is of interest to spell out the physical mechanisms underlying the model. We focus on surface relaxation, in the absence of growth. The steps move as a result of mass conservation under two main kinetic effects [14]: (i) the diffusion of adsorbed atoms (adatoms) on terraces, often simplified via the quasi-steady approximation whereby adatom diffusion equilibrates faster than steps move; and (ii) the atom attachment-detachment at step edges. A variable entering this description is the (discrete) step chemical potential, which is a thermodynamic force equal to the variation of the step free energy with respect to the step positions. In our study, this energy accounts for entropic and nearest-neighbor elastic-dipole step-step interactions. Elements (i) and (ii) permeate the celebrated Burton-Cabrera-Frank (BCF) model [3].

By comparison to real material systems, this setting appears incomplete. For example, terrace diffusion anisotropies are not included. The dipole character of step-step interactions here does not fully describe long-range interactions that may be present, e.g., on the reconstructed Si(001). Diffusion of atoms along step edges, evaporation/condensation, desorption and external material deposition are left out.

In particular, a study of surface reconstructions that accounts for, e.g., the geometry of dimer rows with alternating orientations on adjacent terraces of Si(100) [1,17], would require an in-depth analysis of anisotropic effects in 2+1 dimensions. Furthermore, the reconstruction generates internal stresses which may induce monopole-dipole step-step interactions [38]. This effect is of course not captured by the elastic-dipole model for steps used here [18].

In the spirit of the BCF theory [3], we assume that step motion occurs near thermodynamic equilibrium. This assumption is believed to be adequate for surface relaxation at macroscopic time scales. Material deposition from above and far-from-equilibrium kinetics are not touched upon here. Our methodology should still be applicable within the BCF framework for sufficiently small external deposition flux.

We expect that the present one-dimensional (1D) setting is, in a certain sense, minimal for surface reconstructions. Specifically, our formulation singles out issues intimately connected to averaging. In this vein, we circumvent complications which may arise in a two-dimensional (2D) geometry or richer kinetics.

1.4. Macroscopic limit. We pass to the full continuum in the limit of vanishing step height, a, with fixed step density (surface slope). The ensuing variables of interest are treated as $\mathcal{O}(1)$, a-independent quantities. As in previous treatments of continuum limits for step flow, e.g., [21], the main macroscopic variables are the surface height, the adatom flux, and the continuum-scale step chemical potential.

Our analysis does not avoid certain simplifying hypotheses (and thus bears respective limitations) traditionally present in the study of continuum limits [21]. Step trains are monotone and facets are absent. Microscale effects near facets are known to pose challenging, open problems [20]. We assume that (appropriate) microscale averages of interest vary sufficiently slowly. The full continuum limit is assumed to exist rather than proved to exist. The latter task would require a rigorous study of (e.g., derivation of a priori estimates for) solutions to the discrete step flow.

We employ primarily formal (yet streamline) arguments of classical homogenization [2] similarly to studies of diffusion in layered media [12]. Our homogenization approach is accompanied by a weak formulation for the continuum-scale step chemical potential of the composite stepped surface. On the other hand, mathematical niceties such as the convergence of multiscale expansions [30], although a prerequisite for complete analytical understanding, lie beyond our present scope.

1.5. Article organization and notation. The remainder of this article is organized as follows. Section 2 offers an overview of the model, our assumptions and the homogenization formalism: we introduce the physical setting and give the equations of step motion (section 2.1); introduce aspects of the relevant scales (section 2.2); and describe germane notions of homogenization (section 2.3). Section 3 contains a summary of the main results (especially for readers who wish to skip technical details of derivations). In section 4, we give details of the requisite multiscale expansions, order by order in perturbation, for 1+1 dimensions; and thereby derive the desired evolution laws. In section 5, we briefly discuss the extension of the homogenization approach to the radial setting. In section 6, we summarize our findings, discuss their possible implications, and mention pending issues.

Throughout this article, the terms 'full continuum limit', 'macroscale' and 'macroscopic limit' are used interchangeably. The time dependence is often (but not always) suppressed for notational economy. The symbol $Q^l(x,y)$, where l is an integer, denotes the coefficient of ϵ^l in the ϵ-perturbation expansion for Q, whereas $Q(x,y)^l$ is the usual lth power of Q; such a distinction (of perturbation order from power) should be self-explanatory. By $f=\mathcal{O}(g)$ we imply that $C_1 \leq |f(z)/g(z)| \leq C_2$ for some positive constants C_1 and C_2 as the parameter or variable z approaches an extreme value. Writing $f=o(g)$ amounts to $|f(z)/g(z)| \downarrow 0$.

2. Model and scale separation. Our goal with this section is to clarify the scale separation, and related concepts and notation needed in homogenization.

First, we describe briefly the physical setting of steps and terraces in 1+1 dimensions. Second, we discuss the underlying scales. The smallest scale is the step height, a, and the largest scale is the size of the crystal sample, λ. We define a mesoscale as an intermediate region of size d, $a \ll d \ll \lambda$, which helps formulate and interpret the averaging procedure. Third, we review the basic elements of classical homogenization.

2.1. Microscale model. The geometry consists of N steps descending in the positive x direction; see Figure 2.1. We assume that all steps have the same height, a,

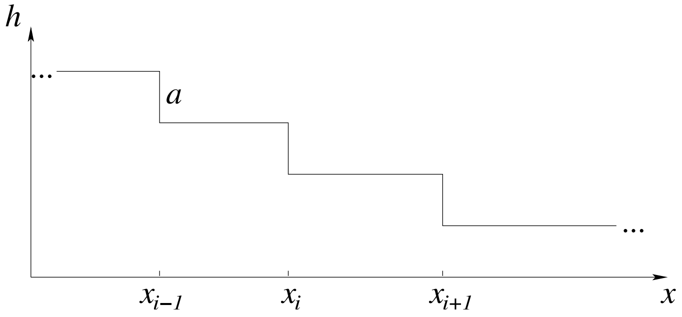

FIG. 2.1. *Schematic (cross section) of steps with atomic height a and positions $x_i(t)$ in one spatial coordinate (x). The ith terrace, $x_{i-1} < x < x_i$, has diffusivity D_i; and the bounding steps have attachment-detachment kinetic rates $k_{\mathrm{u},i-1}$ (up-step at x_{i-1}) and $k_{\mathrm{d},i}$ (down-step at x_i).*

an atomic length. The step positions are $x_i(t)$ where t denotes time; $i = 0, \ldots, N-1$ and $N \gg 1$. Let $x_i(0) > x_{i-1}(0)$. The step ordering is presumed fixed for $t > 0$ because of the dipolar repulsive step interactions [see (2.6)].[2] We conveniently assume that x_i lie on a torus (under periodic boundary conditions). To ensure that the surface slope is kept fixed, we set $w_i(t) := x_i(t) - x_{i-1}(t) = \mathcal{O}(a)$ for the ith-terrace width, w_i. By $x_{N-1}(0) - x_0(0) = \mathcal{O}(\lambda)$ for $N \gg 1$, we (initially) we have the relation

$$\sum_{i=1}^{N-1} w_i(0) = \mathcal{O}(Na) \quad \text{as} \quad N \to \infty. \tag{2.1}$$

The total length is $\lambda = Na = \mathcal{O}(1)$. Define $\epsilon := a/\lambda$ and set $\lambda = 1$ for convenience.

Consider the ith terrace, $\mathcal{T}_{it} = \{x \mid x_{i-1}(t) < x < x_i(t)\}$, and let $U_t := \cup_{i=1}^{N} \mathcal{T}_{it}$, $|U_t| = \mathcal{O}(\lambda)$. The adatom concentration, $\rho_i(x,t) \equiv \rho_i^\epsilon(x,t)$, is defined via[3]

$$\partial_x[D_i(x)\partial_x\rho_i] = \partial_t\rho_i \qquad \text{for } x \in \mathcal{T}_{it} \ , \tag{2.2}$$

where $t \in (0, T]$ and $T = T^\epsilon$ is large enough to account for macroscopic observations (section 2.3). The diffusivities $D_i(x)$ are positive and satisfy

$$0 < D_{\mathrm{m}} \leq D_i(x) \leq D_{\mathrm{M}} \qquad \text{for } \ i = 0, 1, \ldots, N-1 \ , \tag{2.3}$$

regardless of N; D_{m} and D_{M} are constants (and independent of N).

Let $\varrho^\epsilon(\cdot, t)$ represent the extension of the adatom concentration on $\mathcal{U} \supseteq \overline{U}_t$ ($\overline{U}_t$: closure of U_t) for all $t \geq 0$; e.g., $\mathcal{U} = [0,1]$. We must have $\varrho \equiv \rho_i$ for $x \in \mathcal{T}_{it}$ and all $t \geq 0$. Assume that $\varrho^\epsilon(\cdot, t)$ is C^2 (twice continuously differentiable in x) on U_t and bounded on $\mathcal{U}$. At each $x = x_i$ this ϱ^ϵ satisfies the kinetic conditions [3]

$$\mathcal{J}^\epsilon(x_i^-, t) - v_i\varrho^\epsilon(x_i^-, t) = k_{\mathrm{d},i}[\varrho^\epsilon(x_i^-, t) - \rho_i^{\mathrm{eq}}] \ , \tag{2.4a}$$

$$-\mathcal{J}^\epsilon(x_i^+, t) + v_i\varrho^\epsilon(x_i^+, t) = k_{\mathrm{u},i}[\varrho^\epsilon(x_i^+, t) - \rho_i^{\mathrm{eq}}] \ , \qquad v_i = \mathrm{d}x_i/\mathrm{dt} \ . \tag{2.4b}$$

In the above, $Q(x_i^\pm)$ is the restriction of $Q(x)$ (where $Q = \mathcal{J}$, ϱ) at $x = x_i$ from left $(-)$ or right $(+)$. The variable $\mathcal{J}^\epsilon(x,t)$ is the terrace adatom flux defined by Fick's law

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[2] A proof that the step ordering is preserved by the flow is feasible but not pursued here.

[3] Note that we do not apply the quasi-steady approximation at this stage. Accordingly, (2.2) gives rise to convective terms in the overall flux of adatoms impinging on a step edge; cf. (2.4).

$\mathcal{J}^\epsilon(x,t)=-D_i(x)\partial_x\varrho^\epsilon(x,t)$ in $\mathcal{T}_{it}$; this is supplemented with the convective term $v_i\varrho^\epsilon$ at the step edge, where v_i is the step velocity. Note that $\mathcal{J}^\epsilon(\cdot,t)$ is C^1 (continuously differentiable) on U_t and bounded on $\mathcal{U}$. In (2.4), the parameters $k_{\ell,i}$ are positive and bounded kinetic attachment-detachment (sticking) rates for an up- ($\ell=\text{u}$) and a down- ($\ell=\text{d}$) step edge, accounting for the Ehrlich-Schwoebel barrier [7,35]. The discrete variable ρ_i^{eq} represents the equilibrium concentration at the ith step edge.

