Equilibrium step dynamics on vicinal surfaces revisited

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The equilibrium theory for a train of steps is revisited. The analysis is based on a nonlocal Langevin equation derived from the Burton-Carbrera-Frank model. The Ehrlich-Schwoebel effect, diffusion along the step edge, as well as elastic interactions between steps have been incorporated. We discuss several static correlation functions and give an improved estimate for the terrace width distribution. By exploiting the dispersion relation, the time dependence of the step fluctuations has been calculated. In the limit of well separated length scales there are several time intervals where the temporal step correlation function follows a power law with one of the exponents 1/2, 1/3, or 1/4. In the opposite situation, neither power laws nor simple scaling behaviors are obtained. We provide precise conditions on which regime must be expected in a given real situation. Moreover, it is shown that different physical mechanisms can give rise to the same exponent. This study is thus crucial for the discrimination between various physical regimes in a real experiment. The range of validity of the approximation and the crossover times are discussed for steps on Si(111).

I. INTRODUCTION

One of the most active fields of research during the second half of this century has been surface science. This concerns in particular structural and dynamical features ranging from atomic scales to mesoscales. The advent of microscopic visualization techniques, such as scanning tunneling microscopy (STM) and reflection electron microscopy (REM), has opened a large avenue towards the identifications of static and dynamical features on atomic scales. The motivation is due both to potential technological importance in fabricating novel microscopic architectures and to the fact that the systems in question offer at the same time attractive examples of both equilibrium and nonequilibrium statistical mechanics.

Of particular interest are vicinal surfaces. These are obtained by cutting a solid in an orientation (often called misorientation) making some angle (ranging between a fraction of a degree and a few degrees) with respect to a close-packed face of a crystal. The resulting morphology, made of steps separated by terraces, is shown in Fig. 1. Besides the fact that it is often unavoidable to produce steps on a surface, stepped surfaces are often intentionally produced for several purposes.

For example, steps may play an active role as nucleation centers in order to produce layer by layer growth in a step-flow regime (steps advance, or flow, upon addition of particles diffusing on the terrace after they have landed from the incoming flux). Steps can also be a host for foreign particles that preferentially attach along the step, producing thereby a linear chain of atoms (quantum wires).

The first approach towards the identification of physical ingredients that are of vital interest in many static and dynamical surface processes is based on equilibrium statistical analysis. The first noticeable feature of the steps is their meander. The experimental study of step (and terrace width) fluctuations is of a paramount importance in order to access physical surface properties. For example, an isolated step with an extent $L$ wanders at equilibrium on average a distance proportional to $\sqrt{L}$ from its flat configuration. The static mean excursion of an isolated step is given by

$$w = \sqrt{\frac{k_B T L}{2 \gamma}}$$  

Here, $\gamma$ denotes the line stiffness of the step. The experimental study of this quantity leads to the determination of $\gamma$. Note that a similar expression to Eq. (1) is encountered for an ideal polymer chain where $L$ would be the polymer length (or the number of monomers). This is also a typical result for a Brownian motion where the spatial extent $L$ would be represented by the time interval during which the particle trajectory is analyzed. The static features do not depend on the precise transport mechanisms that are involved to create a fluctuation of some size. That is to say, Eq. (1)
will always hold regardless of the specific underlying dynamics responsible for the fluctuation.

This is obviously not the case for kinetic properties (though the system is in global thermodynamical equilibrium), such as how \( w \) behaves in the course of time. The mechanisms that control step fluctuations can be of disparate natures. On the general ground, any step motion is always accompanied with an atomic transport from one point to another. And it will turn out that, although the same atomic motion is under consideration, the (temporal and spatial) scales on which it operates will be decisive for ascertaining the type of behavior (e.g., the step roughness as a function of time) to be expected in an experimental situation. To fix ideas, once a typical scale is given (say that in an experiment one analyses fluctuations in some spatial range), the crucial point is to determine the relevant phenomenon when analyzing the fluctuation. For example, if an atom is strongly bound to the step and that thermal excitation cannot overcome the energy barrier corresponding to diffusion onto the terrace in the temperature range of interest, one expects that (virtually) all atoms move along the steps in order to build up a protrusion. In other limits atoms can make excursions on the terrace and ultimately desorb or visit another terrace. The main requirement is that the information that is extracted from a given experiment should be interpreted properly in that it has to be related to the appropriate dissipation mechanism. It might seem puzzling at this junction that the problem we are addressing, classical as it might appear, is still a source of conflicts and misinterpretations, as will be documented in the present paper.

On theoretical ground, an important step forward was made by Bartelt et al.,\(^3\) who have analysed the dynamical step fluctuations. Their analysis rests on the fact that the step motion is described by a local Langevin equation (relaxational equation) where the step Hamiltonian is composed of a line energy and an interaction energy with the neighboring steps (which are supposed to be rigid; this condition is relaxed here and leads to some modifications to be specified later). Among other results, their model produces for the terrace width variance \( W^2 \sim t^{1/2} \) or \( W^2 \sim t^{1/4} \). The first case holds when the step emits (receives) atoms onto (from) the terrace, whereas the second power law arises if atoms execute a diffusion along the step edge. More recently, Pimpinelli et al.\(^4\) have analysed the time needed to build a protuberance in several cases. They have drawn attention to the fact that the law \( W^2 \sim t^{1/2} \) can have a completely different origin, namely mass exchange between terraces mediated by fast diffusion on terraces. The question of specifying under which circumstance and when precisely a certain mechanism prevails over all the other possible ones needs, however, to be analysed. Moreover, as it will appear, it is not clear that a power-law behavior must always be expected.

In view of this unclear situation, it is important that one can, in a unified picture, develop a general Langevin formalism including attachment and detachment at the step, line diffusion, terrace diffusion, desorption, and elasticity. This will allow us to give precise conditions on whether or when a given physical regime is dominant. The general Langevin equation to be presented here is nonlocal both in space and time. In the present work a “local in time” approximation will be used, which is sufficient at equilibrium for all available experimental data.

A comprehensive connection with previous analyses and with experiments is presented. It will also emerge from this study that power-law behaviors are expected only if some typical length scales are quite different from each other. In the opposite limit, neither a power law nor a simple scaling behavior exists.

The scheme of this paper is as follows. In Sec. II we shall first describe and recall some general features and approximations used in previous analyses. In Sec. III we present the Langevin formalism. Section IV deals with static results, and points out some important features that seem not to be known in the literature. Section V is devoted to an extensive study of dynamical features. Section VI deals with comparison with experiments. A summary is given in Section VII. Details concerning the asymptotic evaluation of integrals are relegated to the Appendix.

**II. PRELIMINARY DISCUSSION**

The aim of this section is to specify some general conditions about the step-step interaction for the model to be presented extensively in the next section. We consider a train of steps on a vicinal surface below the roughening temperature of the terrace. The movement of steps is restricted by the noncrossing condition (the step crossing creates an overhang, which implies an additional energy cost) even in the absence of an explicit step-step interaction. This hinders the fluctuations of a step in the vicinity of another step leading to an entropy reduction, and hence costs free energy.\(^5\) This restriction of noncrossing appears to be one of the main difficulties precluding a full analytical approach. This is not surprising since the problem has a close similarity with a quantum many-body system. There have been theoretical attempts,\(^6\) however, to provide a systematic recursive approximations in order to handle entropic interactions.

For low temperatures the movement of noninteracting steps can be legitimately mapped onto a one-dimensional free fermion gas problem.\(^7\) The resulting distribution function of the terrace width is asymmetric, since it has obviously to vanish for negative step distances, but allows for very large terrace widths. Unfortunately, no analytical solution in closed form is available.

On the other hand, the steps interact via the elastic deformation of the crystal lattice and (possibly) through the dipole moments of the electronic charge distribution at the step edges. In a situation where the elastic stress tensor is different on both sides of a step [the case of Si(001) where the 1 \( \times 2 \) reconstruction occurs], the step-step interaction is expected (at least at large distance) to be logarithmic. The same result holds when a thin film is deposited on a substrate of a different nature. In that case, the steps are a location of forces (since the stress tensor on both sides of the step is different, the total force at the step must be nonzero in order to preserve equilibrium of a small element around the step).\(^7\) For a pure substrate, the step edges are locations of force dipoles. Both interactions (of elastic or electric nature) lead in this case to an \( A/\ell^2 \) dependence in the continuum limit (\( A \) being some constant). Analytical results are only available...
for two specific values of $A$, which have been used by Joóš et al.\textsuperscript{8} to calculate the terrace width distribution function.

One aim of this paper is to present a simple self-contained model with analytical solutions that can be used for estimating step-step interactions without the need for extensive calculations. The simplest model of this kind was introduced a long time ago by Gruber and Mullins.\textsuperscript{9} It consists of only one free step that is trapped between two fixed walls separated by two times the mean terrace width $l$. Here the condition that steps may not cross the walls (only entropic repulsion is present) has been solved. The distribution function reads

$$P(x) = \frac{1}{l} \cos^2 \left( \frac{\pi x}{2l} \right), \quad |x| \leq l. \quad (2)$$

Bartelt et al.\textsuperscript{10} have investigated this model intensively in the case of energetic (in contrast with entropic) step-wall interactions. They approximated the real potential by its harmonic expansion and neglected the entropic “repulsion,” i.e., allow the steps in principle to cross the walls with a small probability. This is justified as long as the interaction is strong enough or the temperature is low enough, so that the average meander of the step is restricted to a small strip of width $W \ll l$. The resulting distribution function is a Gaussian with variance $W$,\textsuperscript{3}

$$W_G(T, l) = \left( \frac{k_B T^2}{8 c(l) \gamma} \right)^{1/4}. \quad (3)$$

$c(l) = U''(l)$ is the second derivative of the interaction potential $U(l)$, $\gamma$ the line stiffness of a free step, and the index $G$ stands for the Gruber-Mullins model. For the elastic potential with $U(l) = A/l^2$, $c(l) = 6A/l^4$. This implies that the width $W$ of the Gaussian distribution is proportional to the mean terrace width $l$.

Experimentally determined terrace width distributions on Si(111) vicinal surfaces at 900 °C (Ref. 1) can be accurately fitted by a Gaussian, which may point to the fact that the energetic repulsion dominates the entropic one. On the other hand, for metals with a low adhesion energy [such as Cu and Ag (Refs. 11–13)], and hence weaker line stiffness and elastic interactions, the entropic repulsion cannot be neglected already at room temperature. The measured distribution is then slightly asymmetric [see, for example, Cu(1,1,n) (Ref. 14)].

It is possible to approximatively describe the entropic effect by an effective temperature-dependent potential that turns out to have the same $1/l^2$ behavior as the energetic one. This corresponds to a renormalization of the coefficient $c$ in Eq. (3). Note, however, that the two interactions do not simply add linearly.\textsuperscript{15–17} Furthermore, one has in principle to use the line stiffness for an interacting step that is larger than for a free one. Because of the weak elastic interaction, this effect is very small [only of the order of 3% for Cu(1,1,11) (Ref. 17)] and can legitimately be disregarded.

The main drawback of the Gruber-Mullins model is that only one step is allowed to meander. At least in the case of silicon, the other simplifications are not crucial as discussed above. We will see that the meandering of all steps leads to a variance $W$ that is 1.34 times larger than the value given by Bartelt et al.\textsuperscript{10,3} [Eq. (3)] An extraction of the value of the elastic coefficient $A$ using the approximation (3) then underestimates $A$ by a factor of about 3.

This effect is in principle hidden in the more detailed and complicated treatment of Barbier et al.\textsuperscript{17,18} based on the transfer-matrix method, but it has not been clearly realized that this stems from the fluctuations of the neighboring steps. As argued above, step-step interactions are important, and therefore it is useful to introduce two important length scales associated with interactions. They will serve later to specify the range of validity of different approximations. We first consider entropic repulsions. We use the analogy between the step meander and a Brownian particle, where the length along the step would be time. This idea leads to the determination of the so-called collision width,\textsuperscript{19}

$$l_{\text{coll}} = \frac{l^2 \gamma}{4 k_B T}. \quad (4)$$

This provides an order of magnitude of the average length between two consecutive step-step (if each were free) collisions. In the noninteracting case this quantity acts as the only characteristic length for spatial correlations in the direction parallel to the steps.

