Surface Diffusion Currents and the Universality Classes of Growth

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We show, through simulations of a variety of one- and two-dimensional solid-on-solid models, that nonequilibrium surface diffusion processes in the presence of a deposition flux are *generically* characterized by a nonvanishing inclination-dependent mass current along the surface. The current stabilizes the surface, leading to Edwards-Wilkinson scaling for the surface fluctuations, if it is a decreasing function of inclination, but induces a growth instability otherwise. Both types of behavior generally occur for the same system at different surface orientations.

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A number of recent theoretical studies [1–7] were devoted to deposition processes in which surface diffusion constitutes the dominant relaxation mechanism. This work was motivated both by the practical interest in deposition techniques such as molecular beam epitaxy (MBE) [8], and by the progress in our general understanding of interfacial fluctuations in growth processes [9,10] which followed the seminal work of Kardar, Parisi, and Zhang (KPZ) [11]. While the KPZ theory applies to situations such as the field induced motion of an interface in the Ising model at low temperatures [9], the microscopic process envisioned in studies of "ideal MBE" [4] consists of atoms impinging randomly onto the growing surface, and then diffusing to highly coordinated sites where they are incorporated into the lattice. The key assumptions of ideal MBE models are the effective absence, on the scales of interest, of (i) desorption and (ii) vacancies and overhangs; (i) implies that the surface relaxation dynamics conserves the mass of the growing film, while (ii) translates mass conservation into volume conservation. Consequently, the large scale fluctuations of the surface can be described by a coarse-grained, noisy continuity equation [1,2]

$$\frac{\partial}{\partial t}h = -\nabla\cdot\mathbf{J}+\eta\,, \tag{1}$$

where $h(\mathbf{x},t)$ is the height of the surface above some d-dimensional reference plane, $\mathbf{J}$ is a function of the derivatives of h, and the Gaussian white noise term $\eta(\mathbf{x},t)$ represents the deposition flux, with $\langle\eta\rangle=\langle\eta^2\rangle-\langle\eta\rangle^2=F$. Violation of either of the two assumptions stated above introduces a term proportional to $(\nabla h)^2$ into (1) and places the model into the KPZ universality class [12].

Here the term *universality class* refers to the dynamic scaling form of the height fluctuations [13], which can be written, using the Fourier amplitudes $S(|\mathbf{k}|,t)=\langle|\hat{h}(\mathbf{k},t)|^2\rangle$ of h, as $S(k,t)=k^{-(d+2\zeta)}f(k^z t)$, where $f(0)=0$ and $f(x\to\infty)=$const for a surface which is flat ($h\equiv 0$) at time $t=0$. The universality class of a growth process is characterized [9] by the values of the roughness exponent ζ and the dynamic exponent z. For height conserving processes described by (1) the scaling relation $z=d+2\zeta$ holds [1,4,5]. Numerically, the exponents are often estimated from the scaling of the surface width $\xi^2(t)=\sum_k S(k,t)\sim t^{2\beta}$, where $\beta=\zeta/z$ [3,7].

Scaling arguments [2] and renormalization group (RG) techniques [4,5] aimed at predicting the exponents ζ and z start from a gradient expansion of the current $\mathbf{J}$ in (1). The central issue, then, is to understand the form of the current that applies to a given microscopic situation. In Mullins' theory of equilibrium surface diffusion [14] the current is the gradient of a local chemical potential $\mu(\mathbf{x},t)$ which, to leading order in h, is proportional to the curvature of the surface. This implies $\nabla\cdot\mathbf{J}\sim\nabla^2(\nabla 2h)$ and leads to the prediction $z=4$, $\zeta=(4-d)/2$ [1]. It has been suggested [2,4] that growth conditions contribute a nonequilibrium term $\mu_{\mathrm{NE}}\sim(\nabla h)^2$ to the chemical potential. This is a relevant term in the RG sense which changes the dynamic exponent to $z=(8+d)/3$ for $d\le 4$ [2,4,5].

In this Letter we show that nonequilibrium conditions *generically* generate a surface diffusion current $\mathbf{J}_{\mathrm{NE}}$ which, to leading order in a gradient expansion, is a function of the local surface inclination ∇h, and therefore *cannot* be derived from a generalized chemical potential $\mu_{\mathrm{NE}}(\nabla h,\nabla^2 h,\ldots)$. The microscopic origin of this current lies in the fact that tilting the surface, $h\to h+\mathbf{x}\cdot\mathbf{m}$, breaks the reflection symmetry in the direction of the tilt and consequently induces a preference for particles to move either uphill or downhill. Writing $\nabla h=m\mathbf{e}$ with $|\mathbf{e}|=1$ have

$$\mathbf{J}_{\mathrm{NE}}(\nabla h)=j(m,\mathbf{e})\mathbf{e}\,. \tag{2}$$

We propose a unified picture of nonequilibrium surface diffusion based on the possible generic shapes of the function $j(m)$ (the dependence on $\mathbf{e}$ will be omitted in most of what follows). Moreover, we demonstrate that the diffusion current can be measured in simulations of deposition onto tilted substrates, allowing us to detect its presence even in cases where its magnitude is too small to

influence the scaling properties of the surface on numerically accessible length and time scales. This provides a new, powerful method to determine the true asymptotic behavior of deposition processes dominated by surface diffusion.