Next, we describe ρ_i^{eq}, which incorporates step energies, in terms of $\{x_i\}$ [14]:

$$\rho_i^{\text{eq}}=\rho_* e^{\mu_i/\vartheta}\sim\rho_*(1+\mu_i/\vartheta)\ , \qquad |\mu_i|\ll\vartheta\ , \tag{2.5}$$

where μ_i is the step chemical potential (a thermodynamic force), ϑ is the Boltzmann energy (i.e, the absolute temperature in units of energy), and ρ_* is a given positive constant.[4] The quantity μ_i expresses step-step repulsions. If steps interact entropically and as elastic dipoles, we have [14,18,26]

$$\mu_i=\frac{\delta E_N[\vec{x}]}{\delta x_i}=\epsilon^{-1}\left[\breve{g}_{i+1}\left(\frac{\epsilon}{w_{i+1}}\right)^3-\breve{g}_i\left(\frac{\epsilon}{w_i}\right)^3\right]\ , \qquad \breve{g}_i=\mathcal{O}(1)>0\ , \tag{2.6}$$

where $\vec{x}:=(x_0,\ldots,x_{N-1})$. The quantity $E_N[\vec{x}]$ is the total step free energy,

$$E_N[\vec{x}]=\frac{1}{2}\sum_{i=1}^{N}\breve{g}_i\left(\frac{\epsilon}{x_i-x_{i-1}}\right)^2\ . \tag{2.7}$$

Alternatively, μ_i can be defined implicitly by the formula [21]

$$\sum_{i=0}^{N-1}v_i\mu_i=\dot{E}_N\ ; \qquad \dot{E}:=\mathrm{d}E/\mathrm{d}t\ , \tag{2.8}$$

where $v_i=\dot{x}_i$ is the ith step velocity. For reasonably arbitrary v_i, this relation is viewed as a weak formulation for μ_i (in the discrete setting), where $\{v_i\}$ is a test sequence (say, discretization of a smooth test function).

The last ingredient is the step velocity law, or mass conservation for adatoms:

$$\dot{x}_i=v_i=-(\Omega/\epsilon)[\mathcal{J}^{\epsilon,+}-\mathcal{J}^{\epsilon,-}-v_i\cdot(\varrho^{\epsilon,+}-\varrho^{\epsilon,-})]\big|_{x=x_i}\ , \tag{2.9}$$

where Ω is the atomic area and $Q^\pm:=Q(x^\pm)$; we henceforth set $\Omega=\epsilon^2$. Equation (2.9) concludes the description of the BCF-type model.

2.2. Mesoscale. The material parameters introduced at the microscale are $\{D_i\}$, $\{k_{\text{d},i},k_{\text{u},i}\}$, and $\{\breve{g}_i\}$. In this section, we define an intermediate scale, which facilitates the interpretation of averages emerging from homogenization (section 4) and is consistent with the existence of the macroscopic limit. The averages of interest are left unspecified in this section.

The main physical idea is that material parameters can vary appreciably within numerous certain regions consisting of consecutive terraces. However, (appropriately defined) material averages over each such region vary slowly across many such regions of the macroscopic sample. The notion of the mesoscale helps express this idea. This statement is mathematically vague at the moment but points to a plausible, and appealing, situation: Despite the presence of inhomogeneities at the microscale, the surface should appear homogeneous at a large enough scale.

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[4] In principle, we could have considered this parameter as varying with i, i.e., use the sequence $\{\rho_{*,i}\}$. However, since these are time-independent parameters, define $\tilde{\rho}_i:=\rho_i/\rho_{*,i}$ and $\tilde{\rho}_i^{\text{eq}}:=\rho_i^{\text{eq}}/\rho_{*,i}$. Because of linearity, the form of step equations (with $\tilde{\rho}$'s) remains intact. Drop the tildes and set $\rho_{*,i}=\rho_*=\mathcal{O}(1)=\text{const.}$ without loss of generality.

2.2.1. Some definitions. Consider M sequences of (consecutive) steps in the step train, where $M \gg 1$, $M < \mathcal{O}(N)$. Let the jth subsequence contain n_j steps labeled by $i \in I_j := \{i_*(j), i_*+1, \ldots, i_\circ(j)\}$ where $i_*(j) := \sum_{k=1}^{j-1} n_k$, $i_\circ(j) := \sum_{k=1}^{j} n_k - 1$ with $n_j \gg 1$ and $n_j = o(N)$ for $j = 1, \ldots, M$; and $i_*(1) := 0$, $i_\circ(M) = N-1$. Each n_j is kept fixed, but the total length of corresponding terraces is allowed to vary by $o(n_j\epsilon)$ during evolution.

So far, we have not specified how the above sequences of steps are chosen. For this purpose, we resort to material parameter averages (to be determined in section 4). For fixed ϵ, let the desired average for every $\{\zeta_i^\epsilon\}_{i\in I_j}$ be denoted $\bar{\zeta}_j^\epsilon$. Aiming at describing changes of these averages across different I_j's, we define notions of slow variation.

Definition 2.1. (Discrete version of difference quotient.) *The* difference quotient *of size $p \in \mathbb{Z}\setminus\{0\}$ for the sequence $\{\bar{\zeta}_j^\epsilon\}_{j=1}^M$ is*

$$\delta^p \bar{\zeta}_j^\epsilon := \frac{\bar{\zeta}_{j+p}^\epsilon - \bar{\zeta}_j^\epsilon}{p}\ . \tag{2.10}$$

Define $\delta^0\bar{\zeta}_j^\epsilon := 0$ for definiteness.

Definition 2.2. (Discrete version of slow variation.) *The sequence $\{\bar{\zeta}_j^\epsilon\}_{j=1}^M$ is* slowly varying *if, for fixed and sufficiently small ϵ, there exist large integers $j_*(M) = o(M)$ and $j_\circ(M)$ with $M - j_\circ = o(M)$ such that, for every $j_* \le j \le j_\circ$,*

$$|p\delta^p\bar{\zeta}_j^\epsilon| = o(\bar{\zeta}_j^\epsilon) \quad \textit{for all } -p_* \le p \le p_\circ\ ;\ M \gg 1\ (0 < j - p_*\ ,\ j + p_\circ < M)\ , \tag{2.11}$$

where $p_*(j), p_\circ(j) = o(M)$ and $n_jp_*, n_jp_\circ = o(N)$; $j_*, j_\circ, p_*, p_\circ \to \infty$ as $\epsilon \downarrow 0$.

The integers j_*, $j_\circ$, p_* and $p_\circ$ in principle depend on ϵ; p_* and $p_\circ$ signify how much one must shift j so that appreciable changes of the average $\bar{\zeta}_j^\epsilon$ tend to occur. Note that, for any large (yet finite) M, these p_* and $p_\circ$ are small compared to M.

Remark 2.3. *We henceforth assume that the sequence $\{I_j\}_{j=1}^M$, where $|I_j| = n_j = o(N) \gg 1$, is such that $\{\bar{\zeta}_j^\epsilon\}$ is slowly varying.*

Definitions 2.1 and 2.2 allude to transferring averages to a continuum setting. Consider ϵ as small but finite; and replace the index j by the (eventually continuous) height variable $h = h_i$ for fixed $i(j) \in I_j$. The use of the coordinate $x = x_i$ is also appropriate if the step density is positive and $\mathcal{O}(1)$. However, the variable h is a natural analog of the step number, i, since the step height is constant [22]. Accordingly, we introduce a $\bar{\zeta}^\epsilon(h)$ such that $\bar{\zeta}^\epsilon(h_{i(j)}) = \bar{\zeta}_j^\epsilon$.

Definition 2.4. (Continuous version of difference quotient.) *The* difference quotient *of size $\nu \in \mathbb{R}\setminus\{0\}$ for a continuous version $\bar{\zeta}^\epsilon(h)$ of $\{\bar{\zeta}_j^\epsilon\}_{j=1}^M$ is*

$$\tilde{\delta}^\nu\bar{\zeta}^\epsilon(h) := \frac{\bar{\zeta}^\epsilon(h+\nu) - \bar{\zeta}^\epsilon(h)}{\nu}\ . \tag{2.12}$$

By direct analogy with Definition 2.2, we propose the following notion.

Definition 2.5. (Continuous version of slow variation.) *The continuous version $\bar{\zeta}^\epsilon(h)$ is* slowly varying *if, for any $\nu = \nu(\epsilon) = \mathcal{O}(p\,n_j\epsilon) = o(1)$ and $p = o(M)$,*

$$|\nu\tilde{\delta}^\nu\bar{\zeta}^\epsilon(h)| = o(\bar{\zeta}^\epsilon(h)) \qquad \text{as } \epsilon \downarrow 0\ . \tag{2.13}$$

Equation (2.11) or (2.13) implies $|\nu(\epsilon)\partial_h\bar{\zeta}^\epsilon(h)| \ll |\bar{\zeta}^\epsilon(h)|$; $\nu(\epsilon)$ expresses the height change across a few mesoscale regions. By assuming that the slow variation persists as $\epsilon \downarrow 0$ (or, $N \to \infty$), we pass to the full continuum limit, $\bar{\zeta}^\epsilon \to \bar{\zeta}^0 =: \zeta^0$. In the following, we use the x coordinate (as an independent variable) in the place of h. By abusing notation, we write $\zeta^0(x)$ in place of $\zeta^0(h(x))$

REMARK 2.6. *The notions of this section can be extended to variables of the step system as well. In other words, the slow variation is assumed to hold for thermodynamic, kinetic and geometric variables, e.g., the step chemical potential, surface free energy per unit length, adatom flux, step velocity, and discrete slopes* ϵ/w_i.

We now define the concept of a mesoscale region in light of the above discussion.

DEFINITION 2.7. (Mesoscale.) *Consider the set* $\{I_j\}$ *for a given monotonic step train, where* $|I_j| = n_j \gg 1$, $n_j = o(N)$, $1 \le j \le M$ *and* $M, N \to \infty$. *Assume* $n_j/n_k = \mathcal{O}(1)$ *for any* j *and* k $(j \neq k)$, *and* $n := \max_j\{n_j\}$. *Any one of the* M *regions* $\cup_{i\in I_j}\mathcal{T}_{(i+1)t}$ *is called* the mesoscale. *Accordingly, a* mesoscale length *is any length* L *of the order of* $d := n\epsilon$ *with* $\epsilon \ll L = \mathcal{O}(d) \ll 1 = \lambda$.

