Surface Diffusion and Fluctuations of Growing Interfaces

Leonardo Golubović[(a)] and Robijn Bruinsma
Physics Department, University of California, Los Angeles, California 90024
(Received 12 April 1990)

We discuss a model for the growth of interfaces relaxing by surface diffusion. We show that thermal noise can produce novel large-scale orientational fluctuations in the active zone of the growth. The resulting growth morphology is related to the structure of membranes and cannot be described by existing theories of self-affine growth.

PACS numbers: 68.55.Jk, 05.70.Ln, 81.15.−z

Growing clusters and films often evolve into fascinating self-similar and fractal patterns. Generally, both the interior and the exterior of the cluster can develop a fractal structure. In a number of cases, however, the growth process is restricted to a thin "active zone" on the outer surface while the interior is dense. Well-known examples are amorphous films grown by ballistic deposition, flame fronts, and the growth of tumors.

A general framework has been proposed[1] to discuss the scaling properties of growing interfaces. The width $w(L,t)$ of the interface was predicted to depend on the sample size L and the growth time t as $w(L,t) = L^{\chi} f(t/L^{z})$, with $f(\infty) = \text{const}$ and $f(x) \sim x^{\chi/z}$ for small x. The exponents χ and z characterize the nature of the growth-induced roughness. Kardar, Parisi, and Zhang[2] (KPZ) proposed an analytic model for growing interfaces. They found that $z = \frac{3}{2}$ and $\chi = \frac{1}{2}$ for one-dimensional interfaces ($d=1$). For $d=2$, only numerical results are available.[3]

In this paper we discuss the effects of *conservation laws* on this scaling. The relaxation of surface features of thin films proceeds, under normal conditions, by surface diffusion which obeys mass conservation.[4] Experimental studies[5] and numerical simulations[6] of these systems reveal the presence of large-scale structures (deep narrow grooves, voids, etc.) not encountered in the KPZ model. Conservation laws are well known to have significant effects on dynamical scaling phenomena such as increasing the dynamical exponent z.[7] We will see that in growth problems, conservative relaxational dynamics leads to an increase of the exponent χ. This has important consequences. "Solid-on-solid" (SOS) models for the cluster surface (such as the KPZ theory) are meaningful only for $\chi < 1$. For $\chi > 1$, the cluster surface would extend into the cluster interior at long enough length scales and develop overhangs which would mean breakdown of the SOS description and destruction of the long-range orientational order of the surface.[8]

The exponent χ increases as we decrease the dimensionality d. We will call the dimensionality d below which $\chi > 1$ the "lower critical dimension" d_L. For the KPZ theory, $\chi < 1$ in any d so the SOS description is always valid. This is in marked contrast to the situation discussed in the following where we will see that d_L exceeds 2, so $\chi > 1$ for $d=1$ and 2.

To study the effect of conservative dynamics on both d_L and χ, we first reexamine the SOS model for growing interfaces. Let $h(\mathbf{r},t)$ describe the height profile of a d-dimensional surface whose growth proceeds along the local surface normal ("Eden model"). By demanding that the growth velocity depends only on the local surface morphology through rotational invariants (such as the curvature), one finds for the growth rate

$$\frac{\partial h}{\partial t} = -K(\nabla^2)^2 h + \nu \nabla^2 h + \tfrac{1}{2}\lambda(\nabla h)^2 + \eta(\mathbf{r},t)\,, \quad (1)$$

to second order in h. Here, λ is the average growth velocity and $\eta(\mathbf{r},t)$ describes the statistical noise of the incoming particle flux. The noise will be assumed to be Gaussian white noise with a correlation function

$$\langle \eta(\mathbf{r},t)\eta(\mathbf{0},0)\rangle = 2D\delta(\mathbf{r})\delta(t)\,. \quad (2)$$

The equation of motion (1) is taken to be in the frame comoving with the interface with average velocity λ.

Equation (1) is a phenomenological model and the quantities K and ν must be identified from physical considerations.[9] In the absence of any conservation law, there is no reason for ν to be zero. In that case, the $\nu\nabla^2 h$ term will dominate the $K(\nabla^2)^2 h$ term as far as the asymptotic, long-length-scale properties are concerned. If we set $K=0$ and assume $\nu > 0$, then Eq. (1) reduces to the KPZ model[2] which produces surfaces with the aforementioned scaling law with $\chi < 1$. The constant ν is proportional to the surface energy. Relaxation by evaporation and recondensation during high-pressure chemical vapor deposition falls, for instance, in this category.