The elastic step-step interaction introduces another length, the elastic length $l_e$,

$$l_e = \sqrt{\frac{\gamma}{2 c(l)}}. \quad (5)$$

It can be interpreted (apart from a numerical factor of order 1) as the distance between step collisions along a virtual wall of height $W$ [Eq. (3)]. This can be seen by replacing $l$ by $W$ in Eq. (4). In a system with strong energetic interactions, $l_e$ would be the only correlation length along the steps. Both lengths are large compared to the mean terrace width $l$ (see later). They depend in the same way on $l$ ($c(l) = U''(l) = 6A/l^4$), but differently on temperature. Their comparison allows statements about the validity of our approach, which neglects the entropic repulsion. It must be kept in mind, however, that separations of the effects are not generally legitimate. Thus, in general, the correlations parallel to the step direction are described by a single length $l_{\text{corr}} = \min(l_e, l_{\text{coll}})$, which results from a combination of both the energetic and entropic interactions.

As discussed later, during the equilibration of the steps several characteristic times show up. These are associated with different length scales of the problem. Spatial step autocorrelations (due to direct interactions discussed above) on scales smaller than $l_{\text{corr}}$ (or temporal correlations at smaller times than their corresponding time scales) are obviously independent of step-step interaction, be they of entropic or elastic nature. In summary, the present description for the equilibration of steps is expected to be valid for not too high temperatures or for materials with strong interactions. These two requirements ensure that no “collision” is probable. On general ground, the present assumption will still be valid for short times even though entropic repulsion may be potentially important. That is to say, many dynamical exponents seen in experiments are independent of step-step interactions (entropic and elastic). Another kind of interaction will appear: this is the diffusive one, which plays an important role in many circumstances.
III. EXTENDED BCF MODEL

Usually step dynamics (at equilibrium) are treated using a simple step Hamiltonian description. A Langevin relaxation equation (the step velocity is a functional derivative of the Hamiltonian, plus a noise term) describes step dynamics. This equation is either of nonconservative (attachment and/or detachment at the step is the limiting factor for the motion) or conservative nature (line diffusion limits step fluctuation). In the general case atoms can also desorb, diffuse on the terrace, and ultimately visit neighboring terraces. It will emerge in this analysis that these situations are clearly met in many real situations. Thus a proper description of step fluctuations should incorporate those effects. That is, a step fluctuation is not only governed by what happens at the step, but also on the environment elsewhere. This means that steps are described by nonlocal equations. Furthermore, the step motion affects the adatom distribution whose response is not necessarily instantaneous. Thus, retarded interactions are involved. In short, after integrating out the diffusion field, the Langevin equation is in general nonlocal both in space and time.

The model is based on the Burton, Cabrera, and Frank (BCF) (Ref. 20) theory extended by inclusion of kinetic attachment together with elastic interactions between steps, diffusion along the step edge, and a consistent fluctuation formalism. We consider a train of monatomic steps separated by terraces of average width $l$, Fig. 1. The $m$th step fluctuates with an amplitude $\xi_m(x,t)$. Its position is $z_m=ml+\xi_m(x,t)$. The adatoms are deposited at a rate $F$ on the surface, diffuse on the terraces with a diffusion constant $D$, and may evaporate after a time $\tau$. The adatoms stick to the step with attachment coefficients $v_+$. The adatom concentration $c(x,z,t)$ (numbers of atoms per unit surface) on a terrace obeys the following stochastic equation:

$$\frac{\partial c}{\partial t} = D\nabla^2 c - \frac{c}{\tau} + F + f - \nabla \cdot \mathbf{q},$$

where we have included the “shot” noise $f$ describing the fluctuations of the deposition rate as well as the conserved noise $\mathbf{q}$ related to fluctuations of the current. In Ref. 21 we have treated this stochastic model in detail and described how to incorporate the noise in a thermodynamically consistent way. Transport along the step edge (not present in Ref. 21) is incorporated here.

In this work we will restrict ourselves to the global equilibrium situation of no net flux across the crystal, i.e., to $F = \langle c \rangle / \tau$. In this case it is in principle easier to neglect first all stochastic terms and to evaluate the dispersion relation of an imposed perturbation of a straight step. The correlations of the effective noise in the Langevin equation for the step position can be determined a posteriori by means of the fluctuation-dissipation theorem.

We have to specify now the boundary conditions at the steps. The original assumption of an instantaneous sticking of atoms in the BCF theory is relaxed here. In a thermodynamic spirit, steps can be viewed as an interface between a two-dimensional gas of adatoms on the terraces described by a local chemical potential $\mu_\xi(r,t)$ and a solid phase with bulk potential $\mu_0$. For an incompressible (here two-dimensional) solid we can define a local chemical potential on its surface,

$$\mu_{\text{eff},m}(x) = \mu_0 + \Omega \frac{\partial \mathcal{F}}{\partial \xi_m(x)},$$

$\mathcal{F}$ is the additional free energy due to the existence of an interface and $\Omega$ is the atomic area. This expression assumes an infinitely sharp interface, which is reasonable for solid-gas interfaces. $\mathcal{F}$ takes the form (in the nearest-neighbor approximation; see later for justification)

$$\mathcal{F} = \sum_{m=-\infty}^{+\infty} \int_0^\infty dx \left[ \gamma \left( 1 + \left( \frac{\partial \xi_m(x)}{\partial x} \right)^2 \right) \right]^{1/2} + A \left( \frac{1}{(1 + \xi_{m+1}(x) - \xi_m(x))^2} \right) + \frac{1}{(1 + \xi_m(x) - \xi_{m-1}(x))^2},$$

where $\gamma$ is the line stiffness of the steps, $\sigma = \sigma + \sigma''$, $\sigma$ is the free energy per unit length (line tension), and $\sigma''$ its second derivative with respect to the step orientation. In the following we will disregard anisotropy effects. The quantity $A$ is the strength of elastic interactions between two steps. Here we assume the standard law where the interaction is inversely proportional to the square of the distance. In principle, the elastic energy depends also on the modulation of the steps. This gives rise to a renormalization of the line stiffness $\gamma$. For homoepitaxy it is shown that this contribution is two to four orders of magnitude smaller than $\gamma$ (a terrace width of $l \sim 100$ lattice constants assumed) and will be neglected here.

The effective chemical potential on the step is then obtained as

$$\mu_{\text{eff},m} = \mu_0 + \Omega R_m,$$

with the abbreviation

$$R_m = \gamma \kappa_m + 2A \left( \frac{1}{(1 + \xi_{m+1}(x) - \xi_m(x))^3} - \frac{1}{(1 + \xi_m(x) - \xi_{m-1}(x))^3} \right),$$

$\kappa_m$ is the step curvature counted to be positive for a convex profile,

$$\kappa_m = -\frac{\partial^2 \xi_m/\partial x^2}{[(\partial^2 \xi_m/\partial x^2)^2 + 1]^{3/2}}.$$
and expanding (see the appendix of Ref. 24) around the equilibrium concentration in front of an isolated straight step $c_{eq}^0$ leads to

$$c_{eq} = c_{eq}^0 \left[ 1 + \frac{\Omega}{k_B T R_m} \right].$$

(13)

Hence, $c_{eq}$ refers to the equilibrium concentration at a step modified by interactions with the neighboring steps as well as by curvature effects.

For small deviations about equilibrium the normal velocity $v_n$ of the step depends linearly on the chemical potential differences as

$$v_n = A_+ (\mu_+ - \mu_{eff}) + A_- (\mu_- - \mu_{eff}) - \frac{\partial f}{\partial s},$$

(14)

where $s$ is the arclength along the step, and where we have left out noise terms for the moment. This type of equation has been widely used in MBE assuming the gas potential $\mu_{eff}$ to be a constant, which corresponds to a situation of infinitely fast diffusion in the gas phase. Equation (14) acquires thus a local character.

The first two terms correspond to a flux of adatoms perpendicular to the step. The flux $J$ of adatoms along the step is defined as

$$J = -B \frac{\partial \mu_{eff}}{\partial s},$$

(15)

with the transport coefficients $A_{\pm}, B$. With the help of Eqs. (13) and (12) and after incorporation of noise, expression (14) can be rewritten in terms of concentrations

$$v_n = \Omega \left( v_+(c_+ - c_{eq} + \eta_+) + v_-(c_- - c_{eq} + \eta_-) + \frac{\partial}{\partial s} \left( B \frac{\partial R_m}{\partial s} + \frac{q}{\Omega} \right) \right),$$

(16)

where $\eta_{\pm}$ are random attachment-detachment forces. In a consistent picture we have to allow for fluctuations of the mass flux along the step edge and have introduced a white noise term $\tilde{q}$ accordingly. $v_{\pm} = A_{\pm} k_B T / (\Omega c_{eq}^0)$ are phenomenological kinetic coefficients having the dimension of a velocity, and $R_m$ is given by Eq. (10). The asymmetry of the attachment kinetics, $v_+ \neq v_-$ expresses the Ehrlich-Schwoebel barrier effect. One expects in general $v_+ \geq v_-$. This is understood by noting that the adatom that sticks to a descending step has to go through a position where it has fewer neighbors (the terrace border). This is unfavorable energetically. This effect leads to several important features out of equilibrium. It must be kept in mind, however, that the ES effect may be reversed in some special situations. This is likely the case when a so-called surfactant atom is present. As long as the system is in equilibrium, the sign of $v_+ - v_-$ does not play a special role. For definiteness, we shall adopt the common situation, $v_+ \geq v_-$.

Let us briefly make some comments about the conservation equation (16). The simple curvature and elasticity dependence of the edge diffusion term in Eq. (16) implicitly assume that the kink distribution is equilibrated on a time scale that is fast on the scales of interest. Hence, kinetic effects resulting from the fast exchange between kink atoms and adatoms adsorbed on the step edge are neglected. Had we relaxed this condition, then Eq. (16) would have been modified in such a way as to include kink and antikink dynamics. Here we assume that temperature is high enough so that the fluctuation $\sqrt{\langle [\xi(t) - \xi(t', x)]^2 \rangle}$ is large as compared to the atomic length $a$. Another remark is in order. Edge diffusion is decoupled from the concentration onto the terraces, contrary to the original approach of Khare et al., which leads to different dynamics at short times. That Eq. (16) is an appropriate formulation can be seen intuitively in the limit of a step in vacuum. There the concentration of adatoms in the gas phase is zero, but there is still the equilibration process described by the diffusion term in Eq. (16). A more recent work by the same authors has cured this problem, and their formulation agrees now with ours.

The velocity can be expressed as a function of the step position as

$$v_n = \frac{\partial \tilde{\xi} / \partial t}{\left[ 1 + (\partial \tilde{\xi} / \partial x)^2 \right]^{1/2}},$$

(17)

Expression (16) generalizes the one already used in a previous publication by the presence of the edge diffusion term.

The present problem is of free-boundary type (Stefan problem), where an additional constraint must be evoked to complete the description (to fix the profile of the “free”—in the sense that it is not known a priori, and not because it does not interact with the rest—boundary, the step). This condition follows from the relation between the perpendicular flux terms of Eqs. (14) and (16) to the normal gradient of the concentration field,

$$\vec{n} \cdot (-D \nabla c + \vec{q})_{\pm} = v_{\pm} (c - c_{eq} + \eta_{\pm}),$$

(18)

where $\vec{n}$ is the unit normal vector at the step pointing from the upper terrace ($-$) into the lower one ($+$).

Finally, we must specify the amplitudes of the Langevin forces. Their correlations take the following forms:

$$\langle q_{m}(r, t) q_{m'}(r', t') \rangle = 2 D c_{eq}^0 \delta_{ij},$$

(19)

$$\langle f_{m}(r, t) f_{m'}(r', t') \rangle = 2 \frac{c_{eq}^0}{\tau} \delta,$$

(20)

$$\langle \eta_{m}(s, t) \eta_{m'}(s', t') \rangle = 2 \frac{c_{eq}^0}{v_{\pm}} \delta(s - s') \delta(t - t') \delta_{mm'},$$

(21)

$$\langle \tilde{q}_{m}(s, t) \tilde{q}_{m'}(s', t') \rangle = \frac{2 k_B T B}{\Omega} \delta(s - s') \delta(t - t') \delta_{mm'},$$

(22)

where $\delta$ is an abbreviation for $\delta(r - r') \delta(t - t') \delta_{m, m'}$, $c_{eq}^0$ is the equilibrium concentration on the terraces, and $s$ denotes the arclength along the step. The average here is performed over the thermodynamical Boltzmann equilibrium distribution.