Before discussing specific models we outline the possible scenarios and their consequences for the stability of the growing surface. Let the height $h(\mathbf{x},t)$ be measured relative to a high symmetry plane of the underlying crystal. Then $j(0)=0$ and j has to be an odd function of m. Expanding j to linear order yields an Edwards-Wilkinson (EW) [15] term $\nu_2\nabla^2 h$ on the right-hand side of (1), with $\nu_2=-j'(0)$. If the current is in the *downhill* direction, $j<0$, we have $\nu_2>0$ and the surface is stable. In fact, all allowed nonlinearities are irrelevant compared to the EW term [5] and we conclude that $z=2$, $\zeta=(2-d)/2$. If, however, the current flows *uphill*, $\nu_2<0$ and the surface is linearly unstable at long wavelengths. Assuming that the small scale fluctuations are suppressed by a Mullins-type equilibrium term $-\nu_4\nabla^2(\nabla^2 h)$, the critical wavelength for the instability is $\lambda_c\approx 2\pi(-\nu_4/\nu_2)^{1/2}$. The asymptotic surface morphology in the unstable case is expected to depend on higher order nonlinearities as well as on the details of the model (see below). The KPZ nonlinearity is capable of balancing a negative EW term such that a positive effective ν_2 is generated on large scales [16], but it is not known whether the weaker nonlinearities compatible with the conservation law (1) have a similar effect.

We emphasize that there is *no physical principle* that would single out one of the two scenarios described above. In fact, in general stable and unstable growth occurs for the same system at different surface orientations. To see this, suppose $\mathbf{e}$ is chosen such that another high symmetry plane is reached at a tilt m_1. Then $j(m_1)=j(0)=0$, and there exists (at least one) inclination m^* where $\nu_2=-j'(m)$ changes sign. The two scenarios then imply that growth is linearly unstable (stable) for $m>m^*$ ($m<m^*$), or vice versa.

An important example of this type of behavior was discussed a long time ago by Schwoebel [17]. He was concerned with the consequences of the experimental observation [18] that atoms diffusing on certain vicinal surfaces prefer to be incorporated at the *ascending* step bordering a terrace, since reaching the descending step would involve traversing a potential barrier. If island formation on the terraces is neglected, this leads to an *uphill* current $j(m)\approx F/m$ which, because $j'(m)<0$, *stabilizes* the surface. The symmetry requirement $j(0)=0$ implies, by continuity, that the $1/m$ behavior must terminate below some slope m^* associated with the onset of nucleation on terraces, and $j'(m)>0$ for $m<m^*$. Hence a singular (≡high symmetry) surface is *destabilized* by the Schwoebel effect, as previously pointed out by Villain [2].

In our numerical simulations, an average surface inclination was imposed through the boundary conditions and the resulting current was recorded by counting the number of diffusion jumps in different directions. The fact that surfaces with $j'(m)<0$ are linearly unstable makes it difficult to measure the current at such inclinations. This problem can be avoided (at least if finite size effects are not too severe; see below) by choosing the system size smaller than the critical wavelength λ_c.

As a first example, Fig. 1 shows results for a restricted solid-on-solid (RSOS) model for nonequilibrium surface diffusion *without* deposition [19]. In this one-dimensional model particles of unit size are transferred between neighboring sites subject to the restriction that the heights h_i satisfy $|h_{i+1}-h_i|\le H$ before and after the move. Depending on the precise way the moves are attempted, the dynamics may or may not satisfy detailed balance (DB) [19]. In the DB case the net current is identically zero [20]. In the non-DB case we observe, for a small system (size $L=20$), a net current which varies smoothly with the imposed slope [Fig. 1(a)], vanishing both at $m=0$ (by symmetry) and at the maximum allowed slope $m=H$. The derivative $j'(0)$ changes sign at $H\approx 3$, in accordance with the transition from stable to grooved surface morphologies at zero average tilt found in [19]. In Fig. 1(b), showing the "stable" case $H=2$ for larger systems, the current is seen to drop sharply to zero at a critical slope m_c, which shows some dependence on L and is *smaller* than the slope m^* at which $j'(m)$ (as determined for $L=20$) changes sign. The drop marks the formation of a grooved state composed of pieces of slope 0 and H, similar to those observed previously for $H>3$ [19] [note that mass conservation requires grooved states to consist of regions of slopes for which $j(m)=0$]. The fact that the instability sets in at a slope where the sur-