2.2.2. Induced average.

At this point, it is advisable to discuss briefly the full continuum limit of averages.

DEFINITION 2.8. (Induced average). *Consider the given function* $f : \mathbb{R} \to \mathbb{R}\setminus\{0\}$ *and averages* $\{\bar{\zeta}_j^\epsilon\}_{j=1}^M$. *The* mesoscale average $\bar{\xi}_j^\epsilon$ of $\{\xi_i^\epsilon\}$ induced by $\{\zeta_i^\epsilon\}$ via f *is*[5]

$$f(\bar{\zeta}_j^\epsilon)\,\bar{\xi}_j^\epsilon := n_j^{-1}\sum_{i\in I_j} f(\zeta_i^\epsilon)\xi_i^\epsilon\ ; \qquad j = 1,\dots,M\ .$$

For our purposes, f can be thought of as Lipschitz continuous; see Appendix A.

It is of interest to point out an implication of Definition 2.8 in the (full continuum) limit $\epsilon \downarrow 0$. In a weak sense, by pairing in a sum $\{f(\zeta_i^\epsilon)\}$ and $\{\xi_i^\epsilon\}$, we have

$$\epsilon\sum_{i=0}^{N-1}\phi_i f(\zeta_i^\epsilon)\,\xi_i^\epsilon = \sum_{j=1}^{M}(n_j\epsilon)\phi(x_{i(j)})f(\bar{\zeta}_j^\epsilon)\bar{\xi}_j^\epsilon \xrightarrow[\epsilon\downarrow 0]{} \int_{\mathcal{U}}\phi(x)f(\zeta^0(x))\xi^0(x)\,\mathrm{d}h(x)\ . \tag{2.14}$$

The set $\mathcal{U}$ is a fixed interval, e.g., $[0,1]$, and will henceforth be omitted in integration.

2.3. Elements of homogenization.

Next, we review basics of homogenization theory needed for our purposes, setting the technical framework for section 4.

First, we clarify the choice of Eulerian spatial variables. Define the fast variable

$$y := (x - \check{x})/\epsilon\ , \tag{2.15}$$

for some reference point $\check{x}$ (to be specified later); thus, make the replacement

$$\partial_x \Rightarrow \partial_x + \epsilon^{-1}\partial_y\ ,$$

treating x and y as independent, and henceforth consider x as the slow spatial variable.

In view of diffusion equation (2.2), it is tempting to define fast and slow time variables as well. However, we wish to restrict attention to macroscopic times, consistent with the quasi-steady approximation [14, 21].

DEFINITION 2.9. (Quasi-steady regime.) *For our purposes, the* quasi-steady regime *is characterized by times* t *such that*

$$\tau = \epsilon^2\, t = \mathcal{O}(1)\ . \tag{2.16}$$

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[5] We use a bar on top for both the starting and the induced averages. These averages should be distinguished by the context through the different symbols for their variables (ζ and ξ).

In section 4, we show that this regime suffices to describe evolution laws consistent with previous continuum limits.[6] Accordingly, we replace

$$\partial_t \Rightarrow \partial_t + \epsilon^2\, \partial_\tau \ ,$$

treating t and τ as independent. In the same vein, consider $T = T^\epsilon = \mathcal{O}(\epsilon^{-2})$ so that τ lies in a fixed interval. Eventually, as $\epsilon \downarrow 0$, we claim that the dependent variables of interest are stationary (i.e., settle to a steady state) in t.

A comment on the assumptions underlying the limit $\epsilon \downarrow 0$ is in order.

REMARK 2.10. *We take* $n_j\epsilon \downarrow 0$ *and* $n_j \to \infty$ *for* $j = 1, 2 \ldots$ *Consistent with other continuum treatments, e.g.,* [21]*, the following hypotheses are made.*

(i) The step density at the mesoscale is fixed, i.e.,

$$\frac{n_j\epsilon}{x_{i_\circ} - x_{i_*}} = \mathcal{O}(1) \qquad \text{as } \ n_j\epsilon \downarrow 0,\ \ n_j \to \infty \ ; \tag{2.17}$$

see section 2.2 *for definitions of* $i_*(j)$ *and* $i_\circ(j)$*. The left-hand side of* (2.17) *approaches the positive surface slope,* $m(x) := \partial_x h$*.*

(ii) The height, h*, chemical potential,* μ_i*, and flux,* $\mathcal{J}$*, are* $\mathcal{O}(1)$ *as* $\epsilon \downarrow 0$*.*

(iii) The material parameters obey

$$k_{\ell,i}\epsilon,\ D_i,\ \breve{g}_i,\ \rho_*,\ \vartheta = \mathcal{O}(1) \qquad \text{for all } i \quad (\ell = \mathrm{u}, \mathrm{d}) \qquad \text{as } \ \epsilon \downarrow 0\ .$$

In particular, steps move in the regime of 'mixed kinetics' [9]*, where*

$$\frac{D_i}{k_{\ell,i}\epsilon} = \mathcal{O}(1)\ . \tag{2.18}$$

Note that the step-step interaction strength $\breve{g}_i$ *is assumed to be independent of* ϵ*. This hypothesis implies that by* (2.7) *the total step free energy, an extensive thermodynamic quantity, scales as* $E_N = \mathcal{O}(\epsilon^{-1}) = \mathcal{O}(N)$*, which is consistent with* $\mu_i = \partial_{x_i} E_N = \mathcal{O}(1)$ *(see section* 4.3*).*

The next element to be discussed is the multiscale expansion. With regard to the concentration $\varrho^\epsilon(x,t)$ of section 2.1, we write[7]

$$\varrho^\epsilon(x,t) = \varrho^0(x,y,\tau,t) + \epsilon\, \varrho^1(x,y,\tau,t) + \epsilon^2\, \varrho^2(x,y,\tau,t) + o(\epsilon^2) \qquad x \in \mathcal{U}\ , \tag{2.19}$$

where the superscript l in ϱ^l ($l = 0, 1, \ldots$) denotes perturbation order and $\varrho^l = \mathcal{O}(1)$ as $\epsilon \downarrow 0$. We treat (2.19) as a global perturbation expansion, valid for all x of interest, subject to conditions (2.4) (which should also be expanded in ϵ). Because of these kinetic conditions, ϱ^l in principle depends on the mesoscale numbers n_j.

To determine ϱ^l, it is necessary to apply dominant balance to (2.2) and enforce conditions (2.4). By (2.19), the multiscale expansion for the adatom flux $\mathcal{J}^\epsilon$ reads

$$\begin{aligned} \mathcal{J}^\epsilon &= -D_i(\epsilon^{-1}\partial_y + \partial_x)\varrho^\epsilon \\ &= -D_i\big[\epsilon^{-1}\partial_y\varrho^0 + (\partial_y\varrho^1 + \partial_x\varrho^0) + \epsilon(\partial_y\varrho^2 + \partial_x\varrho^1)\big] + o(\epsilon) \\ &=: \epsilon^{-1}\mathcal{J}^{(-1)}(x,y,\tau,t) + \mathcal{J}^0(x,y,\tau,t) + \epsilon\mathcal{J}^1(x,y,\tau,t) + o(\epsilon) \qquad x \in \mathcal{U}\ . \end{aligned} \tag{2.20}$$

Expansion (2.20) must be interpreted in the appropriate weak sense [24]. We now state our main requirement.

---

[6] Recall the full-continuum conservation law for adatoms, $\partial_t h + \Omega\partial_x\mathcal{J}^0 = 0$ where h and $\mathcal{J}^0$ are the large-scale height and flux. By $\Omega = \epsilon^2$ and $\partial_x\mathcal{J}^0 = \mathcal{O}(1)$, we have $\partial_\tau h = \mathcal{O}(1)$ for $\tau = \epsilon^2 t$.

[7] The rigorous study of convergence of (2.19) lies beyond our present purposes.

Remark 2.11. (Solvability condition.) *We impose the condition that the coefficients* ϱ^l *in* (2.19) *be bounded in* x, y *and do not grow as* $n_j \to \infty$ *(*M, $N \to \infty$*). Likewise, the coefficients* $\mathcal{J}^l$ *in expansion* (2.20) *for the flux should not grow with* n_j*.*

Naturally, the large-scale limit of step motion corresponds to the leading-order term of (2.19). We give the following (obvious) definition for the sake of completeness.

Definition 2.12. (Full continuum limit.) *The* macroscopic limit *of the adatom concentration emerges from the leading-order term,* ϱ^0*, of expansion* (2.19)*.*

It is expected that ϱ^0 is independent of the fast variables, y and t (see section 4).

At this point, it is necessary to comment on the step velocity.

Remark 2.13. *In view of motion law* (2.9) *and* Definition 2.9*, we set* $\mathrm{d}x_i/\mathrm{d}\tau = \mathcal{O}(1)$ *and thereby infer that*

$$v_i(t) = \epsilon^2\,(\mathrm{d}x_i/\mathrm{d}\tau) = \mathcal{O}(\epsilon^2)\ . \tag{2.21}$$

This scaling with ϵ *is consistent with the level set motion* $\partial_\tau h - u^0 \partial_x h = 0$ *for crystal surfaces with the same parameters in all terraces, where* $u^0(x,\tau) = \mathcal{O}(1)$ *is the continuum limit of an appropriate average of* $\epsilon^{-2} v_i$ *and* $m = |\partial_x h| = \mathcal{O}(1)$*.*

The above considerations will furnish Fick's law of diffusion [19] and the mass conservation statement for composite vicinal surfaces (sections 4.1 and 4.2). These laws involve the height profile, adatom flux and chemical potential.

Thus far, we have not discussed how the large-scale surface free energy, $\mathcal{E}$, and the chemical potential, μ, can be expressed in terms of the surface slope. It suffices to mention that $\mathcal{E}$ and μ will be treated with recourse to summation formulas (2.7) and (2.8), along with Definition 2.8. The details are deferred to section 4.3.

3. Main results.

In this section, we summarize the main results, which include: (i) Fick's law, which relates the large scale adatom flux, $\mathcal{J}^0(x)$, and step chemical potential $\mu(x)$, on the basis of the adatom attachment-detachment conditions (2.4); (ii) a conservation law for the macroscopic height profile, $h(x)$, from the step velocity law (2.9); and (iii) a variational formula for the chemical potential, $\mu(x)$.