Equations (6), (16), and (18) completely describe the dynamics of the vicinal surface. They can be converted into a coupled system of nonlinear integro-differential equations.
for the step positions and the concentration at the step edges by means of the Green’s function of the diffusion equation (6).\textsuperscript{21}

The nonlocal character of the equations arises from the self-interaction of a step as well as from the interaction between different steps. There are also retardation effects due to the fact that diffusion is not necessarily fast. The diffusion field thus keeps some memory of former step positions. In this work retardation effects are neglected. This amounts to making use of the quasistatic approximation. This assumption means that the diffusion field adapts itself instantaneously to the step motion. Formally, this assumption is equivalent to setting the left-hand side of Eq. (6) to zero. This is justified for most practical purposes.

The nonlinearities are on the one hand of geometrical origin, as is seen in the normal velocity (17) and the curvature (11). On the other hand, nonlinearities are due to the moving-boundary character of the problem and not directly visible in the primary set of equations (6), (16), and (18). They can be understood by noting that steps act as one-dimensional sinks of strength $\sim v_s$ for the diffusion field. Knowing the source and sink terms, the field can be determined. By inserting Eq. (18) into Eq. (16), one realizes that this strength follows from the gradient of the diffusion field. Thus the diffusion field is coupled back to itself, a clear nonlinear effect.

Nonlinear effects are clearly important in an out-of-equilibrium situation.\textsuperscript{21} Our main concern here is the study of fluctuations in global equilibrium where the linearization of the full set of equations is legitimate.

A. Order of magnitude of various parameters and link to microscopic quantities

To fix ideas let us discuss now the order of magnitude of various parameters:\textsuperscript{23} $Dc_{eq}^{0} \sim 10^{14} \exp(-[W_d + W_a]k_BT) s^{-1}$ and $\Omega c_{eq}^{0} \tau \sim 10^{-14} \exp(-[W_d + W_a]k_BT) s^{-1}$, where

$W_d, W_a, W_c$ are the diffusion barrier, the energy of an adsorbed atom, and the desorption energy, respectively.

For Si(111), $W_d + W_a \sim 1.1$ eV and $W_c + W_a \sim 4.3$ eV. It will appear useful to estimate the various characteristic length scales. $a_0 = 5.43 \text{ Å}$ is the atomic distance on the Si(111) surface, $a_0 = 5.43 \text{ Å}$ is the bulk lattice constant. The diffusion length is given by $x_s = \sqrt{D\tau} = a \exp((W_c - W_d)/2k_BT)$, where $x_s \sim 10^6 a$ at 900 °C. For the kinetic attachment at the steps characterized by $v_{x, +}$, we find it convenient to introduce length scales $d_{x, +} = D/v_{x, +}$. The kinetic length $d_{x, +}$ is of the order of the atomic distance at this elevated temperature (which expresses the fact that an atom attaches instantaneously to the step when originating from the lower terrace) and $d_{x, -} = a \exp(W_d/k_BT)$ with the Schwoebel barrier $W_d = 0.5$ eV on the Si(111) surface.\textsuperscript{30} This implies $d_{x, -} = 1000 – 10$ atomic lengths for a temperature range from 800 to 1600 K.

Dimensional arguments provide us with orders of magnitude for the line stiffness $\gamma$ and the elastic coefficient $A$. The appropriate macroscopic quantity that characterizes a solid is the Young modulus $E \sim 10^{10}$ Pa, while the atomic distance $a$ is the only length scale. Dimensionally, we must have $\gamma \sim E a^2 \sim 10^{-9}$ J m$^{-1}$ and $A \sim E a^4 \sim 10^{-9}$ J m. This leads to an elastic length $l_e = l_e^2 \sqrt{\gamma/12A} \sim 1(l/a)$, which is large compared to the typical terrace width $l \sim 10^2 – 10^4 a$. This entails that in the range of relevant temperatures $\leq 1400$ °C, we can legitimately assume $l \ll l_e \ll x_s$.

Edge diffusion introduces a new length scale $l_{||}$,

$$l_{||} = \frac{B k_BT}{D_{eq}^{0} \Omega}.$$  

Defined in this way, it allows us to compare the efficiency of the transport along the edge to the transport onto the terraces. The transport onto the terrace becomes more important the higher the concentration of adatoms is on the terraces and the faster they can jump. Both effects reduce $l_{||}$. $l_{||}$ takes a value of the order of the atomic length $a$ when both jump rates along the step and onto the terrace are of the same order. This is the case for Si(111) at high temperatures. At room temperature, the concentration $c_{eq}^{0}$ of adatoms on the terrace can be so low that $l_{||}$ may become larger than the mean terrace width. In that limit most of the step fluctuations would be supported by pure step diffusion. Actually, in order to decide what is really the more efficient way to move a kink atom to another position on the same step, it would require in addition the evaluation of the “evaporation” rate from the step onto the terrace. This phenomenon is associated with the Schwoebel lengths $d_{x, -}$.

There are thus two competing effects: diffusion (line or terrace diffusion) and attachment and detachment kinetics. It is thus important to know which length scale will allow us to discriminate between the two competing phenomena. The length scale that will naturally appear is

$$l_{ed} = \sqrt{l_{||} d}$$  

with $d = \min(d_{x, -}, d_{x, +})$. Note that as in many problems, competition between effects (line diffusion and kinetics at the step) implies a geometrical mean of corresponding length scales. These definitions are appropriate in order to correctly discriminate between the two possible limits, namely whether the transport on the terraces is diffusion limited, $d_{x, -} \ll l_{||}$ (small $D$ and large $v_{x, +}$), or kinetically controlled, $d_{x, +} \gg l_{||}$ (large $D$ and small $v_{x, +}$). As stated above, relaxation on scales smaller than $l_{ed}$ is governed by edge diffusion. This should be true as far as $l_{||}$ is larger than the average kink distance on a step that sets the limit for our phenomenological approach. Which regime is relevant for scales larger than $l_{ed}$ will be specified later.

To interpret Monte-Carlo simulations and experiments, it is essential to connect the phenomenological coefficients of the Langevin approach, such as the line stiffness $\gamma$ and the line transport coefficient $B$, to the microscopic energetics of a vicinal surface. We first note that the line stiffness $\gamma$ of
an isolated step is related to the so-called “diffusivity” $\beta^2$ defined as the mean square of the kink length perpendicular to the step edge by

$$\gamma = \frac{k_B T a_\parallel}{\beta^2(T)}.$$  

(25)

where $a_\parallel$ denotes the lattice constant along the step edge separating kink sites. $a_\perp$ is the atomic distance perpendicular to the step.

This diffusivity is a property of a single step and can straightforwardly be calculated given a microscopic model that specifies the energies of kinks of various lengths. The simplest case would be that kinks can be only one atomic distance $a_\perp$ long and cost the so-called kink creation energy $E_k$. This case corresponds to a Brownian particle that jumps with equal probability $e^{-E_k/b_\parallel T}/Z$ a distance $a_\perp$ to the left or right, or stays at the same point with probability $1/Z$, $Z = 1 + 2 e^{-E_k/b_\parallel T}$. In a Brownian analogy the coordinate along the step plays the role of time. Using the above probabilities one easily obtains for the mean square of the kink

$$\beta^2 = \frac{a_\perp^2 e^{-E_k/b_\parallel T}/Z + a_\perp^2 e^{-E_k/b_\parallel T}/Z + 0}{2 + e^{-E_k/b_\parallel T}}.$$  

(26)

Since this is not quite realistic for higher temperature, one can allow for kinks of multiple atomic length $ma_\perp$. Assuming that the energy of a kink is proportional to its length, $mE_k$, one finds

$$\beta^2 = \frac{2a_\perp^2}{e^{-E_k/b_\parallel T} + e^{-E_k/b_\parallel T} - 2}.$$  

(27)

At low temperatures there are only monatomic kinks, and both models become equivalent where $\beta^2$ describes now the kink concentration $P_k$,

$$P_k \approx \left(\frac{\beta}{a_\perp}\right)^2 = 2 e^{-E_k/b_\parallel T}.$$  

(28)

The static correlation function $G(r)$ measured parallel to the step direction provides a convenient experimental possibility to measure the diffusivity, since it is linear in $r$ for small distances $r$ and depends only on $\beta^2$,

$$G(r) = \langle [\xi(t,x) - \xi(t,x + r)]^2 \rangle = \frac{\beta^2}{a_\parallel} r.$$  

(29)

It saturates for larger distances due to the elastic interaction, a fact that allows the determination of the elastic constant $A$.\(^{17}\) At longer distances this quantity diverges logarithmically, an effect that is typical for two-dimensional behaviors.

Finally, the transport coefficient $B$ has been given by Villain et al.\(^{23}\) by means of a Kubo formula neglecting interactions between adatoms,

$$B = \frac{a_\parallel a_\perp^2}{k_B T \tau_k},$$  

(30)

where $\tau_k$ is the average time for an atom to be emitted from a kink site to the step edge. A similar expression has been derived in Ref. 32. As an example we evaluate $B$ for steps on Cu(100) (atomic distance $a = 2.55 \text{ Å}$). Using the experimental values of Giesen et al.\(^{32}\) in Eqs. (25), (26), and (69) ($A_2$) (see later) we obtain

$$B = \frac{B_0}{k_B T} e^{-E_k/b_\parallel T}$$  

(31)

with $B_0 = 6.5 \times 10^{-12} \text{ m}^3 \text{s}^{-1}$ and $E_k = 10 \ 300 \pm 1630 \text{ K}$.

B. A nonlocal Langevin equation for the step position

In the present work we are only interested in the regression of fluctuations in the linear regime. After linearization, Eqs. (6), (16), and (18) can be diagonalized by Fourier transformation. Finally, after a very tedious calculation,\(^{21}\) an inhomogeneous expression for $\xi_{\text{alk}}$ is obtained,

$$\xi_{\text{alk}} = \Psi_{\text{alk}} \chi_{\text{alk}},$$  

(32)

where the Fourier transformation of $\chi$ (like any other quantity) is defined as

$$\Lambda_{\text{alk}}^{-1} = \int_{-\infty}^{\infty} dt \xi_m(x,t) e^{-\mu(m + k_x\omega + i\omega)}.$$  

(33)

$\chi$ is the linear susceptibility of the steps, and $\Psi$ is a noise resulting from the random forces introduced in Sec. III. The correlations of the noise have been derived directly.\(^{21}\) As mentioned before, at equilibrium they can be obtained in a simpler fashion by means of the fluctuation-dissipation theorem once the dispersion relation of the deterministic version of the model is known. Its property will be specified later.

At equilibrium\(^{30,21}\) (no step flow, and where we have incorporated line diffusion) the inverse of the susceptibility is

$$\chi_{\text{alk}}^{-1} = \mu o + \Lambda_{\text{alk}} o$$  

(34)

with

$$\Lambda_{\text{alk}} o = \left(\frac{\gamma k^2}{4\pi^2} (1 - \cos \Phi) + \frac{Dc_{\text{eq}} \Omega^2 \Lambda_{\text{alk}}}{k_B T} \times 2 \left[ \cosh(\Lambda_{\text{alk}}) - \cos \Phi \right] + \Lambda_{\text{alk}}(d_+ + d_-) \sinh(\Lambda_{\text{alk}}) \right).$$  

(35)

where we have used the notations

$$\Lambda_{\text{alk}} = \sqrt{i o D + k^2 + 1/\tau_y},$$  

(36)

$$R = (d_+ + d_-) \Lambda_{\text{alk}} \cosh(\Lambda_{\text{alk}}) + (d_+ d_- \Lambda_{\text{alk}}^2 + 1) \sinh(\Lambda_{\text{alk}}).$$  

(37)

Note that in real space Eq. (32) represents a nonlocal Langevin equation with a colored noise both in space and time. The nonlocal character is easy to recognize from Eq. (37) since the square root (unlike an algebraic form like $k^2$, which produces the second derivative in real space) does not produce simple derivatives in real space, but rather a series of derivatives of all orders.
In the following we will make use of the quasistatic approximation, \( \omega_0 D \ll k^2 + 1/\lambda_z^2 \). Let us recall that this assumption means that the concentration field relaxes rapidly when responding to a step fluctuation. \( \lambda_{\alpha q} \) and \( \lambda_{k \Phi \omega} \) are now independent of \( \omega \), which allows us to write down immediately the differential equation for \( \xi_{k \Phi \omega}(t) \) by replacing \( i \omega \) by the operator \( \partial / \partial t \) in the inverse susceptibility,

\[
\frac{\partial \xi_{k \Phi \omega}(t)}{\partial t} = -\lambda_{k \Phi \omega} \xi_{k \Phi \omega} + \psi_{k \Phi}(t), \tag{38}
\]

where the noise \( \psi \) is now \( \delta \) correlated in time (see below).

