Phase Ordering and Roughening on Growing Films

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(Received 3 February 2000)

We study the interplay between surface roughening and phase separation during the growth of binary films. Already in $1 + 1$ dimensions, we find a variety of different scaling behaviors, depending on how the two phenomena are coupled. In the most interesting case, related to the advection of a passive scalar in a velocity field, nontrivial scaling exponents are obtained in simulations.

Thin solid films are grown for a variety of technological applications, using molecular beam epitaxy (MBE) or vapor deposition. In order to create materials with specific electronic, optical, or mechanical properties, often more than one type of particle is deposited. When the particle mobility in the bulk is small, surface configurations become frozen in the bulk, leading to anisotropic structures that reflect the growth history, and are different from bulk equilibrium phases [1]. Characterizing structures generated during composite film growth is not only of technological importance, but represents also an interesting and challenging problem in statistical physics.

In this paper, we examine the growth of binary films through vapor deposition, and study some of the rich phenomena resulting from the interplay of *phase separation* and *surface roughening*. Some previous theoretical studies have focused on the stability of the growing film against decomposition and roughening [2,3]; or on crossover phenomena and scaling in a particular computer model [4]. Here, we undertake a first systematic study of the potential couplings between the roughening and ordering of a growing surface. By combining analytical and numerical results, we find a wealth of possible scenarios of critical phenomena in this coupled system, and also unveil unexpected connections to other nonlinear problems of interest, such as the advection of a passive scalar in a turbulent fluid.

Simple models for *layer by layer* growth assume either that the probability that an incoming atom sticks to a given surface site depends on the state of the neighboring sites in the layer below [5], or that the top layer is fully thermally equilibrated [6]. Assuming that the bulk mobility is zero, once a site is occupied, its state does not change anymore. If the growth rules are invariant under the exchange of the two particle types, the phase separation is in the universality class of an equilibrium Ising model. Correlations perpendicular to the growth direction are characterized by the critical exponent ν of the Ising model, and those parallel to the growth direction by the exponent νz_m, with z_m being the dynamical critical exponent of the Ising model.

However, the *layer by layer* growth mode underlying these simple models is unstable, and the growing surface becomes *rough*. In many cases the fluctuations in the height $h(\mathbf{x}, t)$, at position $\mathbf{x}$ and time t, are *self-affine*, with correlations

$$\langle [h(\mathbf{x}, t) - h(\mathbf{x}', t')]^2 \rangle \sim |\mathbf{x} - \mathbf{x}'|^{2\chi}$$
$$\times g(|t - t'|/|\mathbf{x} - \mathbf{x}'|^{z_h}), \quad (1)$$

where χ is the roughness exponent, and z_h is a dynamical scaling exponent. While such scaling has been observed in many models of deposition of a single species (homoepitaxy), there are fewer conclusive studies in the case of binary growth and heteroepitaxy. One example is a computer model introduced in Ref. [4] with local sticking probabilities for two particle species in $1 + 1$ dimensions. The simulations show phase separation into domains (with large sizes consistent with the Ising model), and a very rough surface profile with sharp minima at the domain boundaries. Phase separation in binary growth provides the simplest example of a (nonequilibrium) phase transition in a growing (and rough) film. In this context, we may ask the following questions: (i) Are the surface roughness exponents different in the vicinity of the phase transition point? (ii) Are the critical exponents of the phase transition modified on a rough surface? We shall demonstrate that the coupling of roughening and phase separation leads to a rich phase diagram, and to nontrivial critical exponents already in $1 + 1$ dimensions.

To characterize phase separation, we introduce an *order parameter* $m(\mathbf{x}, t)$, which is the difference in the densities of the two particle types at the surface at position $\mathbf{x}$ and time t. The interplay between the fluctuations in m and the height h is captured phenomenologically by the coupled Langevin equations,

$$\partial_t h = \nu \nabla^2 h + \frac{\lambda}{2} (\nabla h)^2 + \frac{\alpha}{2} m^2 + \zeta_h, \quad (2)$$

$$\partial_t m = K(\nabla^2 m + rm - um^3) + a\nabla h \cdot \nabla m$$
$$+ bm\nabla^2 h + \frac{c}{2} m(\nabla h)^2 + \zeta_m. \quad (3)$$

Here, we have included the lowest order (potentially relevant) terms allowed by the symmetry $m \rightarrow -m$.