3.1. Adatom flux (section 4.1).

The macroscopic adatom flux is

$$\mathcal{J}^0(x,\tau) = -\mathcal{D}_{\mathrm{e}}(x; m(x,\tau))\,\partial_x \varrho^{\mathrm{eq},0}(x,\tau)\ , \tag{3.1}$$

where $\varrho^{\mathrm{eq},0} = \varrho^0$ is the continuum-scale version of ρ_i^{eq} and $\mathcal{D}_{\mathrm{e}}$ is the effective parameter

$$\mathcal{D}_{\mathrm{e}}(x; m(x)) = \frac{D^0(x)}{1 + q^0(x)\,m(x)}\ . \tag{3.2}$$

By the notation of section 2.2, D^0 represents the average

$$D^0(x) = \lim_{\substack{n_j \epsilon \downarrow 0 \\ n_j \to \infty}} \overline{D}_j\ ,\quad \overline{D}_j = \overline{D}_j^{\epsilon} = \left[\left(\frac{1}{y_{i_\circ} - y_{i_*}} \sum_{i \in I_j} \frac{y_{i+1} - y_i}{D_{i+1}(x)}\right)\right]^{-1}\ . \tag{3.3}$$

Similarly, q^0 incorporates an average for the attachment-detachment rates:

$$q^0(x) = \frac{2D^0(x)}{k^0(x)\,\epsilon}\ ,\ k^0(x) = \lim_{\substack{n_j \epsilon \to 0 \\ n_j \to \infty}} \bar{k}_j\ ,\ \bar{k}_j(x) = 2\left\{\left[n_j^{-1} \sum_{i \in I_j}\left(\frac{1}{k_{\mathrm{u},i}} + \frac{1}{k_{\mathrm{d},i}}\right)\right]\right\}^{-1}, \tag{3.4}$$

where the factor of 2 is included so that $k^0 = k$ if $k_{\mathrm{u},i} \equiv k_{\mathrm{d},i} \equiv k$, consistent with [21]. In (3.2), the variable m amounts to the (average) step density (cf. Remark 2.10)

$$m(x,\tau) = \lim_{\substack{n_j\epsilon\downarrow 0\\ n_j\to\infty}} \overline{m}_j(\tau), \quad \overline{m}_j = \left(n_j^{-1}\sum_{i\in I_j} m_i(\tau)^{-1}\right)^{-1} = \left(\frac{n_j\epsilon}{x_{i_\circ} - x_{i_*}}\right) , \tag{3.5}$$

where $m_i(\tau) = \epsilon/w_i(\tau)$ and w_i is the ith terrace width. By section 2.2, we use $x = x_i$ for some $i = i(j) \in I_j$, where x becomes continuous and thus the choice of i is immaterial as $n_j\epsilon \downarrow 0$ (with $n_j \to \infty$). Thus, all sequences in j are viewed as functions of x.

Our derivation of (3.1) is intended to clarify by dominant balance why the concentration entering Fick's law needs to be identified with $\varrho^{\mathrm{eq},0}$. This point was simply stated (but not shown) in [19]. Equations (3.1)–(3.5) are supplemented with

$$\varrho^{\mathrm{eq},0}(x) = \rho_*[1 + \mu(x)/\vartheta] . \tag{3.6}$$

By (3.3), $\overline{D}_j(x)$ is the harmonic average of the terrace diffusivities, D_i, with weights proportional to the terrace widths, w_i. At the risk of redundancy, we repeat that this result is strongly reminiscent of an electric-circuit analog introduced by Nozières [28] (see also, e.g., [14, 31], for related applications). In this electric-circuit picture, the adatom concentration gradient is viewed as a distributed voltage. Hence, the diffusivities play the role of in-series electric conductances per unit length of the surface, and the effective diffusivity in each I_j is the appropriate overall conductance. In this view, the effective circuit parameters are evaluated at the level of a few terraces, without recourse to the generic behavior (e.g., non-growth) of large-scale solutions. This circuit view is distinctly different from our homogenization approach. Furthermore, electric-circuit considerations appear to be static from the outset. In contrast, homogenization incorporates the appropriate handling of time scales.

3.2. Adatom conservation (section 4.2).

We will establish the relation

$$m(x,\tau)u^0(x,\tau) + \partial_x\mathcal{J}^0(x,\tau) = 0 , \tag{3.7}$$

where $u^0(x,\tau)$ denotes the continuum-scale step velocity (see Remark 2.13),

$$u^0(x,\tau) := \lim_{n_j\to\infty} \bar{u}_j, \quad \bar{u}_j = n_j^{-1}\sum_{i\in I_j} u_i , \quad u_i := \epsilon^{-2}v_i^\epsilon(t) = \mathrm{d}x_i^\epsilon/\mathrm{d}\tau , \tag{3.8}$$

and $m(x,\tau)$ is given by (3.5). We will show that

$$\partial_\tau h(x,\tau) = m(x,\tau)\, u^0(x,\tau) \qquad (\tau = \epsilon^2\, t) , \tag{3.9}$$

which describes the motion of the effective level set for the graph $h(x,\tau)$. Thus, (3.7) becomes the familiar conservation law $\partial_t h + \Omega\partial_x\mathcal{J}^0 = 0$ ($\Omega = \epsilon^2$ is the atomic area).

3.3. Chemical potential (section 4.3).

The fully continuum version of the step chemical potential is provided by the (L^2-) variational derivative

$$\mu = \left(\frac{\delta\mathcal{E}[h]}{\delta h}\right)_{L^2} , \tag{3.10}$$

which is implied (weakly) by the statement

$$\dot{\mathcal{E}}(\tau) = \int \mu(x,\tau)\,\partial_\tau h\,\mathrm{d}x . \tag{3.11}$$

The functional $\mathcal{E}(\tau) = \mathcal{E}[h(\cdot,\tau)]$ is the large-scale surface free energy

$$\begin{aligned}\mathcal{E}[h(\cdot,\tau)] &= \lim_{\epsilon\downarrow 0}(\epsilon E_N)\\ &= \frac{1}{3}\int_{\mathcal{U}} g^0(x,\tau)\, m(x,\tau)^3\, \mathrm{d}x\ .\end{aligned} \tag{3.12}$$

The large-scale interaction parameter $g^0(x,\tau)$ is defined via (cf. Definition 2.8)

$$g^0(x,\tau) = \lim_{\substack{n_j\epsilon\downarrow 0\\ n_j\to\infty}} \bar{g}_j(\tau)\ , \quad \bar{g}_j\overline{m}_j^2 := n_j^{-1}\sum_{i\in I_j}(3\breve{g}_i/2)m_i^2 \qquad (\breve{g}_i = \mathcal{O}(1))\ , \tag{3.13}$$

where $\overline{m}_j = (n_j^{-1}\sum_{i\in I_j} m_i^{-1})^{-1}$.

The second line of (3.12) in terms of g^0 is a choice in accord with the elastic-dipole origin of the step interaction energy. Alternatively, it suffices to define $\mathcal{E}[h]$ as $\lim_{\epsilon\to 0}(\epsilon E_N)$ without resorting to the effective parameter g^0. Note that g^0 is given as the average of $\{3\breve{g}_i/2\}_{i\in I_j}$ induced by $m_i = \epsilon/w_i$ via $f(m) = m^2$. Hence, g^0 in principle depends on the surface slope, $m(x,\tau)$. In the special case with a non-composite stepped surface, by $\breve{g}_i = \breve{g} = \text{const.}$ we have $g^0(x) = 3\breve{g}/2$ provided the step densities squared, m_i^2, vary sufficiently slowly within each I_j. Indeed, by $m_{i_\diamond}^2 = m(x(h))^2$ for fixed $i_\diamond \in I_j$, expand $m_{i_\diamond+p}^2 = m(x(h))^2 + \mathcal{O}(p\epsilon)$ for $i = i_\diamond + p \in I_j$ ($p = 0, \pm 1, \ldots$); thus, $n_j^{-1}\sum_{i\in I_j} m_i^2 \to m(x(h))^2$ as $\epsilon\downarrow 0$.

4. Derivation of evolution laws. We proceed to derive in detail the results stated in section 3. We assert that boundary conditions (2.4), which suffice for the usual Fick's law, are decoupled from the step velocity law (2.9) to the desired order of perturbation (at the chosen time scale). Further, we construct the continuum-scale chemical potential on the basis of (2.8) in terms of the step train free energy.

Consider the definitions of section 2.3. By virtue of variables (2.15) and (2.16) along with expansion (2.19), diffusion equation (2.2) reads

$$(\partial_y + \epsilon\partial_x)D_i(x)(\partial_y + \epsilon\partial_x)(\varrho^0 + \epsilon\varrho^1 + \epsilon^2\varrho^2 + \ldots) \tag{4.1}$$

$$= (\epsilon^2\partial_t + \epsilon^4\partial_\tau)(\varrho^0 + \epsilon\varrho^1 + \epsilon^2\varrho^2 + \ldots) \qquad i\in I_j\ , \tag{4.2}$$

where ϱ^l is the lth-order term for the adatom concentration.

Taking $\tau = \mathcal{O}(1)$, we assume that each ϱ^l is stationary in t and subsequently set $\partial_t\varrho^l \equiv 0$. By dominant balance, we find the following cascade of equations:

$$\mathcal{O}(\epsilon^0):\ \partial_y^2\varrho^0 = 0\ , \tag{4.3a}$$

$$\mathcal{O}(\epsilon^1):\ \partial_y^2\varrho^1 + \partial_{yx}\varrho^0 + D_i(x)^{-1}\partial_x\big[D_i(x)\partial_y\varrho^0\big] = 0\ , \tag{4.3b}$$

$$\mathcal{O}(\epsilon^2):\ \partial_y^2\varrho^2 + \partial_{yx}\varrho^1 + D_i(x)^{-1}\partial_x\big[D_i(x)\big(\partial_y\varrho^1 + \partial_x\varrho^0\big)\big] = 0\ldots\ . \tag{4.3c}$$

Equations (4.3) suffice for our purpose of identifying the macroscopic limit.

In subsections 4.1 and 4.2, our computations involve steps and terraces that correspond to some fixed yet arbitrary I_j ($j = 1, \ldots, M$). Set $y := (x - \breve{x})/\epsilon$ where $\breve{x} = x_{i_*}$ is the position of the first step in I_j; thus, $y_{i_*} = (x_{i_*} - \breve{x})/\epsilon = 0$.

The dependence of the coefficient $\varrho^l(x,y)$ on the fast variable, y, in principle stems from kinetic conditions (2.4). These conditions are imposed at the microscale boundaries $y_i = (x_i - \breve{x})/\epsilon$. By subtracting conditions (2.4), we eliminate ρ_i^{eq} and obtain

$$\varrho^+ - \varrho^- = -\epsilon\left(\frac{\mathcal{J}^-}{k_{\mathrm{d},i}\epsilon} + \frac{\mathcal{J}^+}{k_{\mathrm{u},i}\epsilon}\right) + \epsilon v_i\left(\frac{\varrho^-}{k_{\mathrm{d},i}\epsilon} + \frac{\varrho^+}{k_{\mathrm{u},i}\epsilon}\right) \qquad y = y_i\ . \tag{4.4}$$

Equation (4.4) expresses a jump of the adatom concentration across each step edge, including the convective term, ϱv_i.

