Stochastic equations of motion for epitaxial growth

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We report an analytic derivation of the Langevin equations of motion for the surface of a solid that evolves under typical epitaxial-growth conditions. Our treatment begins with a master-equation description of the microscopic dynamics of a solid-on-solid model and presumes that all surface processes obey Arrhenius-type rate laws. Our basic model takes account of atomic deposition from a low-density vapor, thermal desorption, and surface diffusion. Refinements to the model include the effects of hot-atom knockout processes and asymmetric energy barriers near step edges. A regularization scheme is described that permits a (nonrigorous) passage to the continuum limit when the surface is rough. The resulting stochastic differential equation for the surface-height profile generically leads to the behavior at long length and time scales first described by Kardar, Parisi, and Zhang [Phys. Rev. Lett. **56**, 889 (1986)] (due to desorption). If evaporation is negligible, the asymptotic behavior is characteristic of a linear model introduced by Edwards and Wilkinson [Proc. R. Soc. London, Ser. A **381**, 17 (1982)] (due to asymmetric step barriers and/or knockout events). If the latter are absent as well, the surface roughness is determined by an equation independently analyzed by Villain [J. Phys. I **1**, 19 (1991)] and Lai and Das Sarma [Phys. Rev. Lett. **66**, 2348 (1991)] (which includes only deposition and site-to-site hopping). The consequences of reflection-symmetry breaking in the basic microscopic processes are discussed in connection with step-barrier asymmetry and Metropolis kinetic algorithms.

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I. INTRODUCTION AND BACKGROUND

Epitaxial growth is a nonequilibrium process whereby a solid interface advances through the addition of new material that crystallizes in a manner dictated by the lattice structure of the underlying solid. In the laboratory, this phenomenon can be made to occur in many different ways and a complete understanding of its varied manifestations is not close at hand [1]. The most comprehensive discussions of the subject tend to be organized around the two principal experimental growth techniques in use today: molecular-beam epitaxy (MBE) [2] and organometallic-vapor-phase epitaxy [3]. Nonetheless, the basic features common to both are easy to identify and early theoretical studies established cluster nucleation [4] and surface diffusion to preexisting defects [5] as the primary processes which limit growth. The importance of these surface kinetic processes is well established by now [6] and their effect on, e.g., surface morphology and growth shapes has received considerable attention using analytic methods [7].

Computational methodologies have contributed significantly to our understanding of epitaxial growth as well. Monte Carlo simulations using simple lattice models were initiated over 20 years ago [8] and continue unabated today [9]. For the case of molecular-beam epitaxy of GaAs(100), *quantitative* agreement can be obtained between calculated surface-step densities and the signal from reflection high-energy electron diffraction studies in the transient regime [10]. For future reference, we note that this entire body of research presumes microscopic surface kinetic processes that obey an Arrhenius-type rate law—a supposition that is well established both by extensive experimental studies [11] and by full molecular dynamics simulations [12] for the surfaces considered.

The theoretical studies noted above were designed and executed largely for the purposes of direct comparison to experimental epitaxial growth. But, a totally distinct series of Monte Carlo computer simulations of vapor deposition have appeared over the past few years [13] with a very different purpose in mind. Here, interest focuses on testing a notion arising from the study of fractal deposits [14] to the effect that shot noise associated with deposition induces a so-called kinetic roughening [15] of the growing surface. More precisely, if $h(\mathbf{x},t)$ denotes the surface height, it is postulated that the variance $W(L,t)$ satisfies a dynamic scaling [16] hypothesis:

$$W(L,t)=[\langle h^2\rangle-\langle h\rangle^2]^{1/2}\sim L^{\alpha}f(t/L^{\alpha/\beta})\ , \qquad (1)$$

where L is the lateral viewing scale and the scaling function $f(x)\sim x^{\beta}$ for $x\ll 1$ and $f(x)\rightarrow$const for $x\gg 1$. The exponents α and β are supposed to be universal in the sense that their values are expected to depend only on

symmetry and dimensionality rather than on microscopic details. This hypothesis, if true, is very appealing because vapor deposition would then take its place among other driven, nonequilibrium problems as an example of a system that evolves to a nontrivial scale-invariant structure under the influence of input noise [17].

A great stimulus to the analysis of Monte Carlo simulation data from this point of view is the hope that the exponents so obtained might provide a clue to the form of an effective equation of motion for the surface profile valid at long length and time scales. For example, a lattice simulation where every deposited particle is permitted to investigate nearest-neighbor sites and irreversibly settle onto the site with lowest height is observed to obey (1) with particular exponents [18]. The model is presumed to be described by a linear, stochastic, partial differential equation derived by Edwards and Wilkinson (EW) [19] that satisfies (1) with the same exponents [20]. Much subsequent research involving simulations of various "toy" kinetic schemes has precisely this flavor [21]. One class of such schemes generates surfaces that exhibit dynamic scaling with exponents identical to those predicted for a nonlinear Langevin equation proposed by Kardar, Parisi, and Zhang (KPZ) [22]. The exponents differ from those of the EW model and thus the models do not belong to the same universality class.

The question naturally arises: does "real" epitaxial growth exhibit kinetic roughening? If so, to what universality class does it belong? To date, this question has been addressed theoretically from two points of view. The first has been described above: exponents are extracted from simulations that purport to have explicit relevance to epitaxial growth (typically molecular-beam epitaxy) and related to various stochastic equations of motion [23,24]. The present authors have engaged in this exercise as well [25]. Alternatively, the question can be investigated from a purely phenomenological perspective. By far the most valuable discussion of this sort can be found in a trenchant article by Villain [26], who suggests that the long-time and long-wavelength morphological evolution of a growing epitaxial film is best described by the following nonlinear stochastic partial differential equation:

$$\frac{\partial h}{\partial t}=\nu\nabla^2 h+\lambda(\nabla h)^2+K\nabla^2(\nabla^2 h)+\sigma\nabla^2(\nabla h)^2+F+\eta\ . \tag{2}$$

Here, F is the net deposition flux (average deposition flux minus average desorption flux) and $\eta(x,t)$ is a Gaussian random variable with zero mean and shot-noise-type covariance. This formula contains the EW equation ($\lambda=K=\sigma=0$) and the KPZ equation ($K=\sigma=0$) as special cases. Independent proposals for a "conserved" KPZ equation $\nu=\lambda=0$ [27] and the special case $\sigma=0$ [28] can be found in the literature as well. One expects that the presence or absence of the various terms in (2) depends on the presence or absence of various physical processes. For example, there is broad agreement that, during growth, the terms proportional to ν and λ are present whenever thermal desorption is operative and that the terms proportional to K and σ can arise from surface diffusion.

