Unified view of step-edge kinetics and fluctuations

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We study theoretically the equilibrium fluctuations of steps on vicinal surfaces. From an analytical continuum description of the step edge, we find a *single* Langevin equation governing the motion of an isolated step around its equilibrium position that includes attachment/detachment of atoms, diffusion over the terrace, diffusion along the edge, and evaporation. We then extend this approach to treat an array of steps, i.e., a vicinal surface. We also present, in an appendix, an alternative formalism in which detachment to terrace and to step-edge diffusion can take place independently. In established as well as some new limits, and for numerous special cases, we study the wave-vector dependence—both exponent and prefactor—of the relaxation time of fluctuations. From this we recover scaling relations for early-time dependence of the mean-square fluctuations. We discuss how to extract the [mesoscopic] transport coefficients associated with different atomistic mechanisms of surface mass transport and how to distinguish between mechanisms having the same power-law dependence on wavelength in the capillary-wave analysis. To examine the crossovers between limiting regimes, we compute and explore an effective exponent for this power law and show that the crossover occurs over a narrow region of phase space. Furthermore, we find that single-sided approximations are valid only in the limit of extreme Schwoebel barriers. [S0163-1829(98)09008-0]

I. INTRODUCTION

As the fundamental entity of vicinal surfaces, steps have been the subject of substantial experimental investigation. Specifically, a variety of experimental techniques[1–5] have been used to study step-edge fluctuations on metallic and semiconductor surfaces. Some of the important issues addressed have been (i) determining *uniquely* the microscopic mechanisms of surface mass transport, (ii) finding the rate-limiting step in the mass transport, and finally (iii) estimating microscopic energy parameters of the material. We briefly list here some of these studies. Kuipers and co-workers[1] made one of the first observations of these temporal fluctuations of steps on a Au(110) surface using a high-temperature scanning tunneling microscope (STM). They also studied these fluctuations on a Pb(111) surface. In an independent but simultaneous study, Poensgen *et al.*[6] studied the frizzled step edges on Ag(111) and Cu(001) surfaces. The frizziness of the steps was studied as a function of time as well as temperature using scanning tunneling microscopy (STM). Detailed investigation of step fluctuations on Cu(1 1 n), where n = 13,19,79, has also been done, again with STM.[7] They also studied with Monte Carlo simulations the effect of correlation of the kink motion on the fluctuations. An STM study of step fluctuations of steps on Pt(111) was done by Giesen *et al.*[8] and Barbier *et al.*[9] along with L. Masson *et al.*[10] have also reported the roughening of steps on Cu(1 1 11) as well as the quantitative measurement of step-step interactions. Pai and co-workers[11] did a systematic study of step-edge fluctuations on a Ag(110) surface at room temperature using STM, and estimated a single-atom detachment rate at step sites. Recently Wang *et al.*[12] have also studied the spatial and temporal fluctuations of close-packed steps on

Ag(001). Steps on Si(111) have been studied with reflection electron microscopy (REM) at a temperature of 1172 K. Bartelt *et al.*[2] showed in this study that the step fluctuations are rate limited by detachment kinetics on this surface. Alfonso *et al.*[3] studied the same surface at the same temperature to measure the step-edge stiffness and the magnitude of the step-step interactions. On the Si(001)-(2×1) reconstructed surface the dynamics of steps were studied by Pearson *et al.*[13] They found that the mass transport was dominated by kink diffusion. Swartzentruber and Schacht[14] have also studied the kinetics of atomic-scale fluctuations of steps on Si(001) with a variable temperature STM. They found that below 503 K the steps were virtually immobile and that no fluctuations were observed on a time scale of hours. Above 623 K the step movements were so rapid as to make determination of the step position impossible. Bartelt and co-workers[4] did a systematic study of the step-edge fluctuations and equilibrium two-dimensional island shapes on the Si(001) surface. They obtained from this study the step-edge stiffnesses, free energies, and step mobilities of the two types of single-height steps, S_A and S_B, naturally occurring on the Si(001) surface. Their analysis of the step stiffness from thermal fluctuations agreed quantitatively with the equilibrium two-dimensional island shapes they observed, providing a direct connection between equilibrium structure and equilibrium thermal fluctuations.

The theoretical foundations for the relaxation of out-of-equilibrium surface morphologies to equilibrium were laid by the work of Mullins[15,16] in the late 1950s and in the 1960s. More recently Bartelt and co-workers[17,18] applied this work to the equilibrium fluctuations of a single isolated monatomic step on a surface. Pimpinelli *et al.*[19] arrived at some scaling laws in a qualitative manner to relate the relaxation

rates of bumps formed on initially straight steps. Cahn and Taylor[20] generalized Mullins's work for the laws of relaxation of geometrical perturbations of equilibrium surfaces. This in turn inspired work by Bonzel and Mullins,[21] who further generalized Mullins's earlier work[15,16] to include step-step interactions in surface relaxations and to relax the small-slope approximation. Liu and Metiu[22] have also approached this subject from a slightly different perspective, in a Langevin formalism. Recently Blagojević and Duxbury[23,24] derived the results of Bartelt et al.[17,18] by considering various forms of a diffusion kernel. They also were able to apply this analysis to a problem of a vicinal array of steps. In all this previous theoretical work,[15–23] either (i) the analysis has been done for an isolated single step or (ii) step-diffusion-limited kinetics, surface-diffusion-limited kinetics, detachment-limited kinetics, and evaporation-limited kinetics have not been treated together, in a self-consistent manner. We present here a unified view of these four types of kinetics and apply it to multiple steps. In Sec. II we present the derivations for the step-edge velocity in two different configurations: an isolated step and a vicinal array of equidistant steps. In Sec. III we explore the implications of the formal results, cataloguing numerous special limits [including new ones such as (v) in case **A** and (ii) in case **D**]. We also consider carefully the crossover behavior between some of the limiting cases with the goals of assessing the size (in parameter space) of the limiting regimes and the rapidity of the crossover. In Sec. IV we present some general comments, connections with the work of Refs. 20, 23, and 24 and a brief summary and tabulation of limiting cases. Appendix A discusses an alternate formalism to the conventional development given in Sec. II: carrier exchange with the terrace and with the step-edge occur with independent kinetic coefficients. Many of the results derived in the body of the paper can be readily transcribed to this alternative approach, particularly those involving just one or the other of these transport avenues; other implications are left for future papers. Appendix B deals with the case of two adjacent steps, Appendix C lists some mathematical details about the temporal scaling of the mean square step-edge fluctuations, and Appendix D connects some of the notations that are prevalent in the literature. The reader interested only in the results may skip Sec. II.

II. DERIVATIONS FOR THE STEP-EDGE VELOCITY

A. Isolated step—conventional formulation

Consider an isolated step on a surface that undergoes random fluctuations of its position. We assume that the fluctuations are entirely due to the concentration field of adatoms on the pair of terraces adjoining the step, i.e., we neglect any external driving force causing net translation of the step or any step-step interactions that are not brought about by this concentration field. The surface is defined by the function $z = z(x,y,t)$ at time t. The step position at time t is a curve of discontinuity $x_s(y,t)$ on this surface such that the surface position is $z = h$ for $x(y,t) < x_s(y,t)$ and $z = 0$ for $x(y,t) > x_s(y,t)$, where h is the height of the step, assumed to be monatomic. Thus, the step position is given by $x_s(y,t)$, with $\langle x_s(y,t) \rangle_{y,t} = 0$, where $\langle \cdots \rangle_{y,t}$ signifies a spatial (along the step edge) and temporal average. The lattice constants

parallel and perpendicular to the step, in the $\hat{y}$ and $\hat{x}$ directions, are $a_{\parallel}$ and $a_{\perp}$, respectively. The coarse-grained free energy functional[15,21,25,26] of the step edge is

$$F[x_s(y)] = \int_{-L/2}^{L/2} [1 + (x_s{}'(y)^2)]^{1/2} \beta(\theta) dy, \qquad (1)$$

where the integral is over the entire step-edge position given by $x_s(y)$, which runs a distance L in the $\hat{y}$ direction. The prime after the x indicates a derivative with respect to y. The square brackets indicate that the free energy is a *functional* of $x_s(y)$. To avoid clutter in the notation, we henceforth drop the subscript s, denoting the step position simply by $x(y,t)$. The step free energy per unit length [of the step] is denoted by $\beta(\theta)$; it is a function of the local orientation θ of the step edge (i.e., the direction of the normal in the terrace plane), which in turn is given by

$$\theta = (\pi/2) + \tan^{-1} x'(y). \qquad (2)$$

Thus, if the step is perfectly straight, $\theta = \pi/2$. The chemical potential μ_s of the step[15,21,25] is given by

$$\mu_s = \left(\frac{\delta F}{\delta x} \right) \left(\frac{\delta N}{\delta x} \right)^{-1}, \qquad (3)$$

where N is the total number of atoms of the upper terrace minus that when $x(y) \equiv 0$ or in other words it is the number of atoms involved in causing the fluctuation $x(y)$ of the step edge from its mean position of $x(y) \equiv 0$. Hence,

$$N = \int_{-L/2}^{L/2} \frac{x(y)}{\Omega} dy, \qquad (4)$$

where Ω is the surface unit cell area (i.e., in this context the two-dimensional volume of surface mass carriers, namely adatoms and vacancies) involved in mass transport. T is the absolute temperature and k_B is Boltzmann's constant.

We make the linear kinetic assumption[20,27,28] that the velocity of the step is linearly proportional to the change in the chemical potential from its equilibrium value near the step edge. Here the change in chemical potential on both sides of the terrace contributes to the velocity. This is given by

$$\dot{x}(y,t)|_{\pm} = k_{\pm} \left[\frac{\mu(0^{\pm},y) - \mu_s}{k_B T} \right], \qquad (5)$$

where k_- (k_+) is the kinetic coefficient for mass exchange with the upper (lower) terrace,[20,27–29] which causes a step-edge velocity $\dot{x}(y,t)|_{-(+)}$. The dot indicates a partial derivative with respect to time. Strictly speaking the chemical potential on the terraces $\mu(x,y)$ appearing in Eq. (5) has explicit time dependence and obeys the diffusion equation.[20–22,27] In a linear approximation the chemical potential is simply related to the concentration of mass carriers involved in the mass transport [cf. Eq. (3) of Ref. 27]: If $c(x,y)$ is the concentration of the mass carriers on the terraces which takes an equilibrium value c_{su} far away from the step edge, then we have to linear order $\mu(x,y)/k_B T = (c(x,y) - c_{su})/c_{su}$.