4.1. Fick's law. In this subsection, we derive (3.1)–(3.5). For notational economy, we suppress the time dependence in most of our computations.

Since $x_i(t) = x_i^\epsilon(t)$ are moving boundaries, or Lagrangian coordinates, we expand

$$x_i = x_i^0 + \epsilon x_i^1 + \epsilon^2 x_i^2 + \dots \ . \tag{4.5}$$

By Remark 2.13 and expansion (2.20) for the flux, the step velocity law (2.9) reads[8]

$$v_i^\epsilon(t) = \epsilon^2[u_i(\tau) + o(1)] = \left(\mathcal{J}^{(-1),-} - \mathcal{J}^{(-1),+}\right) + o(1) \qquad y = y_i \ , \tag{4.6}$$

where $u_i(\tau) = \mathcal{O}(1)$ and $\mathcal{J}^{(-1),\pm}$ is the restriction of the leading-order flux term $\mathcal{J}^{(-1)}(x,y)$ at $y = y_i^0$ from left ($-$) or right ($+$). Thus, to the lowest order in perturbation, the adatom flux is continuous across step edges, $\mathcal{J}^{(-1),+} = \mathcal{J}^{(-1),-}$. In fact, this property can be extended to the next higher order for $\mathcal{J}$ as well, as shown below.

4.1.1. Lowest (zeroth) order. First, consider (4.3a). The solution reads

$$\varrho^0(x,y) = A_i(x)y + B_i(x) \qquad y_{i-1}^0 < y < y_i^0 \quad (i \in I_j) \ , \tag{4.7}$$

where A_i and B_i are viewed as $\mathcal{O}(1)$ integration constants, to be found via boundary conditions at step edges. By virtue of (4.6), the leading-order mass flux obeys

$$\epsilon\mathcal{J}(x) \to \mathcal{J}^{(-1)}(x,y) = -D_i(x)\partial_y \varrho^0(x,y) = -D_i(x)A_i(x) \qquad y_{i-1}^0 < y < y_i^0 \ . \tag{4.8}$$

Thus, the continuity of flux to this order yields

$$D_i A_i = D_{i-1}A_{i-1} = \ldots = D_{i_*}A_{i_*} \ \Rightarrow \ A_i(x) = \frac{D_{i_*}(x)}{D_i(x)}\, A_{i_*}(x) \ . \tag{4.9}$$

The dominant balance of conditions (4.4) entails

$$\varrho^{0,+} - \varrho^{0,-} = -\left(\frac{1}{k_{\mathrm{u},i}\epsilon} + \frac{1}{k_{\mathrm{d},i}\epsilon}\right)\mathcal{J}^{(-1)} \qquad \text{at} \quad y = y_i^0 \ ,$$

where $\varrho^{0,\pm}$ denotes the restriction of ϱ^0 at y_i^0 from left ($-$) or right ($+$). By (4.7),

$$(A_{i+1} - A_i)y_i^0 + B_{i+1} - B_i = \left(\frac{D_i}{k_{\mathrm{u},i}\epsilon} + \frac{D_i}{k_{\mathrm{d},i}\epsilon}\right) A_i \ .$$

To obtain B_i, we add up the last equations backwards in i in terms of a telescopic sum. The result is

$$B_i = B_{i_*} + A_{i_*} D_{i_*}\left[-\frac{y_i^0}{D_i} + \sum_{p=i_*}^{i-1}\left(\frac{y_{p+1} - y_p}{D_{p+1}} + \sum_{\ell=\mathrm{u,d}} \frac{2}{k_{\ell,p}\epsilon}\right)\right] \quad (i \in I_j,\, i > i_*) \ . \tag{4.10}$$

---

[8] In (4.6) we leave the step number dependence (via i) in the leading-order term for v_i^ϵ. Alternatively, we could have set $v_i^\epsilon(t) = \epsilon^2[u^0(x,\tau) + o(1)]$, anticipating that the step velocity has a macroscopic limit. By adhering to (4.6), we will show that such a u^0 is the mesoscale arithmetic mean of the discrete, $\mathcal{O}(1)$ step velocities u_i in the continuum limit.

By (4.7), (4.9) and (4.10), we find

$$\begin{aligned}\varrho^0(x,y) &= \frac{D_{i_*}(x)A_{i_*}(x)}{D_i(x)}\,(y-y_i^0)+B_{i_*}(x)\\ &\quad + D_{i_*}(x)A_{i_*}(x)\sum_{p=i_*}^{i-1}\left(\frac{y_{p+1}^0-y_p^0}{D_{p+1}(x)}+\sum_{\ell=\mathrm{u},\mathrm{d}}\frac{2}{k_{\ell,p}\epsilon}\right)\ . \end{aligned}\tag{4.11}$$

Now recall Remark 2.11, which implies a solvability condition on ϱ^0. By (2.3) (which exacts the bounds for $D_i(x)$), we assert that

$$0<\frac{y_i^0-y_{i_*}^0}{D_M}\le\sum_{p=i_*}^{i-1}\frac{y_{p+1}^0-y_p^0}{D_{p+1}(x)}\le\frac{y_i^0-y_{i_*}^0}{D_m}\ ,\tag{4.12}$$

where $y_i^0-y_{i_*}^0=\mathcal{O}(n_j)$ for large enough i in I_j. Hence, the sums in (4.11) diverge as $n_j\to\infty$ with $i=i_\circ$, unless

$$A_{i_*}(x)\equiv 0\ .$$

We showed the following (anticipated) property.

Proposition 4.1. *The zeroth-order adatom concentration is*

$$\varrho^0(x,y)=B_{i_*}(x)=:B(x)\ ,\tag{4.13}$$

i.e., independent of the fast variable. Thus, the corresponding flux is $\mathcal{J}^{(-1)}(x,y)\equiv 0$.

4.1.2. First order.

In order to obtain a relation between $\mathcal{J}^0$ and $B(x)$, we proceed to computing the next-order adatom coefficient, $\rho^1(x,y)$. By (4.3b) and Proposition 4.1, we readily obtain

$$\varrho^1(x,y)=C_i(x)y+F_i(x)\qquad y_{i-1}^0+\epsilon y_{i-1}^1<y<y_i^0+\epsilon\, y_i^1\ ,\tag{4.14}$$

suppressing the time variable, s. Thus, the adatom concentration up to $\mathcal{O}(\epsilon)$ is $B(x)+\epsilon[C_i(x)y+F_i(x)]$. The corresponding terrace adatom flux reads

$$\mathcal{J}(x,y)\sim\mathcal{J}^0(x,y)=-D_i(x)[C_i(x)+\partial_xB(x)]\quad(y_{i-1}^0+\epsilon y_{i-1}^1<y<y_i^0+\epsilon\, y_i^1)\ .\tag{4.15}$$

By dominant balance applied to velocity law (2.9) under expansion (4.6), we readily verify the following statement.

Remark 4.2. *The adatom flux* $\mathcal{J}^0(x,\cdot)$ *is continuous across step edges.*

By enforcing the continuity of $\mathcal{J}^0(x,y)$ at each boundary $y=y_i$ we get

$$\begin{aligned}&D_i(x)[C_i(x)+\partial_xB(x)]=D_{i+1}(x)[C_{i+1}(x)+\partial_xB(x)]\\ &\Rightarrow C_i(x)=\frac{D_{i_*}(x)}{D_i(x)}[C_{i_*}(x)+\partial_xB]-\partial_xB\qquad(i,\,i+1\in I_j)\ .\end{aligned}\tag{4.16}$$

We turn attention to boundary conditions (4.4). These are recast to the form

$$\begin{aligned}&[C_{i+1}(x)-C_i(x)](y_i^0+\epsilon y_i^1)+F_{i+1}(x)-F_i(x)+\mathcal{O}(\epsilon)\\ &=\left(\frac{D_i}{k_{\mathrm{d},i}\epsilon}+\frac{D_i}{k_{\mathrm{u},i}\epsilon}\right)[C_i(x)+\partial_xB(x)]+\mathcal{O}(\epsilon)\qquad(i,i+1\in I_j)\ .\end{aligned}\tag{4.17}$$

Thus, by Remark 2.10, the boundary correction terms ϵy_i^1 do not contribute to this order. By using (4.16) and adding up (4.17) via a telescopic sum, we obtain

$$F_i(x) = F_{i_*}(x) + D_{i_*}(x)[C_{i_*}(x) + \partial_x B]\left[\sum_{p=i_*}^{i-1}\left(\sum_{\ell=u,d}\frac{1}{k_{\ell,p}\epsilon} + \frac{y_{p+1}-y_p}{D_{p+1}}\right) - \frac{y_i}{D_i}\right] . \tag{4.18}$$

The substitution of (4.16) and (4.18) into (4.14) yields

$$\begin{aligned}\varrho^1(x,y) = {}& \left(\frac{D_{i_*}(x)}{D_i(x)}[C_{i_*}(x) + \partial_x B(x)] - \partial_x B\right)(y - y_i^0) + F_{i_*}(x) + y_i^0\Biggl\{-\partial_x B \\ & + D_{i_*}(x)[C_{i_*}(x) + \partial_x B](y_i^0)^{-1}\sum_{p=i_*}^{i-1}\left(\frac{y_{p+1}^0 - y_p^0}{D_{p+1}(x)} + \sum_{\ell=u,d}\frac{2}{k_{\ell,p}\epsilon}\right)\Biggr\} ,\end{aligned} \tag{4.19}$$

where $y_{i-1}^0 + \epsilon y_{i-1}^1 < y < y_i^0 + \epsilon y_i^1$ and $i > i_*$. By requiring that ϱ^1 be bounded as i increases, we impose the necessary condition that the coefficient of y_i^0 (enclosed by the curly brackets) vanish when $i - i_* = \mathcal{O}(n_j) \to \infty$ in (4.19). By $i = i_\circ(j)$ we obtain the solvability condition

$$D_{i_*}(x)[C_{i_*}(x) + \partial_x B] \lim_{\substack{n_j\to\infty \\ n_j\epsilon\downarrow 0}}\left[\frac{1}{y_{i_\circ}^0}\sum_{p\in I_j}\left(\sum_{\ell=u,d}\frac{2}{k_{\ell,p}\epsilon} + \frac{y_{p+1}^0 - y_p^0}{D_{p+1}}\right)\right] = \partial_x B . \tag{4.20}$$

By definitions (3.3) and (3.4), this relation is recast to the form

$$D_{i_*}(x)[C_{i_*}(x) + \partial_x B]\left[1 + q^0(x)\lim_{\substack{n_j\to\infty \\ n_j\epsilon\downarrow 0}}\frac{n_j\epsilon}{x_{i_\circ} - x_{i_*}}\right] = D^0(x)\partial_x B ,$$

where the requisite limit is the surface slope (see Remark 2.10); thus,

$$D_{i_*}(x)[C_{i_*}(x) + \partial_x B(x)] = \frac{D^0(x)}{1 + q^0(x)m(x)}\,\partial_x B . \tag{4.21}$$

Equation (4.21) entails the desired Fick's law. By (4.15) and (4.16), the flux is

$$\mathcal{J}^0(x,y) = -D_{i_*}(x)[C_{i_*}(x) + \partial_x B(x)] ,$$

which is independent of the fast spatial variable, y; cf. [19].