On the other hand, in the absence of desorption, it is fair to say that the status of the coefficient ν remains an unsettled issue. Villain argues [26] that $\nu\neq 0$ if asymmetric energy barriers are present in the vicinity of step edges [29]. In that case, one generates the Laplacian term in (2) with a coefficient proportional to the flux F. On the basis of simulation studies that employ Metropolis-type kinetics, it has been claimed [24] that similar behavior is found for ν even for pure surface diffusion without special step-edge barriers. Kang and Evans [30] suggest that this term arises whenever there is "lateral coupling due to realistic adsorption site geometries and deposition dynamics." As an example of the latter, they cite so-called *knockout* processes [31] that involve the replacement of an existing step-edge atom by a freshly deposited atom and thus have the effect of local downward relaxation. The resolution of this point is of some theoretical interest because the asymptotic scaling behavior of the surface roughness will be dominated by this term (and λ) if present. On the other hand, experiments likely will be dominated by crossover effects [32] so that there is a need to at least estimate the sign and relative magnitude of the various coefficients that enter (2).

The purpose of the present paper is to present an analytic derivation of equations of motion for the surface of a single crystal that grows under typical epitaxial conditions. Our treatment begins with a master-equation description of Arrhenius-type microscopic surface kinetic processes assigned to a square lattice solid-on-solid model. Our basic model takes account of atomic deposition, thermal desorption, and surface (height) diffusion. We consider as well the effects of hot-atom downward mobility (knockout) for some freshly deposited atoms and asymmetric barriers to adatom motion in the vicinity of step edges. Some care is taken to assess the extent to which the forms we adopt for these processes represent an adequate model of "real" epitaxial surface dynamics.

Given the transition rates between configurations of a solid-on-solid lattice associated with the foregoing, we make us of contemporary developments in the theory of nonequilibrium processes to pass directly to a set of nonlinear lattice Langevin equations and their associated noise covariances. With a few caveats, direct numerical integration of the latter may be expected to provide an exact alternative to Monte Carlo simulations of the growth model as defined. We next introduce a regularization procedure that permits a (nonrigorous) passage to the continuum limit when the surface is rough. The low-order terms in the resulting stochastic partial differential equation of motion have the form of Eq. (2) with explicit expressions for the coefficients that identify their origin in the basic microscopic processes.

Although the methodology sketched above is similar to one employed previously by Plischke and co-workers [33,34], our conclusions differ. This leads us to analyze the consequences of reflection-symmetry breaking in surface kinetic processes and to conclude that Metropolis-type kinetic schemes can exhibit features not reflective of true surface dynamics. The discussion of our basic model

simplifies and slightly modifies brief accounts of this work presented elsewhere [35]. It is included here for the sake of completeness and clarity. The hot-atom and asymmetric step-edge barrier computations are entirely new. Taken together, our results largely confirm Villain's phenomenological analysis and are in accord with all existing Monte Carlo simulations of which we are aware.

II. THE BASIC MODEL: DEPOSITION, DESORPTION, AND SURFACE DIFFUSION

In this paper we adopt a simple solid-on-solid (SOS) model [36] where a column of height h_j is associated with the jth site of a one-dimensional substrate with lattice constant a. We have shown elsewhere [35] that the generalization to the isotropic two-dimensional case is straightforward and introduces no new physics. On the other hand, the model embodies the assumptions that (i) the growing film has simple cubic symmetry and (ii) vacancies and overhangs are forbidden. These premises are nontrivial and subject to criticism [24,30]. While in principle our methods can be applied to the more general case, the algebraic complications that arise are daunting. We proceed with these caveats duly noted.

Every configuration of the SOS surface $\mathbf{H}$ is specified completely by the collection of column height variables: $\mathbf{H}=\{h_1,h_2,\ldots\}$. The joint probability that the surface has configuration $\mathbf{H}$ at time t is denoted $P(\mathbf{H};t)$. The evolution of this quantity from a particular initial configuration is governed by a birth-death-type master equation

$$\frac{\partial P(\mathbf{H};t)}{\partial t}=\sum_{\mathbf{H}'} W(\mathbf{H}',\mathbf{H})P(\mathbf{H}';t)-\sum_{\mathbf{H}'} W(\mathbf{H},\mathbf{H}')P(\mathbf{H};t)\ , \tag{3}$$

where the sum over configurations $\mathbf{H}'$ is to be interpreted as the joint summation over the heights on every site of the lattice. The quantity $W(\mathbf{H},\mathbf{H}')$ denotes the transition rate from a configuration $\mathbf{H}$ to a configuration $\mathbf{H}'$. The rules that determine these rates reflect the choices one makes for elementary microscopic processes.

For the case of deposition, we suppose that particles ("atoms") are delivered randomly to the surface from a ballistic beam parallel to the SOS columns. Effects associated with diffusive transport in the gas phase [37] and surface self-shadowing [38] are ignored. Moreover, atoms that arrive at the surface are imagined to accommodate immediately and to stick to the top of the column they impinge upon. A refinement that models some effects associated with the heat of condensation will be considered in Sec. III. For the simple case considered here, deposition changes the configuration of the lattice by adding atoms randomly at an average rate of τ^{-1}, where τ is the layer completion time. Thus the transition rate is nonvanishing only between configurations $\mathbf{H}$ and $\mathbf{H}'$ for which $h_j'=h_j+a$ for any site j, with the height of all other lattice sites remaining the same:

$$W_{\rm dep}(\mathbf{H},\mathbf{H}')=\frac{1}{\tau}\sum_k\left[\delta(h_k',h_k+a)\prod_{j\neq k}\delta(h_j',h_j)\right]\ . \tag{4}$$

Arrhenius-type rate expressions are chosen to model the elementary atomistic processes of thermal desorption and surface diffusion. We pause to justify this choice because it appears to be atypical in the statistical physics literature of growth phenomena. First, as noted earlier, there is overwhelming experimental [11] and theoretical [12] evidence in the surface science literature that these processes obey barrier-crossing dynamics of this sort. Although typically merely a mean-field approximation to the true microscopics, various corrections to the basic formulation are well understood [39] and do not alter the essential point that the rates are determined by a so-called *transition state* energy barrier that is not necessarily associated with either the initial state or the final state. Second, it is known [40] that asymptotic surface properties can be affected if, e.g., a Metropolis-type rule [41] is substituted. We demonstrate this explicitly for the present problem in Sec. VI.