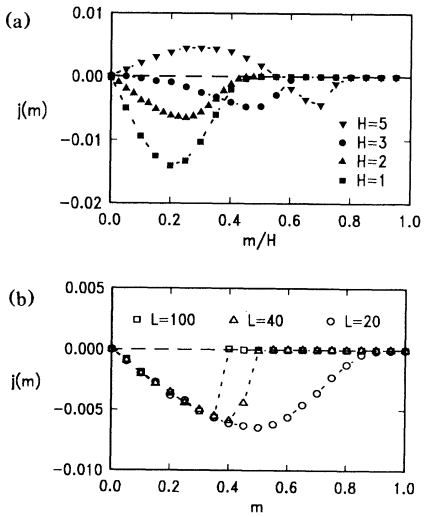

FIG. 1. Surface diffusion currents for the one-dimensional conserved RSOS model [19] with nearest (for $H\ge 2$) and next-nearest (for $H=1$) neighbor hopping. (a) $L=20$, various H; (b) $H=2$, various L. Averages were taken over at least 10^6 attempted moves per site in the steady state.

face is still linearly stable can be understood by following the time evolution. This reveals that the transition to the grooved state occurs through the *nucleation* of a region of maximal slope $h_{i+1}-h_i=H$. Since no moves are allowed in such a region, it is very hard to destroy and will continue to grow once it has reached a certain, critical size. In contrast, a surface prepared at $m>m^*$ develops a periodic structure corresponding to the fastest growing wavelength $\sqrt{2}\lambda_c$ of the linear instability. This is in close analogy with the well-known behavior of phase separating systems in thermal equilibrium [21]: m^* corresponds to the spinodal, m_c is the coexistence boundary, and in the region $m_c<m<m^*$ the surface is metastable. Indeed, in one dimension (1) is very similar to a noisy Ginzburg-Landau model with a conserved order parameter m and thermodynamic potential $\int dm j(m)$ [22].

Next we consider a solid-on-solid model [6] in which random deposition at a rate F is combined with *collective* surface diffusion governed by the Hamiltonian $\mathcal{H}=K\times\sum_i|h_{i+1}-h_i|^n$. For $F=0$ the surface is in thermal equilibrium and $j=0$. At a finite deposition rate a morphological transition was observed as a function of n, from a stable phase with Edwards-Wilkinson scaling at $n=1$ to a grooved phase at $n=4$ [6]. Our data for the diffusion current depicted in Fig. 2 show that this transition fits nicely into the general framework: The current is *uphill* for $n=4$, destabilizing the singular ($m=0$) surface, and *downhill* for $n=1$ (in the intermediate case $n=2$ [6] the current is unmeasurably small; however, we are not aware of any reason why it should vanish entirely). Moreover, for $n=4$ the current is essentially zero up to a slope $m_c(L)$ which is larger than the maximum m^* of $j(m)$ (as estimated from the simulations at small L), indicating a nucleation-driven transition to the grooved state for $m<m_c$ [cf. Fig. 1(b)]. For $m>m_c$ the numerically determined structure factor is consistent with Edwards-Wilkinson behavior on large scales. Similarly, the structure factor for $n=1$ at large slopes shows the breakdown of scaling associated with the formation of a grooved state [6].

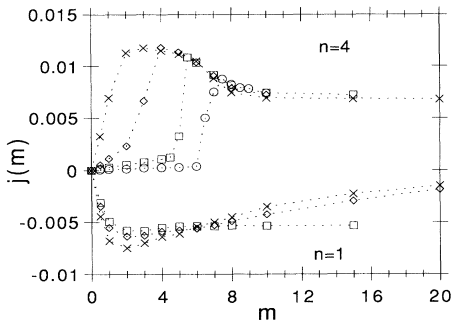

FIG. 2. Surface diffusion currents for the one-dimensional SOS deposition model [6] with $n=1$ (lower data sets) and $n=4$ (upper data sets). The surface was equilibrated in an average deposition flux of one particle per ten MC steps at temperature $T=0$. The substrate sizes are $L=16$ (×), $L=32$ (◇), $L=64$ (□), and $L=128$ (○, for $n=4$).