To solve the diffusion equation, we need an initial condition and boundary conditions for $\mu(x,y,t)$. The boundary

condition is given by the linear kinetic assumption of Eq. (5). As can be seen from the left-hand side of this equation, the step velocity has an explicit time dependence in it. That implies that the boundary conditions on the $\mu(x,y,t)$ are themselves time dependent. Thus, we have to solve self-consistently for the time dependence of the step edge as well as the chemical potential of adatoms on the terraces. This problem, which is difficult (and perhaps impossible) to solve analytically, is also called the classical Stefan problem.[22,30] Following previous work,[20,21,27] we ignore the explicit time dependence of $\mu(x,y,t)$ [i.e., $\mu(x,y,t) \rightarrow \mu(x,y)$], and thus work in the quasistatic approximation. (The range of validity of this common approximation has recently been discussed.[31]) Our problem then reduces to solving the Laplace equation for $\mu(x,y)$:

$$\vec{\nabla}^2 \mu(x,y) = 0. \tag{6}$$

Since the mass conservation is not explicitly present in this Laplace equation, we must account for it separately. The incoming flux at a given point on the step edge consists of a normal component (i.e., along the unit normal in the x direction $\mp \hat{x}$) coming from the upper and the lower terrace and another contribution from motion along the step edge (i.e, along the unit normal in the y direction $\pm \hat{y}$). The two terms on the right-hand side of Eq. (7) describe the flux from the edge and the terrace, respectively. [For notational simplicity, we have assumed—rather unphysically—the same D_{st} on both the upper and the lower side of the step edge. The better approximation of $D_{st} \neq 0$ just on the lower (upper) side for adatom (vacancy) transport leads to more complicated equations with no gain in physical insight.] The left-hand side is the step-edge velocity which is caused by this incident flux. Thus, mass conservation at the step edge leads to the equation

$$\dot{x}(y,t)|_{\pm} = \left(\frac{1}{k_B T}\right)\left[a_\perp D_{st}\mu''(0^{\pm},y) \mp D_{su}\frac{\partial\mu(0^{\pm},y)}{\partial x}\right]. \tag{7}$$

D_{st} is the diffusion constant of atoms moving only along the step edge and D_{su} is the diffusion constant of atoms on the terrace.[29,32–35] Now eliminating the step-edge velocity from Eqs. (5) and (7), we get the boundary condition for $\mu(x,y)$ on the adjoining terraces. This is given by

$$a_\perp D_{st}\mu''(0^{\pm},y) \mp D_{su}\frac{\partial\mu(0^{\pm},y)}{\partial x} = k_\pm[\mu(0^{\pm},y) - \mu_s]. \tag{8}$$

[In Appendix A, we consider an alternative to Eqs. (5), (7), and (8), in which different barriers and kinetic coefficients are associated with terrace and with step-edge diffusion.] The step-edge velocity is now given by

$$\dot{x}(y,t) = \dot{x}(y,t)|_+ + \dot{x}(y,t)|_-. \tag{9}$$

Now to solve the Laplace's Eq. (6), the only quantity still to be computed explicitly is the equilibrium chemical potential μ_s at the step. Thus, Eqs. (1), (2), (3), and (4) yield the exact result

$$\mu_s = -\Omega\tilde{\beta}(\theta)x''[1+(x')^2]^{-3/2}, \tag{10}$$

where $\tilde{\beta}$ is defined by

$$\tilde{\beta} \equiv \beta(\theta) + \frac{\partial^2\beta(\theta)}{\partial\theta^2}. \tag{11}$$

For $x' \ll 1$, we approximate $\tilde{\beta}(\theta) \approx \tilde{\beta}(\pi/2)$. Computing μ_s to linear order in x' we get

$$\mu_s = -\tilde{\beta}(\pi/2)\Omega x''. \tag{12}$$

Henceforth, we will omit the argument of $\tilde{\beta}$, assuming it to be $\pi/2$. We now solve Laplace's equation for the chemical potential on the two adjoining terraces to the step. We assume periodic boundary conditions $\mu(x+L,y) = \mu(x,y)$ and $x(y+L,t) = x(y,t)$. We use along with Eq. (6) the boundary conditions of Eq. (8), and the expression for μ_s of Eq. (12) to get the solution for $\mu(x,y)$. This is then used in Eq. (7) and the step-edge velocity is obtained using Eq. (9). Performing these calculations we get[34,36]

$$\dot{x}(y,t) = -\sum_q \frac{x_q(t)e^{iqy}}{\tau_q}. \tag{13}$$

Here $qL/2\pi = \pm 1, \pm 2 \ldots$, and $x_q(t)$ is given by $x(y,t) = \sum_q x_q(t)\exp(iqy)$. The sum over q is truncated by the condition that $|q| \leq |q|_{\max}$, where $|q|_{\max} \approx 2\pi/a_\parallel$ and $1/\tau_q$ is

$$1/\tau_q = (1/\tau_q^+) + (1/\tau_q^-), \tag{14}$$

where the relaxation times for fluctuations caused by a flux of carriers from the upper $(-)$ and lower $(+)$ side are

$$1/\tau_q^\pm \equiv Sk_\pm q^2\left(\frac{a_q^\pm + b_q^\pm}{1 + a_q^\pm + b_q^\pm}\right). \tag{15}$$

We have used the notation

$$S \equiv \Omega\tilde{\beta}/k_B T \tag{16}$$

for the reduced stiffness, which has dimensions of length, while

$$a_q^\pm \equiv D_{su}|q|/k_\pm, \quad b_q^\pm \equiv D_{st}a_\perp q^2/k_\pm \tag{17}$$

are dimensionless measures of the terrace diffusion and the step-edge diffusion relative to the attachment/detachment rate. Note that $qL/2\pi \neq 0$ because we consider only fluctuations of the step-edge position from its mean position, which was already taken to coincide with the y axis.

To take into account the stochastic nature of the step-edge fluctuations, we add a noise term to Eq. (13). We then have

$$\dot{x}(y,t) = \mathcal{F}[x(y,t)] + \zeta(y,t), \tag{18}$$

where $\mathcal{F}[x(y,t)]$ is a functional of $x(y,t)$, which takes the operational form $-\sum_q \tau_q^{-1}x_q(t)\exp(iqy)$. We will see in Sec. III special cases where $\mathcal{F}$ takes simple forms. Taking the Fourier transform of the step-edge velocity, one gets

$$\dot{x}_q(t) = -\frac{x_q(t)}{\tau_q} + \zeta_q(t). \tag{19}$$

The definition of $\zeta_q(t)$ is given by $\zeta(y,t) = \Sigma_q \zeta_q(t) \exp(iqy)$, with $qL/2\pi = \pm 1, \pm 2 \ldots$. The restrictions that $x_{-q}(t) = x_q^*(t)$ and $\zeta_{-q}(t) = \zeta_q^*(t)$ guarantee that $x(y,t)$ and $\zeta(x,t)$ are real. We take the noise so that $\langle \zeta_q(t) \rangle_t = 0$, for each q, and its two-point correlations are given by

$$\langle \zeta_q(t) \zeta_{-q'}(t') \rangle = f_q \delta_{q,q'} \delta(t-t'). \tag{20}$$

The function f_q is as yet undetermined. From Eqs. (19) and (20), it follows[37] that

$$\langle |x_q(t)|^2 \rangle \equiv \langle x_q(t) x_{-q}(t) \rangle = (f_q \tau_q / 2)(1 - e^{-2t/\tau_q}). \tag{21}$$

We now determine the equilibrium value of these fluctuations in order to determine f_q. Equation (1) can be written to second order in x' following Ref. 26, then expanded in the Fourier modes x_q to obtain the equilibrium fluctuations of $|x_q|^2$:

$$\langle |x_q(t)|^2 \rangle = \frac{k_B T}{\tilde{\beta} L q^2}. \tag{22}$$

Hence taking the limit $t/\tau_q \to +\infty$ in Eq. (21) and comparing with Eq. (22), we get for consistency that

$$f_q = \frac{2\Omega}{\mathcal{S} L \tau_q q^2}. \tag{23}$$

This determines the two-point correlations of the noise term in Eq. (18) completely, through Eqs. (19) and (20), because f_q is now known.

B. Infinite array of steps

We now turn our attention to analyzing the fluctuations of an array of steps. This scenario is more likely to be encountered experimentally than that of an isolated step and hence presents a direct motivation for the study. We shall only consider steps and terraces that are identical in terms of their equilibrium crystal structure. The simplest generalization of the already studied case of an isolated step, namely that of two adjacent steps, is relegated to Appendix B, because it is rather unphysical. Here we consider an infinite array of steps, running on average along the direction $\hat{y}$. The outward normals to all terrace planes point along the positive z axis. The chemical potential of step i, is μ_{si} and is given by Eq. (12). The chemical potential on the terrace bounded by steps i and $i+1$ is $\mu_{i,i+1}$. The geometry is shown in Fig. 1. Each pair of nearest-neighbor steps is separated by a mean distance ℓ. Thus, the mean position at the nth step is given by $x(y) = n\ell$, where $n = 0, \pm 1, \pm 2, \ldots$. *We denote the fluctuations around these mean positions by* $x_n(y,t)$ *for the nth step, so that* $\langle x_n(y,t) \rangle_{y,t} = 0$. In analogy with the case of the single isolated step, the step-edge velocity similar to Eq. (5) is given by

$$\dot{x}_n(y,t) = k_+ \left[\frac{\mu_{n,n+1}(x,y) - \mu_{sn}}{k_B T} \right]_{x=n\ell}$$

$$+ k_- \left[\frac{\mu_{n-1,n}(x,y) - \mu_{sn}}{k_B T} \right]_{x=n\ell}. \tag{24}$$
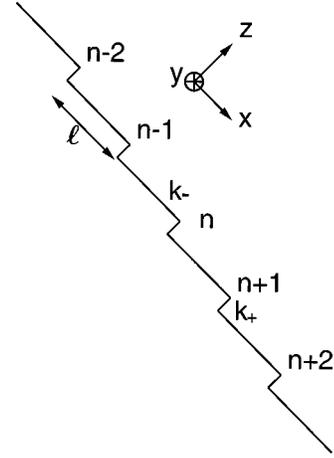
FIG. 1. Schematic representation of an infinite array of steps, illustrating our choice of axes and notation.

The chemical potentials $\mu_{n,n+1}(x,y)$ obey the Laplace equation [Eq. (6)]. This equation is to be solved with the two boundary conditions, similar to that of Eq. (8) for the isolated step case,

$$a_\perp D_{st} \mu''_{n,n+1}(n\ell^+, y) + \frac{D_{su} \partial \mu_{n,n+1}(n\ell^+, y)}{\partial x}$$

$$= k_+ [\mu_{n,n+1}(n\ell^+, y) - \mu_{sn}], \tag{25}$$

and

$$a_\perp D_{st} \mu''_{n,n+1}((n+1)\ell^-, y) + \frac{D_{su} \partial \mu_{n,n+1}((n+1)\ell^-, y)}{\partial x}$$

$$= k_- [\mu_{n,n+1}((n+1)\ell^-, y) - \mu_{sn+1}]. \tag{26}$$

Since all steps are now equivalent, we will just consider the step with $n=1$ and its neighbors on the two sides, $n=0$ and $n=2$. The Laplace Eq. (6) for the chemical potentials μ_{12} and μ_{01} may be solved with the boundary conditions of Eqs. (25) and (26). These solutions may then be substituted into the Eq. (24) for the step velocity. The result is given by[34,36]

$$\dot{x}_1(y,t) = -\sum_q \{A_q x_{1q}(t) - B_q [x_{0q}(t) + x_{2q}(t)]\} q^2 e^{iqy}, \tag{27}$$

where we have defined

$$A_q \equiv \mathcal{S}(k_+ I_q^- + k_- I_q^+)$$

and

$$B_q \equiv \mathcal{S}(k_+ a_q^+ + k_- a_q^-)/[2E_q \cosh(|q|\ell)]. \tag{28}$$

The definitions of E_q, $I_q^\pm(t)$ are

$$E_q \equiv [a_q^+(1+b_q^-) + a_q^-(1+b_q^+)]$$
$$+ [a_q^+ a_q^- + (1+b_q^+)(1+b_q^-)] \tanh(|q|\ell); \tag{29}$$

$$I_q^\pm \equiv \frac{1}{E_q} \{a_q^\mp(1+2b_q^\pm) + [a_q^+ a_q^- + b_q^\mp(1+b_q^\pm)] \tanh(|q|\ell)\}. \tag{30}$$

We make the further definitions

$$x_\Sigma(y,t) \equiv \sum_{n=-\infty}^{\infty} x_n(y,t)$$

and

$$x_\Delta(y,t) \equiv \sum_{n=-\infty}^{\infty} (-1)^n x_n(y,t); \tag{31}$$

and

$$w^2(t) \equiv \langle (x(y,t) - x(y,0))^2 \rangle, \tag{32}$$

as the mean-square fluctuation of each step. The combinations $x_\Sigma(y,t)$ and $x_\Delta(y,t)$ describe the two extreme cases of in-phase/acoustic[27] and [fully] out-of-phase/optical motion of the steps in the array, i.e., $x_{n+1}(y,t) = \pm x_n(y,t)$, for all n, respectively. The two combinations x_Σ and x_Δ are the $\phi = 0$ and $\phi = \pi$ limits, respectively, of the transform $x_\phi(y,t) \equiv \sum_n \exp(in\phi) x_n(y,t)$ discussed recently in Ref. 31. It can be shown from the definitions (31) and Eq. (27) that[36,38]

$$\dot{x}_{\substack{\Sigma \\ \Delta}}(y,t) = -\sum_q (A_q \mp 2B_q) x_{\substack{\Sigma_q \\ \Delta_q}}(t) q^2 e^{iqy}. \tag{33}$$

III. DISCUSSION OF LIMITING CASES AND CROSSOVER

Evidently Eq. (27) [as well as Eq. (59) of Appendix B] describes a set of coupled differential equations. Rather than attempting to decouple these equations, we will content ourselves with describing the important limiting cases, and then the crossover between them. First we consider the limit $|q|\ell \to \infty$. In this limit we expect that each step behaves as an isolated step since all other steps are infinitely far away from it. Indeed, we find that in this limit $B_q \to 0$, so the set in Eq. (27) decouples and reduces to Eq. (13) of the isolated step, with $\mathcal{S}k_\mp q^2 I_q^\pm \to 1/\tau_q^\mp$.

