# Step dynamics on vicinal surfaces using discrete interface models

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(Received 17 November 2005; revised manuscript received 6 January 2006; published 3 March 2006)

Fluctuations of a single step on a vicinal surface are investigated using a discrete model designed to go further in capturing atomistic effects than usually achieved with continuum models. Variants of the model to incorporate the following three types of adatom dynamics are studied: attachment-detachment; periphery (step-edge) diffusion; and terrace diffusion. The dynamic exponent $z$ is determined for each type as well as crossover functions joining three distinct scaling regimes.

DOI: 10.1103/PhysRevE.73.031601 PACS number(s): 81.10.Aj, 68.35.Ct, 05.40.−a, 05.70.Np

Technologists continue to fabricate devices on ever decreasing scales in the nanometer range. This has stimulated much interest amongst surface scientists on the role played by (thermally agitated) nanoscale fluctuations on the surfaces of metals and semiconductors; such fluctuations are expected to pose important physical limitations on the functionality of nanostructures.

Particular interest concerns vicinal surfaces. Such a surface is cut at an angle very close to, but not exactly at, a plane of high crystallographic symmetry so that it consists of a series of atomic-monolayer terraces rather than a single facet. The terraces are separated by steps that themally fluctuate on a time scale of seconds, an effect thus readily and directly observable. For excellent reviews in this field, see, for example, Refs. [1–3].

The purpose here is to report some theoretical work on the roughening dynamics of a single isolated step; the vicinal angle is assumed to be small enough that the average distance between successive steps is sufficiently large to justify ignoring multistep effects. Previous theories have mostly been confined to models which treat the step edge as a continous function of position [4–9] although simulations [4,6,10–13] and related theory [14,15] have been carried out on discrete models. The lack of a detailed and systematic theoretical treatment of lattice models is of some concern in view of the small length scales involved (nanometers) where atomistic effects are likely to show up. Moreover, simulators working with lattice models would benefit from input gained from discrete theories especially since continuum ones sometimes give a poor fit to their data. The model presented here is, indeed, discrete but before presenting it we first describe the differing physical mechanisms by which a step evolves.

A schematic depiction of a step of monoatomic height is shown in Fig. 1 together with indications of the different ways adatoms can attach-detach from the step edge, thus conforming its shape. In addition to the usual periodic potential energy, incorporating energy barriers for adatom hopping between interstitial substrate sites, adatoms are also subject to a deep attractive well at the step edge with repulsive barriers on either side of it. This is the Ehrlich-Schwoebel potential [16–18] and has the effect of confining adatoms to the lower terrace once they get detached from the step edge; the probability of jumping from terrace to terrace being relatively low at moderate temperatures. Figure 1(a) shows atoms evaporating from and depositing to the step edge from the vapor while (b), occurring at high temperatures, shows atoms joining (respectively, leaving) the step edge via a diffusion process from (respectively, to) distant neighboring steps. Both (a) and (b) conform the step in a *nonconserving* and (to a very good approximation) uncorrelated way; the process is often termed *attachment-detachment* and for the purpose of this paper, the resulting dynamics will be referred to as type I. Figure 1(c) shows atoms hopping between neighboring sites along the step edge without making further excursions. This process, often termed *periphery diffusion*, *locally conserves* the number of atoms in the upper terrace and the resulting dynamics will be referred to as type II. Finally, Fig. 1(d) shows an atom detaching from the step and rejoining it further away via a diffusion process on the lower terrace. This process, termed *terrace diffusion* in the literature, is also conserving but in a *nonlocal* way and the resulting dynamics will be referred to as type III.