Consistent with the foregoing, we model the rate for an atom to desorb from site j by

$$\Omega_j=k_0'\exp[-\beta(U_S+n_jE_N)]=\frac{1}{\tau'}\exp(-\beta n_jE_N)\ , \tag{5}$$

where $\beta=1/k_BT$ and the prefactor k_0' is an attempt frequency that may be taken to be the vibrational frequency of an adatom. The desorption barrier is a configuration-dependent quantity parametrized by a contribution U_S from the substrate and a contribution E_N from each of the n_j lateral nearest neighbors. Note that the choice (5) manifestly satisfies detailed balance if (as is appropriate) we identify U_S+nE_N as the negative of the binding energy of a surface atom with n lateral nearest neighbors. Since a desorption event reduces the height of a column by one lattice unit, the transition rate for this process between configurations $\mathbf{H}$ and $\mathbf{H}'$ can be written

$$W_{\rm des}(\mathbf{H},\mathbf{H}')=\sum_k\left[\Omega_k\delta(h_k',h_k-a)\prod_{j\neq k}\delta(h_j',h_j)\right]\ . \tag{6}$$

To complete our basic model, we treat surface diffusion as a process whereby an atom hops away from site j at a rate

$$\Lambda_j=k_0\exp[-\beta(E_S+n_jE_N)]=\frac{1}{\tau_0}\exp(-\beta n_jE_N)\ . \tag{7}$$

The substrate contribution to the barrier E_S is guaranteed to be smaller in magnitude than U_S and the prefactors k_0 and k_0' are not in general equal. One checks that detailed balance is satisfied as before. To maintain the SOS constraint, we further stipulate that an atom hopping at the rate (7) lands (with equal probability) onto the *top* of a nearest-neighbor column. Our model thus exhibits height diffusion rather than true surface diffusion [24]. On the other hand, extensive Monte Carlo simulations performed by us reveal that the fraction of particles that make multiple-height jumps is *extremely* small for the physically relevant case of a two-dimensional substrate at typical experimental growth rates and temperatures. We therefore accept the possible error introduced in the interest of making analytic progress.

The above makes clear that a surface diffusion event reduces the height of a given column by one lattice unit and increases the height of a nearest-neighbor column by one lattice unit. The transition rate between such configurations thus takes the form

$$W_{\text{diff}}(\mathbf{H},\mathbf{H}')=\frac{1}{2}\sum_k \left\{ [\Lambda_k \delta(h_k',h_k-a)\delta(h_{k+1}',h_{k+1}+a)+\Lambda_{k+1}\delta(h_k',h_k+a)\delta(h_{k+1}',h_{k+1}-a)] \prod_{j\neq k,k+1} \delta(h_j',h_j) \right\} . \tag{8}$$

The factor of $\frac{1}{2}$ in (8) arises from the assumption of equal likelihood of hopping to the left or to the right (isotropic hopping). At this point, one could substitute the transition rates in (4), (6), and (8) into (3) to arrive at a complete master-equation description of the assumed dynamical model. We have done this elsewhere [35] and it is a useful step if one wishes to compute deterministic equations of motion for quantities such as the average height $\langle h_k(t)\rangle$ and the correlation function $\langle h_k(t)h_j(t)\rangle$ [33,34]. Here, however, we choose to proceed directly to a Langevin equation for each stochastic column height variable and the associated noise correlations. The traditional route to this goal involves the passage from a truncation of the Kramers-Moyal form of the master equation to a Fokker-Planck equation in appropriately scaled variables and finally to a linear Langevin equation [42]. Instead, we make use of a limit theorem [43] (not discussed in Ref. [42]) that is valid for nonlinear problems such as ours.

Our application follows the clear exposition of the matter by Fox and Keizer [44] who show that one requires only the first and second moments of the transition rates:

$$K_i^{(1)}=\sum_{\mathbf{H}'}(h_i'-h_i)W(\mathbf{H},\mathbf{H}') , \tag{9}$$

$$K_{ij}^{(2)}=\sum_{\mathbf{H}'}(h_i'-h_i)(h_j'-h_j)W(\mathbf{H},\mathbf{H}') . \tag{10}$$

Then, provided the system size is large and the intrinsic fluctuations are not too large (as might occur if the deterministic dynamics were chaotic), the equations we seek are

$$\frac{dh_i}{dt}=K_i^{(1)}+\eta_i(t) , \tag{11}$$

where the Gaussian random variables η_i have zero mean

$$\langle \eta_i(t)\rangle=0 \tag{12}$$

and covariances

$$\langle \eta_i(t)\eta_j(t')\rangle=K_{ij}^{(2)}\delta(t-t') . \tag{13}$$

It is straightforward to calculate the required moments of the transition rates for deposition (4), desorption (6), and surface diffusion (8). The results can be written as

$$K_i^{(1)}=a^{-1}D_S\Delta^2\lambda_i-\frac{a}{\tau'}\lambda_i+\frac{a}{\tau} \tag{14}$$

and

$$K_{ij}^{(2)}=a^2\left[\frac{\lambda_i}{\tau'}+\frac{1}{\tau}\right]\delta_{ij}+D_S[\delta_{ij}\Delta^2\lambda_i-(\lambda_i+\lambda_j)\Delta^2\delta_{ij}] . \tag{15}$$

In these formulas, δ_{ij} is the Kronecker delta, D_S is the diffusion constant

$$D_S=\frac{a^2}{2\tau_0} , \tag{16}$$

and

$$\lambda_i=\exp(-\beta n_i E_N) . \tag{17}$$

The discrete second derivative operator defined by

$$\Delta^2\lambda_i=\lambda_{i+1}-2\lambda_i+\lambda_{i-1} \tag{18}$$

acts on the first index of δ_{ij} in (15). Note that the covariances include contributions from desorption and surface diffusion in addition to the deposition-induced shot noise. The stochastic equations defined by (14) and (15) constitute one of the central results of this paper. Their physical meaning and consequences for the problem of kinetic roughening will be discussed in Sec. V.