Evaporation condensation (3dEC), case A of Ref. 19. So far we have neglected [3d] evaporation of the adatoms from the terraces. This may easily be incorporated in our derivation of Eq. (13). We now define $\Lambda_q \equiv \sqrt{x_s^{-2} + q^2}$, where x_s is the diffusion length given by $\sqrt{D_{su}\tau_e}$, and τ_e is the mean time for evaporation of an atom with surface diffusion constant D_{su}.[29,35] Now we need to replace Eq. (6) by[19,27]

$$[\vec{\nabla}^2 - x_s^{-2}]\mu(x,y) = 0. \tag{34}$$

All the boundary conditions remain the same as before. Finally, in Eqs. (13), (18), (27), and (33) [as well as Eq. (B1) of Appendix B and Eq. (58)], we can simply replace all $D_{su}|q|$ by $D_{su}|\Lambda_q|$ and $|q|\ell$ by $|\Lambda_q|\ell$. Note that no change should be made in the terms $D_{st}q^2$ or terms involving just q without a corresponding D_{su} or ℓ. In the limit where the steps are infinitely far apart, (i.e., $|\Lambda_q|\ell \to \infty$), Eq. (27) [and, later, Eq. (B1)] reduces to Eq. (13) with $|q|$ replaced by $|\Lambda_q|$ where appropriate. This is as expected since each step is now isolated from the others, and ℓ disappears from all the physically relevant expressions. This form of Eq. (13) provides a generalization of the velocity equations of Bonzel and Mullins[21] and of Cahn and Taylor[20] for the single isloated

step. Another alternative to this is Eq. (13) with the use of Eq. (A1) (with the replacement of all $D_{su}|q|$ by $D_{su}|\Lambda_q|$). We now consider five distinct sets of limits, for all of which we demand that there be no step-edge diffusion (i.e., $D_{st} = 0$), and that the diffusion length x_s be small (i.e., $|q|x_s \ll 1$ or equivalently $|\Lambda_q| \approx x_s$): (i) $D_{su}/(x_s k_\pm) \ll 1 \ll \ell/x_s$, (ii) $(x_s k_\pm)/D_{su} \ll 1 \ll \ell/x_s$, (iii) $\ell/x_s \ll 1 \ll D_{su}/(x_s k_\pm)$ such that $D_{su}\ell/(x_s^2 k_\pm) \gg 1$, (iv) $\ell/x_s \ll D_{su}/(x_s k_\pm) \ll 1$ and (v) $D_{su}/(x_s k_\pm) \ll \ell/x_s \ll 1$. Applying the limits in (i) to Eq. (13) (with $|\Lambda_q|$ replacing $|q|$ where appropriate), reduces it to[39]

$$\dot{x}(y,t) = 2\mathcal{S}(D_{su}/\tau_e)^{1/2} x''(y,t). \tag{35}$$

This is analogous to Eq. (7) of Ref. 19. With the limits in (ii), the form of Eq. (35) is unchanged except that $2(D_{su}/\tau_e)^{1/2}$ is replaced by $(k_+ + k_-)$. We note that this is the same as Eq. (38) of case B. (See below.) This should not be surprising since in both cases the kinetics is detachment limited ($D_{su}|\Lambda_q|/k_\pm \gg 1$); the only difference is that in the present case we use $|\Lambda_q| \approx 1/x_s$ while for deriving Eq. (38) we shall use the limit of $|\Lambda_q| = |q|$. The limits in (iii) are similar to those in (ii), except that we now apply them to steps close together. Hence Eq. (33) gives in these limits[39] $\dot{x}_{(\Delta,\Sigma)}(y,t) = \mathcal{S}(k_+ + k_-)x''_{(\Delta,\Sigma)}(y,t)$, which just suggests that even though the steps are close together ($|q|\ell \ll 1$) the terrace diffusion is so fast ($a_q^\pm|q|\ell \gg 1$) that each individual step fluctuation is just determined by the detachment kinetics, which is the rate-limiting step. It is as if the presence of the neighboring steps is not ''felt.'' With the limits in (iv) in Eq. (33), we get[39]

$$\dot{x}_\Sigma(y,t) = \mathcal{S}(\ell/\tau_e) x''_\Sigma(y,t)$$

and

$$\dot{x}_\Delta(y,t) = 4\mathcal{S}\left(\frac{k_+ k_-}{k_+ + k_-}\right) x''_\Delta(y,t). \tag{36}$$

The result for $x_\Sigma(y,t)$ in Eq. (36) is the result analogous Eq. (8) of Ref. 19, as expected. The result in $x_\Delta(y,t)$ however, is interesting in that the out-of-phase kinetics of the steps is determined by the ''parallel-resistor combination'' of $k_\pm$ [i.e., $1/(k_+^{-1} + k_-^{-1})$, or half the harmonic mean]. Even though the kinetics is diffusion limited, the $k_\pm$ appear in the equation for $x_\Delta(y,t)$. We now consider the limits in (v) in Eq. (33) to get[39]

$$\dot{x}_\Sigma(y,t) = \mathcal{S}\left[\frac{D_{su}(k_+ + k_-)}{\tau_e k_+ k_-}\right] x''_\Sigma(y,t)$$

and

$$\dot{x}_\Delta(y,t) = 4\mathcal{S}(D_{su}/\ell) x''_\Delta(y,t). \tag{37}$$

This limit was not considered in Ref. 19. The result for $x_\Sigma(y,t)$ is unique in that all three types of transport coefficients corresponding to terrace diffusion (D_{su}), evaporation (τ_e) and attachment/detachment at the step edge ($k_\pm$) appear in it. Comparing the limits in (iv) and (v), we see that the only difference is that $\ell \ll D_{su}/k_\pm$ in the former while $\ell \gg D_{su}/k_\pm$ in the latter. Not surprisingly one obtains the

result for $x_\Sigma(y,t)$ in Eq. (37) by just replacing ℓ with $D_{su}(k_+^{-1}+k_-^{-1})$ in Eq. (36). Similarly the equation for $x_\Delta(y,t)$ in Eq. (37) is obtained by replacing $k_+^{-1}+k_-^{-1}$ with ℓ/D_{su} in Eq. (37). One may now use $m=2$ and $A_2=2\mathcal{S}(D_{su}/\tau_e)^{1/2}$, $\mathcal{S}\ell/\tau_e$, $4\mathcal{S}k_+k_-/(k_++k_-)$, $\mathcal{S}D_{su}(k_++k_-)/(\tau_ek_+k_-)$, or $4\mathcal{S}D_{su}/\ell$ [extracted from Eqs. (35–37)] in Eq. (C4) of Appendix C to obtain the scaling of the mean square fluctuations $w^2(t)$, $w_\Sigma^2(t)$, and $w_\Delta^2(t)$ [all defined by Eq. (32)] at early times.

Isolated-step evaporation-condensation (EC), case B of Ref. 19. In this case the rate-limiting step for mass transport around an isolated step is the random attachment/detachment of adatoms (or vacancies) at the edge of the boundary. This happens in three distinct limits: (i) If periphery motion along the step edge is completely prohibited (i.e., $D_{st}=0$) and if the detaching (attaching) atoms or vacancies go to (come from) a reservoir of adatoms or vacancies on the terraces, then $a_q^\pm\gg1$. (ii) If terrace diffusion is completely absent (i.e., $D_{su}=0$) and if the detaching (attaching) atoms or vacancies from kink sites go to (come from) a one-dimensional reservoir of adatoms or vacancies along the step edge itself where they move rapidly, then $b_q^\pm\gg1$, and we still obtain the same result of Eqs. (38) and (39). A more careful discussion of this situation is provided in Sec. IV. We emphasize that this limit also gives q^2 kinetics even though there is no diffusion on the terraces ($D_{su}=0$). (iii) Finally, motion may be allowed along both the terraces and the step edge though detachment/attachment of the mass carriers is still the rate-limiting step in transport (i.e., $a_q^\pm\gg1$ and $b_q^\pm\gg1$). Each of these cases gives the identical simplification that the term of unity in the two denominators of Eq. (14) may be ignored, to get $1/\tau_q=2\mathcal{S}kq^2$, where we define $k\equiv(k_++k_-)/2$. Using this expression for τ_q in Eq. (18), we find $\mathcal{F}$ to be[39]

$$\mathcal{F}[x(y,t)]=2\mathcal{S}kx''(y,t) \tag{38}$$

with

$$\langle\zeta(y,t)\zeta(y',t')\rangle=4k\Omega\delta(y-y')\delta(t-t'). \tag{39}$$

Then in this limit the scaling of the mean-square fluctuations of Eq. (32) with time is given by letting $m=2$ and $A_2=2\mathcal{S}k$ in Eq. (C4). These are the same expressions as obtained by Bartelt *et al.*[17] Thus, our general formulation of the problem presented here falls in the case of detachment-limited kinetics considered in Ref. 17, as expected.

Isolated-step terrace diffusion (ISTD), case C of Ref. 19. In this case there is no step-edge diffusion ($D_{st}=0$). There are three possiblities one of which is that (i) the mass transport is rate limited by diffusion on both the upper and lower terrace ($a_q^\pm\ll1$). This limit is considered for an isolated step. Then the dominant term in the two denominators of Eq. (14) is the constant term of unity. This leads to $1/\tau_q=2\mathcal{S}D_{su}|q|^3$, which when used in Eq. (18) gives a nonlocal form of $\mathcal{F}$ given by[18,39]

$$\mathcal{F}[x(y,t)]=\frac{4D_{su}\mathcal{S}}{\pi}\int_{-\infty}^{\infty}\left[\frac{\partial^2 x(y'',t)}{\partial y''^2}\right]_{y'}g(y-y')dy' \tag{40}$$

with

$$\langle\zeta(y,t)\zeta(y',t')\rangle=8D_{su}\Omega g(y-y')\delta(t-t'), \tag{41}$$

where $g(y)\equiv(c^2-y^2)/(c^2+y^2)^2$ defines $g(y)$; c is a convergence factor on the order of a lattice constant, i.e., $c\approx a_\parallel$. Then the mean-square fluctuations are given by using $m=3$ and $A_3=2\mathcal{S}D_{su}$ in Eq. (C4). All these are the same expressions as obtained by Bartelt, Einstein, and Williams[18] Thus, our general formulation of the problem presented here falls in the case of terrace-diffusion-limited kinetics of Ref. 18, as expected. As an alternative to the limit (i) $a_q^\pm\ll1$ we may consider a case where there mass transport occurs only from one side of the step edge and is rate limited by diffusion on the other terrace, so that either (ii) $k_-=0$, $a_q^+\ll1$ or (iii) $k_+=0$, $a_q^-\ll1$. In both these cases as well we get the limit of Eq. (18) of the form of Eqs. (40) and (41) except that $2D_{su}$ is replaced by D_{su} in both equations and in the expression for $1/\tau_q$ above.