The quantity of interest, amenable to the experimental measurement, is the equilibrium autocorrelation function $G(t)$ for step-edge fluctuations, defined as follows. If $y_i(t)$ is

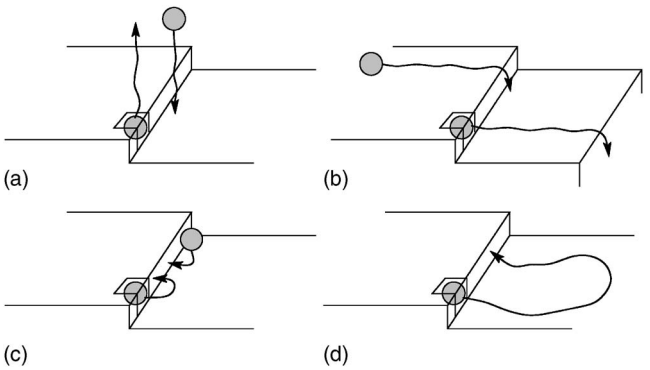


FIG. 1. Different types of step dynamics with (a) and (b) being type I (nonconserving and uncorrelated); (c) being type II (local conserving); and (d) being type III (nonlocal conserving). The shaded circles depict adatoms.

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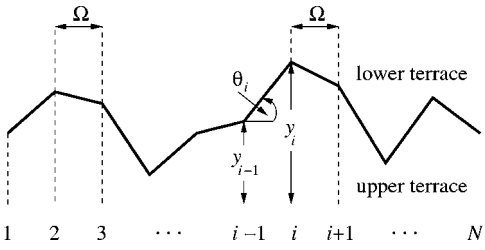


FIG. 2. Plan view of a vicinal step illustrating the discrete model.

the displacement of the step edge at location $i$ and time $t$, then

$$G(t) \coloneqq \lim_{s\to\infty}\langle[y_i(s+t)-y_i(s)]^2\rangle, \tag{1}$$

for $t>0$. For a step of infinite length, it is found that $G(t) \sim t^{1/z}$, for large $t$, where $z$ is the dynamic exponent whose value depends on the type of dynamics dominating step-edge fluctuations. Previous work (see Refs. [1–3] and references therein) has concluded that for type I, $z=2$; type II, $z=4$; and for type III, $z=3$. Alternatively, and often of interest to simulators, is the growth through kinetic roughening of the mean-squared width, $w^2(t)=\langle y_i^2(t)\rangle$, of an initially flat (nonequilibrium) interface, $y_i(0)=0$ for all $i$. Theoretically, it's been found that $G(t)$ and $w(t)$ are simply related through $w^2(t)=G(2t)$ [4,7], a relationship also confirmed below for the following lattice model.

We coarse-grain the step on a scale $\Omega$ and then model the location of the step edge by an $N$-component column vector, $y=(y_1,\ldots,y_N)^T\in\mathbb{R}^N$, as shown in Fig. 2. Periodic boundary conditions $y_{i+N}=y_i$ are imposed. Each coarse-grained step-edge configuration has an effective energy given by the Hamiltonian

$$\mathcal{H}(y)=\Omega\sum_{i=1}^{N}\Sigma(\theta_i)\sec\theta_i, \quad \tan\theta_i=\frac{(y_i-y_{i-1})}{\Omega}, \tag{2}$$

where $\Sigma(\theta)$, the step tension at angle $\theta$, is expressible in terms of microscopic parameters. In what follows it will be sufficient to replace $\mathcal{H}(y)$ by its expansion to quadratic order in $y$, the "discrete Gaussian model":

$$[\mathcal{H}(y)-N\Omega\Sigma(0)]/k_BT \approx \tilde{\sigma}y^TAy/2\Omega, \tag{3}$$

with $\tilde{\sigma}\coloneqq\tilde{\Sigma}(0)/k_BT$ and $\tilde{\Sigma}(0)$ is the step stiffness, $\tilde{\Sigma}(0)=\Sigma(0)+\Sigma''(0)$. The $N\times N$ matrix $A$ has elements, $(A)_{ij}=A_{ij}$, with: $A_{ii}=2$; $A_{ij}=-1$ for $i=j\pm1$ mod $N$; and $A_{ij}=0$ otherwise. In other words, $A=-\Delta_{\text{per}}$, where $\Delta_{\text{per}}$ is the one-dimensional periodic lattice Laplacian.