Equation (2) is the Kardar-Parisi-Zhang (KPZ) equation [7] for surface growth, plus a coupling to the order parameter. Equation (3) is the time dependent Landau-Ginzburg equation for a (nonconserved) Ising model, with three different couplings to the height fluctuations. The Gaussian, delta-correlated noise terms, ζ_h and ζ_m, mimic the effects of faster degrees of freedom. A different set of equations was proposed by Léonard and Desai [2] for phase separation during MBE. Their equations reflect the MBE conditions of random particle deposition (in contrast to sticking probabilities that depend on the local environment) and a conserved order parameter which evolves by surface diffusion. They do not include the KPZ nonlinearity. Computer simulations of corresponding $1 + 1$ dimensional systems are presented in [2,3].

Dimensional analysis indicates that the couplings appearing in Eqs. (2) and (3) are relevant, and may lead to new universality classes. We shall leave the renormalization group analysis of these equations to a more technical paper, and focus here instead on computer simulations in $1 + 1$ dimensions. The quantities evaluated in the computer simulations are the height correlation function in Eq. (1) and the order parameter correlation functions perpendicular and parallel to the growth direction. Allowing for the possibility of different dynamic exponents, z_m and z_h, for the order parameter and the height variables, we fit to the scaling forms:

$$
\begin{aligned}
G_m^{(x)}(x - x') &\equiv \langle m(x,t)m(x',t)\rangle \\
&= |x - x'|^{\eta-1} g_m^\perp(|x - x'|/\xi), \\
G_m^{(t)}(t - t') &\equiv \langle m(x,t)m(x,t')\rangle \\
&= |t - t'|^{(\eta-1)/z_m} g_m^\|(|t - t'|/\xi^{z_m}).
\end{aligned}
\qquad (4)
$$

Rather than discretizing Eqs. (2) and (3) and integrating them numerically, we decided to perform a computer simulation using a "brick wall" restricted solid-on-solid model (see Fig. 1). As this model has the same symmetries and conservation laws as the Langevin equations, it is expected to be in the same universality class. Starting from a flat surface, particles are added such that no overhangs are formed, and with the center of each particle atop the edge of two particles in the layer below. We use two types of particles, A and B (black and grey in the figures). The probability for adding a particle to a given surface site, and the rule for choosing its color, depends on the local neighborhood. When A particles are more likely to be added to A dominated regions, and vice versa, the particles tend to phase separate and form domains. In this case, the order parameter correlation length ξ is of the order of the average domain width. By changing the growth rules, it is possible to study cases in which some (or all) of the

FIG. 1.   The "brick wall" model used in the simulations.

couplings a, b, c, and α vanish, and thus to gain a complete picture of the different ways in which the height and the order parameter influence each other.

The decoupled case, $\alpha = a = b = c = 0$, is implemented using the following updating rules: A surface site is chosen at random, and a particle is added if it does not generate overhangs. Its color is then chosen depending on the colors of its two neighbors in the layer below. If both neighbors have the same color, the newly added particle takes this color with probability $1 - p$, and the other color with probability p (where p is much smaller than 1). If the two neighbors have different colors, the new particle takes either color with probability $1/2$. Neighbors within the same layer are not considered.

Since the probability of adding a particle to a given surface site does not depend on its color, the surface grows exactly as with only one particle type, and is characterized by the KPZ exponents $\chi = 1/2$ and $z_h = 3/2$. Similarly, the choice of particle color at a given site is not affected by the height profile. The height profile determines only the moment at which a site is added, since the no-overhang condition requires both neighbors in the previous layer to be occupied. If we equate layer number with time, domain walls move to the right or left with probability $1/2$ during one time unit, and a pair of new domain walls is created with probability p. This is identical to the Glauber model for a one-dimensional Ising chain with coupling J and at temperature T, with $p = \exp(-4J/kT)$. The correlation length ξ perpendicular to the growth direction is, consequently, $\xi = \exp(2J/kT) = 1/\sqrt{p}$, and the correlation time is $\tau = \exp(4J/kT) = 1/p$. The dynamical critical exponent for the order parameter is thus $z_m = 2$. Note that the "time" used for the order parameter (namely, layer number) is different from real time, which is for each particle the moment when it is added to the growing surface. However, this difference becomes negligible for sufficiently small p since the thickness of the surface over the correlation length, $\sqrt{\xi}$, is much smaller than the characteristic time, ξ^2, for order parameter fluctuations. Simulations indeed confirm that the order parameter and height evolve completely independently. A typical profile is shown in Fig. 2a; the corresponding scaling analysis conforms to expectations, and is not presented here.