PROPOSITION 4.3. *The macroscopic limit of the adatom flux reads*

$$\mathcal{J}^0(x) = -\frac{D^0(x)}{1 + q^0(x)m(x)}\,\partial_x B , \tag{4.22}$$

where D^0 and q^0 are defined in (3.3) *and* (3.4)*; $B(x)$ is discussed in section* 4.1.3.

4.1.3. Equilibrium concentration. Next, we show that the $B(x)$ in (4.22) can be identified with the continuum-scale version, $\varrho^{\mathrm{eq},0}$, of ρ_i^{eq}, which is affine in the large-scale chemical potential, μ. For this purpose, we revisit kinetic conditions (2.4). By adding up these equations, we obtain the relation

$$\epsilon\left(\frac{\mathcal{J}^-}{k_{\mathrm{d},i}\epsilon} - \frac{\mathcal{J}^+}{k_{\mathrm{u},i}\epsilon}\right) + \epsilon v_i\left(\frac{\varrho^+}{k_{\mathrm{u},i}\epsilon} - \frac{\varrho^-}{k_{\mathrm{d},i}\epsilon}\right) = (\varrho^+ + \varrho^-)\big|_{y_i} - 2\rho_i^{\mathrm{eq}} . \tag{4.23}$$

Let us first comment on the left-hand side of (4.23). By the hypothesis $k_{\ell,i}\epsilon = \mathcal{O}(1)$ (Remark 2.10), the first term is $\mathcal{O}(\epsilon)$. In view of Remark 2.13, the second term is $\mathcal{O}(\epsilon^3)$. Thus, by formally writing

$$\rho_i^{\mathrm{eq}} = \varrho^{\mathrm{eq},0}(x,y) + o(1) \ , \tag{4.24}$$

we apply the usual dominant-balance argument to order $\mathcal{O}(\epsilon^0)$ to infer

$$2B(x) = \varrho^{0,+} + \varrho^{0,-} = 2\varrho^{\mathrm{eq},0} \ \Rightarrow \ \varrho^{\mathrm{eq},0}(x) = B(x) \ . \tag{4.25}$$

Relation (4.25) is consistent with defining the induced average $\bar{\varrho}_j^{\mathrm{eq}}$,

$$\bar{\phi}_j \ \bar{\varrho}_j^{\mathrm{eq}} = n_j^{-1} \sum_{i \in I_j} \rho_i^{\mathrm{eq}} \phi_i \ , \tag{4.26}$$

and subsequently taking $\bar{\varrho}_j^{\mathrm{eq}} \to \varrho^{\mathrm{eq},0}(x)$ and $\bar{\phi}_j \to \phi(x)$ as $\epsilon \downarrow 0$. Indeed, multiply (4.23) by ϕ_i, sum over $i \in I_j$, and then expand in ϵ. Accordingly, (4.25) is interpreted in the weak sense, where $\{\phi_i\}$ is an appropriate test sequence. Equation (4.26) is used for $\phi_i = u_i$ in conjunction with the continuum-scale chemical potential (section 4.3).

Proposition 4.3 and relation (4.25) yield formulas (3.1)–(3.5). By $\mu_i = \mu(x) + o(1)$ and (2.5) we express $\varrho^{\mathrm{eq},0}$ in terms of μ, winding up with (3.6). Alternatively, define μ via $\bar{\phi}_j \bar{\mu}_j := n_j^{-1} \sum_{i \in I_j} \phi_i \mu_i$, invoke the linear relation between μ_i and ρ_i^{eq}, and take $\bar{\mu}_j \to \mu$.

4.2. Mass conservation law and level set motion. In this subsection, we derive (3.7) and (3.8) with recourse to step velocity law (2.9). This law involves a jump of the flux, $\mathcal{J}(x,\cdot)$, at each step edge ($y = y_i$). Recall that the flux is continuous to the first two orders in ϵ. In fact, the jump is revealed to the next higher order. We follow two alternate routes. One method is to apply a solvability condition in the spirit of section 4.1. Another route is a weak formulation [21].

Further, we employ a weak formulation in order to derive the level set motion law (3.9), which introduces the time derivative $\partial_\tau h$. This law emerges in connection to the continuum-scale chemical potential; see (4.42).

4.2.1. Perturbation expansion. We continue the homogenization argument of section 4.1. We resort to (4.3c), solve for $\varrho^2(x,y)$ and the flux coefficient $\mathcal{J}^1$, and thereby determine the discontinuity of $\mathcal{J}$ at each step edge to order $\mathcal{O}(\epsilon)$. A solvability condition for $\mathcal{J}^1$ then yields the desired formula.

Equation (4.3c) along with Proposition 4.3 and Definition (3.2) yield

$$\begin{aligned} &\partial_y[D_i(x)\partial_y \varrho^2(x,y)] = -\partial_x\{D_{i_*}(x)[C_{i_*}(x) + \partial_x B]\} - D_i(x)\partial_x C_i \\ &\Rightarrow \varrho^2(x,y) = yG_i(x) + H_i(x) - \left\{\partial_x C_i + D_i(x)^{-1}\partial_x[\mathcal{D}_{\mathrm{e}}(x)\partial_x B]\right\}\frac{y^2}{2} \ . \end{aligned} \tag{4.27}$$

The adatom concentration on the ith terrace up to $\mathcal{O}(\epsilon^2)$ reads

$$\begin{aligned} \varrho(x,y) &= \varrho^0(x,y) + \epsilon\varrho^1(x,y) + \epsilon^2\varrho^2(x,y) + o(\epsilon^2) \\ &= B(x) + \epsilon[C_i(x)y + F_i(x)] + \epsilon^2\{yG_i(x) + H_i(x) \\ &\qquad - [\partial_x C_i + D_i(x)^{-1}\partial_x(\mathcal{D}_{\mathrm{e}}(x)\partial_x B)]y^2/2\} + o(\epsilon^2) \ . \end{aligned} \tag{4.28}$$

The respective surface flux on the ith terrace is

$$\begin{aligned} \mathcal{J}(x,y) &= -D_i(x)[(\partial_y\varrho^1 + \partial_x\varrho^0) + \epsilon(\partial_y\varrho^2 + \partial_x\varrho^1)] + o(\epsilon) \\ &= -D_i(x)[C_i(x) + \partial_x B] - \epsilon\{D_i(x)[G_i(x) + \partial_x F_i] - y\partial_x(\mathcal{D}_{\mathrm{e}}(x)\partial_x B)\} + o(\epsilon) \ . \end{aligned} \tag{4.29}$$

The $\mathcal{O}(\epsilon^0)$ term is of course continuous at each y_i. At $y = y_i$ this flux has the jump

$$\epsilon(\mathcal{J}^{1,+} - \mathcal{J}^{1,-}) = -\epsilon[D_{i+1}(x)(G_{i+1}(x) + \partial_x F_{i+1}) - D_i(x)(G_i(x) + \partial_x F_i)] \ .$$

The next task is to determine the coefficient $G_i(x)$. By virtue of (4.6), step velocity law (2.9) with $\Omega = \epsilon^2$ is recast to the form

$$u_i + o(1) = -[\mathcal{J}^{1,+} - \mathcal{J}^{1,-} + o(1)] + \epsilon^2 u_i \, [\varrho^{1,+} - \varrho^{1,-} + o(1)] \qquad y = y_i \ .$$

Note that the convective term in the right-hand side of this equation does not contribute to the lowest order. Thus, we obtain the distinguished limit

$$u_i = -(\mathcal{J}^{1,+} - \mathcal{J}^{1,-})\big|_{y_i^0} \Rightarrow u_i = D_{i+1}(G_{i+1} + \partial_x F_{i+1}) - D_i(G_i + \partial_x F_i) \ . \tag{4.30}$$

This result amounts to adatom mass conservation: the discontinuity in the flux is balanced by the step velocity. The convective terms do not contribute because of the slow time scale. Equation (4.30) leads to a telescopic sum for G_i. Thus, we get

$$G_i(x) = \frac{D_{i_*}(x)}{D_i(x)}[G_{i_*}(x) + \partial_x F_{i_*}(x)] - \partial_x F_{i_*} + D_i(x)^{-1} \sum_{p=i_*}^{i-1} u_p \ . \tag{4.31}$$

By (4.29) and (4.31), the coefficient of the $\mathcal{O}(\epsilon)$ term for the flux reads

$$\begin{aligned} \mathcal{J}^1(x,y) &= -D_{i_*}(x)[G_{i_*}(x) + \partial_x F_{i_*}(x)] + (y - y_i^0)\partial_x\big(\mathcal{D}_{\rm e}(x)\partial_x B\big) \\ &\quad + y_i^0 \left[\partial_x\big(\mathcal{D}_{\rm e}\partial_x B\big) - (y_i^0)^{-1} \sum_{p=i_*}^{i-1} u_p\right] \ . \end{aligned} \tag{4.32}$$

Now let $i \in I_j$ approach $i_\circ(j) = i_*(j) + n_j - 1$. By requiring that $\mathcal{J}^1$ does not grow as $n_j \to \infty$, we assert that

$$\partial_x\big(\mathcal{D}_{\rm e}(x)\partial_x B\big) = \lim_{\substack{n_j \to \infty \\ n_j \epsilon \to 0}} \left[\left(\frac{n_j}{y^0_{i_\circ(j)}}\right)\left(n_j^{-1}\sum_{p=i_*}^{i_\circ} u_p\right)\right] \Rightarrow -\partial_x \mathcal{J}^0(x) = m(x)\, \bar{u}(x) \ ,$$

with recourse to (2.17) and (3.8).

Proposition 4.4. *The macroscopic limit, $\mathcal{J}^0$, of the surface flux obeys* (3.7), *where the continuum-scale step velocity is the (mesoscale) arithmetic mean* (3.8).