The dynamics is modeled by the following Master equation, describing the evolution of the conditional joint probability density, $P(y,t)$, for this process:

$$\frac{\partial P(y,t)}{\partial t}=\int_{\mathbb{R}^N}\mu_G(dr)\{W(y|y-r)P(y-r,t) - W(y+r|y)P(y,t)\}, \tag{4}$$

where the probability rate for the transition from $y$ to $y'$ is given by

$$W(y'|y)=\mathcal{N}_\Omega\exp\{[\mathcal{H}(y)-\mathcal{H}(y')]/2k_BT\}, \tag{5}$$

which allows detailed balance to be satisfied, with the Gibbs distribution, proportional to $\exp[-\mathcal{H}(y)/k_BT]$, as the equilibrium distribution, and $\mathcal{N}_\Omega$ is some factor which can be absorbed into the time scale.

The jumps $r\in\mathbb{R}^N$ are distributed according to the multivariate Gauss measure, $\mu_G(dr)$, defined through the characteristic function

$$\int_{\mathbb{R}^N}\mu_G(dr)\exp(i\vartheta^Tr)=\exp(-\vartheta^TB\vartheta/2), \tag{6}$$

with $B$ being the covariance matrix (which may be singular); the element $B_{ij}$ describing how the jumps at locations $i$ and $j$ are correlated. Clearly, $B$ is always symmetric. It is important to note that if $\Sigma_{i=1}^N B_{ij}=0$ *for all* $j$ then the dynamics is *conserving*, i.e., $\Sigma_{i=1}^N y_i$ is kept constant. If the matrix $B$ does not satisfy this property then the dynamics is *nonconserving*.

Results are often expressed in terms of the spectra of the matrices $A$ and $B$: $Au(q)=\alpha(q)u(q)$, $Bu(q)=\beta(q)u(q)$, where $\alpha(q)$ and $\beta(q)$ are the eigenvalues corresponding to the eigenvector $u(q)$ with components $u_j(q)=N^{-1/2}\exp(iqj)$, $j=1,2,\ldots,N$, and $q\in S_N$ with $S_N\coloneqq\{0\leqslant q<2\pi: e^{iqN}=1\}$. The eigenvalues of $A$ are $\alpha(q)=2(1-\cos q)$.

The following choices are taken for $B$.

*Type I.* For nonconserving and uncorrelated jumps we take the diagonal matrix $B=\Gamma_2\mathbb{1}$, where $\Gamma_2$ is some constant which sets the time scale. For this case, the eigenvalue $\beta(q)=\Gamma_2$ for all $q$.

*Type II.* For local-conserving jumps, the simplest choice for $B$ is $B=\Gamma_4A$, with $A$ given as above. This corresponds to nearest-neighbor adatom hopping along the step edge. Here, we have $\beta(q)=\Gamma_4\alpha(q)$.

*Type III.* For non-local-conserving dynamics, by far the most complicated case to model, we take an approach motivated by ideas contained in Refs. [7,9]. The jumps in the interface at positions $i$ and $j$ get correlated as a result of diffusing adatoms making excursions in the lower terrace between these positions. The Ehrlich-Schwoebel potential is accounted for by preventing adatoms from jumping onto the upper terrace. On the lower terrace the adatoms are posited on sites, $(j,k)$, of a square lattice—the interstitial sites on the substrate—with $j$ (respectively $k\geqslant0$) being the lattice coordinate running parallel (respectively perpendicular) to the step edge located at $k=0$, which is assumed to be an absorbing barrier. The atoms jump between nearest-neighbor sites with the following jump-probability rate: $w_\parallel$ for sites parallel to the step edge with $k>1$; $w'_\parallel$ for parallel sites with $k=1$ (i.e., adjacent to the step edge); $w_\perp$ for sites perpendicular to the step edge with the final site having $k\geqslant1$; and $w'_\perp$ for jumping into site $k=0$, where the atom is *absorbed* into the