The situation $\alpha > 0$ with $a = b = c = 0$ can be implemented by updating sites on top of particles of different colors less often by a factor $r < 1$ compared to sites above particles of the same color, a situation that is similar to the one studied in [4]. As the order parameter is not affected by the height variable, its dynamics is still the same as that of an Ising model, with $z_m = 2$. Because growth is slower at domain boundaries than within domains, the domain boundaries sit preferentially at the local minima of the height profile, with a mound over each domain (see Fig. 2b). This means that the surface roughness exponent is $\chi = 1$ on length scales up to ξ. Changes in the height profile on this scale result from

(a)   (b)
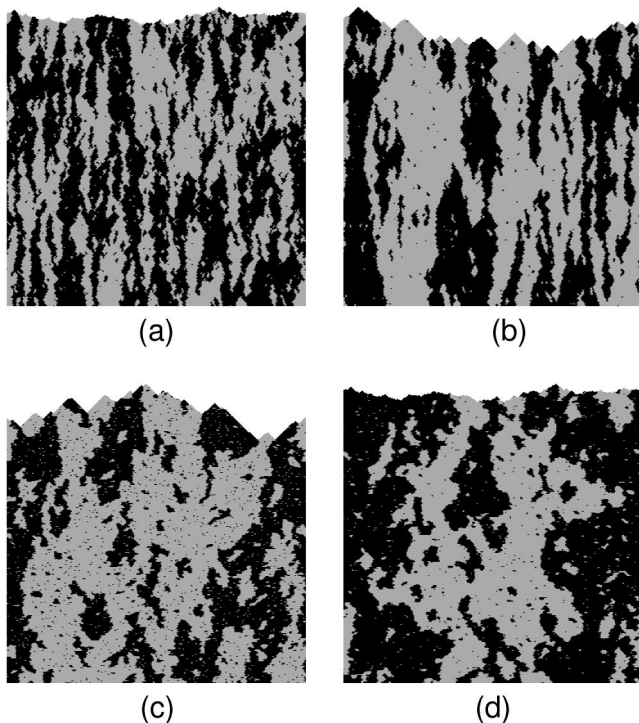
(c)   (d)

FIG. 2. Snapshot of the last 400 layers of simulations, for $L = 200$ sites. (a) The decoupled case with $p = 1/90$, and $r = 1$. (b) For $p = 1/200$, and $r = 1/20$, the height is coupled to the domains, but not vice versa. (c) The fully coupled case, using the same parameters as (b), but with updating rules that include neighbors in the same layer. (d) With $r = 1$, and the updating rules of (c), the domains are influenced by the height, but not vice versa. (Note that the profiles in (a) and (d) are identical since we used the same random numbers.)

domain wall diffusion, and the dynamic exponent is therefore $z_h = 2$. On length scales much larger than ξ, the average order parameter is zero, implying that KPZ exponents of $\chi = 1/2$ and $z_h = 3/2$ are regained. The crossover in the roughness is described by a scaling form:

$$\langle [h(x, t) - h(x', t)]^2 \rangle = |x - x'|^2 g(|x - x'|/\xi),$$

with a constant $g(y)$ for $y \ll 1$, and $g(y) \sim 1/y$ for $y \gg 1$.