As it will be demonstrated later, under the assumption that all length scales are well separated, the dispersion relation (35) can be approximated by a power law in certain regions \( i \) of the momentum space,

\[
\frac{\partial \xi_{k \Phi}(t, x)}{\partial t} = \begin{cases} 
\frac{d_i}{\pi} \int_{-\infty}^{\infty} \frac{d^2 \xi_{k \Phi}(t, x + z) + \frac{\partial^2 \xi_{k \Phi}(t, x - z)}{\partial x^2} - 2 \frac{\partial^2 \xi_{k \Phi}(t, x)}{\partial x^2}}{z^2} dz + \psi_{k \Phi}(t, x), & n = 3 \\
-\frac{d_i}{\pi} \frac{\partial^4 \xi_{k \Phi}(t, x)}{\partial x^4} + \psi_{k \Phi}(t, x), & n = 4.
\end{cases}
\tag{40}
\]

For even \( n \) the equation is local. The nonlocal case \( n = 3 \) has been obtained by means of the integral representation of \( |q| \):

\[
|q| = \frac{1}{\pi} \int_{-\infty}^{\infty} \sin^2 q z \, dz. \tag{41}
\]

Physically the nonlocal character is traced back to the fact that a step fluctuation with wave number \( q \) will perturb the diffusion field on a strip of width \( 1/|q| \) on the terrace. Any other point located on the step will be sensitive to that perturbation and its motion depends on the field elsewhere (nonlocality).

Equation (38) can easily be integrated to give the dynamical spectrum of the meander,

\[
E = \sum_{m = -\infty}^{\infty} \int_{0}^{L} dx \left[ \frac{\gamma}{2} \left( \frac{\partial \xi_{m}}{\partial x} \right)^2 + \frac{3A}{2M^2} (\xi_m - \xi_{m+1})^2 + (\xi_m - \xi_{m-1})^2 \right]. \tag{44}
\]

We obtain in Fourier space

\[
E = \frac{2\pi}{\delta} \int_{-\infty}^{\infty} dk \left[ \frac{\gamma}{2} k^2 + \frac{6A}{\delta^2} \right]. \tag{45}
\]

Assigning an energy \( k_B T / 2 \) to each mode \( k, \Phi \), leads to expression (43).
This means that at equilibrium the amplitude of the resulting noise \( \psi \) must be colored in such a way that Eq. (43) will be satisfied. This is indeed the case and has been explicitly shown in Ref. 21. Equation (43) therefore provides us directly with the noise correlations,

\[
\langle |\psi_{k\Phi}|^2 \rangle = 2Bk_B T k^2 + 2D_c \text{eq} \Omega^2 \lambda_k \left( \frac{2[\cosh(\Lambda_k l) - \cos \Phi] + \Lambda_k (d_++d_-) \sinh(\Lambda_k l)}{(d_++d_-)\Lambda_k \cosh(\Lambda_k l) + (d_+d_-^2+1) \sinh(\Lambda_k l)},
\]

(46)

with \( \Lambda_k = \sqrt{k^2 + 1/4x_s^2} \).

In order to investigate the temporal behavior of the vicinal surface, we shall evaluate two relevant quantities that characterize the meander \( \zeta_m(x,t) \) of the steps in a train. These are the step roughness \( w(t) \) and the interstep correlations \( I(p,t) \) perpendicular to the steps. The RMS roughness is defined by

\[
w^2(t) = \langle \zeta_m(x,t)^2 \rangle = \frac{1}{N} \sum_{m=1}^{N} \frac{1}{L} \int_0^L dx \langle \zeta_m(x,t)^2 \rangle
\]

(47)

in a train of \( N \) steps of length \( L \). The definition of the interstep correlations \( I(p,t) \) is

\[
I(p,t) = \langle [\zeta_m(x,t) - \zeta_{m+p}(x,t)]^2 \rangle
\]

\[
= \frac{1}{N} \sum_{m=1}^{N} \frac{1}{L} \int_0^L dx \langle [\zeta_m(x,t) - \zeta_{m+p}(x,t)]^2 \rangle
\]

(48)

\[
= \frac{2}{N} \int_{-\infty}^{\infty} d\Phi \int_0^{2\pi} \frac{d\Psi}{2\pi} \langle \xi_{k\Phi}(t)^2 \rangle [1 - \cos(p\Phi)].
\]

The function \( I(p=1) \) is particularly important since it is equal to the mean square width \( W^2 \) of the terrace width distribution, a quantity that is of practical use in experiments.

The average is to be performed, in principle, over the distribution function of the random variable \( \zeta_m \). Here the variances \( \langle \xi_{k\Phi}^2 \rangle \) are determined by using Eq. (46), which itself follows from the variance of the "elementary" noises Eq. (22).

Bartelt et al.\(^3\) considered one fluctuating step between two straight steps \( \zeta_{m-1} = \zeta_{m+1} = 0 \). In this case, \( W \) is always equal to the roughness \( w \) of the step. However, as explained later on, for a train of steps the ratio \( (W/w)^2 \) is time dependent. It takes the value 2 at small times when step fluctuations are still uncorrelated, and then goes to zero for \( t \to \infty \) due to the logarithmic divergence of the roughness \( w \) when all steps are allowed to move (see below). In what follows, we will keep the same initial condition of initially straight steps at \( t=0 \) as Bartelt et al.\(^3\).

Of more practical interest, especially for the interpretation of dynamical STM and REM experiments, is the time correlation function \( G(t-t') \) of a train of steps that has already reached its stationary configuration,
The evaluation of the integrals will allow us to identify the time range where a power-law behavior should effectively be expected.

IV. STATIC CALCULATIONS

First we would like to discuss the static case, $t \to \infty$, where the steps are completely equilibrated. We assume that the train of steps consists of a finite number $N$ of steps of infinite length, which imposes cutoff angles $\Phi_{\text{min}} = 2\pi/N$, $\Phi_{\text{max}} = 2\pi - 2\pi/N$ (recall that $\Phi$ is the phase shift between two neighboring steps). The case where $N$ is very large whereas the finite size effects are due to the finite length of the steps is of less importance and will not be considered here. From Eqs. (50) and (47) follow the stationary terrace width variance $W_\infty^2$ [Eq. (48), for $p = 1$] and the step roughness $w^2$ [Eq. (47)],

$$W_\infty^2 = \int_{-\infty}^{\infty} \frac{dk}{2\pi} \int_{2\pi/N}^{2\pi} d\Phi \frac{k_B T}{\gamma k^2 + \frac{12A}{l^4} (1 - \cos \Phi)}$$

$$= \frac{k_B T l^2}{2\pi \gamma A} \ln \tan \left( \frac{\pi}{2N} \right),$$

(53)

$$W^2_\infty = \int_{-\infty}^{\infty} \frac{dk}{2\pi} \int_{0}^{2\pi} d\Phi \frac{k_B T (1 - \cos \Phi)}{\gamma k^2 + \frac{12A}{l^4} (1 - \cos \Phi)}$$

$$= \frac{4\sqrt{2}}{2\pi} \frac{k_B T l^2}{\sqrt{48A \gamma}} = \frac{4\sqrt{2}}{\pi} W^2_G,$$

where $W^2_G$ corresponds to the (Gruber-Mullins) situation considered by Bartelt et al., where one step fluctuates between two straight steps ($\xi_{m-1} = \xi_{m+1} = 0$),

$$W^2 = W^2_G = \int_{-\infty}^{\infty} \frac{dk}{2\pi} \frac{k_B T}{\gamma k^2 + \frac{12A}{l^4}}$$

$$= \frac{k_B T l^2}{\sqrt{48A \gamma}}.$$  

(54)

Thus, this restriction underestimates the terrace width fluctuations $W$ by a factor of $4\sqrt{2}/\pi = 1.8$ compared to the realistic case where all steps are allowed to meander.

Finally $W$ can be calculated beyond the nearest-neighbor approximation taking interactions between all steps into account,

$$W^2_{\text{all}} = \int_{-\infty}^{\infty} \frac{dk}{2\pi} \int_{0}^{2\pi} d\Phi \frac{2k_B T (1 - \cos \Phi)}{\gamma k^2 + \frac{12A}{l^4} \left( \frac{1}{\sqrt{48}} \Phi (2\pi - \Phi) \right)^2}$$

$$= \frac{1}{\pi} \frac{k_B T l^2}{\sqrt{48A \gamma}} = 0.950 32W^2_\infty,$$

(55)

We see that a nearest-neighbor approximation is legitimate, since it overestimates $W$ only by 3%.

It is important to have a more precise relation for the evaluation of the elastic constant $A$ from measurements of $W$. The expression for $A$, defined by the energy interaction per unit length $U_{el} = A/l^2$, is obtained from Eq. (55),

$$A = \alpha \frac{(k_B T)^2}{\gamma} \left( \frac{l}{W_{\text{all}}} \right)^4,$$

(56)

$$\alpha = 0.061 \ldots,$$

where $W_{\text{all}}$ is the mean square of the terrace width distribution measured in a system of many fluctuating steps. As stated above, evaluations of $A$ based on the Gruber-Mullins approximation (54), which neglects the meandering of all steps, would lead to $\alpha = 1/48$ and hence underestimate $A$ by a factor of $\approx 3$.

The situation of many interacting steps raises naturally the question of which value of the phase $\Phi$ would statistically be selected. It is natural to expect the $\Phi = 0$ mode to dominate. The reason is that this is a Goldstone mode costing no elastic energy. However, the synchronization of the steps is not an instantaneous event, but rather operates on some time scale. It is therefore of great interest to estimate that time. Thus, if during the observation time the phases are not statistically explored, then the notion of a static thermodynamical limit must be called into question when using the above limits. The next section, which is devoted to dynamical calculation, will deal with that question.

V. DYNAMICAL CALCULATIONS

The static properties at equilibrium are fixed by the fluctuation-dissipation theorem and depend only on the energy of the system. Any Langevin equation based on this energy with arbitrary dynamics will lead to the same static results.

In contrast, in the dynamical case the Langevin approach becomes useful. The model is based on the microscopic kinetics and contains all physically relevant transport processes that intervene in the mechanisms by which the equilibration is accomplished. These different transport phenomena are connected with specific length scales. These length scales are in turn associated with time scales within which some dissipation mechanisms are efficient. For instance, building up a macroscopic step fluctuation in a fixed time $t$ requires the transport of atoms, which, at short times, can only be provided by nearby parts of the same step. At these small times, this diffusional transport occurs only in a small strip in front of the step. Atoms leaving this strip have no chance to contribute to the fluctuation within the required time. As time elapses, this strip thickens and atoms start to “see” the neighboring step.

Hence, the effect of a finite terrace width $l$ can only be visible after a time $\tau$ where a considerable amount of the diffusing atoms needed to create the fluctuation have been emitted or reflected (strong Schoebel effect) by the next step.
For times shorter than $\tau$ (weak elastic interactions assumed) a train of steps has the same properties as a single step. In particular, the results should depend neither on the mean terrace width $l$ nor on the phase shift $\Phi$ between steps.