III. HOT-ATOM EFFECTS

We consider in this and the next sections two refinements to our basic model. The first amounts to a generalization of the simple deposition dynamics assumed in the basic model. According to (4), every atom sticks to the top of whatever column it impinges upon. As noted in Sec. I, there is theoretical evidence [31] that the heat of condensation can induce atom exchange mechanisms if a freshly deposited atom lands upon a step-edge site. In particular, one observes "knockout" processes [30] where such a "hot" atom both laterally displaces and occupies the original site of the atom it landed upon. One gains confidence in the likelihood of such a scenario from the experimental observation of very similar exchange processes at step edges that involve only thermally diffusing species [45].

For deposition onto SOS step-edge sites, the hot-atom knockout process is indistinguishable from a downward bias deposition process where the arriving atom "falls over" the edge and attaches to the top of a nearest-neighbor column. The local rules needed to incorporate this effect are similar to those used in a Monte Carlo simulation noted earlier [18] except that the final deposition site is chosen randomly as one of the nearest-neighbor columns when both are lower than the original deposition site. Thus, when knockout is present, the transition rate for deposition (4) is replaced by

$$W(\mathbf{H},\mathbf{H}')=\tau^{-1}\sum_k \left\{ w_k^{(1)}\delta(h'_k,h_k+a)\prod_{j\neq k}\delta(h'_j,h_j)+w_k^{(2)}\delta(h'_{k-1},h_{k-1}+a)\prod_{j\neq k-1}\delta(h'_j,h_j) \right.$$
$$\left. +w_k^{(3)}\delta(h'_{k+1},h_{k+1}+a)\prod_{j\neq k+1}\delta(h'_j,h_j) \right\}, \tag{19}$$

where

$$w_k^{(1)}=\Theta(h_{k+1}-h_k)\Theta(h_{k-1}-h_k)\ , \tag{20}$$

$$w_k^{(2)}=\Theta(h_{k+1}-h_k)[1-\Theta(h_{k-1}-h_k)]+\tfrac{1}{2}[1-\Theta(h_{k+1}-h_k)][1-\Theta(h_{k-1}-h_k)]\ , \tag{21}$$

and

$$w_k^{(3)}=\Theta(h_{k-1}-h_k)[1-\Theta(h_{k+1}-h_k)]+\tfrac{1}{2}[1-\Theta(h_{k+1}-h_k)][1-\Theta(h_{k-1}-h_k)]\ . \tag{22}$$

In these formulas, the unit step function $\Theta(x)$ is defined by

$$\Theta(x)=\begin{cases}1 & \text{if } x\geq 0\\ 0 & \text{if } x<0\ .\end{cases} \tag{23}$$

The factors (20)–(22) merely test for the relative heights of nearest-neighbor sites so that the identity

$$w_k^{(1)}+w_k^{(2)}+w_k^{(3)}=1 \tag{24}$$

guarantees that the average deposition rate per site remains equal to τ^{-1}. From these, one immediately writes down the corresponding transition moments

$$K_i^{(1)}=\frac{a}{\tau}[w_i^{(1)}+w_{i+1}^{(2)}+w_{i-1}^{(3)}]\ , \tag{25}$$

$$K_{ij}^{(2)}=a\delta_{ij}K_i^{(1)} \tag{26}$$

needed to construct the equation of motion including knockout events. If desired, one can specify that such events occur only with a probability p.

IV. ASYMMETRIC STEP-EDGE BARRIERS

The second generalization of our basic model to be considered pertains to the well-known experimental fact [29] that the energy barrier for a diffusing adatom to approach a step edge from above generally differs from the energy barrier to approach the step edge from below. The barrier asymmetry can be unobservably small [46], but when appreciable it engenders a surprising variety of theoretical consequences [47]. In the context of surface roughening, the effect has been examined both phenomenologically [26] and with Monte Carlo computer simulations [48].

To fix ideas, consider a recent field-ion microscope study of iridium atom diffusion on the Ir(111) surface by Wang and Ehrlich [29]. The adiabatic potential energy surface in the vicinity of a step evidently is quite complex for this surface of this material. Here we focus only on the fact that, relative to the diffusion barrier on a flat terrace, an adatom experiences an enhanced (reduced) barrier to join the step edge from a nearest-neighbor site above (below) the step (Fig. 1). We denote the magnitude of the *change* in the transition state barrier to step attachment from the upper (lower) terrace as E_U (E_L) so that $E_U>0$ and $E_L<0$ on Ir(111).