To mimic the influence of surface roughness on the order parameter [nonzero a, b, or c in Eq. (3)], the color of a newly added particle is made dependent not only on those of its two neighbors in the layer below, but also on the colors of its two nearest neighbors on the same layer, if these sites are already occupied. With probability $1 - p$, the newly added particle takes the color of the majority of its 2, 3, or 4 neighbors, and with probability p it assumes the opposite color. If there is a tie, the color is chosen at random with equal probability. The height variable now affects the order parameter in two ways:

(i) Domain walls are driven downhill.—The reason is that the neighbor on the hillside of a site being updated is more likely to be occupied than the one on the valley side. The newly added particle is thus more likely to have

the color on the hillside. [This corresponds to $a > 0$ in Eq. (3).]

(ii) New domains are predominantly formed on hilltops.—This is because domains on hilltops can expand more easily than those on slopes or in valleys, indicating $b > 0$ in Eq. (3). Another consequence is that for the same p, the correlation length ξ is much larger than in the decoupled case, as is apparent in Figs. 2c and 2d.

For the fully coupled case depicted in Fig. 2c, we find essentially the same scaling behavior as in Fig. 2b, i.e., a height profile slaved to the Glauber dynamics of the domains. The most interesting case, shown in Fig. 2d, is when the height profile is independent of the domains ($\alpha = 0$), evolving with KPZ dynamics, while the order parameter is influenced by the roughness. The dynamic exponent z_m for the order parameter was first obtained by collapsing the correlation functions using Eqs. (4), as shown in Fig. 3. These curves imply that $\eta = 1$, $\xi \propto p^{-0.542}$, and $\tau \propto \xi^{z_m} \propto 1/p$, giving $z_m \simeq 1/0.542 \simeq 1.85$.

The same nontrivial value for z_m is obtained by a completely independent measurement of the dynamics of domain coarsening following a quench from a "high temperature" (p close to 0.5) to zero temperature ($p = 0$). Figure 4 shows the domain density as a function of time for a system of size $L = 16\,384$. The resulting $z_m \simeq 1.85$ is in agreement with the value from the scaling collapse.

The following simple argument fails to provide the exponent $z_m \simeq 1.85$. Consider a Langevin equation, $\dot{x} = \eta(t)$, for the position x of a single domain wall at time t. Since the motion of the domain wall is strongly influenced by the height profile, the noise $\eta(t)$ must have long-range correlations $\langle \eta(t)\eta(t') \rangle = D|t - t'|^\alpha$, reflecting the
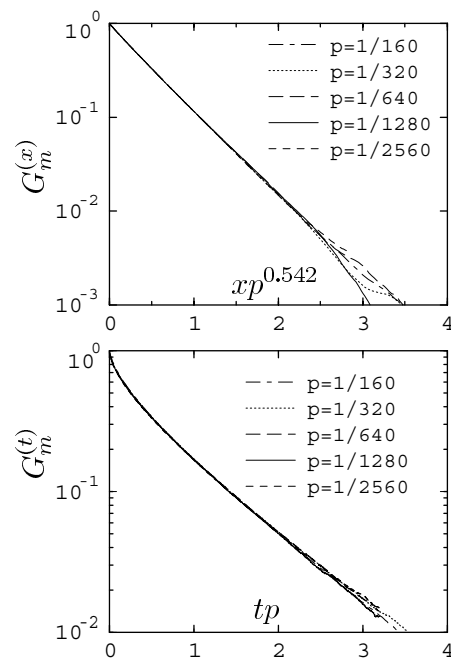

FIG. 3. Scaling collapse of correlations $G_m^{(x)}$ and $G_m^{(t)}$ in Fig. 2d. For each p, the data is an average over 7500 widely separated layers, and for systems of size up to 8192.
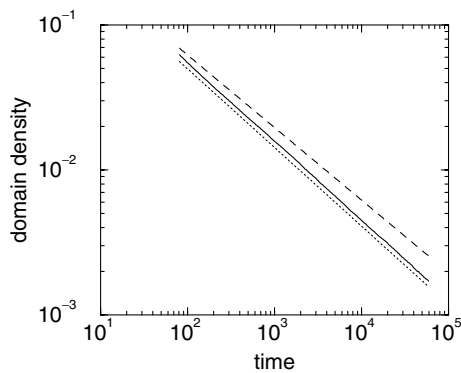
FIG. 4. Domain density (i.e., number of domains, divided by the system size) as a function of time for $L = 16\,384$, averaged over 100 samples. The dotted line is a power-law fit (slightly shifted for better visibility) with exponent of $1/z_m = 0.542$. For comparison, a power law with exponent -0.5 is also shown (dashed line).

