Kinetic Roughening of Laplacian Fronts

Joachim Krug[a]
Theoretische Physik, Ludwig Maximilians Universität, Theresienstrasse 37, D-8000 München 2, Germany

Paul Meakin
Central Research and Development Department, E. I. du Pont de Nemours and Company, Experimental Station E356/153, Wilmington, Delaware 19880-0356
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The kinetic fluctuations of a stable interface driven by the gradient of a Laplacian field are investigated. In three and more dimensions the interface width is finite. In two dimensions the width diverges logarithmically with time and system size. Its scaling form is derived in agreement with simulations of diffusion-limited erosion (antidiffusion-limited aggregation). A crossover to algebraic roughness with an extended intermediate scaling regime is predicted for diffusion with a drift towards the interface. Capillary effects are discussed in relation to recent experiments on fluid displacement in porous media.

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A moving interface can be kinetically roughened by microscopic fluctuations in the local velocity. Relevant examples are deposition processes, where the fluctuations are due to shot noise in the particle flux, and two-fluid displacements in random porous media. If the interfacial dynamics is *local,* the motion can be described by a stochastic partial differential equation for the interface position.[1] This approach predicts universal dynamic scaling properties for the interfacial fluctuations, in agreement with a large body of computer simulations.[2-4] However, in many physical processes the velocity of the interface is proportional to the gradient of some field (e.g., concentration, temperature, pressure, or electrostatic potential) satisfying the Laplace equation, and hence the dynamics is intrinsically *nonlocal.* Two cases must be distinguished according to whether the interface moves in the direction of increasing or decreasing field strength. In the first case, the Saffman-Taylor[5] and Mullins-Sekerka[6] instability drives a planar interface into a macroscopically modulated state determined by subtle pattern selection mechanisms.[7] Here we address the second case of a macroscopically stable, planar front that is kinetically roughened on a mesoscopic scale.

Unstable Laplacian interfaces at large noise levels are simulated by the Witten-Sander model[8] of diffusion-limited aggregation (DLA), in which particles follow random-walk trajectories and accrete to the deposit at the point of first contact. Similarly, the basic model for a stable Laplacian front is diffusion-limited erosion (DLE),[9] the reverse process of DLA. Here an initially flat substrate is eaten away by particles that diffuse toward the surface and annihilate with a substrate particle where they hit. In the quasistatic, low-density limit the particle concentration satisfies the Laplace equation and the surface moves in the direction of decreasing concentration. The time scale is set by the advancement of the front.

A situation rather close to DLE arises in electrolytic polishing[10] where the diffusion of "acceptor" molecules to the anode has been identified as the rate-limiting process.[11,12] The acceptors can be complex forming ions such as CN^- or water molecules that are needed for the formation of hydrated cations.[12] In the steady state each acceptor arriving at the anode immediately recombines with a metal ion which is thereby removed from the sample. It is well known[10] that electrolytically polished surfaces are exceedingly smooth on all length scales. Indeed we demonstrate, in accordance with previous numerical results,[9] that fluctuations are not capable of roughening a stable Laplacian front in three (or more) spatial dimensions. In two dimensions the roughness of the interface diverges logarithmically with time and substrate size. We compute the scaling form[13] for the interface width and compare it to simulation data.[9]

Our analysis is readily extended to DLE with a spatial bias perpendicular to the interface. Clearly, for a strong *attractive* bias towards the interface the model reduces to ballistic deposition[14] which is well represented by the local theory.[1] We show that even a weak bias dominates on large length scales, leading to algebraic roughness in two and three dimensions, and predict crossover scales and scaling functions. In contrast, a *repulsive* bias implies bounded fluctuations in all dimensions.

Two-dimensional DLE has been studied previously as a model for viscous fluid displacement in porous media, in the limit where the displaced fluid has zero viscosity.[15,16] The role of the Laplacian field is then played by the pressure in the displacing fluid. On short length scales, or for slow displacement, capillary forces dominate that are not included in the simple DLE model. Provided such effects can be lumped into an effective interfacial tension[17] we show that they lead to algebraic roughness on short length scales, with the logarithmic behavior reappearing asymptotically. The relevance of these results to recent experiments[18,19] on fluid displacement in two-dimensional porous media is discussed.