Care must be taken when these effects are incorporated into a transition rate to replace (8) in order to guarantee that detailed balance is respected. For example, we require the change E_U to be present both for an adatom about to jump down and join a step edge from the upper terrace and for a detaching step-edge atom about to jump up onto the upper terrace. Similarly, we require the change E_L to be present both for an atom about to join the step edge from the lower terrace and for a step-edge atom about to detach onto the lower terrace. The final result is a modification of (8) to

$$W_{\text{diff}}(\mathbf{H},\mathbf{H}')=\tfrac{1}{2}\sum_k \left\{ [\lambda_k \exp(-\beta\mathcal{E}_k^+)\delta(h'_k,h_k-a)\delta(h'_{k+1},h_{k+1}+a) \right.$$
$$\left. +\lambda_{k+1}\exp(-\beta\mathcal{E}_{k+1}^-)\delta(h'_k,h_k+a)\delta(h'_{k+1},h_{k+1}-a)]\prod_{j\neq k,k+1}\delta(h'_j,h_j) \right\}, \tag{27}$$

where

$$\mathcal{E}_k^+=E_U[\Theta(h_{k+1}-h_k)+\Theta(h_k-h_{k+1}-2a)]$$
$$+E_L[\Theta(h_{k+2}-h_{k+1}-a)\Theta(h_k-h_{k-1}-a)$$
$$+\Theta(h_{k-1}-h_k)\Theta(h_{k+1}-h_{k+2})]$$
$$\times\delta(h_k,h_{k+1}+a) \tag{28}$$

and

$$\mathcal{E}_k^-=E_U[\Theta(h_{k-1}-h_k)+\Theta(h_k-h_{k-1}-2a)]$$
$$+E_L[\Theta(h_{k-2}-h_{k-1}-a)\Theta(h_k-h_{k+1}-a)$$
$$+\Theta(h_{k+1}-h_k)\Theta(h_{k-1}-h_{k-2})]$$
$$\times\delta(h_k,h_{k-1}+a)\ . \tag{29}$$

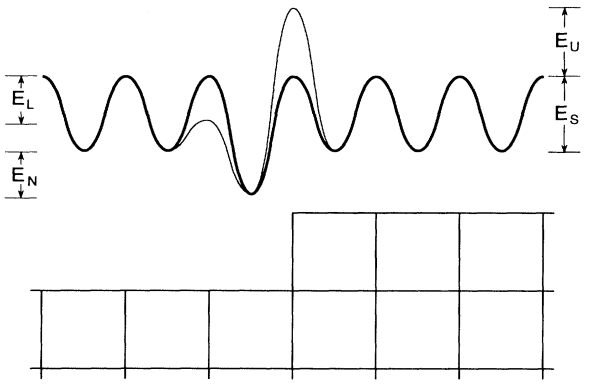

FIG. 1. Model potential energy surface experienced by an added "test" atom as it moves across the illustrated SOS step geometry.

The Langevin equations that reflect the effects of asymmetric step-edge barriers follow immediately from the transition moments of (27):

$$a^{-1}K_i^{(1)}=\tfrac{1}{2}\Delta^+[\Lambda_i\exp(-\beta\mathcal{E}_i^-)]-\tfrac{1}{2}\Delta^-[\Lambda_i\exp(-\beta\mathcal{E}_i^+)], \tag{30}$$

$$\begin{aligned}2a^{-2}K_{ij}^{(2)}=&-[\Lambda_i\exp(-\beta\mathcal{E}_i^+)+\Lambda_i\exp(-\beta\mathcal{E}_i^-)]\Delta^2\delta_{ij}\\&-(\Delta^-\delta_{ij})\Delta^-[\Lambda_i\exp(-\beta\mathcal{E}_i^+)]\\&-(\Delta^+\delta_{ij})\Delta^+[\Lambda_i\exp(-\beta\mathcal{E}_i^-)]\ ,\end{aligned} \tag{31}$$

where the discrete left and right derivatives of an indexed quantity f_i are defined by

$$\Delta^+f_i=f_{i+1}-f_i\ ,\quad \Delta^-f_i=f_i-f_{i-1}\ . \tag{32}$$

The reader can verify that (30) and (31) reduce to the diffusion contributions to (14) and (15) whenever $\mathcal{E}_i^+$ and $\mathcal{E}_i^-$ vanish, e.g., for an isolated adatom on a flat terrace.

V. CONTINUUM EQUATIONS OF MOTION

Numerical integration of the lattice Langevin equations (11)–(13) with the various transition moments computed above provides a formally exact alternative to direct Monte Carlo simulations that employ the corresponding kinetic rules. Unfortunately, the computational labor is still considerable due to the complexity of the noise covariances. For this reason, it is appropriate to inquire whether the results we have obtained lend themselves to a passage to the continuum limit from which macroscopic physical consequences might be extracted. We have discussed one way to proceed in a previous publication [49]. There it is shown that the quantity λ_i defined in (7) can be rewritten

$$\lambda_i=\sum_{n=0}^{2}c_i(n)\exp(-n\beta E_N)\ , \tag{33}$$

where $c_i(n)$ denotes the probability that site i is surrounded by n lateral nearest neighbors. The transition moments of the basic model are *linear* in these variables so the continuum limit is straightforward to obtain. It turns out that the macroscopic variable $\lambda(x)$ that emerges can be identified with the chemical *activity* [50] of the surface. We have explored some of the consequences of this fact elsewhere [49,51].

In the present paper, we adopt a less exact procedure that has the virtue of leading directly to continuum stochastic equations of motion that can be analyzed by dynamic renormalization group methods [52]. The basic idea [33,34] is to *regularize* the lattice Langevin equation by replacing the various nonanalytic quantities that enter with analytic quantities and then to retain only the leading-order terms. Let us note immediately that by far the most crucial step in this procedure is the presumption that the discrete terrace and step morphology of the surface can be replaced by an analytic function $h(x)$. This can only be done with confidence if the surface is either thermally rough or kinetically rough. Since we confine ourselves here to surfaces far from equilibrium, the regularization we employ can be valid (if at all) only for an epitaxial interface *in motion*. In that case, asymptotic roughness is guaranteed [53]. Alternatively [26], one can assume that the surface is rough under these conditions and then check that the theory is self-consistent.