For a system obeying mass conservation, relaxation proceeds via surface diffusion.[4] The surface diffusion current $\mathbf{j}$ is proportional to $\nabla\mu$, with μ the chemical potential.[4] The chemical potential is a scalar so it can only depend on rotationally invariant quantities.[10] The lowest-order invariant is the interface curvature $\nabla^2 h$, so $\mu \propto \nabla^2 h$. By mass conservation, $\partial h/\partial t$ is proportional to $\nabla\cdot\mathbf{j}$ and thus to $\nabla^4 h$. This argument produces only the *first* term of Eq. (1). In other words, for conservative relaxation, rotational invariance implies $\nu = 0$.[10] The constant K is proportional to the surface diffusion constant as well as the surface energy. An example is growth by sputter deposition.[5] From now on we will refer to ν as a "surface tension," while a surface controlled by $K\nabla^4 h$ relaxation will be called "tensionless" (even though K is actually proportional to the surface energy[9]).

The surface diffusion term is much less efficient in relaxing large-scale surface features than the surface tension term of the KPZ model. We thus would expect to encounter more disordered surfaces. It is, however, important to note that the noise—as defined in Eq. (2)—as well as the quadratic, lateral growth nonlinearity in Eq. (1),[2] *violates* the conservation law. It is only the surface relaxation which is conservative; the incoming flux, which produces both the noise and the nonlinear term in Eq. (1), obeys no such law.[11] Thus, even though the initial, *bare* value of ν is zero, there must be an *effective*, noise-induced nonzero ν at large length scales. One would thus naively expect the KPZ model to remain valid asymptotically.

We will start our analysis with the special case $\lambda=0$, in which case Eq. (1) is harmonic. In this harmonic regime, Eq. (1) becomes a Langevin equation which is readily solvable. The result is that, after a transient period, the surface configurations are controlled by a Boltzman distribution. The probability $P(\{h(\mathbf{r})\})$ for a given height profile $h(\mathbf{r})$ to be realized is, for $\nu=0$, given by

$$P(\{h\}) \propto \exp\left\{-\frac{1}{2}\frac{K}{D}\int d^d r (\nabla^2 h)^2\right\}. \tag{3}$$

This particular distribution function has been examined in detail in the context of the fluctuations of *membranes* and we can take over the analysis.[12] The fluctuations of membranelike surfaces are characterized most conveniently by the orientational correlation function $G(r)$ defined as

$$G(r) \equiv \langle [\nabla h(\mathbf{r}) - \nabla h(\mathbf{0})]^2 \rangle. \tag{4}$$

For an asymptotically flat surface, $G(\infty)$ is a constant, while for an orientationally disordered, crumpled surface, $G(\infty)$ diverges. By (3), one finds for $G(r)$, in the limit $r=|\mathbf{r}|\to\infty$,

$$G(r) \propto \begin{cases} (D/K) r^{2-d}, & d < d_L^H, \\ (D/K)\ln(r/a), & d = d_L^H, \\ \text{const}, & d > d_L^H, \end{cases} \tag{5}$$

where $d_L^H=2$ is the (harmonic) lower critical dimension and a, a short distance cutoff. Thus, for $d > d_L^H$ the surface is asymptotically flat while below or at d_L^H it loses its orientational order with increasing r. Within harmonic theory, the growth exponents are $\chi_H=(4-d)/2$ and $z_H=4$ for $d > d_L^H$. For $d \le d_L^H$, we can characterize the degree of orientational order by introducing the *persistence length* ξ_p such that for $r \gg \xi_p$ the orientational correlations are lost while for $r < \xi_p$ the surface is relatively flat.[12] Thus, ξ_p can be estimated from (5) by setting $G(\xi_p)\approx 1$. This yields

$$\xi_p \approx \begin{cases} a\exp(\text{const}\,K/D), & d = d_L^H, \\ (K/D)^{1/(2-d)}, & d < d_L^H. \end{cases} \tag{6}$$

For distances $r > \xi_p$, the SOS description is actually inconsistent. Numerical simulations of membranelike surfaces indicate that such surfaces become highly convoluted for $r > \xi_p$ with overhangs, grooves, etc.[12] Let us stress that the SOS description, though breaking down for $r > \xi_p$, can still be used to estimate the persistence length ξ_p in excess of which overhangs and inlets will appear.