We conclude that the morphological transition observed in this model as a function of n is essentially analogous to that of the RSOS model as a function of H, the main effect of the deposition flux being to break DB relative to the Hamiltonian $\mathcal{H}$. Nevertheless, comparing Figs. 1 and 2 two important differences are noted. First, the deposition model shows severe finite size effects, particularly for $n=1$, which prevent us from obtaining a quantitatively reliable estimate of $j(m)$ in the asymptotic ($L\to\infty$) limit. We attribute this to the fact that the roughness exponent for the linear theory with $\nu_2=0$, which, due to the smallness of $j(m)$, is expected to govern the surface fluctuations on intermediate length scales, is $\zeta=\frac{3}{2}$ in one dimension. Consequently, the slope fluctuations *diverge* with system size as $L^{\zeta-1}$, and it becomes difficult to fix the local slope through the global boundary conditions. Second, the current does *not* decay to zero for large slopes. Since the inclinations appearing in the grooved states are associated with the zeros in $j(m)$ (see above), this implies that the grooved states of this model do not have well-defined slopes in the limit $L\to\infty$, in accordance with the findings of [6].

Our final example is the class of models introduced by Wolf and Villain (WV) [1], in which a freshly landed atom relaxes immediately to the site with the largest coordination number within a distance l from the deposition site. It can be shown [20] that $j(m)\equiv 0$ in one dimension if no distinction is made between *kink* sites with coordination number $N_i=2$ and *trapping* sites with $N_i=3$. Consequently, any net current in the original WV model (which does distinguish between $N_i=2$ and 3) is solely due to the rather rare events in which a particle has a choice between a trapping site and a kink site. Not surprisingly, the currents measured in our simulations of the one-dimensional WV model are very small, and hampered by severe finite size effects. The asymptotic ($L\to\infty$) regime could only be accessed for rather large slopes ($m=2$) where we obtain a *downhill* current $j\approx -0.0012$ for $l=1$. Using the linear theory $\partial h/\partial t=\nu_2\nabla^2 h-\nu_4\nabla^4 h+\eta$ we estimate that the time-dependent surface width $\xi(t)$ should cross over from Mullins scaling, $\beta=\frac{3}{8}$ [1], to EW behavior ($\beta=\frac{1}{4}$) for $t>t_c\sim\nu_2^{-2}\sim 10^6$, which is well beyond the scope of available numerical work [1,3,7]. The sign of the current can be reversed by introducing (for $l\geq 2$) a "Schwoebel" parameter which governs the probability for an atom to be reflected at a descending step.

If the severe finite size effects in the one-dimensional deposition models can indeed be attributed to the fact that $\zeta>1$ for $\nu_2=0$, it should be an advantage to go to the two-dimensional case, where the slope fluctuations diverge only logarithmically ($\zeta=1$). In Fig. 3 we show the diffusion current measured for a two-dimensional version of the WV model. The data have only a moderate size dependence, and the current is downhill, as in $d=1$.

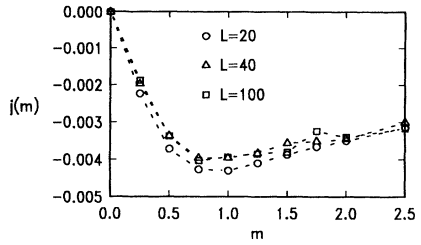

FIG. 3. Surface diffusion current for a two-dimensional version of the Wolf-Villain model [1] with nearest neighbor hopping. The current was averaged over between 10^5 (for $L=80$) and 1.6×10^6 (for $L=20$) monolayers.

We estimate that $\nu_2\approx0.0075$, corresponding to a crossover time $t_c\approx2\times10^4$. Simulations of a large system ($L=400$) up to $t=10^3$ gave the estimate $\beta\approx0.2$, which is consistent with other numerical work in two dimensions [7] and suggests that the model is governed by the nonlinear Villain-Lai-Das Sarma equation [2,4] on intermediate time and length scales.

In conclusion, we have outlined a new approach to surface diffusion controlled deposition processed which focuses on the inclination-dependent diffusion current. This quantity contains the full information about the large scale behavior of the surface, similar to the inclination-dependent growth rate in KPZ-type processes [9,12]. Our central prediction is the *generic* occurrence of surface diffusion induced growth instabilities of the kind found previously [6,19] in specific models. The generality of the phenomenon suggests that it may be relevant to real deposition processes. Since real surfaces are not subject to the constraints of our models, it seems plausible to conjecture that the generation of large surface gradients by the linear instability implies, in effect, the breakdown of the solid-on-solid assumption (ii) of ideal MBE, leading to amorphous growth and ultimately to KPZ behavior [12]. This provides a new perspective on the question about the universality class of MBE: Since the amorphous growth associated with $\nu_2<0$ is no longer epitaxial, *the only consistent large scale description for MBE is the Edwards-Wilkinson theory,* which predicts essentially flat surfaces ($\zeta=0$) in $d=2$. Theoretically, the outstanding problem is the prediction of the sign and magnitude of the current for a given microscopic rule. In particular, it is of interest to know if the smallness of the current and the associated large crossover scales found in most of the cases studied here is an artifact of the models or a feature of real deposition processes.

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