Our regularization procedure [35] proceeds in several steps. Note first that the number of lateral nearest neighbors of a SOS surface atom at site j can be written

$$n_j=\Theta(h_{j+1}-h_j)+\Theta(h_{j-1}-h_j)\ . \tag{34}$$

Then, since the exponential of a step function is also a step function but with a different step jump, viz.,

$$\exp[-\beta E_N\Theta(x)]=1-\gamma\Theta(x)\ , \tag{35}$$

with

$$\gamma=1-\exp(-\beta E_N)\ , \tag{36}$$

the lattice activity (17) that enters the transition moments for our basic model can be written

$$\lambda_i=[1-\gamma\Theta(\Delta^+h_i)][1-\gamma\Theta(-\Delta^-h_i)]\ . \tag{37}$$

Next, we approximate the step function by an analytic function, e.g., a shifted hyperbolic tangent, which is expanded in a Taylor series:

$$\Theta(x)\approx 1+\sum_{k=1}^{\infty}A_kx^k\ . \tag{38}$$

Inserting (38) into (37) of course generates an infinite series. Some of the terms can be rearranged exactly since, e.g.,

$$\Delta^2f_i=\Delta^+f_i-\Delta^-f_i\ . \tag{39}$$

We then fit an interpolating function $\hat{h}(x,t)$ through the points h_i so that

$$h_{i\pm1}(t)-h_i(t)=\sum_{n=1}^{\infty}\frac{(\pm a)^n}{n!}\frac{\partial^n\hat{h}(x,t)}{\partial x^n}\bigg|_{x=ia}\ . \tag{40}$$

The final step is a coarse-grained spatial average that replaces $\hat{h}(x,t)$ by a function $h(x,t)$ that is smooth at the

macroscopic scale.

If the foregoing can be justified, the continuum stochastic equation that corresponds to our basic Arrhenius model is obtained by retaining the first few terms in the expansion when (14) is inserted into (11):

$$\frac{\partial h}{\partial t}=\nu\frac{\partial^2 h}{\partial x^2}+\lambda\left[\frac{\partial h}{\partial x}\right]^2+\frac{\partial^2}{\partial x^2}\left[K\frac{\partial^2 h}{\partial x^2}+\sigma\left[\frac{\partial h}{\partial x}\right]^2\right]+F+\eta\ . \tag{41}$$

The coefficients are given by

$$\begin{aligned}\nu&=\frac{a^3}{\tau'}\gamma(1-\gamma)A_1\ ,\\ \lambda&=\gamma\frac{a^3}{\tau'}[2(1-\gamma)A_2+\gamma A_1^2]\ ,\\ K&=-\left[a^3D_S+\frac{1}{12}\frac{a^5}{\tau'}\right]\gamma(1-\gamma)A_1\ ,\\ \sigma&=-aD_S\gamma[2(1-\gamma)A_2+\gamma A_1^2]\ ,\\ F&=\frac{a}{\tau}-\frac{a}{\tau'}(1-\gamma)^2\ ,\end{aligned} \tag{42}$$

and to lowest order, the noise covariance matrix is

$$\langle\eta(x,t)\eta(x',t')\rangle=a^3\left[\left(\frac{1}{\tau}+\frac{1}{\tau'}\right)\delta(x-x')-2D_S\nabla^2\delta(x-x')\right]\delta(t-t')\ . \tag{43}$$

Our result (41) is seen to be identical to (2). Moreover, (42) demonstrates that ν and λ arise exclusively from desorption while surface diffusion contributes only to K and σ. Given the presence of shot noise in (43), we thus confirm previous calculations [33] and phenomenological arguments [26,27] to the effect that the generic scaling behavior of the surface roughness during epitaxial growth is determined by the KPZ equation (due to desorption) but that a "conserved" KPZ equation is relevant if evaporation can be neglected. To be consistent with the phenomenology, the regularization scheme must have $A_1>0$. Equally importantly, the discussion above (34) makes clear that every A_k must depend implicitly on F since it is the shot noise in (43) that guarantees roughness away from equilibrium. Indeed, the fact that both nonlinear terms rigorously are *absent* in equilibrium [51] implies a relationship between A_1 and A_2 when $F=0$.

The contribution of hot-atom knockout processes to the continuum equation of motion is obtained similarly from (20)–(22) and (25). For present purposes, the important result is that these downward bias events generate a Laplacian in the equation of motion with the coefficient

$$\nu_{\rm dep}=\frac{a^3}{\tau}A_1\ , \tag{44}$$

but do *not* contribute a KPZ nonlinearity. Inessential contributions to K and σ appear as well. Our naive regularization procedure applied to the case of asymmetric step barriers generates a Laplacian in the equation of motion as well. But in this case, the corresponding coefficient $\nu_{\rm step}$ is easily seen to take the form of an infinite series that explicitly involves every one of the quantities A_k in (38). Accordingly, a more sophisticated argument is required to extract the dependence of $\nu_{\rm step}$ on the microscopic parameters E_U and E_L.

For this purpose, we focus on the role of *left-right asymmetry* in the microscopic transition rates and suppose that surface diffusion alone is operative. It is evident from (8) that the transition rate for *any* description of site-to-site hopping can be written generically as

$$W(\mathbf{H},\mathbf{H}')=\sum_k W_k\ , \tag{45}$$

where W_k is the transition rate associated with each site. The latter of course depends on the heights in a neighborhood of the site k. Now decompose each site rate into a rate W_k^+ for a jump to the right and a rate W_k^- for a jump to the left so that $W_k=W_k^++W_k^-$. For reasons that will become clear below, we note that W_k^- and W_k^+ are related by $W_k^-=\mathcal{R}W_k^+$ where $\mathcal{R}$ is a reflection operator that transforms $h_{j\pm n}$ into $h_{j\mp n}$. Thus, neglecting the deposition and noise terms, the equation of motion takes the form

$$\frac{\partial h_k}{\partial t}=W_{k-1}^+-W_k^++W_{k+1}^--W_k^-\ . \tag{46}$$

To pass to the continuum limit, the finite differences in (46) are expanded in terms of analytic functions precisely as was done in (40). The final spatial coarse-grained average quantities $W^\pm$ we require are expressed most simply as

$$W^\pm[h(x,t)]=\frac{a}{L}\sum_{k\in x}W_k^\pm\ , \tag{47}$$

where the sum extends over the L/a sites collectively denoted by the macroscopic variable x. The result is

$$\frac{\partial h(x,t)}{\partial t}=a^2\nabla\{W^-[h(x,t)]-W^+[h(x,t)]\}+\cdots\ . \tag{48}$$

For any reasonable microscopic dynamics, the transition rate $W^+[h]$ can be expected to have the form

$$W^+[h]=A\nabla h+B(\nabla h)^2+\cdots+C\nabla^2 h+\cdots+D\nabla h\nabla^2 h+\cdots\ , \tag{49}$$

since there can be no dependence on the absolute height of the surface. $W^-[h]$ is obtained upon application of the reflection operator:

$$W^-[h]=-A\nabla h+B(\nabla h)^2+\cdots+C\nabla^2 h+\cdots-D\nabla h\nabla^2 h+\cdots\ . \tag{50}$$

To demonstrate that a Laplacian is present in the equa-

tion of motion, it is sufficient to show that

$$A=\left.\frac{\partial W^{+}[h]}{\partial h'}\right|_{h'=0}\neq 0\ , \tag{51}$$

where $h'=\nabla h$.