We now turn to the behavior of the nonlinear theory, Eq. (1) with $\lambda\neq 0$, using a renormalization-group (RG) analysis similar to that of KPZ.[2] The RG equations were obtained by eliminating fluctuations in the momentum shell $\Lambda e^{-l} < q < \Lambda$ (with Λ the momentum cutoff) and by performing the rescaling $r=e^l r'$, $t=e^{lz}t'$, and $h=e^{l\chi}h'$. To one-loop order we find the following:

$$dD/dl = (z-2\chi-d+g_d\lambda^2/4)D, \tag{7a}$$

$$dK/dl = [z-4+a(d)g_d\lambda^2/4]K, \tag{7b}$$

$$d\lambda/dl = (\chi+z-2)\lambda, \tag{7c}$$

$$d\nu/dl = (z-2)\nu + (8-d)K\Lambda^2 g_d\lambda^2/4d, \tag{7d}$$

where

$$a(d) = (5d^2-22d-16)/4d(d+2)$$

and $g_d = s_d(2\pi)^{-d}\Lambda^{d-8}DK^{-3}$ (s_d is the d-dimensional unit-sphere area). In contrast to KPZ, in deriving Eqs. (7) we considered the regime with small tension, $|\nu| \ll K\Lambda^2$, since $\nu(l=0)=0$ as discussed above. By choosing χ and z to keep D and K constant, we obtain from Eqs. (7a)-(7d)

$$d\lambda/dl = \lambda\{(8-d)/2 - [3a(d)-1]g_d\lambda^2/8\}. \tag{7e}$$

It immediately follows from Eq. (7e) that for $l\to\infty$, $\lambda(l)$ scales to zero only for $d>8$. The harmonic theory is thus valid at long length scales only for $d > d_U = 8$ and we can identify $d_U=8$ as the "upper critical dimension." The RG flow lines for $d \lesssim 8$ are shown in Fig. 1(a). We find two fixed points at the boundary between the KPZ and an unstable phase: For $\lambda^*=\nu^*=0$, we recover the harmonic fixed point with $\chi=\chi_H=(4-d)/2$ and $z=z_H=4$. In addition to this, for $8-d=\varepsilon>0$ there appears a new, anharmonic fixed point, with $\lambda^*=\text{const}\sqrt{\varepsilon}$ and $\nu^*=0$, to $O(\varepsilon)$.[13] This anharmonic fixed point is more stable than the harmonic fixed point as is evident from Fig. 1(a). The associated scaling exponents are, to $O(\varepsilon)$,[14]

$$\chi = -2+2\varepsilon, \tag{8a}$$

$$z = 4-2\varepsilon. \tag{8b}$$

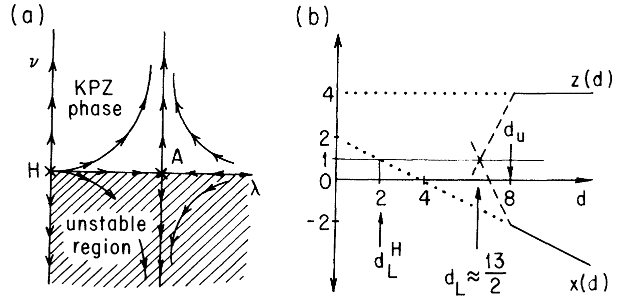

FIG. 1. (a) Renormalization-group flows in the (λ, ν) plane for $d \lesssim 8$. H, harmonic fixed point; A, anharmonic fixed point. To lowest order in $\varepsilon = 8-d$, $\nu^* = 0$. (b) Exponents z and χ as functions of d. For $d > 8$, $z = 4$ and $\chi = (4-d)/2$ exactly (solid lines). Dotted lines: extrapolation of these values to $d < 8$. From $\chi(d_L^H) = 1$, one finds $d_L^H = 2$. Dashed lines: leading-order corrections in ε for $d < 8$. For $d < 8$, $z + \chi = 2$ *exactly*, so $z(d_L) = \chi(d_L) = 1$ at d_L, giving $d_L = \frac{13}{2}$ to lowest order in ε.