We consider a d-dimensional system of linear extension L in the $d-1$ transverse coordinates $\mathbf{x}_{\|}=(x_1,\ldots,x_{d-1})$, and of infinite extension in the x_d direction. On a somewhat coarse-grained scale the interface position can be parametrized by a single-valued height function $x_d=h(\mathbf{x}_{\|},t)$. The Laplacian field $\phi(\mathbf{x},t)$ satisfies $\nabla^2\phi=0$ for $x_d<h$ and vanishes for $x_d>h$. The field is continuous at the interface. The normal interface velocity is proportional to the normal field gradient. This implies

$$\partial h/\partial t=-D[\partial\phi/\partial x_d-\nabla_{\|}\phi\cdot\nabla_{\|}h]_{x_d=h}\,, \tag{1}$$

where, in the case of DLE, D is the particle diffusion coefficient. A straightforward linear analysis[5,12] shows that a small periodic perturbation $h_{\mathbf{q}}(t)e^{i\mathbf{q}\cdot\mathbf{x}_{\|}}$ superimposing a flat front $h=Vt$ moving at velocity $V>0$ decays as $e^{-\sigma(\mathbf{q})t}$, where

$$\sigma(\mathbf{q})=V|\mathbf{q}|\,. \tag{2}$$

This dispersion relation has been verified in experiments on electrolytic polishing of periodically modulated surfaces[11,12] and also in simulations of DLE.[9,16] In the presence of randomness perturbations are constantly generated. To model the noisy interface motion we add a random force to the relaxational dynamics[20] and obtain a stochastic equation of motion for the Fourier components of $h(\mathbf{x}_{\|},t)$,

$$\partial h_{\mathbf{q}}(t)/\partial t=-\sigma(\mathbf{q})h_{\mathbf{q}}(t)+\eta_{\mathbf{q}}(t)\,. \tag{3}$$

The nonlocality of the problem is reflected by the fact that $\sigma(\mathbf{q})$ is a nonlocal operator in real space. The $\eta_{\mathbf{q}}(t)$ are Fourier components of Gaussian white noise in space and time, with zero mean $\langle\eta_{\mathbf{q}}(t)\rangle=0$ and covariance

$$\langle\eta_{\mathbf{q}}(t)\eta_{\mathbf{q}'}(t')\rangle=(\Delta/L^{d-1})\delta_{\mathbf{q}+\mathbf{q}'}\delta(t-t')\,. \tag{4}$$

On dimensional grounds we may write $\Delta=Va_r^d$, where a_r is a length scale associated with the randomness.

Using the field $\phi(\mathbf{x},t)$ calculated from the linear analysis we find that the second term on the right-hand side of (1) leads to a nonlinearity $\sim V(\nabla h)^2$ which is known to be of crucial importance for local interface dynamics.[1,4] Here simple power counting shows that such a term is irrelevant compared to the nonlocal linear term,[21] and hence the linearization is justified. The nonlinearity becomes relevant in the presence of an attractive bias, as will be demonstrated below.

The linear equation (3) is trivially solvable. The solution appropriate for an initially flat interface $[h_{\mathbf{q}}(0)=0]$ is

$$\langle|h_{\mathbf{q}}(t)|^2\rangle=L^{-(d-1)}[\Delta/2\sigma(\mathbf{q})](1-e^{-2\sigma(\mathbf{q})t})\,. \tag{5}$$

A dynamically scale-invariant interface can be characterized by two scaling exponents ζ and z, which describe, respectively, static and dynamic scaling properties.[1,4] For a rough interface the typical amplitude of transverse excursions on a scale l parallel to the interface is proportional to l^{ζ}.[22] If the interface evolves from a flat initial state, it is rough on scales $l\le t^{1/z}$ after a time t. The general scaling form for the Fourier amplitudes is then[23] $\langle|h_{\mathbf{q}}(t)|^2\rangle\sim|\mathbf{q}|^{-d+1-2\zeta}g(|\mathbf{q}|^z t)$. Comparing with (5) the scaling exponents in the present case are identified as $\zeta=(2-d)/2$ and $z=1$. We note that the result for ζ is shifted by one dimension $(d\to d+1)$ with respect to an equilibrium interface roughened by capillary waves.[22]

For $d=2$, $\zeta=0$ and the interface is logarithmically rough. The quantity of interest is the rms interface width ξ as a function of time and system size,[13] which is obtained[23] by summing (5) over the $L-1$ modes $q=(2\pi/L)n$, $n=-L/2+1,\ldots,-1,1,\ldots,L/2$ (L is taken even). We find

$$\xi(L,t)^2=\frac{\Delta}{2\pi V}[\ln(L/a)-f(Vt/L)]\,, \tag{6}$$

where a is a short-range (lattice) cutoff and $f(x)=-\ln(1-e^{-4\pi x})$ is a universal scaling function. At short times $(t\ll L/V)$ the width increases as $\xi(t)^2\approx(\Delta/2\pi V)\ln(4\pi Vt/a)$ while in the stationary regime $(t\gg L/V)$ it saturates at $\xi_\infty(L)^2\approx(\Delta/2\pi V)\ln(L/a)$.