step edge [20]. The off-diagonal elements $B_{ij}$ of the covariance matrix are related to the probability, $w_{ji}(\tau_0)$, that an atom initially at position $i$ adjacent to the step edge passes through position $j$ next to the step edge at least once over a sufficiently large time interval, $\tau_0$, i.e., the coarse-grained time scale. If $f(j,k|j_0,k_0;t)$ is the probability that an atom starting at an initial position $(j_0,k_0)$ on the terrace touches the position $(j,k)$ *for the first time* after time $t$, then we have

$$w_{ji}(\tau_0) = \int_0^{\tau_0} dt\, f(j,1|i,1;t). \tag{7}$$

If $p(j,k|j_0,k_0;t)$ is the probability of finding an atom at $(j,k)$ after time $t$ given that it is initially at $(j_0,k_0)$, then

$$p(j,k|j_0,k_0;t) = \int_0^t d\tau\, f(j,k|j_0,k_0;\tau)p(j,k|j,k;t-\tau). \tag{8}$$

Using the shorthand $p=p(j,k|j_0,k_0;t)$, $p$ solves the following:

$$(\partial_t - w_\perp\Delta_\perp - w_\parallel\Delta_\parallel)p|_{k>1} = 0, \tag{9}$$

$$(\partial_t + w_\perp + w'_\perp - w'_\parallel\Delta_\parallel)p|_{k=1} = w_\perp p|_{k=2}, \tag{10}$$

with an initial condition $p|_{t=0}=\delta_{j_0,j}\delta_{k_0,k}$. Here, $\Delta_\parallel$ is the periodic lattice Laplacian ($=\Delta_{\rm per}$) acting on $j$ and $\Delta_\perp$ is the lattice Laplacian acting on $k$ [21]. The typical time scale of the step-edge motion (seconds) is much larger than that of the diffusive motion of individual atoms on the terrace (nanoseconds). Thus, we can set the coarse-grain scale $\tau_0$ to be arbitrarily large, and propose that the off-diagonal elements of $B$ are given by $B_{ij}=-w_{ij}(\infty)-w_{ji}(\infty)$ for $i\neq j$. The diagonal elements follow by imposing the condition for conservation, $\Sigma_{i=1}^N B_{ij}=0$, for all $j$. Equations (9) and (10) can be treated using spectral methods from which, by Laplace transforming (8)), $w_{ji}(\infty)$ and the matrix $B$ can be obtained. Results are more simply expressed in spectral form

$$\beta(q) = \frac{K_N\Upsilon[\alpha(q)]}{w'_\perp + \Upsilon[\alpha(q)]}, \tag{11}$$

where

$$\Upsilon(\alpha) := (w_\parallel\alpha)^{1/2}\left(w_\perp + \tfrac{1}{4}w_\parallel\alpha\right)^{1/2} + \left(w'_\parallel - \tfrac{1}{2}w_\parallel\right)\alpha, \tag{12}$$

and [22]

$$K_N^{-1} = \frac{1}{2N}\sum_{q\in S_N}\frac{w'_\perp}{w'_\perp + \Upsilon[\alpha(q)]}. \tag{13}$$

This form for $\beta(q)$ implies that the off-diagonal elements of the Toeplitz matrix, $B$, have the following properties: Writing $B_{i,i+n}=-b_{|n|}$, we have (i) $\lim_{N\to\infty}b_n\sim n^{-2}$ as $n\to\infty$ (provided $w'_\perp>0$); (ii) $b_n\to 2$ as $w'_\perp\downarrow 0$ for all $1\leqslant n<N$ [since $w_{ji}(\infty)=1$ when $w'_\perp=0$ due to recurrence of random walkers off a reflecting barrier].