Diffusion from step to step (DSS), case D of Ref. 19. In this case we consider an infinite array of steps in which the steps are close to each other [i.e., $|q|\ell\ll1$, implying $\tanh(|q|\ell)\approx\sinh(|q|\ell)\approx|q|\ell$ and $\cosh(|q|\ell)\approx1$]. No step-edge diffusion is allowed ($D_{st}=0$). We now consider three distinct limits: The mass transport is terrace diffusion limited ($a_q^\pm\ll1$) such that (i) $a_q^\pm\ll|q|\ell\ll1$, or such that (ii) $|q|\ell\ll a_q^\pm\ll1$. Alternatively, mass transport is rate limited by detachment kinetics so that (iii) $|q|\ell\ll1\ll a_q^\pm$ in such a way that $a_q^\pm|q|\ell\gg1$. Limits (i), (ii), and (iii) here are reminiscent of the limits (v), (iv), and (iii), respectively, considered under case A, except that now $|q|$ replaces $1/x_s$. Taking the limits (i) of Eq. (33) gives[39]

$$\dot{x}_\Delta(y,t)=\left(\frac{4\mathcal{S}D_{su}}{\ell}\right)x_\Delta''(y,t)$$

and

$$\dot{x}_\Sigma(y,t)=-\mathcal{S}D_{su}^2\left(\frac{k_++k_-}{k_+k_-}\right)x_\Sigma''''(y,t). \tag{42}$$

The result for $x_\Delta(y,t)$ is analogous to Eq. (12) of Ref. 19, as expected. Unlike case B for an isolated step, here q^2 behavior results from terrace-diffusion-limited kinetics. The result for $x_\Sigma(y,t)$ is analogous to Eq. (44), discussed below. This behavior arises because mass is conserved for the step array as a whole in the terrace-diffusion-limited kinetics considered here. However, as opposed to the derivation of Eq. (44), we have in this case $k_-\neq0$, which appears in the prefactor. If we consider the limits in (ii) in Eq. (33), we get

$$\dot{x}_\Delta(y,t)=\frac{4\mathcal{S}k_+k_-}{k_++k_-}x_\Delta''(y,t)$$

and

$$\dot{x}_\Sigma(y,t)=-\mathcal{S}D_{su}\ell x_\Sigma''''(y,t). \tag{43}$$

This limit, analogous to limit (iv) in case A, also was not considered in Ref. 19. Interestingly, in this case as well, the result for $x_\Sigma(y,t)$ is analogous to Eq. (44) with even matching terms ($D_{su}\ell$) in the prefactor. If we apply the limits[39] in (iii) to Eq. (33) then we get identical results for $x_\Sigma(t)$ and

$x_\Delta(t)$ of the form $\dot{x}_{(\Delta,\Sigma)}(t) = \mathcal{S}(k_+ + k_-)x''_{(\Delta,\Sigma)}(t)$. These are identical to the results obtained in limit (iii) of case A leading to the same conclusions.

We thus see that the three distinct limits (i), (ii), and (iii), give the same q^2 kinetics for $x_{\Delta q}(t)$ whenever step-edge diffusion is disallowed ($D_{st}=0$) and the steps are sufficiently close together ($|q|\ell \ll 1$), albeit with different prefactors $4\mathcal{S}D_{su}/\ell$, $4\mathcal{S}k_+k_-/(k_++k_-)$, and $\mathcal{S}(k_++k_-)$, respectively. For $x_{\Sigma q}(t)$ we get q^4 whenever the kinetics is terrace-diffusion limited ($a_q^\pm \ll 1$) and q^2 kinetics whenever it is detachment limited in a way that $a_q^\pm|q|\ell \gg 1$. With $w_{\Delta,\Sigma}^2(t)$ defined by Eq. (32), one can use $m=2$ and $A_2 = 4\mathcal{S}D_{su}/\ell$ or $4\mathcal{S}k_+k_-/(k_++k_-)$ to get $w_\Delta^2(t)$, and $m=4$ and $A_4 = \mathcal{S}D_{su}^2(k_++k_-)/(k_-k_+)$ or $\mathcal{S}D_{su}\ell$ to obtain $w_\Sigma^2(t)$ from Eq. (C4), in the limits (i) and (ii) considered here. Though here as well as in cases A and B we have q^2 kinetics, the allowed motion of the mass carriers as well the rate-limiting process in mass transport are quite different, as is evidenced by the different expressions for A_2 in all these cases. Experimentally these cases may simply be distinguished by measuring not only the single-step fluctuations but also those of $x_\Sigma(y,t)$ and $x_\Delta(y,t)$.

Perfect Schwoebel effect terrace diffusion (PSTD), case E of Ref. 19. We now consider the limit of the perfect Schwoebel barrier[40] (i.e., $k_-=0$). No motion along the step edge is allowed ($D_{st}=0$) and the rate-limiting step in mass transport is terrace diffusion ($a_q^+ \ll 1$), and the steps are close to each other [$|q|\ell \ll 1$, implying $\sinh(|q|\ell) \approx |q|\ell$ and $\cosh(|q|\ell) \approx 1$]. Since $B_q \to 0$, the terms in $x_{2q}(t)$ and $x_{0q}(t)$ in Eq. (27) vanish: since neighboring steps now do not exchange any mass, the fluctuations of one should not depend on the fluctuations of the other when the fluctuations are small. The dependence on ℓ of the fluctuations in the position of the step just indicates that the step is interacting with a perfect reflector at a distance ℓ away on the lower terrace. With these results used in Eq. (27) for $x_1(y,t)$, we get

$$\dot{x}_1(y,t) = -\mathcal{S}D_{su}\ell x_1''''(t). \tag{44}$$

This equation shows that q^4 kinetics may be found in an array of steps even though the rate-limiting process in mass transport is terrace diffusion (with no mass transport along the step edge). [See case F for a different situation.] Now using $m=4$ and $A_4 = \mathcal{S}D_{su}\ell$ in Eq. (C4), we get the mean-square step fluctuations of the step as a function of time. Thus, just like the q^2 kinetics, this q^4 kinetics also does not support a unique microscopic mass transport mechanism. Though both the PSTD limit of case E and the PD limit of case F give the same scaling $1/\tau_q \sim q^4$ in q, there is one crucial difference. In the former case the prefactor depends on the interstep separation ℓ and in the later case it does not. This enables one, in principle, to measure these fluctuations in the same sample at regions of different local orientations. Also, independent checks need to be obtained for the remaining prefactor of $D_{su}\ell$ or $a_\perp D_{st}$, as the case may be. It is not necessary that there be a perfect Schwoebel barrier as considered here or that carrier motion be restricted to the step edge as in case F for q^4 kinetics to be observed. This is clear from the limits (i) and (ii) of case D, where the in-phase step kinetics in an array exhibits q^4 behavior even in the absence

of a Schwoebel barrier and with terrace motion of carriers allowed, as observed from Eqs. (42) and (43) for $x_\Sigma(y,t)$.

Periphery or edge diffusion (PD), case F of Ref. 19. We now consider the limit in which the motion of adatoms is allowed only along the step edge ($D_{su}=0$), (i) but is the rate-limiting step in mass transport ($b_q^\pm \ll 1$). Then Eqs. (13) and (27) [as well as Eq. (B1)] take identical forms, as expected since for perfect periphery diffusion the presence of an adjacent step should become irrelevant, at least in the limit of small fluctuations. The relaxation time is now given by $1/\tau_q = 2\mathcal{S}a_\perp D_{st}q^4$ and hence

$$\mathcal{F}[x(y,t)] = -2\mathcal{S}a_\perp D_{st}x''''(y,t), \tag{45}$$

with

$$\langle \zeta(y,t)\zeta(y',t')\rangle = 4a_\perp \Omega D_{st}\delta''(y-y')\delta(t-t'). \tag{46}$$

Then the mean square fluctuations are given by Eq. (C4) by using $m=4$ and $A_4 = 2\mathcal{S}a_\perp D_{st}$. These are again the same expressions as obtained by Bartelt et al.[17] Thus, our general formulation of the problem presented here falls in the case of edge-diffusion-limited kinetics, as discussed in Ref. 17, as expected. If the rate-limiting periphery motion is allowed only along the lower or upper terrace of the step edge then the limit (i) $b_q^\pm \ll 1$ is replaced by (ii) $k_-=0$, $b_q^+ \ll 1$ or (iii) $k_+=0$, $b_q^- \ll 1$, then the factor of 2 should be removed from Eq. (45) and from the expression for τ_q above; the 4 in Eq. (46) should be replaced by 2, as is found in Refs. 32 and 33, where they consider limit (ii).

Direct sublimation (3dS) from the step edge, case G. We now allow direct sublimation (condensation) of atoms [$3d3$] at the step edge into (from) the vapor. Equation (34) must then be written in $3d$, with $\mu = \mu(x,y,z)$. All the boundary conditions remain as before. This leads in Fourier space to the wave vectors q_z and q_x relating to q, the wave vector of fluctuations along the step edge by the equality $q_z^2 + q_x^2 = -q^2 - x_s^{-2} \equiv -\Lambda_q^2$. Then the solution of Eq. (13) remains essentially unchanged in form, and we just need to replace ($D_{su}|q|$) by ($D_{va}|q_z| + D_{su}|q_x|$), where D_{va} is the diffusion constant of atoms in the vapor.[35] Let Ω_v be the $3d$ atomic volume in the vapor. For notational clarity we identify our D_{va}/Ω_v with $D_v\rho_v$ of Eq. (A.7) of Ref. 19. In the thus-modified Eq. (13), we take the limits $D_{su}=D_{st}=0$ and $D_{va}|q_z|/k_\pm \ll 1$, so that in Fourier space we obtain a result analogous to that of Eq. (A.7) of Ref. 19 for an isolated step,[41]

$$\dot{x}_q(t) = -\mathcal{S}D_{va}|q_z|q^2 x_q(t). \tag{47}$$

The result of Eq. (47) is essentially the $3d$ equivalent (in Fourier space) of Eq. (40) of case C. Here the step kinetics is limited by diffusion in a saturated $3d$ vapor instead of a $2d$ vapor as in case C.