In Fig. 1 we compare the predicted scaling form (6) to simulation results for DLE that span more than two decades in system size. In the simulations time is measured in units of the average surface position, so $V=1$. The noise amplitude $\Delta\simeq1.2$ was estimated from the data for $\xi_\infty(L)$ obtained by Meakin and Deutch.[9] To eliminate the short-range cutoff we plot $\xi^2-\xi_\infty^2$ as a function of $\log_{10}(t/L)$. Apart from statistical fluctuations for large L that are due to the small number of independent runs, the agreement between theory and simulation is excellent.

In three dimensions the sum over the modes (5) is

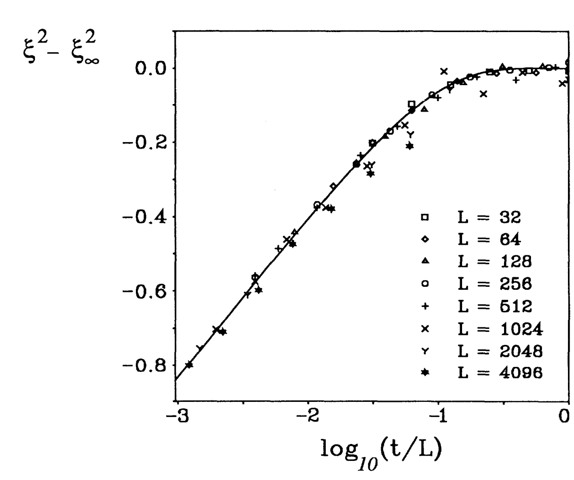

FIG. 1. Numerical results (symbols) and theoretical scaling function (solid curve) for the surface width in diffusion-limited erosion on the square lattice. The model is described in Ref. 9. The numerical data were averaged over a number of independent runs ranging from 4000 for $L=32$ to 20 for $L=4096$.

dominated by the short-range cutoff and the interface width ξ tends to a (nonuniversal) constant $\xi_\infty \sim \sqrt{\Delta/Va}$ as $t,L \to \infty$. The negative roughness exponent $\zeta = -\frac{1}{2}$ shows up in the finite-size corrections to the width. We find $\xi_\infty^2 - \xi^2 \sim 1/t$ for $Vt \ll L$ and $\xi_\infty^2 - \xi^2 \sim 1/L$ for $Vt \gg L$. Moreover, the correlations in the interface position have a power-law decay,

$$\langle h(\mathbf{x}_\parallel,t)h(\mathbf{x}_\parallel+\mathbf{r},t)\rangle - (Vt)^2 \sim |\mathbf{r}|^{2\zeta} = 1/|\mathbf{r}|$$

for $t \to \infty$. At finite times this behavior is limited to length scales below a correlation length $r_c = 2Vt$, where a more rapid decay $\sim t^2/|\mathbf{r}|^3$ sets in.

Suppose now that the diffusing particles have a small drift velocity u perpendicular to the interface. The Laplace equation in the region $x_d < h$ is then changed to $D\nabla^2\phi - u\,\partial\phi/\partial x_d = 0$ and a term $u\phi|_{x_d=h}$ is added to the right-hand side of (1). Repeating the linear analysis[24] we obtain the dispersion relation

$$\sigma(\mathbf{q}) = V\{[\mathbf{q}^2 + (u/2D)^2]^{1/2} - u/2D\}\,, \tag{7}$$

which deviates from (2) for $|\mathbf{q}| \ll |u|/D$, corresponding to length scales $l \gg l_u = D/|u|$. For a repulsive bias $(u<0)$ $\sigma(\mathbf{q}) \to V/l_u$ for $\mathbf{q} \to 0$ which implies that the logarithmic roughness in two dimensions is limited to length scales $l < l_u$. For an attractive bias $(u>0)$ the dispersion relation is quadratic for small $\mathbf{q}$, $\sigma(\mathbf{q}) \approx Vl_u\mathbf{q}^2$, corresponding to the local diffusion operator $\nabla^2 h$ in real space. Compared to this term the nonlinearity arising from the lateral motion in (1) may no longer be neglected.[1,4] Hence on length scales $l \gg l_u$ the interface is described by a *local* equation of the type suggested by Kardar, Parisi, and Zhang (KPZ),[1]

$$\partial h/\partial t = V[l_u\nabla^2 h + (\nabla h)^2] + \eta\,. \tag{8}$$

For small drift, $l_u \gg a, a_r$, there is a substantial intermediate scaling regime where the linear term in (8) dominates and the scaling exponents take on the values $\zeta_0 = (3-d)/2$ and $z_0 = 2$ familiar from capillary-wave theory[22] and sedimentation processes.[25] The crossover scales can be estimated using known results[1,4,26] for the KPZ equation.