dynamics of surface. This choice leads to $z_m = 2$ for $\alpha > 1$, and $z_m = 2/(2 - \alpha)$ for $\alpha < 1$. From the scaling behavior of the KPZ equation, we find $\alpha = 2/3$ for a colored noise dominated by the slope fluctuations, and therefore $z_m = 3/2$. This is identical to the dynamical critical exponent z_h for the height variable, which means that the height imposes its characteristic time scale on the order parameter. This result for z_m would presumably be correct if the domain walls were uniformly distributed along the surface. However, due to their tendency to move downhill, they are preferentially found near valleys. A different scaling of the slope fluctuations in the valleys may be the reason for the nontrivial value of z_m. Indeed, for short times, before the domain walls have moved to their preferred positions, the exponent $3/2$ is seen in the simulations.

The dynamics of domain walls on a growing KPZ surface bears some resemblance to the advection of a passive scalar in a turbulent velocity field, which is characterized by nontrivial exponents and multiscaling [8]. If we neglect interactions between domain walls, and treat them as independent "dust particles" floating on the KPZ surface, the Langevin equation for the particle density ρ is

$$\partial_t \rho = K\nabla^2 \rho + a(\nabla h \cdot \nabla \rho + \rho \nabla^2 h) + \zeta_\rho. \quad (5)$$

The second term describes the *advection* of particles along a velocity field $\vec{v} = \nabla h$. Indeed this transformation maps the KPZ equation into the Burgers equation for a vorticity-free, compressible fluid flow [7]. Equation (5) is a special case of Eq. (3) for m, with $r = u = c = 0$, $b = a$, and with a conserved noise ζ_ρ. [Together with Eq. (2) for the height profile, it is also a special case of the equations used to describe the dynamic relaxation of drifting polymers [9].] In the remainder, we give the results of computer simulations for this case. The rules for the motion of dust particles are identical to those for domain walls. However, each particle is treated as if the others were not
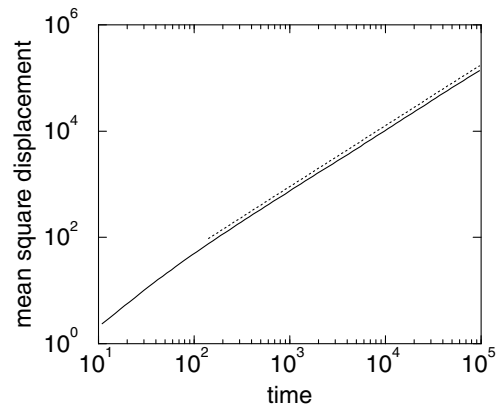

FIG. 5. Mean square displacement of a single domain wall in a system of size $L = 4096$. The power-law fit (dotted line) has an exponent 1.1467, corresponding to $z_\rho \simeq 1.74$.

present. This means, in particular, that there is no creation or annihilation of particles.

Figure 5 shows the mean square displacement of a single dust particle in a system of size $L = 4096$. To obtain good statistics, we averaged over 512 independent and non-interacting particles, and used more than 40 runs. The best fit is obtained for $z_\rho \simeq 1.74$, distinct from the previous $z_m \simeq 1.85$, implying that the exponents depend on whether or not the domain walls (or dust particles) are conserved. In contrast to the advection of a passive scalar in a turbulent velocity field, we find no sign of multiscaling. A fit of the density-density correlation function to $\langle \rho(x)\rho(0) \rangle \sim 1/x^{2(1-\chi_\rho)}$ gives an exponent $\chi_\rho \simeq 0.85$.

In summary, the interplay between surface roughening and phase separation leads to a variety of novel critical scaling behaviors. At one extreme, the height profile adapts to the dynamics of critical domain ordering. At the other, the dynamics of domain wall motion is influenced by the roughness, exhibiting new and nontrivial scaling behaviors.

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