Crossover between some of the limits. To study the crossover between the cases B, C, and F of an isolated step, we define an effective exponent z_q by

$$z_q \equiv -\frac{\partial \log(\tau_q)}{\partial \log(|q|)}, \tag{48}$$
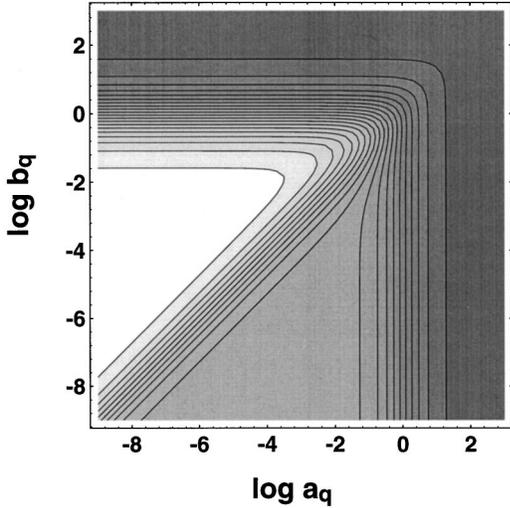
FIG. 2. Contour plot, with gray-scale shading, of the effective exponent z_q of Eq. (50) as a function of the logarithms of the dimensionless quantities related to the ratios of transport along steps and across terraces to the detachment rate: $\log_{10} b_q$ and $\log_{10} a_q$. To aid comparison with experimental numbers, all logarithms in this and subsequent figures are common. Use of logarithmic scales not only allows us to display a greater range of independent variables but corresponds to how the data is plotted in extraction of effective exponents. Note that plateaus at the limiting-case, integer values 2 (gray), 3 (light gray), and 4 (white)—indicative of EC of case B, TD of case C, and PD of case F, respectively—consume most of the parameter space; the crossover regions are rather narrow. Contour curves are spaced at intervals of 0.10 from 2.05 through 3.95.

referring back to Eq. (15). For simplicity of notation we define, $a_q \equiv a_q^+$, $b_q \equiv b_q^+$, $p_q \equiv a_q + b_q$. We denote by r the asymmetry in the attachment/detachment rates: $r \equiv k_- / k_+$. It is convenient to recast this asymmetry as an *effective Ehrlich-Schwoebel barrier* $\epsilon \equiv -k_B T \ln(r)$,[23,24,42] although this identification can be quite misleading if the extra barrier at the step edge is small compared to the thermal energy.[43] Then we get

$$z_q = 2 + \left[\frac{p_q + b_q}{p_q(1+p_q)} \right] \left[\frac{[r(1+p_q)]^2 + (r+p_q)^2}{(r+p_q)[2r+(1+r)p_q]} \right]. \quad (49)$$

In *both* of the special cases (i) no Schwoebel barrier,[40] $k_- = k_+$ (i.e., $r=1$, $\epsilon=0$) and (ii) perfect Schwoebel[40] barrier, $k_- = 0$ (i.e., $r=0$, $\epsilon=\infty$), the bracket on the right in Eq. (49) reduces to unity . Then z_q is simply

$$z_q = 2 + \frac{p_q + b_q}{p_q(1+p_q)} \equiv 2 + \frac{a_q + 2b_q}{(a_q+b_q)(1+a_q+b_q)}. \quad (50)$$

A contour plot of z_q of Eq. (50) as a function of a_q and b_q is shown in Fig. 2. When k_+ is small, i.e., a_q or b_q is large, we have a large plateau with $z_q = 2$ and electronic capture (EC)

behavior (case B). For $a_q \ll b_q \ll 1$, there is a plateau with $z_q = 4$, since terrace diffusion becomes unimportant and step-edge diffusion limits transport (case F). Likewise, for $b_q \ll a_q \ll 1$, there is a plateau with $z_q = 3$, since terrace diffusion limits transport (case C). The crossover regions are rather well confined and smooth, so that if one measures the effective exponent over a decade or so, it should have a well-defined value (i.e., the log-log plot should be linear) only if one is in one of the plateau regions. Conversely, if one finds an effective exponent other than 2, 3, or 4, the fits should not be good, and there should be indications of monotonic variation. With two or more decades of data, one reaches one of the three integer plateau regions. If any of these statements are inconsistent with the data, then either the experiment is flawed or the theory has left out some crucial ingredient of the system.

Between the two extreme values of r, the rightmost bracket in Eq. (49) does not reduce to unity, but nonetheless it turns out to be relatively unimportant. In Fig. 3(a) we display a contour plot of it as functions of r and p_q. For p_q larger than about unity, the term decreases smoothly [like $1 - (1 + p_q^{-1})r$, to leading order], to reach a minimum value of essentially $2(\sqrt{2}-1) \approx 0.828$, then rises smoothly again to unity [like $1 - [(1+p_q)/2](1-r)$, to leading order]. For large p_q the minimum occurs at about $r = \sqrt{2} - 1 \approx 0.42$ ($\epsilon/k_B T = 0.87$), but decreases smoothly [to $r \approx 0.32$ ($\epsilon/k_B T = 1.1$)] as p_q decreases to about 2, then turns down strongly to zero. Overall, the factor appears most significant for positive values of a_q and b_q, regions in which $z_q \approx 2$ (EC plateau), so that the r-independent factor is tiny.

To check what happens at smaller r, we show in Fig. 3(b) by gray scaling the difference in z_q produced by the r-dependent factor. The largest decrease is about -0.3, indicated by the darkest shading. Evidently for small [but not tiny] values of r the decrease occurs primarily in the region $-2 \lesssim \min(a_q, b_q) \lesssim 0$, which is in the crossover region between EC and the other two plateaus. (The details can be readily investigated but are not particularly enlightening.) The main conclusion is that the well-defined integer values should not be be affected significantly. From the contour lines in Fig. 3(b), we see that the principal effect of asymmetry is a modest decrease in the size of the plateau regions of PD and TD behavior, with crossover setting in for smaller values of b_q and a_q, respectively. For practical purposes, this role of asymmetry seems negligible.

To consider the crossover among cases B, C, D, and E we use the definition

$$z_q \equiv 2 + \left(\frac{\partial \log(A_q + 2B_q)}{\partial \log(|q|)} \right), \quad (51)$$

as applied to $x_{\Delta,q}(t)$ in Eq. (33), with the restriction $D_{st}=0$ ($b_q=0$). Even with this restriction, the expression is complicated. For compactness, we define $c_q \equiv \cosh(|q|\ell)$ and $s_q \equiv \sinh(|q|\ell)$. Then

$$z_q = 2 + \frac{2r[r(1+c_q)+(1+r)a_q s_q](s_q-|q|\ell) - 2ra_q^2(|q|\ell c_q + s_q) + (1+r^2)a_q^2(c_q s_q + |q|\ell)}{[(1+r)a_q c_q + (a_q^2+r)s_q][2r(1+c_q)+(1+r)a_q s_q]}. \quad (52)$$
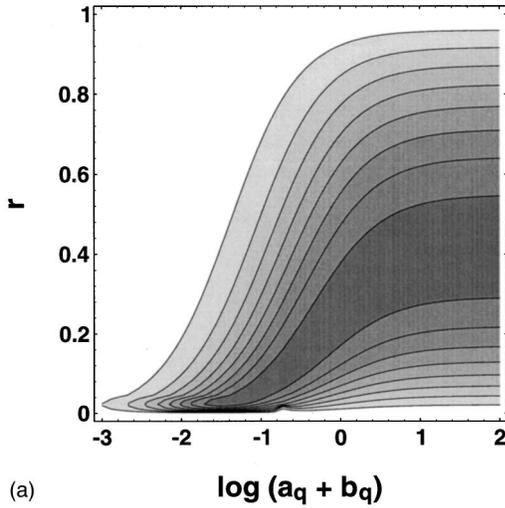
(a)
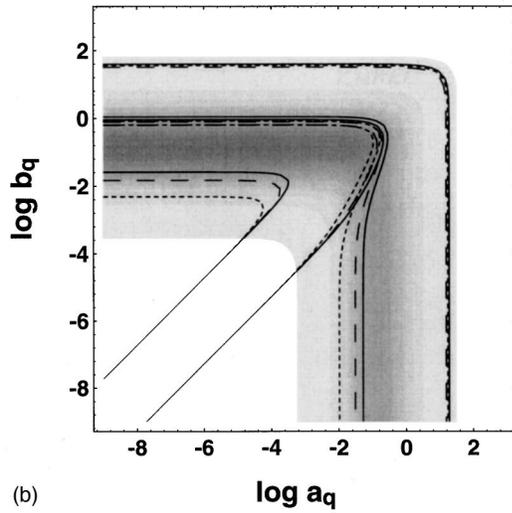
(b)

FIG. 3. (a) Contour plot, with gray-scale shading, of the multiplicative factor in brackets on the extreme right of Eq. (49). Note that this factor depends on a_q and b_q only through the sum $p_q \equiv a_q + b_q$. For most of phase space, this expression is nearly unity (indicated by white). Contour lines are at 0.98, 0.96, . . . 0.86, and 0.84. The minimum value of the factor is $2(\sqrt{2}-1) \approx 0.828$. See text for more details. The overall effect of the factor turns out to be negligible, as seen in (b). (b) Two distinct aspects of the reduction of z_q by the multiplicative factor plotted in (a). (i) Modification of contours of constant z_q of Fig. 2 due to the r-dependent factor. For clarity and emphasis, only the four contours bounding the crossover regions are depicted: $z_q = 3.95, 3.05, 2.95,$ and 2.05. The unbroken curves, taken directly from Fig. 2, are for $r = 1$. The long-dashed and short-dashed curves are for $r = 0.4$ and $r = 0.1$, respectively, in Eq. (49). (ii) The gray-scale shading indicates, for the particular value $r = 0.1$, the *difference* in the value of z_q due to the r-dependent factor, viz. z_q as given by Eq. (49) minus z_q as given by Eq. (50). White indicates essentially no difference. The darkest shading indicates about -0.29.

Analytically, we see that in the limit $|q|\ell \rightarrow \infty$ we recover the behavior for isolated steps: the terms in Eq. (52) that survive (besides, of course, the constant term of 2) are the terms in c_q^2, s_q^2, or $c_q s_q$. Then Eq. (52) reduces to Eq. (49) with $b_q = 0$. Furthermore, in this limit as can be seen from Eq. (49) the limit $a_q \gg 1$ leads to $z_q = 2$, while the limit $a_q \ll r$ gives $z_q = 3$, corresponding to cases B and C, respectively.[44]
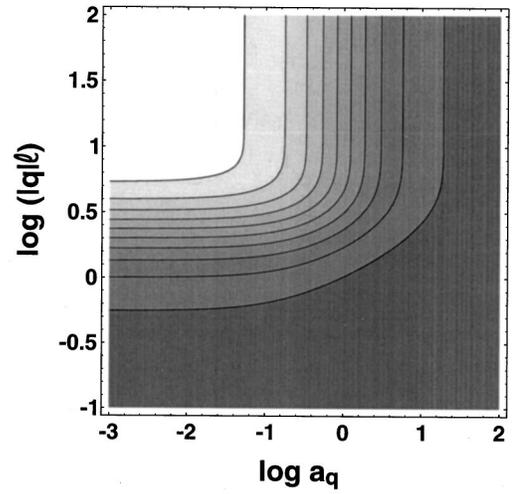
FIG. 4. Contour plot with gray-scale shading, of z_q illustrating the features of Eq. (52) for no step-edge transport ($b_q = 0$) and no Ehrlich-Schwoebel barrier ($r = 1$). The crossover region is relatively narrow. The value of $|q|\ell$ near the crossover is generally insensitive to a_q. The white plateau corresponds to case C behavior with $z_q = 3$. On the dark plateau $z_q = 2$. Contour curves are spaced at intervals of 0.10 from 2.05 through 2.95.