In *two dimensions* the static correlations are not affected by the nonlinearity.[1] Hence the crossover from logarithmic $[\xi_\infty \sim \ln(L/a)^{1/2}]$ to algebraic $[\xi_\infty \sim L^{1/2}]$ divergence of the stationary width can be computed directly from the dispersion relation (7). The dynamic exponent changes[1,4] from $z_0 = 2$ to $z = \frac{3}{2}$ at a time scale $t_c = l_c^{(2)}/V$ with $l_c^{(2)} \sim l_u^5/a_r^4$. In *three dimensions* the logarithmic roughness associated with the static exponent $\zeta_0 = 0$ becomes visible on length scales $l > l_c^{(1)} \sim a \times \exp(l_u/a) \gg l_u$. On even larger scales[26] $l > l_c^{(2)} \sim a \times \exp(l_u^3/a_r^3)$ the nonlinearity leads to algebraic roughness with[2-4] $\zeta \simeq 0.4$. Clearly, these giant length scales are not expected to be observable for moderate drift velocities. Finally, we note that in dimensions $d > 3$ a phase transition occurs[1,3,4,26] as a function of the dimensionless coupling $\bar{\lambda} \sim (a_r/l_u)^3$ from a smooth phase at small $\bar{\lambda}$ to a rough phase with $\zeta > 0$ at large $\bar{\lambda}$.

Next we turn to the corrections due to capillary forces acting on a fluid interace in a porous medium. If the displacing fluid wets the medium, which is the case of experimental relevance,[18,19] it has been suggested[17] that the capillary pressure drop across the interface can be related to the interfacial curvature through an effective coarse-grained interfacial tension γ^*. Including the effective tension in the linear stability analysis adds a (positive) term proportional to $|\mathbf{q}|^3$ to the dispersion relation (2).[5] Computing the stationary roughness in two dimensions we now obtain

$$\xi_\infty(L)^2 = \frac{\Delta}{4\pi V}[\ln(1+L^2/l_{\mathrm{Ca}}^2) - \ln(1+a^2/l_{\mathrm{Ca}}^2)] \tag{9}$$

with a crossover length scale $l_{\mathrm{Ca}} = 2\pi\sqrt{\kappa/\mathrm{Ca}}$, where $\mathrm{Ca} = \mu V/\gamma^*$ is the effective capillary number, μ is the viscosity of the displacing fluid, and κ is the permeability of the porous medium. For $a \ll L \ll l_{\mathrm{Ca}}$ a new scaling regime with $\zeta = 1$ $(\xi_\infty \sim L)$ appears, while on larger length scales the logarithmic scaling (6) is recovered. In the case of unstable displacement l_{Ca} is the critical wavelength for the onset of the instability.[5]

The value of the effective interfacial tension γ^* is unknown in general.[17] For a rough comparison with the experiments of Rubio *et al.*[18] we set γ^* equal to the microscopic (molecular) interfacial tension γ. The crossover scale l_{Ca} then turns out to fall within the range of length scales used to estimate the roughness exponent ζ. According to (9), this would imply a distinctly curved log-log plot for $\xi_\infty(L)$ and an effective exponent that decreases systematically with increasing capillary number. Instead $\zeta \simeq 0.73$ is found independently of Ca.[18] On the other hand, a recent reanalysis[27] of the data of Rubio *et al.*[18] does show a clear crossover behavior that is well fitted by our formula (9) with $\gamma^* = \gamma$ and $a_r \simeq 39$ in units of the bead size. Although the agreement could be accidental,[28] we do believe that the applicability of an effective interface tension[17] in this situation remains an open question that should be systematically addressed in future experiments.

In summary, we have described a novel universality class of kinetic roughening with a large variety of physical applications. While the limiting case of purely Laplacian dynamics admits a simple analytic solution, the crossover to other displacement mechanisms on large or short length scales is quite complex and certainly merits further theoretical and experimental work.

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[a]Present address: IBM Thomas J. Watson Research Center, P.O. Box 218, Yorktown Heights, NY 10598.

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