In what follows, we will distinguish between temporal correlation functions measured in the stationary state and the approach to this state from a configuration of initially straight steps. As stated before, most of the dynamical measurements by STM or REM have access to the equilibrium quantity $G(t-t')$, 

$$G(t-t') = \frac{1}{2} \left[ (\xi(t,x) - \xi(t',x))^2 - \langle \xi(t,x) \xi(t',x) \rangle \right],$$

which is not equal to $\langle \xi(t,x)^2 \rangle$ corresponding to the non-equilibrated situation $\langle \xi(0,x)^2 \rangle = 0$. This distinction has been clearly pointed out only very recently $^{33}$ [see for instance Eq. (12) in Ref. 3].

We will give here the expressions for the temporal behavior of the roughness $w(t)$ and the root mean square of the terrace width distribution $W(t)$ with the initial condition $w(0) = W(0) = 0$ as well as the time correlation function $G(t-t')$ in the stationary state.

For a transparent discussion we first consider separately the contributions coming from different phases $\Phi$ and define the quantities

$$G_0(t, \Phi) = \int_{-\infty}^{\infty} \frac{dk}{2\pi} \langle |\xi_k \Phi(t) - \xi_k \Phi(0)| \rangle_{\text{stat}},$$

$$w^2_0(t, \Phi) = \int_{-\infty}^{\infty} \frac{dk}{2\pi} \langle |\xi_k \Phi(t)|^2 \rangle,$$

which ultimately have to be integrated in order to average over all possible phase fluctuations,

$$G(t) = \int_{0}^{2\pi} \frac{d\Phi}{2\pi} G_0(t, \Phi),$$

$$w^2(t) = \int_{0}^{2\pi} \frac{d\Phi}{2\pi} w^2_0(t, \Phi).$$

The initial configuration in the evaluation of $w(t)$ and $W(t)$ will be taken to be a train of straight steps. However, $G(t)$ is, by its very definition, an evaluation of the step configuration that is at its stationary (or permanent) regime even at $t = 0$.

Let us first quote some special cases. $w^2_0(t, \Phi = 0)$ represents the contribution of the in-phase mode where the effective elastic interaction vanishes. On the other hand, $w_0(t, \Phi = \pi/2)$ is equal to the roughness $w$ of a single step fluctuating between two rigid ones since the condition $\xi_{m+1} = \xi_{m-1} = 0$ in Fourier space is equivalent to setting $\cos \Phi = 0$. The equilibration in this case has been analysed by Bartelt et al. $^{3}$ for two special situations, as follows.

(a) The step equilibrates thanks to particle exchange with the terrace. The model corresponds to model $A$ for a nonconserved order parameter (in the terminology of critical phenomena) $^{34}$ or to the so-called Edwards-Wilkinson model. The terraces act here as infinite particle reservoirs. The diffusion on the terraces is fast compared to the kinetics at the steps, so that diffusion on the terrace is not a limiting factor. For small times a power-law behavior for the roughness was found,

$$w^2(t) \sim t^{1/2}.$$  

(60)

At later times the roughness saturates due to the elastic interaction with the two fixed walls and reaches asymptotically the value given by Eq. (54).

(b) The step can only equilibrate by transport of particles along the step edge. This case is observed at low temperatures. The corresponding dynamics is described by model $B$ for a conserved order parameter. It yields a much slower approach to the equilibrium spectrum

$$w^2(t) \sim t^{1/4}.$$  

(61)

Although $w^2_0$ and $G_0$ are equal in the static limit $t \to \infty$, they differ at finite times. From Eqs. (50), (51), and (58) we have

$$w^2_0(t, \Phi) = G_0(2t, \Phi).$$

(62)

Therefore, we can concentrate only on the functions $G$, $G_0$. From Eqs. (48) and (58) it follows that

$$G_0(t, \Phi) = e \int_{0}^{1 - \exp \left\{ -bt \Lambda (k^2 + l_{\text{eff}}^2) \right\} \frac{2(\chi h - \cos \Phi) + \Lambda (d_+ + d_-) s h}{(d_+ + d_-) \Lambda c h + (1 + d_+ + d_-) \Lambda^2 s h + l_{\text{||}}^2}} \frac{k^2 + l_{\text{eff}}^2}{dk}$$

with the definitions

$$e = \frac{k_B T}{\gamma \pi}, \quad s h = \sinh (\Lambda l),$$

$$b = \frac{D_{c_{\text{eq}}} \Omega^2 \gamma}{k_B T}, \quad c h = \cosh (\Lambda l),$$

$$l_{\text{||}} = \frac{B k_B T}{D_{c_{\text{eq}}} \Omega}, \quad l_{\text{eff}} = \frac{l_{\text{||}}}{\sqrt{1 - \cos \Phi}},$$

Recall that $l_{\text{||}}$ is the elastic length, $l_{\text{eff}}$ an effective elastic length, and $l_{\text{||}}$ the edge diffusion length. In the following we
will distinguish between a weak Schwoebel effect corresponding to \(d_-, d_+ \ll l\) and a strong Schwoebel effect, \(d_+ \to \infty, d_- \ll l\).

### A. Weak Schwoebel Effect

We assume the following separation of length scales \(l_\parallel, d_+ \ll l \ll x_s\), which is appropriate for the realistic case of weak desorption.

The integral (63) is split into five parts:

\[
G_0(t, \Phi) = e(I_1 + I_2 + I_3 + I_4 + I_5)
\]

with

\[
I_1 = \int_0^{g/l} J(k)dk,
\]

\[
I_2 = \int_{g/l}^{g/l} J(k)dk,
\]

\[
I_3 = \int_{g/l}^{g/d} J(k)dk.
\]

\[
G(t) = \begin{cases} 
  e \Gamma \left( \frac{3}{4} \right) \left( \frac{l_\parallel}{b} \right)^{1/4} t^{1/4} & \text{for } t \leq \max \left( \frac{l_\parallel d^2}{b^2}, \frac{l_\parallel}{b} \right) \\
  e \sqrt{\pi} \left( \frac{1}{d_-} + \frac{1}{d_+} \right)^{1/2} t^{1/2} & \text{for } \max \left( \frac{l_\parallel d^2}{b^2}, \frac{l_\parallel}{b} \right) \ll t \leq \max \left( \frac{d^3}{b}, \frac{l_\parallel}{b} \right) \\
  e \Gamma \left( \frac{1}{2} \right) \left( 2b \right)^{1/2} t^{1/2} & \text{for } \max \left( \frac{d^3}{b}, \frac{l_\parallel}{b} \right) \ll t \leq \frac{l_\parallel}{b} \\
  2 \sqrt{\frac{2}{\pi}} \frac{2b}{\left( 1 + d_- + d_+ \right)^{1/2}} t^{1/2} & \text{for } \frac{l_\parallel}{b} \ll t \ll \frac{l_\parallel}{b} \\
  \text{no power law due to elasticity} & \text{for } t \gg \frac{l_\parallel}{b} 
\end{cases}
\]

We refer to the behavior at \(t \ll l^3/b\) as short time or isolated step regime. Indeed in that regime the steps do not feel each other. As a consequence \(G_0\) depends neither on the phase nor on the terrace width \(l\). At later times, the phase \(\Phi\) appears explicitly in \(G_0\). It turns out that \(G_0\) possesses an additional regime with the same exponent \(1/4\) originating from terrace diffusion and is different from the one due to true edge diffusion displayed above \((A_1)\). That regime is potentially present in the time interval between \(C_1\) and \(D_1\) [Eq. (65)] but not shown here for the following reason. It is shown in the Appendix that this behavior plays no role for the integrated quantity \(G(t)\) since it appears only in a narrow band in phase space, \(1 - \cos \Phi \ll 1\), and the corresponding value of \(G_0\) is small compared to those stemming from other phases. Therefore, for all practical purposes only the \(t^{1/4}\) regime of pure edge diffusion \((A_1)\) is relevant in \(G(t)\).

Because the above results are based on an asymptotic analysis, it is important to check their validity by numerical evaluation of Eq. (63). This will allow us at the same time to see when a power-law assumption breaks down. We present only typical examples: scanning the whole parameter space would be too tedious and without real interest. We have evaluated the integral (63) numerically for the parameters \(l_\parallel = 0, d_- = d_+ = 0.01l, l_c = 245l,\) and \(x_s = 9000l\); see Fig. 2. To test the reliability of our analysis we considered separately the transition from the regime \(B_1\) (65) to \(C_1\), Fig. 2(a), and from \(C_1\) over \(D_1\) to \(E_1\), Fig. 2(b). The crossover times calculated from Eq. (65), \(\tau_{BC} = (d/l)^3 = 10^{-6}, \tau_{CD} = 1,\) and \(\tau_{DE} = (l_c/l)^3 = 6 \times 10^4\), agree very well with the plot. \(\tau = bt/l^3\) is the dimensionless time. Only after one to two decades away from the crossover times is the actual behavior of \(G(t)\) well described by the power-law expressions of Eq.
FIG. 2. Small Schwoebel effect, weak desorption: The dimensionless correlation function $\Gamma(\tau) = G(t)/el$ versus the scaled time $\tau = bt/l^3$ [$l$ is the terrace width; for definitions of $e$, $b$, see Eq.(64)] for well separated length scales $d_0 \sim l \ll l_c, l_\parallel = 0$. (a) At small time the crossover from the kinetic regime $G \sim \tau^{1/2}$ to the nonlocal one $\sim \tau^{1/3}$ at $\tau \sim (d_0/l)^3$ is seen. For comparison, the asymptotic approximations for the first $1/2$ regime (dotted line) [Eq.(65), $B_1$] and the $1/3$ one (thin solid line) [Eq.(65), $C_1$] are given. (b) at later times with the crossover from the nonlocal regime to the $1/2$ behavior due to adatom exchange between steps at $\tau \sim 1$ and the non-power-law regime due to elastic interactions for $\tau \sim (l_c/l)^2$. The approximations for the nonlocal $1/2$ regime (dotted line) [Eq.(65), $C_1$] and for the second $1/3$ one [Eq.(65), $D_1$] are plotted. Parameters: $d_0 = d_\perp = 0.01l, l_c = 245l, x_\perp = 9000l$.

This means that the three lengths $d_0, l_c, l_\parallel$ have to differ at least by one decade in order to observe a power-law behavior. This analysis is also a call for careful experimental studies.

Figure 3 shows the dependence of $G_0$ on time and phase $\Phi$ in order to visualize the influence of the phase. The behavior for $\Phi = 0$ is very different from that corresponding to phases $1 - \cos \Phi \sim 1$. The two extreme curves correspond to the in-phase mode (filled circles) and the out-of-phase one (full line). The curve with filled circles shows the $1/4$ mode mentioned above (and not shown in the previous summary).

This regime is theoretically there. Indeed, a small desorption (as assumed here) together with the in-phase mode (if a particle leaves a terrace, another one comes in) should imply dynamics of a conserved quantity, leading to the $1/4$ regime. However, and as shown in Fig. 3, the other phases have more pronounced contributions (the scale is logarithmic), as explained above. As a consequence, a transition to the $1/2$ regime takes place immediately after the $1/3$ one.

B. Strong Schwoebel effect

The Schwoebel length $d_\perp$ is assumed to be infinitely large. This is the so-called one-sided model where interlayer mass transport is inhibited. For $d_\perp \to \infty$ the exponent $\lambda$ in Eq.(63) takes a simple form,

$$
\lambda = bt\Lambda \left(k_0^2 + \frac{1}{l_\parallel^{2\text{eff}}} \left(l_\parallel k_0^2 \lambda + \frac{\sinh \Lambda l}{\cosh \Lambda l + d_\perp \Lambda \sinh \Lambda l}\right) \right).
$$

An asymptotic analysis in the limit $l_\parallel, d_\perp \ll l \ll x_\perp, l_\parallel$, similar to the one presented before, yields
The phase $\Phi$ appears in $\lambda$ [Eq. (66)] only via $l_{\text{eff}}$. Therefore, in the absence of elastic interactions $G_0$ would not depend on the phase at all. This contrasts with the case of a weak Schwoebel effect where the exponent $\lambda$ remains phase-dependent, Eq. (63). That dependence expresses the step interactions via the diffusion field. That this interaction is absent in the strong Schwoebel effect is not a surprise. Indeed, since in that limit a step does not exchange mass with other steps, the relative phase does not matter.\(^{35}\) Hence, in all power-law regimes, $t^{\frac{7}{12}} \leq \min \{ x_s^4 l_c^3 (bl), x_s^3 l_c^4 (bl) \}$, steps are uncorrelated, and $G_0 = G$. Correlation takes place only at late times due to elasticity.