Let us now apply the foregoing to the case of asymmetric barriers to adatom attachment to steps. For simplicity, consider first the case where $E_U\neq 0$ and $E_L=0$. The derivative in (51) is computed as follows. First calculate $W^{+}[h]$ for a segment of surface of length L with $N+1$ up steps and n down steps, for which the slope $h'=a/L=\alpha$. Then do the same for a segment of length L with N up steps and $N+1$ down steps, for which $h'=-a/L=-\alpha$. By definition,

$$\left.\frac{\partial W^{+}[h]}{\partial h'}\right|_{h'=0}=\lim_{\alpha\to 0}\frac{W^{+}[h'=\alpha]-W^{+}[h'=-\alpha]}{\alpha-(-\alpha)}\ . \tag{52}$$

The two surfaces defined above differ only in that one has an excess up step and the other has an excess down step. Therefore, for purposes of calculating the difference in (52), we may focus attention on a single step. Let H_1^{+} and H_2^{+} denote the configurations depicted in Figs. 2(a) (an up step) and 2(b) (an up step plus an adatom), respectively, and let H_1^{-} and H_2^{-} be the left-right reflected versions of H_1^{+} and H_2^{+}. Only these configurations need be considered since they alone involve the extra step barrier for a hop to the right (cf. Sec. IV). Then

$$\begin{aligned}W^{+}[h'=\alpha]-W^{+}[h'=-\alpha]=\frac{\alpha}{\tau_0}\{&P^{+}(H_1^{+})\exp[-\beta(E_N+E_U)]+P^{+}(H_2^{+})\\&-P^{-}(H_1^{-})\exp(-\beta E_N)-P^{-}(H_2^{-})\exp(-\beta E_U)\}\ ,\end{aligned} \tag{53}$$

where P^{+} (P^{-}) is the probability that the up (down) step has the given configuration. The factor α comes from taking the spatial coarse-grain average. Now use the fact that

$$\begin{aligned}&P^{+}(H_1^{+})=P^{-}(H_1^{-})=P(H_1)\ ,\\&P^{+}(H_2^{+})=P^{-}(H_2^{-})=P(H_2)\ .\end{aligned} \tag{54}$$

Taking the limit $\alpha\to 0$, one obtains

$$\begin{aligned}\left.\frac{\partial W^{+}[h]}{\partial h'}\right|_{h'=0}=&\frac{1}{2\tau_0}[P(H_1)\exp(-\beta E_N)-P(H_2)]\\&\times[\exp(-\beta E_U)-1]\ .\end{aligned} \tag{55}$$

A similar argument for the case $E_L\neq 0$ and $E_U=0$ yields

$$\begin{aligned}\left.\frac{\partial W^{+}[h]}{\partial h'}\right|_{h'=0}=&\frac{1}{2\tau_0}[P(H_1)\exp(-\beta E_N)-P(H_3)]\\&\times[1-\exp(-\beta E_L)]\ ,\end{aligned} \tag{56}$$

where H_3^{+} is the configuration depicted in Fig. 2(c).

Therefore, combining (48)–(51) with (55) and (56), the coefficient of the Laplacian in the equation of motion for the case of asymmetric step barriers can be written

$$\begin{aligned}\nu_{\text{step}}=&\frac{a^2}{\tau_0}[P(H_1)\exp(-\beta E_N)-P(H_2)]\\&\times[1-\exp(-\beta E_U)]\\&+\frac{a^2}{\tau_0}[P(H_1)\exp(-\beta E_N)-P(H_3)]\\&\times[\exp(-\beta E_L)-1]\ .\end{aligned} \tag{57}$$

As it should, this formula reproduces the result $\nu_{\text{step}}=0$ obtained earlier for our basic model ($E_U=E_L=0$). Moreover, even in the presence of asymmetric step barriers, $\nu_{\text{step}}=0$ *at equilibrium* since detailed balance guarantees that the first bracketed factor in each term vanishes identically. But under more general conditions these factors will not be zero. In particular, a deposition flux may be expected to enhance $P(H_2)$ and $P(H_3)$ relative to $P(H_1)$. In agreement with phenomenological ar-

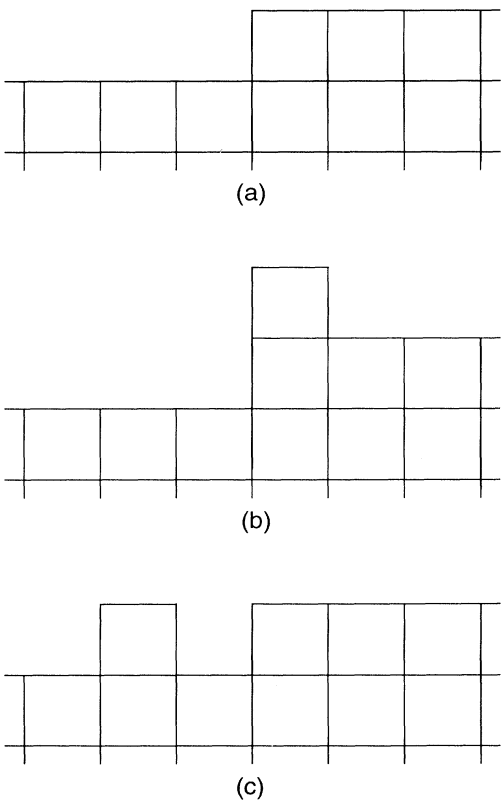

FIG. 2. Configurations used to demonstrate the effect of reflection-symmetry breaking for asymmetric step barriers: (a) H_1^{+}: single step; (b) H_2^{+}: step plus adatom that can join the step from the upper terrace; (c) H_3^{+}: step plus adatom that can join the step from the lower terrace.

guments [26,54], (57) then predicts that positive values of E_U (E_L) roughen (smooth) the surface during growth while negative values of E_U (E_L) smooth (roughen) the surface during growth.