We now apply the van Kampen expansion for large $\Omega$ [19]. This starts by writing

$$y(t) = \Omega\varphi(t) + \Omega^{1/2}\tilde{y}(t), \tag{14}$$

where $\tilde{y}(t)\in\mathbb{R}^N$ is the fluctuation about the mean location, $\varphi(t)\in\mathbb{R}^N$, of the step edge. With the initial condition $\varphi(0)=\varphi^0$ and $\tilde{y}(0)=0$ we obtain the following (deterministic) macroscopic law for $\varphi(t)$:

$$d\varphi/dt \approx -\tfrac{1}{2}\tilde{\sigma}BA\varphi, \tag{15}$$

to leading order in $\varphi$. The linearization leading to (15) is consistent with the quadratic approximation of $\mathcal{H}(y)$ in Eq. (3). However, one can easily construct the full *nonlinear* macroscopic law, without making these approximations, which is useful, for example, in the study of thermal smoothing dynamics of some initial shape $\varphi^0$. The fluctuating vector $\tilde{y}(t)$ is distributed according to the conditional joint probability density $\tilde{P}=\tilde{P}(\tilde{y},t)$, that satisfies the following Fokker-Planck equation:

$$\frac{\partial\tilde{P}}{\partial t} = \frac{1}{2}\tilde{\sigma}\frac{\partial}{\partial\tilde{y}_i}[(BA\tilde{y})_i\tilde{P}] + \frac{1}{2}B_{ij}\frac{\partial^2\tilde{P}}{\partial\tilde{y}_i\,\partial\tilde{y}_j}, \tag{16}$$

using the summation convention for repeated indices and, again, the terms of higher order in $\varphi$ are neglected.

The main result, obtained from solving Eq. (16), is the mean-squared width of an initally flat interface ($\varphi^0=\mathbf{0}$),

$$w^2(t;N) = \langle y_{\rm cm}^2(t)\rangle + \Omega\sum_{q\in S_N\setminus\{0\}}\frac{1-\exp[-\tilde{\sigma}\alpha(q)\beta(q)t]}{N\tilde{\sigma}\alpha(q)}, \tag{17}$$

where $y_{\rm cm}(t)=N^{-1}\Sigma_{i=1}^N y_i(t)$, the "center of mass" of the interface, has $\langle y_{\rm cm}^2(t)\rangle=\Omega\beta(0)t/N$, which is nonzero *only* for type I [since $\beta(0)\neq 0$ only for type I]. From a different starting point and physical context, a result similar to Eq. (17), for type I only, was reported in Ref. [14] but with fixed-end boundary conditions, $y_1=y_N=0$ (as opposed to periodic), giving $\langle y_{\rm cm}^2(t)\rangle=0$.

The value of the dynamic exponent, $z$, was found to depend on the type of dynamics as before. Thus, in the following formulas, dynamics of types I, II and III corresponds to having $z=2$, 4, and 3, respectively. For all types of dynamics, and for a given $N$, $w^2(t;N)$ grows through three distinct regimes in the following order.

*a. Initial diffusive growth*. For early time, one finds that $w^2\sim t$ as $t\to 0$, and the step-edge displacements $y_i$ fluctuate like *independent* random walkers.

*b. Subdiffusive late-stage roughening*. This is for $t\to\infty$ and $t/N^z\to 0$ where one finds $w^2\sim t^{1/z}$. Here, the interface is still far from equilibrium but time has progressed long enough for correlations to develop between distant step-edge displacements with the interfacial correlation length $\xi_\parallel\sim t^{1/z}$ in this regime.

*c. Equilibration*. This occurs as $t/N^z\to\infty$ and $N\to\infty$ so that at these *very* late times the interface achieves its equilibrium roughness, $w^2-\langle y_{\rm cm}^2\rangle\sim N$.

The transition from regimes *a* to *b* is best understood by considering an interface of *infinite* extent. With the defini-

tion, $w(t) \coloneqq \lim_{N\to\infty} w(t;N)$, the $N\to\infty$ limit of Eq. (17) gives the integral

$$w^2(t) = \frac{\Omega}{2\pi}\int_0^{2\pi} dq \frac{1-\exp[-\tilde{\sigma}\alpha(q)\beta(q)t]}{\tilde{\sigma}\alpha(q)}, \qquad (18)$$

as the crossover function between regimes $a$ and $b$; the regimes themselves being identified from the following limits [with $\Gamma(\cdot)$ denoting the gamma function]:

$$w^2(t)/\Omega \approx \begin{cases} \Gamma_z^0 t & \text{as } t \to 0, \\ \dfrac{1}{\pi\tilde{\sigma}}\Gamma(1-1/z)(\tilde{\sigma}\Gamma_z t)^{1/z} & \text{as } t \to \infty . \end{cases} \qquad (19)$$