Figure 4 shows the temporal correlation function $G(t)$ obtained by numerical evaluation of Eq. (63) at intermediate and late stages for parameters: $d_- = \infty$, $d_+ = 0.01l$, $x_s = 30l$, $l_c = 9000l$, $l_l = 0$. For times $t \leq \tau_{\text{CD}} = 1$ the nonlocal regime ($C_2$) with exponent 1/3 is visible, followed by the regime of effective line diffusion ($D_2$), atoms are reflected by the neighboring step and therefore will on the average move along the steps) with exponent 1/4. Finally, at $t \gg \tau_{\text{DE}} = (x_s / l)^4 = 8 \times 10^3$ the $t^{1/2}$ regime ($E_2$) due to exchange with the vapor is reached.

The effect of the edge diffusion is examined in Fig. 5. There the behavior of $G(t)$ at very short times is displayed. Two cases are considered: (a) The Schwoebel length $d_+$ is larger than the line diffusion length $l_l$. This case leads to a kinetic regime $\Gamma \sim t^{1/2}$ ($B_2$) taking place between the two regimes, one due to true edge diffusion, $t^{1/4}$ ($A_2$), and the other to nonlocal effects, $t^{1/3}$ ($C_2$). The following parameters are used: $l_l = 10^{-6}l$, $d_+ = 0.05l$. (b) The Schwoebel length $d_+$ is smaller than the line diffusion length $l_l$: $l_l > l_l$. This regime is practically absent. The line diffusion controlled mode $\sim t^{1/4}$ exhibits a direct crossover to the $\sim t^{1/3}$ behavior.

Finally, let us draw attention to a rather extreme regime. This corresponds to the situation where $l_l \gg l_l$, which might be relevant for steps on Cu(100) at room temperature. The only power-law regime that survives is the one of true line diffusion ($A_2$). At $t_c = l_l^2 / l_l = l_l^2 (B \gamma \Omega)$, it should exhibit a crossover to the non-power-law regime controlled by elastic interactions.

\[ G(t) = \begin{cases} 
  \epsilon \Gamma \left( \frac{3}{4} \right) \left( \frac{bl}{d} \right)^{1/4} l^{1/4} & \text{for } t \leq \max \left( \frac{l_l^2}{d}, \frac{l_l^3}{b} \right) \\
  \epsilon \frac{b}{d} \left( \frac{l}{d} \right)^{1/2} l^{1/2} & \text{for } \max \left( \frac{l_l^2}{d}, \frac{l_l^3}{b} \right) \leq t \leq \max \left( \frac{d^3}{l_l^2}, \frac{l_l^3}{b} \right) \\
  \epsilon \Gamma \left( \frac{2}{3} \right) \left( \frac{b l}{d} \right)^{1/3} l^{1/3} & \text{for } \max \left( \frac{d^3}{l_l^2}, \frac{l_l^3}{b} \right) \leq t \leq \frac{l_l^3}{b} \\
  \epsilon \frac{b l}{d^2} \left( \frac{l}{d^2} \right)^{1/2} l^{1/2} & \text{for } \frac{l_l^3}{b} \leq t \leq \min \left( \frac{x_s^4 l_c^3}{b^3 l}, \frac{x_s^3 l_c^4}{b^3 l} \right) \\
  \text{no power law due to elasticity} & \text{for } t \gg \min \left( \frac{x_s^4 l_c^3}{b^3 l}, \frac{x_s^3 l_c^4}{b^3 l} \right) 
\end{cases} \]

C. Discussion of the different regimes

The physical origin of all the different power-law regimes has already been pointed out in a very illuminating way by Pimpinelli \textit{et al.}\(^{5}\) However, neither the role of the elasticity nor that of the phase shift between steps has been discussed yet. It seems thus difficult to guess which of the many regimes given in Ref. 4 should apply to a concrete experiment. Furthermore, one cannot expect \textit{a priori} the ideal situation of Ref. 4, which rests on the assumption that each mechanism is operating in a certain range of length scale without any interference with other mechanisms. This is a rather restrictive situation. Generally, there are many competing mechanisms.
and consequently the temporal correlation does not necessarily follow a power law. Power laws are special cases and can only be observed under the following conditions. (a) The interaction between steps, characterized by the elastic length \( l_e \), must be weak. \( l_e \) fixes the maximum time up to which a power-law behavior might occur: \( t_{\text{max}} = 1l_e^2/\beta \) for the weak and \( t_{\text{max}} = 1l_e^4/\beta \) for the strong Schwoebel effect. (b) Length scales must be well distinct (for example, \( d_\perp, d_\parallel < l, x, l_e \) to find \( G \propto t^{1/3} \)). If these conditions are fulfilled, the temporal correlation function \( G(t) \) does indeed exhibit a sequence of power laws. The present unified approach is capable of establishing the sequential temporal order of different regimes and specifying their crossover times.

Let us now discuss several possible situations. We will distinguish between short times \( t \ll l_e^4/\beta \), where there is no difference between the behavior of an isolated step and a train of steps, and large times where step-step interactions are important.

**Short times**

In this regime the temporal correlation is qualitatively the same for both the strong and the weak Schwoebel effect. The only difference lies in the fact that a weak Schwoebel effect implies (except if the regime is of pure edge diffusion) a slightly larger fluctuation since both terraces near a step provide particles (compare for instance \( C_1 \) and \( C_2 \), which differ by a factor of \( 2^{1/3} \)).

A remark is in order. The validity of our continuum approach requires that the observation time be not too small in the sense that \( \sqrt{G(t)} \) must be large in comparison to the atomic distance \( a \). For this reason the equilibration of kink positions as seen at very short times in Monte Carlo simulations\(^3\) is not accessible. In contrast to that work, our description leads always to a \( t^{1/4} \) regime at very short times, which expresses hopping of atoms along the step edge. This phenomenon occurs at low temperatures (room temperature for \( \text{Cu} \)), where atoms are already mobile along the step edge, but cannot overcome the barrier to evaporate onto the terraces. We refer to this regime as true line diffusion, which has to be distinguished from the one of effective line diffusion at later times (encountered when a strong ES effect is assumed).

By inserting Eqs. (25) and (30) into Eq. (65) \( (A_1) \), we can rewrite the correlation function in the true line diffusion regime as (which was already given by Masson et al.)\(^18\)

\[
G(t) = \frac{1}{\pi^2} \Gamma \left[ \frac{3}{4} \right] (\beta^2)^{3/4} \left( \frac{t}{\tau_k} \right)^{1/4}
\]  

\[\text{[note our different definition of } G(t) \text{].}\]

Bartelt et al.\(^3\) have calculated \( G(t) \) in a nonstationary case under the assumption of initially straight steps, which leads to a prefactor \( 2^{3/4} \) times smaller.

In order to decide how long this regime would persist, we must compare the efficiency of hopping along the step edge with other potentially competing transport mechanisms. If we exclude an unphysical fast exchange between the step and the vapor (which would correspond to a very short diffusion length \( x_e \), the only remaining alternative of interest is mass exchange with the terrace. That is to say, an atom is detached from the step, makes short excursions on the terrace, remaining either in the vicinity of the step or making a large excursion away, before it is again incorporated onto the step at a different position.

We must thus distinguish between two cases: (i) The transport is diffusion limited \( \{d = \min(d_\perp, d_\parallel) < l || \} \), meaning that the transport on the terrace is slow, but the detachment from the kink site onto the terrace is fast (that is to say, excursion away from the step is the slowest mode). (ii) The transport is controlled by kinetics \( \{d > l || \} \) and most of the travel time is consumed by the slow detachment onto the terrace, whereas diffusion on the terrace is fast.

The edge diffusion length \( l || \) [Eq. (23)] measures how fast line diffusion is in comparison to terrace diffusion. It does not contain information about kinetics. Small \( l || \) means that
edge diffusion is slow and only important at very short times. Edge diffusion plays no role whatsoever if \( I_{||} \) is of the order of the atomic distance.

The attachment and detachment kinetics at the step edge is described by the two small lengths \( d_- \) and \( d_+ \). The Schwoebel lengths can be interpreted as the distances an adatom has additionally to diffuse near a step in order to be finally accepted for attachment. Hence, \( d_- = d_+ = 0 \) means instantaneous kinetics.

For short times considered here, the excursion of atoms on the terrace traveling from one part of the step to a different position on the same step occurs on average only in a small strip in front of the step. Atoms that leave the strip cannot come back in the required time and, hence, do not contribute to the fluctuation. This means that interaction with neighboring steps plays no role at these short times.

For the kinetically dominated case (ii), the \( t^{1/4} \) behavior of pure line diffusion ceases to operate and a regime with \( G \sim t^{1/2} \) takes place. Remembering the definition of \( b \sim D \) and \( d_+ = D / v_+ \), one sees that the diffusion coefficient does not enter expression \( \langle B_1 \rangle \), Eq. (65), as one would intuitively expect. This regime is characterized by a completely random exchange of adatoms between the step and the terrace. This exchange can be viewed as random only if the supply of new particles is ensured by a fast enough terrace diffusion. The terraces are in some sense felt by the steps as homogeneous reservoirs. If fast diffusion is not ensured, neighboring step positions become correlated by deviations of the adatom concentration from the equilibrium value \( c_0^0 \). This corresponds to case (i) introduced above where terrace diffusion is the slowest (and thus the prevailing) mechanism. As a consequence of this insufficient particle supply, the fluctuation increases with an exponent \( 1/3 \) when the \( t^{1/4} \) regime is over [instead of a prior regime \( t^{1/2} \) as in case (ii)]. To see that, recall that situation (i) corresponds to \( l_{||} \gg d_+ \), in which case the \( t^{1/2} \) interval due to kinetics vanishes \( \langle B_1 \rangle \). This behavior can be compared to that of case (ii) where an intermediate transition to \( t^{1/2} \) occurs before reaching the \( 1/3 \) regime.

The regime with \( G \sim t^{1/3} \) refers to a nonlocal situation, and is finally reached in both situations (i) and (ii). In this time range \( G(t) \) is dominated by fluctuations of larger wave-lengths between max\( (d, l_{||}) \) and the terrace width \( l \). Building up a perturbation of this lateral extent \( \alpha \) (i) consumes too many adatoms. They have to be provided by other parts of the step thanks to diffusion on the terrace. On these scales the adatom concentration cannot be considered anymore as constant. Different positions at a step are coupled via the diffusion field. To summarize, there is the following sequence of exponents at small times: case (i) \( 1/4, 1/3 \) and case (ii) \( 1/4, 1/2, 1/3 \).

Large times

At larger times the transport of adatoms on the terrace is influenced by the existence of the neighboring steps. In the case of a strong Schwoebel effect \( d_- \gg l \), adatoms are reflected by the neighboring step. The exponent of the correlation function decreases, since the transport on the terrace is hindered. On a large scale \( \gg l \) they perform an effective motion along the step. Hence, the time dependence is the one typical for line diffusion \( W^2 \sim t^{1/4} \) [regime \( D_2 \), Eq. (67)]. As time elapses atoms will acquire a reasonable probability to regain the atmosphere. A new regime where \( G(t) \sim t^{1/2} \) is established \( \{ E_2 \}, \) Eq. (67)]]. The exponent of \( G(t) \) increases again, because atoms transported through the vapor phase around the crystal can now contribute. The transport in the gas phase is ballistic and very fast.