4.2.2. Weak formulation for mass conservation. The step velocity law (2.9) is written in the form

$$\epsilon^2 u_i = -\epsilon\, [\mathcal{J} + \mathcal{O}(\epsilon^2)]_i \ ; \qquad [Q]_i := Q(y_i^+) - Q(y_i^-) \ . \tag{4.33}$$

Following [21], we multiply both sides of (4.33) by ϕ_i and sum over i to obtain

$$\begin{aligned} &\epsilon \sum_{i=0}^{N-1} \phi_i u_i = -\sum_{i=0}^{N-1} [\mathcal{J}]_i \phi_i + \mathcal{O}(\epsilon) \\ \Rightarrow &\sum_j (n_j\epsilon)\phi_{i_*(j)}\left(n_j^{-1}\sum_{i\in I_j} u_i\right) = -\sum_j \phi_{i_*(j)} \sum_{i \in I_j} [\mathcal{J}]_i + o(1) \\ \Rightarrow &\int \phi(x)\, \bar{u}(x) m(x)\, {\rm d}x = -\int \phi(x)\, {\rm d}\mathcal{J}^0(x) \qquad \text{as } \epsilon \downarrow 0 \ , \ n_j\epsilon \downarrow 0 \quad \forall \phi \ , \end{aligned} \tag{4.34}$$

which implies (3.7) in the weak sense. Note that we employ a discretized version $\phi_i = \mathcal{O}(1)$ of smooth $\phi(x)$ such that $\phi_{i_\circ(j)} - \phi_{i_*(j)} = o(\phi_{i_*})$ for all j; i.e., ϕ_i varies slowly (in i) within each I_j. This detail becomes immaterial in the continuum limit.

4.2.3. Level set motion law. Next, we derive geometric law (3.9) with recourse to a weak formulation. For this purpose, define

$$I_N := \epsilon \int_0^{T_0} \sum_i u_i \phi_i \, \mathrm{d}\tau \ ; \quad u_i = \mathrm{d}x_i/\mathrm{d}\tau \ , \tag{4.35}$$

where the sequence $\{\phi_i\}$ consists of smooth, compactly supported functions $\phi_i : (0, T_0] \to \mathbb{R}$. We will show that, in the continuum limit,

$$\int_0^{T_0} \int \phi \, h_\tau \, \mathrm{d}x \mathrm{d}\tau = \int_0^{T_0} \int \phi \, u^0 \, m \, \mathrm{d}x \mathrm{d}\tau \ , \tag{4.36}$$

for any test function $\phi : \mathcal{U} \times (0, T_0] \to \mathbb{R}$. Note that u^0 denotes the macroscopic limit of $\bar{u} = n_j^{-1} \sum_{i \in I_j} u_i$, and $h_\tau := \partial_\tau h$.

First, consider the case with homogeneous steps and terraces, i.e., when the material parameters remain unchanged across terraces. By (4.35), we have

$$I_N \to \int_0^{T_0} \int u^0 \, \phi \, \mathrm{d}h = \int_0^{T_0} \int u^0 \, m \, \phi \, \mathrm{d}x \mathrm{d}\tau \qquad \text{as } N \to \infty, \, N\epsilon = \mathcal{O}(1) \ , \tag{4.37}$$

where u^0 is the continuum limit of $\{u_i\}$.

On the other hand, by integration by parts, we assert that

$$I_N = -\epsilon \int_0^{T_0} \sum_i x_i \frac{\mathrm{d}\phi_i}{\mathrm{d}\tau} \, \mathrm{d}\tau \to \int_0^{T_0} \int x \, (\phi_\tau|_h) \, h_x \, \mathrm{d}x \mathrm{d}\tau \ , \tag{4.38}$$

where $h_x = -|h_x|$ and $\phi_\tau|_h$ denotes the partial derivative of ϕ with respect to τ with *fixed* h. By changing variables (from Lagrangian to Eulerian coordinates), we write

$$\phi_\tau|_h = \phi_\tau|_x - \phi_x \, \frac{h_\tau}{h_x} \ .$$

Thus, after an integration by parts in τ and another one in x, (4.38) yields

$$\begin{aligned} I_N &\to - \int_0^{T_0} \int x h_{x\tau} \phi \, \mathrm{d}x \mathrm{d}\tau - \int_0^{T_0} \int x \phi_x h_\tau \, \mathrm{d}x \mathrm{d}\tau \\ &= \int_0^{T_0} \int [(x\phi)_x - x\phi_x] h_\tau \, \mathrm{d}x \mathrm{d}\tau \ . \end{aligned} \tag{4.39}$$

The comparison of (4.37) and (4.39) implies (4.36).

Alternatively, for the above case of a non-composite stepped surface, write $h(x_i(t), t) =$ const. for each terrace (level set of h). The differentiation of this equation with respect to t yields the desired law in the continuum limit [21]. However, this argument becomes questionable for a composite surface.

In the case of a composite stepped surface, we need to slightly modify the manipulation of (discrete) sum I_N in (4.37) and (4.38). In particular, for appropriate test sequences, we have

$$I_N = \int_0^{T_0} \sum_{j=1}^{M} (n_j \epsilon) \phi_j \left(n_j^{-1} \sum_{i \in I_j} u_i \right) \mathrm{d}\tau \to \int_0^{T_0} \int \phi \, u^0 \, \mathrm{d}h \mathrm{d}\tau \ . \tag{4.40}$$

On the other hand, I_N equals

$$I_N = \int_0^{T_0} \sum_{j=1}^{M} (n_j\epsilon)\phi_j \left(n_j^{-1} \sum_{i\in I_j} \frac{\mathrm{d}x_i}{\mathrm{d}\tau} \right) \mathrm{d}\tau = -\int_0^{T_0} \sum_{j=1}^{M} (n_j\epsilon) \frac{\mathrm{d}\phi_j}{\mathrm{d}\tau} \left(n_j^{-1} \sum_{i\in I_j} x_i \right) \mathrm{d}\tau$$

$$\to -\int_0^{T_0} \int (\phi_\tau|_h) x \,\mathrm{d}h \mathrm{d}t \ . \tag{4.41}$$

The remainder of the derivation leading to (4.36) follows directly from (4.39).

4.3. Step chemical potential and free energy. Next, we focus on the derivation of (3.10)–(3.13). To obtain the continuum-scale chemical potential, μ, in terms of $\mathcal{E} = \lim_{\epsilon\to 0}(\epsilon E_N)$, we use (2.8). By $\tau = \epsilon^2 t$ and $u_i = \epsilon^{-2} v_i + o(1)$, we have

$$\epsilon \frac{\mathrm{d}E_N}{\mathrm{d}\tau} = \epsilon \sum_{i=0}^{N-1} u_i \mu_i = \sum_{j=1}^{M} (n_j\epsilon) \left(n_j^{-1} \sum_{i\in I_j} u_i \mu_i \right)$$

$$\Rightarrow \dot{\mathcal{E}}(\tau) = \int u^0(x,\tau)\, \mu(x,\tau)\, \mathrm{d}h \quad \text{as } n_j \to \infty,\ n_j\epsilon \downarrow 0 \ . \tag{4.42}$$

Formula (3.11) follows by $\int \cdot\, \mathrm{d}h = \int \cdot\, m\, \mathrm{d}x$ and $u^0 m = \partial_\tau h$.

Recall that the microscale free energy E_N is described by (2.7). To make a connection to the known continuum-scale surface free energy of a non-composite stepped surface [10,14], we have recourse to an induced average of $\{\breve{g}_i\}$. By Definition 2.8, we obtain the formal expression

$$\epsilon E_N(t) = \frac{\epsilon}{2} \sum_{j=1}^{M} \sum_{i\in I_j} \breve{g}_i m_i^2 = \frac{1}{3} \sum_{j=1}^{M} (n_j\epsilon) \bar{g}_j\, \bar{m}_j^2$$

$$\to \frac{1}{3} \int g^0(x,\tau)\, m(x,\tau)^2 \,\mathrm{d}h =: \mathcal{E}(\tau) \qquad \text{as } \epsilon \downarrow 0 \ , \tag{4.43}$$

assuming that the average $\bar{g}_j$ varies slowly across mesoscale regions.

5. Extension: Radial setting.

In this section we discuss the radial case, indicating that the homogenization procedure follows directly from the (1+1)-dimensional case described above. Our main assumption is the existence of a mesoscale (in the spirit of section 2.2) linking the microstructure of terraces to the macroscale.

The surface consists of concentric, circular steps of radii $r_i(t)$ with $r_{i+1} > r_i$, $i = 0, \ldots, N-1$. The ith terrace is $\mathcal{T}_{it} = \{r \,|\, r_{i-1}(t) < r < r_i(t)\}$, characterized by diffusivity $D_i(r)$, while the kinetic rates at step i are $k_{\mathrm{d},i}$ and $k_{\mathrm{u},i}$. Suppose that the polar coordinate r does not lie close to the top step (at r_0). The adatom diffusion equation (in polar coordinates) now reads

$$\frac{1}{r} \frac{\partial}{\partial r} \left[r D_i(r) \frac{\partial \varrho^\epsilon}{\partial r} \right] = \frac{\partial \varrho^\epsilon}{\partial t} \qquad r \in \mathcal{T}_{it} \ . \tag{5.1}$$

By $\varsigma = (r - r_{i_*})/\epsilon$ and $\tau = \epsilon^2 t$, the multiscale expansion for ϱ^ϵ is

$$\varrho^\epsilon(r,t) = \varrho^0(r,\varsigma,\tau,t) + \epsilon \varrho^1(r,\varsigma,\tau,t) + \epsilon^2 \varrho^2(r,\varsigma,\tau,t) + \ldots \ ; \quad \varrho^l = \mathcal{O}(1) \tag{5.2}$$

as $\epsilon \downarrow 0$. The corresponding expansion for the mass flux stems from $\mathcal{J}^\epsilon = -D_i(r)(\partial_r + \epsilon^{-1}\partial_\varsigma)\varrho^\epsilon$, $r \in \mathcal{T}_{it}$. By $\partial_t \Rightarrow \partial_t + \epsilon^2 \partial_\tau$ and $\tau = \mathcal{O}(1)$, we hypothesize that the dependent variables are stationary in the fast time scale, t, and thus set $\partial_t = 0$. In the remainder of this section, we suppress the τ-dependence.