This is a very interesting result because, as shown in Fig. 1(b), it indicates that the harmonic theory *underestimates* the actual value of χ. Recall that $\chi = 1$ at the lower critical dimension. From Eq. (8), we estimate that $\chi = z = 1$ at $d = d_L = \frac{13}{2}$, as indicated in Fig. 1(b).[14] Although this result may be affected by higher-order corrections in ε, it strongly suggests that the lower critical dimension will exceed the harmonic-theory estimate $d_L^H = 2$. One- and two-dimensional surfaces are thus *below* the lower critical dimension. The inequality $d_L > d_L^H = 2$ is also plausible on physical grounds: The relevant quadratic nonlinearity in Eq. (1) is due to lateral growth effects[2] which always make interfaces rougher. They thus should increase the value of χ with respect to the harmonic-theory estimate χ_H, so d_L obtained from $\chi(d_L) = 1$ has to be above d_L^H obtained from $\chi_H(d_L^H) = 1$.

The orientational correlation function $G(r)$, Eq. (4), is, for $d < d_L$, given by

$$G(r) \sim (D/K) L_G^{2-d} (r/L_G)^{2(\chi-1)}, \tag{9}$$

so that the persistence length ξ_p, as estimated by $G(\xi_p) \approx 1$, is

$$\xi_p \sim L_G (L_G^{d-2} K/D)^{1/2(\chi-1)}. \tag{10}$$

Here $L_G = (K^3/\lambda^2 D)^{1/(8-d)}$ is the Ginzburg length associated with crossover from harmonic behavior [Eq. (5)] to anharmonic behavior [Eq. (9)].

Our anharmonic fixed point is, however, unstable against deviations of the surface tension ν away from its fixed-point value $\nu^* = 0$ [to $O(\varepsilon)$] as depicted in Fig. 1(a).[13] The associated eigenvalue is $y = 2 - 2\varepsilon$. For $\nu > 0$, $\nu(l)$ steadily increases with l, implying that, at large enough length scales, the interface will crossover to the surface-tension-dominated behavior discussed by KPZ.[2] For $\nu < 0$, $\nu(l)$ becomes more and more negative with increasing l. In this regime the interface is unstable against finite-wave-vector modulation. Thus, only for a particular critical value of the bare surface tension $\nu(l=0)$, say ν_c, do we have the "tensionless" scaling behavior associated with the exponents of Eq. (8). We saw that ν_c is zero within the present $O(\varepsilon)$ calculation.[13] However, as noted before, the nonconservative terms in the equation of motion (1) will generate a noise-induced fluctuation contribution to the surface tension, so in general ν_c must be nonzero.

What are the implications of these results for the interface morphology? If the noise level is weak or λ small, ν_c will be nonzero but still small. On the other hand, for a system with conservative relaxation, the bare value of the surface tension, $\nu(l=0)$, is zero. This means that over a considerable range of length scales we should be close to the "tensionless" critical region and that one should be able to observe a rich crossover behavior: At scales up to the Ginzburg length L_G, the interface structure will be described by the harmonic theory, i.e., Eq. (5) should be valid. For length scales longer than L_G, anharmonic effects become important, i.e., we cross over to Eqs. (8) and (9). Eventually, the noise-induced surface tension becomes important—RG flows move $\nu(l)$ away from the small-surface-tension region $\nu \approx \nu_c$. If $\nu(l)$ is positive for large l, we cross over to the KPZ behavior, while for $\nu(l)$ negative, the surface should exhibit finite-wave-vector instabilities.

To examine which one of these possibilities is realized for a system with zero bare surface tension, $\nu(l=0) = 0$, we performed a numerical simulation of Eq. (1) in $d = 1$,[15] which indicated that $\nu(l)$ flows into the unstable region of negative ν values. This would mean that for zero "bare" surface tension $\nu(l=0)$, e.g., as in sputter deposition, the noise generates a *negative* effective surface tension, destabilizing the interface. In other words, as we decrease ν in Eq. (1), the KPZ phase becomes unstable at a small *positive* value ν_c of $\nu(l=0)$.

We believe that this result is of a general nature. The fact that $\nu(l \to \infty) \neq 0$ while $\nu(l=0) = 0$ is due to the nonlinear term in Eq. (1) which, as mentioned, always *destabilizes* the surface. Recall that it increases the exponent χ with respect to the harmonic theory, both in the present calculation and in the KPZ model. As a result, this term is likely to produce a destabilizing negative effective surface tension if initially $\nu(l=0) = 0$—although this claim would have to be confirmed by a calculation to order ε^2.[13] If true, it would imply that for conservative dynamics with $\nu(l=0) = 0$, the asymptotic properties are not controlled by the KPZ fixed point but rather by the unstable region of Fig. 1(a).