On the other hand in the limit $|q|\ell \ll 1$, we have $\sinh(|q|\ell) \approx |q|\ell$ and $\cosh(|q|\ell) \approx 1$, which reduces Eq. (52) to

$$z_q = 2 + \frac{2(1-r)^2 a_q^2 |q|\ell}{[(1+r)a_q + (a_q^2 + r)|q|\ell][4r + (1+r)a_q|q|\ell]}. \tag{53}$$

Note that in the limit $r = 1$, we get $z_q = 2$ in Eq. (53), corresponding to case D. In the symmetric case $r = 1$, Eq. (52) reduces to

$$z_q = 2 + \frac{\sinh(|q|\ell) - |q|\ell}{\sinh(|q|\ell) + a_q(\cosh(|q|\ell) + 1)}, \tag{54}$$

for which we show a contour plot of z_q as a function of a_q and $|q|\ell$ in Fig. 4. For $|q|\ell \gg 1$ and $a_q \ll 1$ there is a plateau at $z_q = 3$, corresponding to the TD behavior of case C. There is a smooth descent to $z_q = 2$, characteristic of EC, in the other three quadrants. The crossover occurs over roughly a decade along either axis. The two quadrants with $a_q \gg 1$ correspond to case B, while the remaining quadrant with $a_q \ll 1$ and $|q|\ell \ll 1$ is case D, in which the long-wavelength TD fluctuations on a step relatively close to its neighbors have the signature of EC fluctuations because a power of $|q|$ is surplanted by $1/\ell$. The general behavior seen in Fig. 4 holds until remarkably small r. Qualitatively, the descent from the plateau occurs at decreasing values of a_q and ripple develops on the lower plateau along the diagonal $a_q|q|\ell \approx 1$. For $r = 1/2$ ($\epsilon/k_B T = 0.69$), this ripple at its largest corresponds to $z_q \approx 2.05$. By $r = 0.1$ ($\epsilon/k_B T = 2.3$), this ripple has increased to $z_q \approx 2.5$ [see Fig. 5(a)]. Only for very small r does qualitatively new behavior occur. The ripple broadens and grows, and its center shifts to smaller values of a_q as r decreases. By $r \approx 10^{-5} - 10^{-6}$ ($\epsilon/k_B T = 11.5 - 13.8$), a plateau at $z_q = 4$ has formed in the quadrant $a_q \ll 1$ and $|q|\ell \ll 1$ (see Fig. 5(b)]. This region corresponds to case E. Referring to Eq. (53), we see that for $r = 0$, $a_q \ll 1$, we get $z_q = 4$ analyti-

(a)

log a_q

(b)

log a_q
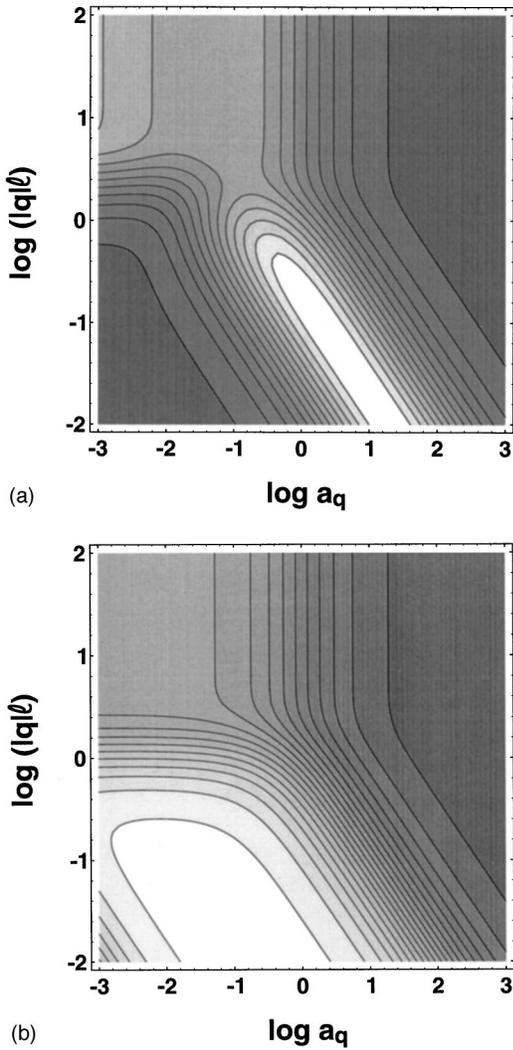
FIG. 5. (a) As in Fig. 4, contour plot of z_q illustrating the features of Eq. (52) for no step-edge transport ($b_q = 0$) but with a rather large Ehrlich-Schwoebel barrier ($r = 10^{-2}$). Contour curves are spaced at intervals of 0.10 from 2.05 through 3.25. A new ridge (white sliver, with $z_q \approx 3.3$) divides the plateau at $z_q = 2$. (For $r \approx 0.2$ this feature already appears—at somewhat larger a_q—as a modest "foothill.") The plateau at $z_q = 3$ (now darker than in Fig. 2 due to the gray-scale change necessitated by z_q above 3) has been pushed largely off the plot on the upper left, to smaller a_q; the last contour curve near the upper-left edge is for $z_q = 2.95$. (b) Similar to (a), but with asymmetry sufficient (here $r = 10^{-6}$) to produce behavior close to the "perfect Schwoebel barrier" (case E) limit of Pimpinelli *et al.*[19] Contour curves are spaced at intervals of 0.10 from 2.05 through 3.95. The new plateau at $z_q = 4$ (white region) is indicative of this extreme case. As $r \to 0$ this plateau extends to progressively smaller values of a_q. The plateaus associated with TD at $z_q = 3$ (light gray, upper-left quadrant) and with EC at $z_q = 2$ (darker gray, right side) are clearly seen.

cally. We recognize now, however, that such behavior will occur only for extremely small r, with virtually no attachment to steps from their upper side.

IV. COMMENTS AND SUMMARY

Remarkably, only the $|q|^3$ kinetics of case C is associated with a unique microscopic mass transport mechanism of

"slow" terrace diffusion $a_q^{\pm} \ll 1$ in an isolated step geometry. The other two limits of q^2 and q^4 kinetics are found in multiple cases such as cases A, B, and D for the former and cases D, E, and F for the latter, so that further information is needed to establish the transport mechanism. Two of the isolated-step cases, B and F, correspond to models A and B, respectively, of dynamical critical phenomena.[45] However, for case C we have found no simple correspondence[46] with these models.

In taking the limits considered in cases A through F, it is important to distinguish the physically allowed motion of the mass carriers from the rate-limiting process in the motion. Since there has been considerable confusion about this idea, we examine arguably the most striking example. We consider the limit $D_{su} = 0$ in Eq. (13), which implies that the motion of carriers is restricted to the periphery or edge of the isolated step. This restriction does not imply that the system exhibits behavior characteristic of case F: even though detachment onto the terrace is explicitly prohibited because $D_{su} = 0$, we can still get case B, i.e., EC. This result follows from Eq. (13) or the extrapolation of Fig. 2 to the limit $\ln a_q \to -\infty$, when the rate-limiting process is detachment kinetics, i.e., $b_q^{\pm} \gg 1$. On the other hand, if the rate-limiting process is diffusion along the periphery, i.e., $b_q^{\pm} \ll 1$, then we do get case F. Hence, the nomenclature EC or PD stands not for the allowed motion but for the rate-limiting process in the motion. In other words, the EC limit can also occur when the motion of carriers is only along the periphery and disallowed on the terraces. In this case motion along the step edge of an atom between detaching from a kink site and reattaching onto another kink site on the edge occurs "very fast." This may also be seen in terms of an effective "hop length" of the [mass] carriers, as we will see in the following paragraph. In general one may expect intermediate behavior between these two extreme limits. Such an analysis for the fluctuating boundary of adatom and vacancy islands has already been shown in Ref. 33. This intermediate case has recently been observed in the case of monolayer adatom island edges on Cu(001) and Ag(001) by Pai *et al.*[47] Earlier explanations[32,33,48] of the motion of large vacancy islands on Ag(111) suggested that mass transport in this system was terrace-diffusion limited. Recent evidence[49] indicates that the microscopic mass transport is restricted to the island periphery and that an explanation similar to the case of Cu(001) and Ag(001) adatom islands[47] may describe the motion more correctly. Similar distinctions between "allowed motion" and "rate-limiting motion" apply to the other limits of cases A, C, D, and E as well.

Recently, Blagojević and Duxbury[23,24] have described step motion and fluctuations from a new perspective. They have derived some of the limits we have considered in terms of the probability distribution $P(y)$ that an adatom emitted from a step will reattach to it a distance y away. Furthermore they have related parameters in the continuum theory such as D_{su} and $k_{\pm}$ to microscopic energy parameters, which describe the potential energy surface in which the mass carriers (adatoms or vacancies) move. We briefly show here some of the connections between their approach and the present work. In Eq. (13) we take the limits $D_{su} = 0$ and $k_- = 0$. We define the length $\xi \equiv \sqrt{a_{\perp} D_{st}/k_+} = q^{-1}\sqrt{b_q^+}$. The $q\xi$ depen-

dence of the qth component of Eq. (13) then becomes[50]

$$(q\xi)^2/[1+(q\xi)^2]$$

$$=\sum_{n=1}^{\infty}(-1)^{n+1}(q\xi)^{2n}$$

$$=\sum_{n=1}^{\infty}[(-1)^{n+1}(q\xi)^{2n}\Gamma(2n+1)]/[(2n)!], \qquad (55)$$

where the $\Gamma(z)$ is the gamma function, which for integer values of z reduces to $(z-1)!$.[53] Using this form we get

$$(q\xi)^2/[1+(q\xi)^2]$$

$$=-\xi^{-1}\int_0^{\infty}\sum_{n=1}^{\infty}(-1)^n[(2n)!]^{-1}(qy)^{2n}e^{-y/\xi}dy$$

$$=2\int_0^{\infty}[1-\cos(qy)]P(y,\xi)dy, \qquad (56)$$

where $P(y,\xi)\equiv(2\xi)^{-1}\exp(-y/\xi)$. Inserting Eq. (56) into Eq. (13) we see that its qth component is identical to Eq. (11) of Ref. 23 with the choice of $P(y)$ made above, which is also one of their special choices. Thus, we further identify our $\widetilde{\beta}$ as their $\widetilde{\Sigma}$, and our Ωk_+ as their Γ_{se} (their hopping parameter).

Consider now the alternative limit $D_{st}=0$ and $k_-=0$ (or $k_+=0$) of Eq. (13) so that

$$1/\tau_q=\mathcal{S}k_{\pm}q^2a_q^{\pm}/(1+a_q^{\pm})$$

$$=2\mathcal{S}k_{\pm}q^2\int_0^{\infty}(1-e^{-|q|x})P(x,D_{su}/k_{\pm})dx. \quad (57)$$

The $\pm$ signs in the equation correspond to the limits $k_{\mp}=0$. Now consider the corresponding limit in Eq. (28) of Ref. 23, which in their notation implies $\alpha_U=0$ (or $\alpha_L=0$). Using this limit in their Eq. (28) and substituting the result in their Eq. (11) we get Eq. (57) provided we replace $\exp(-|q|a_{\perp})$ by unity in their Eq. (28). Then we get the identifications as before that our $\widetilde{\beta}$ is their $\widetilde{\Sigma}$, and our $\Omega k_{\pm}=\Gamma_h$. Furthermore we get our D_{su} as their $(a_{\perp}^2\nu)\exp(-E_0/k_BT)$, our $k_{\pm}/D_{su}$ as their α_U^L and our Ω as their $a_{\perp}a_{\parallel}$. Duxbury[51] points out that the *ad hoc* replacement of $\exp(-|q|a_{\perp})$ by unity to match their result with Eq. (57) signifies one of the real differences between our approach and that of Ref. 23. He states that this difference originates because of the different ways of solving Laplace's equation on the terraces. In Ref. 23, an atom is placed an atomic spacing away from the step edge, and the diffusion is treated from that source. In our work and similar earlier treatments,[15,17,21,27,33,34] the diffusion is studied in the presence of a step which is perturbed from its original flat shape by an infinitesimal amplitude. The method of Ref. 23 then introduces new terms such as $\exp(-|q|a_{\perp})$, which are due to the distance an atom jumps when it detaches from a step edge. There are no such terms in our analysis or earlier work.[15,17,21,27,33,34] Though we have shown the similarity of our approach and that of Ref. 23 in the limit of no terrace diffusion ($D_{su}=0$), we have not been able to do so in the most general form of Eq. (13). As mentioned above[51,52] there is a real difference in the treatment of terrace diffusion in the

two approaches. Even if this were not the case, it is not clear how to show the equivalence of these two approaches for the most general case, when *neither* D_{su} nor D_{st} vanish.