For a weak Schwoebel effect, atoms that are emitted from a step can now leave the original terrace within the observation time and contribute to build up the fluctuation of another step. Hence, the exponent increases \( \sim 1/2 \) to \( 1/2 \) \( D_1 \), Eq. (65)]. On a time scale larger than that needed to travel from one step to the next, the regime is similar to the kinetic one with random exchange with a reservoir of particles. The role of the reservoir is now played by the neighboring steps. This corresponds to the regime observed experimentally on Si(111). We note that here the second \( t^{1/2} \) regime \( \sim E_2 \) is due to a different physical mechanism (exchange with the vapor), and contrasts with the case of a weak Schwoebel effect \( D_1 \) where the mechanism is due to interlayer transport. This regime (like \( E_2 \)) could potentially be present in the weak Schwoebel effect as well. However, as long as the diffusion length \( x_+ \) is larger than the terrace width, it would be relevant only for the in-phase modes with \( \cos (\theta) \approx 1 \) (see the crossover times in the Appendix). The physical reason is that for in-phase modes there is no fluctuation visible since if a terrace loses a particle it gains a new one at the same time. Thus vapor becomes the effective reservoir though \( x_+ \) is large. However, the contribution of the in-phase mode does not show up in the integrated quantity \( G(t) \), which means that the main contribution to the second \( t^{1/2} \) mode comes from the exchange between steps and not with the vapor. Consequently, the diffusion length does not appear in the expressions (65). This differs from the case of the strong Schwoebel effect, where a contribution in \( G(t) \) due to exchange with the vapor is noticeable at large enough times, since interlayer transport is forbidden.

We have to remember that all these power-law regimes are expected (besides the condition of length-scale separations) only if the elastic length \( l_\epsilon \) is larger than all the other lengths. Hitherto, this is what has been assumed. \( l_\epsilon \) fixes a time above which \( G(t) \) ceases to follow a power law. This quantity ultimately diverges logarithmically like the static roughness \( w_2^\infty \) [Eq. (53)].

VI. COMPARISON WITH EXPERIMENTS

Experimentally, so far only the exponents 1/2 and 1/4 for the dynamic correlation function \( G(t-t') = \langle \zeta(t) \zeta(t') \rangle \) have been reported. 1/2 has been found at high temperature \( \sim 850 \) °C on Si(111) [Ref. 37] and on Au(110). The latter case is quite special because in the \( 2 \times 2 \) missing-row reconstruction of this surface the motion of a kink in a \( [1 \bar{1} 0] \) step involves the detachment of two atoms. The exponent 1/4 has been seen on various metals such as Cu(100), Pt(111), and Pb(111) [Ref. 38] at room temperature, and has been interpreted as due to true edge diffusion [\( A_1 \), Eq. (65)] or \( A_2 \), Eq. (67)]. This is consistent with different energetic calculations based on the effective-medium theory.21,13 They all agree that the activation energies for hopping along the step edge are considerably smaller than for jumping onto the terraces. Furthermore, experimen-
tally no dependence of $G(t-t')$ on the mean terrace step width $l$ has been observed for these systems. This rules out the other mechanism encountered for a strong Schwoebel effect leading to the same exponent $1/4$ \cite{D2, Eq. (67)}, where $l$ enters $G(t')$ as a result of atom reflection on neighboring steps. On a coarse-grained picture, this mechanism is also a diffusion along the step, but now in a strip of width $l$ and not of width of the order of the atomic distance as before, so that $G$ could depend on $l$.

Thus, it appears that for steps on the close-packed metal surfaces, the prevailing effect of mass transport along the step is the rule. It must be emphasized that although the covered temperature range in the experiments was quite large, no indication towards a crossover to a different exponent at later times seems to have been detected. It is tempting to speculate that the adatom concentration on the terrace is so low that the line diffusion length $l_{\text{diff}}$ persists to remain much larger than the mean terrace width. Hence no dissipation mechanisms other than edge diffusion would prevail, so that only this unique power-law regime survives. It should then only succumb to an elasticity dominated regime at later times (presumably very large), where a simple power-law regime ceases to exist.

**Steps on Si(111) at high temperature**

This system has given rise to extensive investigations and seems to provide a canonical example of stepped semiconductor surfaces. It has thus seemed to us worthwhile to devote a separate discussion to major observations. Alfonso et al.\textsuperscript{1} have investigated the fluctuations of a single pinned step on Si(111) at 900 °C and have extracted the line stiffness of a step $\gamma = 1.1 \times 10^{-10}$ J m by means of the well known expression for the roughness $w$ of a free step,

$$w^2 = \frac{k_B T}{12 \gamma L},$$

where $L$ is the length of the step. As pointed out by Pimpinelli et al.\textsuperscript{4}, this leads to an underevaluation of $\gamma$ by a factor 2, since the pinning of the step has to be taken into account. This leads to

$$w^2 = \frac{k_B T}{6 \gamma L},$$

and thus the value for $\gamma \approx 2.2 \times 10^{-10}$ J m is more appropriate. Furthermore, the elastic constant $A = 4.6 \times 10^{-30}$ J m has been estimated from the terrace width distribution. As mentioned above, values of $A$ obtained by the expressions of the Gruber-Mullins model have to be multiplied by a factor $\approx 3$, if one considers a train of meandering steps as is the case here. The corrected $A = 5.9 \times 10^{-30}$ J m differs not very much from the reported value, because the two errors for $\gamma$ and the terrace width partially cancel. As mentioned before, this correction is necessary only if the notion of static properties involves an average over the phases. Otherwise, one has to know which statistics are performed experimentally before a definite statement.

Pimpinelli et al.\textsuperscript{4} have measured the formation time $\tau$ of a bump on a step of wavelength $L$ for several terrace widths between 300 Å and 7000 Å. $\tau$ ranges from 0.1 to 100 s and was found to be approximately proportional to the terrace width $l$ and to $L^2$. As shown later\textsuperscript{30} this dependence on $l$ even for small $l$ down to 300 Å allows an estimate for the Schwoebel length $d_\rightarrow$. Evaluating the dispersion relation in the appropriate limit ($l\ll l_{\text{eff}}\ll k\ll l_{\text{diff}}, d_\rightarrow < l, d_+ < l$) yields to leading order in $k$

$$\lambda_k \sim \frac{D_c^0 \Omega^2 \gamma^2}{k_B T} 2(1 - \cos \Phi) \frac{1}{d_{\rightarrow} + l} k^2. \quad (71)$$

$\tau \sim 1/\lambda_k$ can depend on the terrace width only if $d_{\rightarrow}$ is smaller than the smallest $l = 300$ Å. With

$$d_{\rightarrow} = a \exp \frac{E_S}{k_B T}, \quad (72)$$

where $a$ is the atomic distance on Si(111), $a = 3.33$ Å, an upper bound for the Schwoebel barrier $E_S$ of 0.47 eV was found. This value seems very reasonable and is consistent with measurement by Latyshev et al.\textsuperscript{39} who observed a potential difference ranging from 0.2 eV to 0.6 eV.

The dependence of $\tau$ on $L^2$ and $l$ led the authors\textsuperscript{4} to the conclusion that the exchange of atoms between different steps, and supported by surface diffusion, should be the appropriate equilibration mechanism \cite{D1, Eq. (65)}. This allowed them to provide an estimate for the product

$$D_c^0 \sim 10^8 \text{ s}^{-1}. \quad (73)$$

At the same time, Bartelt et al.\textsuperscript{37} performed measurements of the correlation function $G(t-t') = \langle \xi(x,t) \xi(0,0) \rangle$ over a po- lared $G = \gamma t^{1/2}$ and claimed that the relevant equilibration process should be due to the slow kinetics at the step edge, which is one of the two possible mechanisms leading to the exponent $1/2$ \cite{B1, Eq. (65)}.

Although the measured quantities are quite different, this conclusion is in our opinion in contradiction with that of Pimpinelli et al.\textsuperscript{4} First, Bartelt et al. have only considered one value for the terrace width $l = 1400$ Å and could therefore not really rule out the mechanism found by the other group, which has the same dependence on $L$ but depends additionally on $l$.

At equilibrium the formation time of a bump should be equal to the relaxation time of a macroscopic fluctuation because of invariance under time reversal. From the Langevin approach it can be seen that the power $\alpha$ of $G \sim t^{\alpha}$ is connected to the exponent of $k$ of the leading order term in the dispersion relation by $\lambda \sim k^{1/\alpha}$, i.e., $\tau \sim L^{1/\alpha}$. The choice of the approximation for $\lambda$ depends on the observation time $t$. This is what we are going to analyze now.

Both groups have measured the same exponent $\alpha = 1/2$. The temporal range where $G(t-t')$ has been investigated, 0.5–15 s, together with that of the terrace width $l = 1400$ Å are of the same order as those explored when measuring the bump formation time. The relevant mechanism should then be the same in both experiments. Below we argue that it is indeed exchange between steps that is responsible for this observation. We show below that it is not possible to gain information about the step kinetics (i.e., atoms attachment and detachment at the step) in the investigated time and temperature range.
It seems to us necessary to reinterpret the measurement of \( G(t-t') \) as a result of exchange of particles between neighboring steps. In order to establish this, we first note that the kinetic regime proposed by Bartelt et al. must be restricted to \( t \ll d_\parallel / b \). In the range of explored temperature, \( d_\parallel \) should be of the order of a few atomic distances \( a \) or at least \( d_\parallel = d_\perp \). Using \( D_{c_0} = 10^8 \text{ s}^{-1} \) and \( a \sim 3 \text{ Å} < d_\parallel \leq 300 \text{ Å} \) provides us with an upper time limit for the observation of the kinetic mechanism (exchange with the terraces):

\[
\tau_1 \sim \frac{d_\parallel^2}{b} = 10^{-9} - 10^{-3} \text{ s}, \tag{74}
\]

where the values \( T=1173 \text{ K}, \Omega \sim a^2, \gamma = 2.2 \times 10^{-10} \text{ J m}^{-1} \) have been used. This time interval is several orders of magnitude lower than the typical observation time in the experiment of Ref. 37, which lies between 0.5 and 15 s. Furthermore, we note that this experiment has really been performed in the “late time” limit, where step-step interactions due to diffusion on terraces are most likely pronounced. It can already be seen that even the time limit \( \tau_2 \) for the nonlocal regime \( G \sim t^{1/2} \),

\[
\tau_2 \sim \frac{l^3}{b} = 0.17 \text{ s} \quad \text{with} \quad l = 1400 \text{ Å}, \tag{75}
\]

is considerably smaller than the experimental time window. In agreement with Ref. 4, we find that it is the \( G \sim t^{1/2} \) regime due to particle exchange between different steps that has been observed. Its upper temporal limit

\[
\tau_3 \sim \frac{l l^2}{b} = 170 \text{ min} \tag{76}
\]

\([l^2 = l^2 \sqrt{\gamma/(12A)} = 245l, A = 5.9 \times 10^{-30} \text{ J m}] \) is much bigger than the experimental time. This means, on the other hand, that saturation effects due to the elastic interaction play no role during the experimental window.

A second way to justify our criticism of the interpretation of Bartelt et al.\(^37\) is supplied by Fig. 5, used to extract the \( k^2 \) dependence of the dispersion relation \( \lambda_k = 1/\tau \). In the plotted range of wave numbers (wavelengths between 7 and 45 times the terrace width \( l \)), the fluctuations are clearly dominated by the step-step exchange mechanism (if one excludes unphysically large kinetic lengths \( d_\perp \sim l \)). This can simply be seen by approximating \( \lambda \) in this range of wave numbers. Expression (71) characteristic for the step-step exchange is appropriate when one has the values of different quantities entering that formula.