The perturbation scheme can be pursued order by order in ϵ along the lines of section 4.1. To zeroth order, (5.1) yields $r^{-1}\partial_\varsigma[rD_i(r)\partial_\varsigma\varrho^0] = 0$ in $\varsigma_{i-1}^0 < \varsigma < \varsigma_i^0$, by which $\varrho^0(r,\varsigma) = A_i(r)\varsigma + B_i(r)$. Note that the form of the zeroth-order concentration is preserved in passing from 1+1 dimensions to the radial geometry. The integration constants A_i and B_i are determined by the atom attachment-detachment conditions at $\varsigma = \varsigma_{i-1}^0, \varsigma_i^0$: the mass flux is continuous (in view of the step velocity law) but the concentration has a jump proportional to $\mathcal{J}^{(-1)}$ at each step edge. By requiring that ϱ^0 does not grow in the limit $n_j \to \infty$, we find $\varrho^0(r,\varsigma) = B(r)$; cf. (4.13). To the next higher order in ϵ, we have $r^{-1}\partial_\varsigma[rD_i(r)\partial_\varsigma\varrho^1] = 0$ which entails $\varrho^1(r,\varsigma) = C_i(r)\varsigma + F_i(r)$, as in the (1+1)-dimensional case (section 4.1.2). The enforcement of kinetic conditions at the bounding steps ($\varsigma = \varsigma_{i-1}^0 + \epsilon\varsigma_{i-1}^1$, $\varsigma_i^0 + \epsilon\varsigma_i^1$) yields C_i and F_i. The solvability (non-growth) condition on ϱ^1 entails a relation between $\partial_r B$, $D_{i_*}(r)$ and $C_{i_*}(r)$ analogous to (4.20). Hence, the macroscopic Fick law reads

$$\mathcal{J}^0(r,\varsigma) = -\frac{D^0(r)}{1+q^0(r)m(r)}\partial_r B(r)\ , \tag{5.3}$$

where $m(r)$ is the continuum-scale surface slope, $D^0(r)$ and $q^0(r)$ are given by (3.3) and (3.4) with y replaced by ς; and $B(r)$ is identified with the continuum limit of ϱ_i^{eq}, i.e., the variable $\varrho^{\mathrm{eq}}(r) \sim \rho_*[1+\mu(r)/\vartheta]$ where μ is the continuum-scale chemical potential.

The remaining continuum laws can be determined along the lines of sections 4.2 and 4.3. Accordingly, the mass conservation law for adatoms reads $m(r)u^0(r) + \partial_r\mathcal{J}^0(r) = 0$, which is found by detailed balance as in the (1+1)-dimensional case (section 4.2). The (geometric) level set motion law again reads $m\,u^0 = \partial_\tau h$ where h is the large-scale height, $m = |\partial_r h|$. The continuum-scale chemical potential is $\mu = \delta\mathcal{E}[h]/\delta h$; $\mathcal{E}$ is the continuum limit of the mean energy ϵE_N where [13, 21]

$$E_N[\vec{r}] = \sum_{i=0}^{N-1}\left[\breve{g}_{1,i} + \breve{g}_{3,i}\,\frac{2r_{i+1}}{r_i + r_{i+1}}\left(\frac{\epsilon}{r_{i+1}-r_i}\right)^2\right] \tag{5.4}$$

corresponds to the total step free energy. In (5.4), $\vec{r} = (r_0, \dots, r_{N-1})$, and $\{\breve{g}_{1,i}\}$ and $\{\breve{g}_{3,i}\}$ are $\mathcal{O}(1)$ parameters expressing the step line tension and elastic-dipole (and entropic) step-step interaction, respectively.

6. Conclusion.

We examined the consistency of macroscopic laws for crystal surface relaxation with the discrete, BCF-type step flow when terraces and steps are characterized by sequences of distinct material parameters. The main assumption is the existence of an intermediate scale, the mesoscale, which helps express the slow spatial variation of microscale averages and the property that the crystal surface appears homogeneous at the macroscale.

Our methodology relies on classical homogenization, by which the adatom concentration and mass flux are globally expressed in terms of appropriate multiscale expansions. Two features of these expansions are: (i) the inclusion of step boundaries through enforcement of boundary conditions at the fast spatial variable, in the spirit of [12]; and (ii) the use of a macroscopic time scale consistent with the quasi-steady approach, under the hypothesis that the variables are stationary in the fast time scale.

The form of macroscopic laws is the same as in the case of a non-composite stepped surface [29, 34]. These laws comprise Fick's law of surface diffusion (with an appropriate mobility), the mass conservation statement for surface height, and a variational formula for the chemical potential. The homogenization approach shows *how* the large-scale surface mobility turns out to be a function of the slope, with the effective diffusivity and kinetic rate given as appropriate harmonic averages. The precise dependence of the mobility on the slope, emerging from a solvability condition for the adatom concentration, is viewed as a result conceptually *distinct* from the electric-circuit analog of a stepped surface [28].

Regarding the large-scale chemical potential, μ, we assumed that the discrete step free energy has a well-defined continuum limit; and expressed this limit in terms of the induced average of microscale parameters for step-step interactions. The only justification for the choice of this average is the need to provide a continuum formula consistent with the known formula in the case of a non-composite stepped surface.

Our formulation is restricted to one spatial coordinate. In 2+1 dimensions, the adatom concentration can be expressed conveniently in a local coordinate system with axes normal and parallel to step edges [21]. It can be conjectured that the one-dimensional result found for Fick's law can be generalized to include the (2+1)-dimensional-flux component *normal* to step edges. The tangential flux component would have to be treated differently. We expect that the form of macroscopic laws in 2+1 dimensions would not be different from the one derived in [21] for a non-composite stepped surface. The details of the averaging, however, would require a more elaborate application of homogenization. This problem is left for future work.

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Appendix A. On slowly varying averages. In this appendix, we discuss the notion of slow variation of section 2.2. First, we formulate an example of a condition on $\{\zeta_i^\epsilon\}$ (material parameters or variables) such that respective averages, $\bar{\zeta}_j^\epsilon$, are slowly varying (see Definition 2.2). Further, we study implications for a class of induced averages $\bar{\xi}_j^\epsilon$ (Definition 2.8). The dependence on the small scale, ϵ, is suppressed.

Without loss of generality, we take $n_j = n = o(N) \gg 1$ for $j = 1, \ldots, M$, i.e., equal number of steps in each I_j. (Recall: $N = Mn$, $M, N \to \infty$). Prescribe a sequence $\{\zeta_i\}_{i=0}^{N-1}$ such that its subsequences within numerous adjacent I_j's are close in the l^1-sense. Specifically, for any $0 < \sigma \ll 1$ there exist positive integers $j_*(M) = o(M)$ and $j_\circ(M)$ with $M - j_\circ = o(M)$ such that, for each $j_* \le j \le j_\circ$,

$$\text{(A.1)} \qquad \sum_{i\in I_j} |\zeta_{i+k} - \zeta_i| < \sigma \sum_{i\in I_j} |\zeta_i| \qquad \text{for all } |k| < k_\circ(\sigma) \quad \text{if } M > M_\circ(\sigma)\ ,$$

where $k_\circ = p_\circ n = o(N)$ and $p_\circ = o(M)$; in particular, $\sigma \downarrow 0$ as $M_\circ, np_\circ \to \infty$. We will refer to such a sequence $\{\zeta_i\}$ as 'admissible'. This construction (trivially) includes bi-phasic surface reconstructions of Si(001), where $\{\zeta_i\}$ may contain, e.g., alternating diffusivities [38]. We henceforth restrict attention to admissible $\{\zeta_i\}$.

In light of the above prescription, we state and prove two propositions regarding slow variation of averages. The function f invoked below is ϵ-independent.

Proposition A.1. *Suppose* $f : \mathbb{R} \to \mathbb{R}$ *is Lipschitz continuous with* $f(\zeta) > 0$ *for* $\zeta \neq 0$*; and* $\{\zeta_i\}_{i=0}^{N-1}$ *is bounded and admissible as* $N \to \infty$*. In particular, assume that* $0 < \alpha \le |\zeta_i| \le \beta$ *for each* i*. For* $j = 1, \ldots, M$*, define*

$$\text{(A.2)} \qquad \bar{\zeta}_j := n^{-1} \sum_{i\in I_j} f(\zeta_i)\ .$$

Then, $\bar{\zeta}_j$ *are slowly varying.*

Proof. The proof follows directly from Definition 2.2. First, we note that $\bar{\zeta}_j$ have a fixed, positive greatest lower bound. Let $\bar{\alpha} = \min_j\{\bar{\zeta}_j\}$. Accordingly, we obtain an estimate for the quotient $\delta^p\bar{\zeta}_j$:

$$|\delta^p\bar{\zeta}_j| \le (pn)^{-1}\sum_{i\in I_j}|f(\zeta_{i+k(p)})-f(\zeta_i)| \le \mathrm{Lip}[f]\,(pn)^{-1}\sum_{i\in I_j}|\zeta_{i+k(p)}-\zeta_i|$$

$$\le C\sigma\left(n^{-1}\sum_{i\in I_j}|\zeta_i|\right)\le C\sigma\ ,\qquad k=np\ , \tag{A.3}$$

by virtue of the boundedness of $|\zeta_i|$, provided $j_* \le j \le j_\circ$ and $-p_\circ < p < p_\circ$, by the prescription of $\{\zeta_i\}$. Thus, for sufficiently large M, the σ can be made small enough so that $|p\delta^p\bar{\zeta}_j| \ll \bar{\alpha}$ for appropriate j and p. □

Proposition A.2. *Suppose* $f:\mathbb{R}\to\mathbb{R}$ *is Lipschitz continuous with* $f(\zeta)>0$ *for* $\zeta\neq 0$*; and* $\{\zeta_i\}_{i=0}^{N-1}$ *and* $\{\xi_i\}_{i=0}^{N-1}$ *are bounded and admissible. In particular, assume that* $0<\alpha\le|\zeta_i|\le\beta$ *for each* i*. Then, the averages* $\bar{\xi}_j$ *of* $\{\xi_i\}$ *induced by* $\{\zeta_i\}$ *via* f *are slowly varying.*

Proof. Consider Definition 2.8 for induced averages. Without further ado, we estimate $\delta^p[f(\bar{\zeta}_j)\bar{\xi}_j]$ for $p\neq 0$:

$$|\delta^p[f(\bar{\zeta}_j)\bar{\xi}_j]| \le (np)^{-1}\sum_{i\in I_j}\big(|f(\zeta_{i+np})||\xi_{i+np}-\xi_i| + |f(\zeta_{i+np})-f(\zeta_i)||\xi_i|\big)$$

$$\le Cn^{-1}\sum_{i\in I_j}\big(|\xi_{i+np}-\xi_i|+|\zeta_{i+np}-\zeta_i|\big)\le C\sigma\, n^{-1}\sum_{i\in I_j}\big(|\xi_i|+|\zeta_i|\big)\le C\sigma\ , \tag{A.4}$$

where we used the Lipschitz continuity of f; $\sigma=\max(\sigma_\xi,\sigma_\zeta)$ and σ_ℓ is the constant entering the admissibility definition of $\{\ell\}$ for $\ell=\xi,\,\zeta$. By Proposition A.1, $f(\bar{\zeta}_j)$ is also slowly varying. □

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