An intuitive feeling for Eq. (56) is obtained simply. The mass transport of a step at a point is determined by the difference in chemical potential there and that at a point a distance $\pm y$ along the step edge. This difference in q space is proportional to $1-\cos(qy)$. We may think of the probability of having this difference at a distance y as given by $P(y)$. Again if $\xi\to\infty$ then $P(y)$ goes to a constant and the transport is just curvature $[x''(y,t)]$ driven (i.e., q^2 kinetics, case B). If instead $\xi\to a_{\parallel}$ then for $qa_{\parallel}\gg1$ the transport is driven by the second derivative of the curvature (i.e., q^4 kinetics, case F). Alternatively we may regard ξ as an effective hop length along the periphery of an atom after it detaches from a kink site on the step edge. Let us suppose that it may only attach to another kink site on the edge. Then $P(y)$ becomes the probability of finding a kink at a distance y along the step edge from a kink at the origin (i.e., at $y=0$). If we assume now that the probability of finding n kinks in a length ξ of the step is a Poisson distribution with a mean of unity,[54] then it would automatically lead to the form of $P(y)$ chosen by us. Similar arguments for a closed (circular) geometry can equally well be used to explain the noninteger exponents observed in the diffusion of large adatom islands[47] on Cu(001) and Ag(001) surfaces or of vacancy islands on Ag(111).[48,49] In the notation of Ref. 33, R_{st} would be the equivalent of ξ in a circular, closed geometry.[47] In that continuum theory,[33] this extra length scale [ultimately the denominator in Eq. (55)] can produce a noninteger scaling exponent (α), in contrast to simpler theories.

Table I lists the special cases addressed explicitly in this paper. Identical entries in the fourth and fifth column in a row indicate that the steps appear as ''effectively'' decoupled (since it implies $B_q/A_q\ll1$ in these limits) and are described by a single step equation, as in case E and in limits (iii) of cases A and D. However, in case E the interstep distance ℓ does enter as a prefactor, indicating the indirect effect of the neighboring steps.

We have shown in a unified picture how the various microscopic mass-transport mechanisms come into play in causing step-edge fluctuations of a single isolated step as well as those of steps in a vicinal array. We have considered special limits [including some new ones such as (v) in case A and (ii) in case D] where only certain types of mass transport are allowed and of these allowed ones a particular one is rate limiting. In these limits a scaling of the relaxation time τ_q of a fluctuation of wave number q (wavelength $\lambda=2\pi/|q|$) was found. There is, however, no scaling in general away from these limits, as is evidenced from the complicated forms of Eqs. (13) and (27). We have, however, studied the crossover between several of these limits.

In analyzing both the limiting behavior and the crossover, it has been convenient to rely on the dimensionless ratios $r=k_-/k_+$, $|q|\ell$, $a_q^{\pm}$, and $b_q^{\pm}$. In an actual experiment, one will certainly vary q and might also change ℓ or temperature. Since q is implicitly involved in the formation of the latter two dimensionless ratios, experimental trajectories will be complicated but manageable in the various figures describing crossover. Temperature manipulation produces a greater problem, since characterization of the thermal varia-

TABLE I. List of limiting cases discussed in the text. The first column describes the cases and the corresponding limits considered in Sec. III. D_{su} and D_{st} are the diffusion constants for motion along the surface (terrace) and the step edge, respectively (Refs. 29 and 35); $k_{\pm}$ is the kinetic coefficient for mass exchange between the step and the lower/upper terrace; and ℓ is the mean spacing between steps. These are defined more precisely in Sec. II (Refs. 29 and 35). The key dimensionless ratios are $|q|\ell$, where $|q|$ is the capillary wave number (wavelength $\lambda = 2\pi/|q|$), and $a_q^{\pm}$ and $b_q^{\pm}$, which are measures of the terrace diffusion and the step-edge diffusion, respectively, relative to the attachment/detachment rate, as defined in Eq. (17). Other parameters in cases A and G are defined in the appropriate subsection of Sec. III, and reduced stiffness S is defined by Eq. (16). The second column cites the relevant equation numbers for that case, with any modification of prefactors if necessary. The remaining columns indicate the determined prefactor and the exponent of $|q|$ of the scaling of τ_q^{-1}: The third column indicates the limit of τ_q^{-1}/Sq^2 from Eq. (13). The fourth and fifth columns indicate the limits of $(A_q \pm 2B_q)/S$ from Eq. (33), for the out-of-phase [optical] and in-phase [acoustic] combinations, respectively, of steps in an infinite array. Stars indicate the absence of relevant entries in that cell. As described in Appendix A, the table can be readily recast into the variables of the alternative formalism by making the appropriate replacement of $k_{\pm}$ by $k_{\pm}^{su}$ or $k_{\pm}^{st}$, with $a_q^{\pm}$ and $b_q^{\pm}$ being replaced by their primed counterparts.

Case: Limits	Equation	τ_q^{-1}/Sq^2	$(A_q+2B_q)/S$	$(A_q-2B_q)/S$
A (3dEC): $D_{st}=0$, $|\Lambda_q| \approx x_s$				
(i) $D_{su}/(x_s k_{\pm}) \ll 1 \ll \ell/x_s$	(35)	$2(D_{su}/\tau_e)^{1/2}$	$\star$	$\star$
(ii) $x_s k_{\pm}/D_{su} \ll 1 \ll \ell/x_s$	(35) w/ $2(D_{su}/\tau_e)^{1/2} \to k_+ + k_-$	$k_+ + k_-$	$\star$	$\star$
(iii) $\ell/x_s \ll 1 \ll D_{su}/(x_s k_{\pm})$	(36) w/ $4(k_+^{-1}+k_-^{-1})^{-1}$			
such that $D_{su}\ell/(x_s^2 k_{\pm}) \gg 1$	& $\ell/\tau_e \to k_+ + k_-$	$\star$	$k_+ + k_-$	$k_+ + k_-$
(iv) $\ell/x_s \ll D_{su}/(x_s k_{\pm}) \ll 1$	(36)	$\star$	$4(k_+^{-1}+k_-^{-1})^{-1}$	ℓ/τ_e
(v) $D_{su}/(x_s k_{\pm}) \ll \ell/x_s \ll 1$	(37)	$\star$	$4D_{su}/\ell$	$D_{su}(k_+^{-1}+k_-^{-1})/\tau_e$
B (EC): $a_q^{\pm} \gg 1$ or $b_q^{\pm} \gg 1$				
(i) $D_{st}=0$, $a_q^{\pm} \gg 1$	(38), (39)	$k_+ + k_-$	$\star$	$\star$
(ii) $D_{su}=0$, $b_q^{\pm} \gg 1$	(38), (39)	$k_+ + k_-$	$\star$	$\star$
(iii) $b_q^{\pm} \gg 1$, $a_q^{\pm} \gg 1$	(38), (39)	$k_+ + k_-$	$\star$	$\star$
C (ISTD): $D_{st}=0$				
(i) $k_-=0$, $a_q^+ \ll 1$	(40), (41) w/ $D_{su} \to D_{su}/2$	$D_{su}|q|$		
(ii) $k_+=0$, $a_q^- \ll 1$	(40), (41) w/ $D_{su} \to D_{su}/2$	$D_{su}|q|$	$\star$	$\star$
(iii) $a_q^{\pm} \ll 1$	(40), (41)	$2D_{su}|q|$		
D (DSS): $D_{st}=0$, $|q|\ell \ll 1$				
(i) $a_q^{\pm} \ll |q|\ell$	(42)	$\star$	$4D_{su}/\ell$	$D_{su}^2(k_-^{-1}+k_+^{-1})q^2$
(ii) $|q|\ell \ll a_q^{\pm} \ll 1$	(43)	$\star$	$4(k_+^{-1}+k_-^{-1})^{-1}$	$D_{su}\ell q^2$
(iii) $a_q^{\pm} \gg 1$	(43) w/ $4(k_+^{-1}+k_-^{-1})^{-1}$			
such that $a_q^{\pm}|q|\ell \gg 1$	& $D_{su}\ell q^2 \to k_+ + k_-$		$k_+ + k_-$	$k_+ + k_-$
E (PSTD): $k_-=0$, $D_{st}=0$				
& $|q|\ell \ll 1 \ll 1/a_q^{\pm}$	(44)	$\star$	$D_{su}\ell q^2$	$D_{su}\ell q^2$
F (PD): $D_{su}=0$				
(i) $b_q^{\pm} \ll 1$	(45), (46)	$2a_{\perp}D_{st}q^2$		
(ii) $b_q^-=0$, $b_q^+ \ll 1$	(45), (46) w/ $D_{st} \to D_{st}/2$	$a_{\perp}D_{st}q^2$	$\star$	$\star$
(iii) $b_q^+=0$, $b_q^- \ll 1$	(45), (46) w/ $D_{st} \to D_{st}/2$	$a_{\perp}D_{st}q^2$		
G (3dS): $D_{st}=D_{su}=0$				
& $D_{va}|q_z|/k_{\pm} \ll 1$	(47)	$D_{va}|q_z|$	$\star$	$\star$

tion requires an explicit microscopic model of the transport process. In our treatment, such a model is neither necessary nor even desirable; our approach is to extract transport coefficients from [other mesoscopic] experiments, so that there is no dependence on *a priori* insights. Likewise, in our formalism it does not matter whether transport is by adatoms (as is usually assumed implicitly) or by vacancies [as found recently[55] for Cu(001)]. Thus, many microscopic subtleties are transcended at this level.

The present theory is limited in several ways. It is linear in the fluctuations. It uses a small-slope approximation for the chemical potential. We have also neglected the possibly significant effect of step permeability. This phenomenon is

important in the relaxation of biperiodic gratings on Si(001), as demonstrated recently.[5] The possibility of atoms just crossing a step from one terrace to the next without being attached to the step edge has also been proposed earlier.[56] In Appendix A we have introduced a formalism that treats independently the mass exchange associated with terrace and with step-edge diffusion. Some transcriptions of the results in the body of our paper (in this alternative approach) can be readily written, while others remain for future publications. Generalization of our and related approaches may be taken in at least three possible directions: (i) Other linear physically relevant terms such as a constant force acting on the adatoms and vacancies on the terraces (such as may be thought of as

acting under the influence of a direct current being passed through the crystal under study[57]) or a linearized step-step interaction term.[2,17,31,58] (ii) Another generalization would be to consider the effect of large fluctuations through the exclusion of the small slope approximation[21] and including other nonlinear terms.[21,59] (iii) One may also relax the quasistatic assumption.[22] These and other generalizations, though studied to some extent,[2,17,21,22,58,59] have not yet been fully explored.

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APPENDIX A: ALTERNATIVE FORMALISM

The boundary condition expressed in Eq. (8) is not unique, even in a linear theory. In an alternative and arguably physically more appealing description, the kinetic coefficient $k_{\pm}$ is different for an atom detaching from a kink site onto a neighboring terrace and an atom detaching from a kink site onto a more mobile position but still along the step edge. On a microscopic level, there should be (and are, physically) different activation barriers associated with detaching from the step or just becoming mobile along it, with a resultant discontinuity in the chemical potential at the step edge. This picture is more general than that considered previously in Refs. 21, 33, and 34. We denote the two kinetic coefficients by $k_{\pm}^{su}$ and $k_{\pm}^{st}$, respectively. Then we get two equations to replace each of Eqs. (5), (7), and (8). To illustrate, Eq. (8) is replaced by the equations $a_{\perp}D_{st}\mu''(0^{\pm},y)=k_{\pm}^{st}[\mu(0^{\pm},y)-\mu_s]$ and $\mp D_{su}\partial\mu(0^{\pm},y)/\partial x=k_{\pm}^{su}[\mu(0^{\pm},y)-\mu_s]$. The discontinuity of $\mu(x,y)$ as $x\rightarrow 0$ requires the use of these *two* boundary conditions instead of the single one of Eq. (8). The form of Eq. (13) remains the same but with the replacement of Eq. (15) by

$$1/\tau_q^{\pm}\equiv Sq^2\left[\frac{k_{\pm}^{su}a_q'^{\pm}}{1+a_q'^{\pm}}+\frac{k_{\pm}^{st}b_q'^{\pm}}{1+b_q'^{\pm}}\right], \quad (A1)$$

with the definitions $a_q'^{\pm}\equiv D_{su}|q|/k_{\pm}^{su}$ and $b_q'^{\pm}\equiv a_{\perp}D_{st}q^2/k_{\pm}^{st}$. Eqs. (18)–(23) maintain their form under these changes. For the boundary conditions for an infinite array, Eqs. (25) and (26), similar modifications are then required. This reformulation entails the following changes in Eqs. (27)–(33) and Eq. (B1): (i) setting $b_q^{\pm}=0$, (ii) replacement of $a_q^{\pm}$ by $a_q'^{\pm}$, (iii) replacement of $k_{\pm}$ by $k_{\pm}^{su}$, and (iv) in the definition of A_q, addition of the term $Sk_{\pm}^{st}b_q'^{\pm}/(1+b_q'^{\pm})$.