For type III, $\beta(q)$ in Eq. (18) is given by Eq. (11) but with $K_N$ replaced by $K_\infty \coloneqq \lim_{N\to\infty} K_N$. The time scales are set by $\Gamma_z$ and $\Gamma_z^0$, where $\Gamma_2^0=\Gamma_2$, $\Gamma_4^0=2\Gamma_4$, $\Gamma_3^0=K_\infty-2$, and $\Gamma_3=K_\infty\sqrt{w_\parallel w_\perp}/w'_\perp$. Note, in particular, that $w'_\perp$ (adatom hopping rate into the step edge) has a strong influence on time scales. This is because the lower $w'_\perp$ is, the further adatoms can hop in the lower terrace before being adsorbed into the step edge, an effect which enhances correlations between interfacial jumps and is quantified in the following limiting behavior: $K_\infty \sim 1/[w'_\perp \ln(1/w'_\perp)]$ as $w'_\perp \downarrow 0$ and $K_\infty = 2 + O(1/w'_\perp)$ as $w'_\perp \to \infty$. We stress that lattice affects are crucial in getting the correct functional form of the crossover function in Eq. (18); a continuum description (with an *ad hoc* "momentum" cutoff) will not suffice. For type I, it is possible to express $w^2(t)$ explicitly in terms of the modified Bessel functions $I_0(\cdot)$ and $I_1(\cdot)$ [14].

To understand the crossover from regimes $b$ to $c$, the scaling limit, $t\to\infty$, $N\to\infty$ with $t/N^z$ finite, is taken, giving

$$w^2(t;N) - \langle y_{\rm cm}^2(t)\rangle \approx \frac{N\Omega}{2\tilde{\sigma}}\Phi_z(\tilde{\sigma}\Gamma_z t/N^z), \qquad (20)$$

where the crossover scaling function is

$$\Phi_z(x) = \frac{1}{6} - \sum_{n=1}^{\infty} \frac{\exp[-(2\pi n)^z x]}{(\pi n)^2}. \qquad (21)$$

Since both length and times are large throughout this crossover, similar scaling functions can be derived from continuum theories although care has to be taken to ensure the correct prefactors and metric factors in Eq. (20), a somewhat haphazard task when working directly from continuum theories. Note that

$$\Phi_z(x) = -2x\delta_{z,2} + \frac{2}{\pi}\Gamma(1-1/z)x^{1/z} + \cdots, \qquad (22)$$

as $x\to 0$ (i.e., regime $b$), implying that the $t/N^z \to 0$ limit of Eq. (20) coincides exactly with the $t\to\infty$ limit of Eq. (19), as required.

To summarize, we have developed a lattice theory of step dynamics on vicinal surfaces which should go some way in capturing microscopic and atomistic effects. The methods used will also prove valuable in many other applications concerning interfacial dynamics.


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[20] The jump rates are given by an Arrhenius equation: e.g., $w_\parallel = \nu_\parallel e^{-\Delta E_\parallel/k_{\rm B}T}$, where $\nu_\parallel$ is the attempt frequency and $\Delta E_\parallel$ is the energy barrier for the jump. Analogous expressions hold for $w'_\parallel$, $w_\perp$, and $w'_\perp$.
[21] Note that $\Delta_\perp p|_k \coloneqq p|_{k+1} + p|_{k-1} - 2p|_k$ and similarly for $\Delta_\parallel$, the latter having a period-$N$ boundary condition.
[22] It is important to note that $w'_\perp + \Upsilon[\alpha(q)]$ can never be negative and is equal to zero *only* at $q=0$ when $w'_\perp = 0$ where the barrier becomes reflecting.