Instead of using the above value for \( D_{c_0} \), we can alternatively extract it by means of Eq. (65) \((D_1)\) from the data of Bartelt et al. by using their information about \( l/\tau \) (Fig. 5 in Ref. 37). They have obtained \( l/\tau = \lambda_k \) from a Fourier transformation of \( G(t) \), which represents an average over all modes. The appropriate approximation for \( \lambda_k \) is given by Eq. (71), where we have to replace \( 1 - \cos \Phi \) by the phase average of \( \sqrt{1 - \cos \Phi} = \sqrt{8/\pi} \).

\[
\bar{\lambda}_k = 2 \sqrt{\frac{8}{\pi}} \frac{D_{c_0} \Omega^2}{k_B T} \frac{k^2}{d_{\perp} + d_\parallel + l}. \tag{77}
\]

We obtain a value

\[
D_{c_0} = (0.8 \pm 0.3) \times 10^8 \text{ s}^{-1}, \tag{78}
\]

where we have assumed \( d_\parallel = 300 \text{ Å} \sim 0.2l \). This value agrees perfectly with the estimate of Pimpinelli et al. It must be noted also that our attempt to fit the measured quantity \( G(t-t') \) (Fig. 3 of Ref. 37) by using the above estimated value of \( D_{c_0} \) failed and led to \( G \) one order of magnitude too small. This disagreement can formally be resolved by keeping the product \( D_{c_0} \gamma \) constant and assuming a value for the line stiffness \( \gamma \) much smaller than the corrected value \( \gamma = 2.2 \times 10^{-10} \text{ J m}^{-1} \) from Alfonso et al.\(^1\) used up to now in our calculations. Unfortunately, the value of \( \gamma \) one has to use, \( \gamma = 3 \times 10^{-11} \text{ J m}^{-1} \), is even significantly smaller than the minimal corrected value of \( 1.2 \times 10^{-10} \text{ J m}^{-1} \) given by Alfonso et al.\(^1\). This points to the fact that one would have to significantly alter collected data for \( \gamma \) in order to remedy the discrepancy. At present we do not have any explanation for this fact.

Let us reiterate that, apart from the numerical prefactor, the fit with the theoretical expression \((D_1)\) for the step-step exchange mechanism fits very well. Figure 6 shows \( G(t) \) calculated by numerical integration of Eq. (63) with the parameters known for Si(111) in the time regime measured by Bartelt et al. This means at the same time that (see Fig. 6) in this time interval \( G(t) \) depends neither on the kinetic parameters, \( d_{\perp}, d_\parallel \), nor on the line diffusion length or the elastic length, \( l_\parallel, l_\perp \).

In summary, we have shown that it is possible to go beyond the estimate of \( D_{c_0} \) on Si(111) by Pimpinelli et al. by measuring the time correlation function at one point \( x \) of a fluctuating step \( G(t-t') = \langle \xi^2 \rangle - \langle \xi(t,x) \xi(0,0) \rangle \) in a train of steps. Furthermore, we have argued that it is necessary to reinterpret the observed power-law behavior of the correlation function \( G(t-t') \sim t^{1/2} \) on Si(111), as being due to the exchange of atoms between steps via diffusion on the ter-
races and not as due to the slow kinetics at the step edge. Further experiments studying the temperature and terrace-width dependence of the fluctuations are clearly warranted to clarify the mentioned contradiction in the fit of $G$ with previously established material parameters.

VII. CONCLUSION

We have presented the equilibrium theory for a vicinal surface in the framework of a nonlinear Langevin equation based on the Burton-Carbrera-Frank model (BCF). The original BCF theory has been extended by the Schwoebel effect, diffusion along the step edges, and elastic interactions between steps as well as by a thermodynamically consistent incorporation of fluctuations. This formulation allows the exact calculation of the dynamics of step fluctuations under many competing relaxation processes. If all these mechanisms act on scales of the same order, the resulting correlation functions are not given anymore by simple power laws in time. This is an interesting fact in itself. This work is also a call for further experimental investigations. In the opposite case of well distinct scales, we recover several known time regimes with power-law behaviors $\sim t^{\alpha}$, $\varphi=1/2,1/3,1/4$ and calculated the prefactors analytically. Additional to earlier approaches, mainly based on simple local step dynamics, we were able to define the order of the sequences in the course of time by using a unified picture. Furthermore, the crossover times between the different power-law regimes are given and deviations from scaling are estimated by numerical integration of the full Langevin equation.

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APPENDIX

We present here an asymptotic calculation of the temporal correlation function $G(t) = \langle \zeta(t)^2 \rangle - \langle \zeta(t) \zeta(0) \rangle$ in the limit of perfectly distinct length scales. As an example, we consider the case of a weak Schwoebel effect without line diffusion, $l||=0$. It will appear that extension to any other situation is accomplished along the same lines. For this purpose the following hierarchy of length scales is assumed, $d_+ < d_0 < d_3$, which represents well the real situation on Si(111) surfaces at $T=900^\circ$C. We will refer to the different pieces $I_3-I_4$ in the integral (63) in order to separate the contributions from different dissipation mechanisms.

Short times

For short times $t$ the particles that contribute to the equilibration of the step can make excursions only over short distances. Therefore the temporal behavior of $G_0$ in the limit $t \rightarrow 0$ is determined by the large $k$ behavior of the integrand. This information is stored in $I_4$ [Eq. (63)], which contains wavelengths shorter than all occurring length scales (we assume here $l||=0$). We thus extract the largest power of $k$ in $I_4$. The exponent in Eq. (63) behaves for $k \rightarrow \infty$ as

$$\lambda = -bt k^2 \left( \frac{1}{d_-} + \frac{1}{d_+} \right).$$

(A1)

Defining the transformation

$$u = \left( bt \left( \frac{1}{d_-} + \frac{1}{d_+} \right) \right)^{1/2} k$$

(A2)

and taking the limit $t \ll d^3/b$, we recognize the only contribution to $w_0^2$ comes from $I_4$ yielding asymptotically

$$I_4 = \left( bt \left( \frac{1}{d_-} + \frac{1}{d_+} \right) \right)^{1/2} u_2 \sim t^{1/2} \text{ at } t \ll \frac{d^3}{b}.$$

(A3)

Throughout the asymptotic calculations we will need the following constants:

$$u_2 = \int_0^\infty \frac{1 - e^{-u^2}}{u^2} du = \Gamma \left( \frac{1}{2} \right) = \sqrt{\pi},$$

$$u_3 = \int_0^\infty \frac{1 - e^{-u^3}}{u^2} du = \Gamma \left( \frac{2}{3} \right) \approx 1.35175,$$

$$u_4 = \int_0^\infty \frac{1 - e^{-u^4}}{u^2} du = \Gamma \left( \frac{3}{4} \right) \approx 1.22541.$$ (A4)

The calculation of the other integrals $(I_2, I_3)$ follows the same scheme with the difference being that one has to perform two limits corresponding to the two boundaries of the integrals. By definition, all terms containing the Schwoebel lengths $d_+ , d_-$ in $I_3$ can be neglected, since the main contribution to $I_3$ comes from wave numbers $k$ with $kd \ll 1$. Furthermore, most wave numbers in the integration range obey $kl \gg 1$ leading to $sh \sim ch \sim 0.5 \exp(kl)$, $\Lambda \sim k$, $k^2 + 1/2_{eff} \sim k^2$. The exponent can then be approximated by

$$\lambda = -2bt |k|^3,$$

(A5)

which motivates the transformation $u = (2bt)^{1/3} k$ for extracting the leading behavior. The boundaries of the integral follow the transformation $(k_{\min} = g/l, k_{\max} = g/d) \rightarrow [u_{\min} = g(2bt)^{1/3}, u_{\max} = g(2bt)^{1/3}]$.

$I_3$ dominates the behavior of $w_0^2$ when $u_{\min} \ll 1$ and $u_{\max} \gg 1$. This condition provides the time interval for the validity of the approximation. In this limit $I_3$ becomes

$$I_3 = (2bt)^{1/3} u_3 \sim t^{1/3} \text{ at } \frac{d^3}{b} \ll t \ll \frac{d^3}{b}.$$ (A3)

Up to now the results are independent of the terrace width $l$ and the phase shift $\Phi$. At these short times, $t \ll l^3/b$, steps are completely uncorrelated. Each step behaves as a free one and the mean square width of the terrace distribution $W^2$ is just the sum of the roughness $w^2$ of the two steps forming the
terrace, $W^2 = 2w^2$. As time proceeds, other length scales become relevant in the order given by the assumed hierarchy: $d_- \sim d_+ \ll l \ll l_x \ll x_s$.

**Large times**

For larger times, $t \gg l^3/b$, step-step interactions become important. The interactions are either mediated by the diffusion field or directly by the elastic distortions. The asymptotic behavior involves both the phase $\Phi$ and the terrace width $l$. Mathematically, this information is stored in the remaining integrals $I_2, I_1$ with small wave numbers, $kl \ll 1$, allowing a small argument expansion of $ch, sh$.

In the wave-number range of $I_2$ the exponent behaves as

$$
\lambda \sim -\frac{btk^2}{l + d_+ + d_-} [2(1 - \cos \Phi)] + \frac{1}{3} l^2 \left( k^2 + \frac{1}{x_s^2} \right) (2 + \cos \Phi) \right].
$$

(A6)

For the out-of-phase modes $s = (1 - \cos \Phi) = O(1)$ we obtain again a $G_0 \sim t^{1/4}$ behavior by means of the transformation $u = \{(bt/l)[2s + (3 - s)t^2/3x_s^2]\}^{1/2}$. But for in-phase modes $s \ll 1$ an additional regime $G_0 \sim t^{1/4}$ appears (transformation $u = [bt(3-s)/3]^{1/4}k$). The two possible asymptotics are

$$
I_2 = \frac{l^{3/4}}{b} \leqslant t \leqslant \frac{l^3}{b[2s + (ll_x)^2]^2} \quad \text{and}
$$

$$
I_2 = \sqrt{\pi} \frac{bt}{l + d_+ + d_-} \left[ 2s + \frac{(3-s)t^2}{3x_s^2} \right]^{1/2} \sim t^{1/4}
$$

(A7)

Let us discuss now the limiting case of mode $\Phi = 0$, where all steps execute in-phase motions. The crossover time from the $t^{1/4}$ regime to $t^{1/2}$ is set by the diffusion length $x_s$. This means that the latter regime is caused only by the exchange of particles with the vapor and not with the next step. This becomes clear by noting that a train of in-phase steps is equivalent to one step with periodic boundary conditions in the direction parallel and perpendicular to the step. Hence, particle transport from step to step is not possible. That is to say that, for example, if a step collects a particle from the upper terrace, then the terrace ahead of that step loses at the same time a particle in favor of the next descending step, and so on. For $x_s = \infty$, the number of particles on one terrace is conserved, which explains the line diffusion exponent 1/4. There is no saturation by elastic effects at very large times contrary to the other modes, $G_0(t)$ diverges $\sim t^{1/2}$ (for an infinitely large system).

In this non-power-law saturation regime, time is so large that it is sufficient to keep only the contribution of the zero wave number in the exponent,

$$
G_0(t, \Phi) = e^{\int_0^t 1 - e^{-bt\lambda_k}} \frac{dk}{k^2 + l_{0\text{eff}}^{-2}}
$$

which gives the continuation to times above the limits considered for $I_2$.

Finally, in order to obtain $G(t)$, $G_0$ has to be integrated over all phases, Eq. (59). The expressions for small times $t \ll l^3/b$ remain unchanged, since they are independent of the phase. Later, at $t \gg l^3/b$, we note that the contribution of the in-phase modes $G_0 \sim (btl)^{1/4}$ is small compared to the one from the other modes $G_0 \sim (btl)^{1/2}$. One sees from Eq. (A7) that there is no $t^{1/4}$ regime for these out-of-phase modes ($\pi/2 \ll \Phi \lessgtr 3\pi/2$), which represents half of all possible modes.

Therefore, at times $l^3/b \ll t \ll l_l^2/b$ the main contribution to the averaged quantity $G(t)$ comes from the out-of-phase modes. The $t^{1/4}$ regime of in-phase fluctuations is not visible in $G(t)$, Eq. (65).

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Assume that the step of interest is located around $z = \xi_0$. In the one-sided model the field on the next step is subject to the condition \( \partial c / \partial z|_{z = \xi_0} = 0 \), which reduces in the linear regime to \( \partial c / \partial z|_{z = \xi_0} = 0 \), so that no information of the configuration of the next step is left; the phase shift is thus unimportant.

This has been pointed out to us by T. Einstein after this paper was submitted; see Blagojević and P. M. Duxbury, in *Dynamics of Crystal Surface and Interfaces*, edited by P. M. Duxbury and T. Pence (Plenum, New York, 1997), p. 1; T. L. Einstein and S. V. Khare, in *Dynamics of Crystal Surface and Interfaces*, edited by P. M. Duxbury and T. Pence (Plenum, New York, 1997), p. 83; see also Ref. 29.

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If $\xi_0$ is large enough, the phase shift is not important.

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