Notice that in the limit $k_{\pm}^{su}=k_{\pm}^{st}=k_{\pm}$, Eq. (A1) does *not* simply reduce to Eq. (15); this is an alternative formulation rather than a mere extension of the conventional approach of

Sec. II. All the limits considered in Sec. III and Table I remain unaffected with the replacement of $k_{\pm}$ by $k_{\pm}^{su,st}$ as appropriate. As already discussed in case A of Sec. III, we can include the effect of evaporation from the terraces into the $3d$ vapor by just replacing $|q|$ in the definition of $a_q'^{\pm}$ by $|\Lambda_q|$ and also $|q|\ell$ everywhere by $|\Lambda_q|\ell$. In case G (only) we substitute for $k_{\pm}$ a third kinetic coefficient $k_{\pm}^{va}$ corresponding to the direct sublimation of an atom from the step edge into $3d$ vapor. Figures 4 and 5 remain unaffected, but with the understanding that $k_{\pm}^{su}$ and $a_q'^{\pm}$ replace $k_{\pm}$ and $a_q^{\pm}$, respectively. Figures 2 and 3 will change, as will their corresponding Eqs. (49) and (50). Equation (A1) [and the resulting Eq. (13)] serves as an alternative to the velocity equation derived by Bonzel and Mullins in the Appendix of Ref. 21.

APPENDIX B: TWO ADJACENT STEPS

As an intermediate case between single isolated steps and a vicinal array of steps one can consider two adjacent steps separated by a mean distance ℓ.[24,34] The mean position of the first step will be given by $x=0$ and that of the second one by $x=\ell$. The upper terrace of the first step extends to $x=-\infty$ from $x\approx 0$. The lower terrace of the second step extends to $x=+\infty$ from $x\approx\ell$. This scenario of two noninteracting steps separated by a fixed distance ℓ cannot occur in equilibrium. We present this case for comparison with a similar result of Ref. 24. One may write appropriate boundary conditions for this case and solve the problem as done in Ref. 34. However, the approach we follow here is to use the solution in the form of Eq. (27) of the problem for a vicinal infinite array of steps and then, specialize it to the case of two steps of the above-mentioned geometry. To do this we first observe that the 0th step has to be infinitely far away from the first step and the 3rd one has to be infinitely far away from the second step. This is simply achieved by letting $\ell\rightarrow\infty$ in the terms containing $B_qx_{0q}(t)$ or $k_-I_q^+$ in the equation for the first step and in the terms containing $B_qx_{3q}(t)$ or $k_+I_q^-$ in the equation for the second step. In this limit we get $Sq^2k_{\pm}I_q^{\mp}=1/\tau_q^{\pm}$ and $B_qx_{0q}(t)=B_qx_{3q}(t)=0$. We then get the equations for the two steps as[34,36]

$$\dot{x}_{\underset{2}{1}}(y,t)=-S\sum_q e^{iqy}\left\{\left[k_{\pm}I_q^{\mp}q^2+\left(\frac{1}{\tau_q^{\mp}}\right)\right]x_{\underset{2q}{1q}}(t)\right.$$
$$\left.-\left[\frac{D_{su}|q|^3}{E_q}\right]x_{\underset{1q}{2q}}(t)\right\}. \quad (B1)$$

Then limits can be taken as described in the text.

APPENDIX C

In this appendix we derive the early-time scaling relationship of the mean-square width $w^2(t)$ defined by Eq. (32), with time t. Such a scaling is possible only in cases where τ_q of Eq. (14) scales with $|q|$, so that we may write $\tau_q^{-1}=A_m|q|^m$, where $m=2,3,4$, and A_m is a constant. Using the Fourier expansion of $x(y,t)=\Sigma_q x_q(t)\exp(iqy)$, we may write

$$w^2(t) = \sum_q \sum_{q'} \langle [x_q(t) - x_q(0)][x_{q'}(t) - x_{q'}(0)]$$

$$\times \exp[i(q+q')y] \rangle. \qquad (C1)$$

Using this with Eqs. (20)–(23), we get

$$w^2(t) = \sum_q \langle |(x_q(t)|^2 \rangle = \frac{k_B T}{\widetilde{\beta} L} \sum_q \left[\frac{1 - \exp(-2tA_m|q|^m)}{q^2} \right], \qquad (C2)$$

In deriving Eq. (C2) we assumed that the step was initially straight and lying along the y axis, implying thereby that $x_q(0) = 0$ for all q. Taking the limits $tA_m/L^m \to 0$ and $a_{\parallel}^m/(tA_m) \to 0$, we get

$$w^2(t) = \frac{\Omega}{m\pi\mathcal{S}}(2tA_m)^{1/m} \int_0^\infty \left[\frac{1 - \exp(-x)}{x^{(m+1)/m}} \right] dx \qquad (C3)$$

which, after integration by parts, gives

$$w^2(t) = \frac{\Omega}{\pi} \left(\frac{2tA_m}{\mathcal{S}^m} \right)^{1/m} \Gamma\left(\frac{m-1}{m} \right), \qquad (C4)$$

where $\Gamma(z)$ is the gamma function[53] and $\Gamma(1/2) = \sqrt{\pi} = 1.77245...$, $\Gamma(2/3) = 1.3541...$, and $\Gamma(3/4) = 1.2254...$.[53] As an alternative to the definition of Eq. (32) we may use $w^2(t_1, t_2) = \langle [x(y,t_2) - x(y,t_1)]^2 \rangle$. Here as well it is assumed that the step is initially straight [i.e., $x(y,0) = 0$], and that $t_1, t_2 \to \infty$, so that the step fluctuations have reached equilibrium at these times. The time $t \equiv |t_2 - t_1|$ is assumed to be small. In this case Eq. (C2) is modified so that we replace T by $2T$ and $2A_m$ by A_m and make the identification of $w^2(t)$ with $w^2(t_1, t_2)$. Then the only modification in the final result of Eq. (C4) is that $2^{(1/m)}$ is replaced by 2.[23,60] This explains the apparent difference by a factor of $2^{3/4}$ between the results of Refs. 10 and 17.

APPENDIX D: NOTATIONS

The literature contains an unfortunate plethora of notations for identical quantities. We here connect our notation with some of that present in the previous literature. We denote symbols used in Ref. 32 by KB, Ref. 33 by KE, Ref. 21 by BM, Ref. 27 by BZ, Ref. 17 by BG, and Ref. 18 by BE. The symbols in the present work shall carry no superscript. We note that there is a subtle difference between the notation of KB and KE. First there are the identitical definitions in both papers $c_{su}^{(KB)} = c_{su}^{(KE)}$, $D_{su}^{(KB)} = D_{su}^{(KE)}$. However, $c_{st}^{(KB)}$ is the actual carrier density along the step edge, and $D_{st}^{(KB)}$ is the actual diffusion constant along the edge. In KE, $c_{st}^{(KE)}$ is defined as $[c_{su}a_{\perp}]^{(KE)}$ leading to $D_{st}^{(KE)} = [D_{st}c_{st}/(c_{su}a_{\perp})]^{(KB)}$. Thus $c_{st}^{(KE)}$ and $D_{st}^{(KE)}$ are mere definitions and should not be confused with the actual physical carrier concentration along the step edge which is $c_{st}^{(KB)}$ and the actual diffusion constant of these carriers along the step edge $D_{st}^{(KB)}$. Also to connect the present work with that of KB and KE we have the identifications $k_{\pm} = [c_{su}\Gamma_{\pm}]^{(KE)}$, $D_{su} = [D_{su}c_{su}\Omega]^{(KB)}$ and $a_{\perp}D_{st} = [D_{st}c_{st}\Omega]^{(KB)}$ or in other words with $\Omega = a_{\parallel}a_{\perp}$ we get $D_{st} = [D_{st}c_{st}a_{\parallel}]^{(KB)}$. Footnote 34 of KE also needs modification. We identify $\widetilde{\beta} = [h\zeta]^{(BM)}$. BM consider mass transport on both sides of the step in a single expression in their Eq. (A.10). This is also true of Eq. (6) of BG. It is therefore not possible to compare our $k_{\pm}$ with their notation. Still, we use here $k_- = 0$ for sake of comparison to get $k_+/(k_B T) = [m/\Omega]^{(BM)}$. They use $\Omega^{(BM)}$ as a three-dimensional volume while Ω is two dimensional. We identify $\Omega = [\Omega/h]^{(BM)}$, $D_{su} = [\Omega \nu D^t/h]^{(BM)}$, and $a_{\perp}D_{st} = [\Omega \nu a_0 D^s/h]^{(BM)}$. Similarly comparing our Eq. (38) (with $k_- \neq 0$) with Eq. (6) of BG we get $2k\Omega = \Gamma_a^{(BG)}$ and comparing our Eq. (45) with their Eq. (17) we get $2a_{\perp}\Omega D_{st} = [\Gamma_h]^{(BG)}$, with periphery diffusion along the lower and upper step edge being allowed. If it is allowed only on the lower step edge then $a_{\perp}\Omega D_{st} = [\Gamma_h]^{(BG)}$. BZ do not consider transport along the step edge so that they have no terms involving our D_{st}. Our $D_{su} = [D_s c_{eq}^0 \Omega]^{(BZ)}$, $k_{\pm} = [k_{\pm}c_{eq}^0 \Omega]^{(BZ)}$, $\tau_e = [\tau/(c_{eq}^0 \Omega)]^{(BZ)}$, and also $[c_{su}]^{(KB)} = [c_{eq}^0]^{(BZ)} = [c_0]^{BE}$. Similarly $D_{su} = [D_s c_0 \Omega]^{(BE)}$. For completeness we reiterate that our D_{va}/Ω_v is the same as $D_v \rho_v$ of Eq. (A.7) of Ref. 19.

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[39] In deriving limiting forms of Eqs. (13), (18), and (27) such as Eq. (35) and others, we have used inequalities and limits involving $|q|$. There are bounds on $|q|$ so that $|q|_{min} < |q| < |q|_{max}$, where $|q|_{max} \approx 2\pi/a_\parallel$ originating from the discreteness of the lattice and in the limit that the total length of the step diverges (i.e., $a_\parallel/L \to 0$) one gets $|q|_{min}=0$. Hence the sums over q are terminated on the positive (negative) side by by $+(-)|q|_{max}$. So to take Fourier transforms of equations in q space to get derivative(s) of y in real space as on the right-hand side of Eq. (35) and others is only an approximation and hence is defined here to mean only a formal representation for such truncated sums over q. Also, in deriving equations such as Eq. (38) we encounter limits of the form $a_q^\pm \gg 1$ and/or $b_q^\pm \gg 1$. To have these limits valid for all q we need to take the limit $Lk_\pm/(2\pi D_{su}) \to 0$ and/or $L^2 k_\pm/(4\pi^2 D_{st}) \to 0$ before taking the limit $a_\parallel/L \to 0$.

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[42] We define ϵ as an effective Ehrlich-Schwoebel barrier in our continuum picture. The connection of this to the discrete or microscopic barrier is explored by Blagojević and Duxbury (Refs. 23 and 24).

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[44] Note that $0 \le r \le 1$ generally holds. Hence $a_q \gg 1$ implies $a_q \gg r$ and $a_q \ll r$ implies $a_q \ll 1$. In the unexpected case that $1 < r$ we can do the same analysis in $1/r$ instead of r for which $0 \le 1/r \le 1$ would then be true. We note that all expressions for z_q maintain their form under the change from r to $1/r$, a_q to a_q/r, and b_q to b_q/r.

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