The true morphology of such surfaces at very large length scales cannot be addressed within the present methods, although the crossover behavior discussed in this work will dominate in a considerable range of length scales. The asymptotic morphology is expected to have a convoluted, membranelike appearance, but testing this will require numerical studies which go beyond the SOS model.

We would like to thank M. Kardar, J. Rudnick, and A. Zangwill for helpful discussions. This work was supported by U.S. Defense Advanced Research Projects Agency under Grant No. Army DAAL 03-89-K-0144 and by NSF Grant No. DMR 8922027.

Note added.—After this work was submitted we received a number of preprints considering fluctuations of surfaces relaxing by surface diffusion.[16]

---

[a]On leave from Boris Kidrič Institute of Nuclear Sciences, Institute of Theoretical Physics, Vinča, P.O. Box 522, Beograd, Yugoslavia.

[1]F. Family and T. Viscek, J. Phys. A **18**, L75 (1985).

[2]M. Kardar, G. Parisi, and Y.-C. Zhang, Phys. Rev. Lett. **56**, 889 (1986).

[3]See, e.g., J. G. Amar and F. Family, Phys. Rev. Lett. **64**, 543 (1990).

[4]C. Herring, J. Appl. Phys. **21**, 301 (1950); in *Structure and Properties of Solid Surfaces,* edited by R. Gomer and C. S. Smith (Univ. of Chicago, Chicago, 1953), pp. 5–72, see discussion on p. 64.

[5]B. A. Movchan and A. V. Demchishin, Phys. Met. Metallogr. U.S.S.R. **28**, 83 (1969); J. A. Thornton, Annu. Rev. Mater. Sci. **7**, 239 (1977). For a recent review, see R. Bruinsma, R. P. U. Karunasiri, and J. Rudnick, in *Proceedings of the NATO Advanced Research Workshop on Kinetics of Ordering and Growth on Surfaces,* edited by M. Lagally (Plenum, New York, 1990).

[6]See P. Meakin, CRC Crit. Rev. Solid State Mater. Sci. **13**, 143 (1987).

[7]S.-k. Ma, *Modern Theory of Critical Phenomena* (Benjamin, Reading, 1976).

[8]Similar interfacial arguments were used for random-field ferromagnets. See, e.g., G. Grinstein and S.-k. Ma, Phys. Rev. Lett. **49**, 685 (1982).

[9]W. W. Mullins, J. Appl. Phys. **28**, 333 (1957).

[10]Note that under conditions of conservative relaxation, the $\nu\nabla^2 h$ term can be produced only by a chemical potential $\mu(\mathbf{r},t)\propto h(\mathbf{r},t)$ which breaks the rotational invariance.

[11]T. Sun, G. Guo, and M. Grant, Phys. Rev. A **40**, 6763 (1989), consider a different model in which both the dynamics and the noise are conservative. The resulting interfaces are thus considerably less rough than those discussed here.

[12]For a general review, see *Statistical Mechanics of Membranes and Surfaces,* edited by D. Nelson, T. Piran, and S. Weinberg (World Scientific, Singapore, 1989). The notion of a persistence length for surfaces was introduced by P. G. de Gennes and C. Taupin, J. Phys. Chem. **86**, 2294 (1982).

[13]It would appear that $\nu^* \sim \varepsilon$ according to Eq. (7d) since $\lambda^{*2} \sim \varepsilon$ at the fixed point. However, because of the prefactor $8-d$ in Eq. (7d), $\nu^* \sim \varepsilon^2$ and we must go to order ε^2 to ascertain not only the magnitude but also the *sign* of ν^*.

[14]The exponents χ and z satisfy exactly the relationship $\chi+z=2$ for $d \leq 8$. This is a consequence of the Galilean invariance of Eqs. (1) and (2) which implies that Eq. (7c) is exact (see Ref. 2), so at the *true* d_L, $\chi=z=1$.

[15]L. Golubović and R. P. U. Karunasiri (to be published).

[16]D. Wolf and J. Villain (to be published); J. Villain (to be published); S. Das Sarma and P. Tamborenea, following Letter Phys. Rev. Lett. **66**, 325 (1991).