We conclude finally that knockout processes and asymmetric barriers to step approach *both* induce roughness characteristic of the linear EW model for a growing epitaxial interface if desorption is negligible. Hot-atom effects always lead to smoothing ($\nu > 0$) while the sign of the Laplacian term depends on the precise nature of the barriers and flux conditions for the case of asymmetric diffusion in the vicinity of steps.

VI. DISCUSSION

The results of the preceding section illustrate that the stochastic equation of motion (2) can be derived analytically when a simple regularization procedure is applied to lattice Langevin equations derived from Arrhenius surface kinetics. Already at the level of our basic model (Sec. II), low-order nonlinearities are present during growth that are rigorously *absent* in the corresponding stochastic equation of motion that governs such evolution very near and at equilibrium [51]. In particular, $\lambda \neq 0$ if desorption is present and $\sigma \neq 0$ if surface diffusion is present. This result is consistent with two known results. First, mass conservation guarantees that $\lambda = 0$ if desorption is absent because this term manifestly cannot be written as the gradient of a surface current [21]. Second, Rácz *et al.* [34] have shown that the coefficient $K = 0$ for surface diffusion rates that are invariant under the combined transformation $\{h_i\} \rightarrow \{-h_i\}$ and $\{h_{i+j}\} \rightarrow \{h_{i-j+1}\}$. This result is relevant to typical Metropolis-type kinetic schemes where the transition rate depends on the difference in a potential energy functional before and after a diffusion event. But it does *not* contradict the present results because the stated symmetry is not respected by the Arrhenius kinetics adopted here.

On the other hand, the discussion of Sec. V makes clear that the behavior of surface diffusion rates under reflection symmetry can have a profound effect on the presence or absence of a Laplacian in the equation of motion. If this symmetry is respected (as in our basic model), the Laplacian is rigorously absent. But it turns out that such symmetry breaking is only a *necessary* condition for the presence of a term in (2) proportional to $\nabla^2 h$. It is not *sufficient* because the asymmetry may arise from terms of higher order than ∇h in (55) and (56). As it happens, the Laplacian *does* occur for Arrhenius-type asymmetric kinetics considered here. But the foregoing immediately explains why Monte Carlo simulations of "surface diffusion" that employ a Metropolis-type kinetic rule often *but not always* exhibit behavior consistent with the EW model when the surface is out of equilibrium [24,55]. In all such algorithms, the transition probability depends on the total energy of the system before and after the diffusion event. Inevitably, there will be configurations that generate the asymmetric hopping rates discussed here. But it does not appear straightforward to isolate the symmetry breaking term in the general case as we have done here. Indeed, one easily checks that Metropolis kinetic schemes [55] that make use of SOS Hamiltonians of the form

$$H = \frac{E_N}{2} \sum_{i=1}^{N} |h_i - h_{i+1}|^n \tag{58}$$

produce *no* asymmetry at all for the physically relevant configurations that involve the Arrhenius barriers E_U and E_L discussed in Sec. III. For this reason, we do *not* accept the notion that such algorithms "capture the essential physics of surface diffusion during MBE" simply because the symmetry breaking is associated with step configurations. Quite apart from the lack of any experimental evidence that Metropolis-type kinetics occurs in nature, there is the undeniable fact that the asymmetry and its sign arise entirely from a rather arbitrary choice of a model Hamiltonian.

VII. SUMMARY AND CONCLUSION

In this paper, we have derived equations of motion for the time evolution of a solid surface under typical epitaxial growth conditions. Our basic model was defined to include atomic deposition and Arrhenius-type desorption and surface (height) diffusion as the fundamental microscopic processes operative for a solid-on-solid crystal. Refinements to the model included hot-atom knockout processes that induce downward bias for some freshly deposited atoms and asymmetric diffusion barriers for the approach of an adatom to a step edge. Standard methods from the theory of stochastic processes then were used to convert a master-equation description of the dynamics into a set of Langevin equations (one for each column height variable) and their associated noise covariances. Solution of these equations constitutes a formal alternative to Monte Carlo simulation of the dynamical processes.

A nonrigorous regularization procedure was employed to transform the set of lattice Langevin equations to a single nonlinear, stochastic, partial differential equation of motion for the surface profile. In agreement with previous phenomenological treatments, the final equation obtained for our basic model included terms of the KPZ variety that arise from thermal desorption and additional terms of the conserved KPZ variety that arise from surface diffusion. The refinements to the basic model we examined for deposition and surface diffusion were found to generate a Laplacian but no KPZ nonlinearity in the equation of motion. The origin of this behavior was traced to a breaking of reflection symmetry in the basic transition rates for diffusion. These results provide a consistent interpretation to all Monte Carlo simulation results of which we are aware.

What relevance do the results summarized above have to experimental studies of surface roughness? In the literature, one finds exponents quoted for different systems that are consistent with the predictions of the KPZ model [56], the conserved KPZ model [57], the EW model [58], and unstable growth [59]. To rationalize this state of affairs, we must recognize that all the processes

discussed in Secs. II–IV are present in any real system to some degree. Thus, strictly speaking, only unstable EW ($\nu<0$) and KPZ behavior ($\nu>0$) can be present asymptotically. But for some systems, e.g., Si(100) and GaAs(100) grown by molecular-beam epitaxy, desorption is almost negligible and the coefficient λ [cf. (42)] will be very small [60]. In that case, experiments performed over limited time scales are more likely to be dominated by crossover effects [32] and thus to exhibit scaling behavior consistent with the stable EW or conserved KPZ models. In conjunction with surface science estimates of the elementary microscopic parameters, the estimates of the coefficients of the various terms in (2) provided in this paper may permit one to judge the expected behavior for any particular material system and deposition conditions.

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