

generated by a softly repulsive contact potential

$$\frac{b}{2} \int dx dx' \mathcal{V}(\mathbf{r}(x) - \mathbf{r}(x')). \quad (117)$$

The relevance of this term is also controlled by the scaling dimension $y_b = \nu z - 1 - (d - 2)\nu$, and therefore this effect is marginal in three dimensions at the nonlinear fixed point, in contrast with both Rouse and Zimm models where self-avoidance becomes relevant below four dimensions. Unfortunately, one is ultimately forced to consider nonlocal *and* nonlinear terms based on similar grounds, and such terms are indeed relevant below four dimensions. In some cases, local or global arclength conservation may be an important consideration in writing down a dynamics for the system. However, a local description is likely to be more correct in a more complicated system with screening effects (motion in a gel that screens hydrodynamic interactions) where a first principles approach becomes even more intractable. Therefore, this model is an important starting point towards understanding the scaling behavior of polymers under a uniform drift, a problem with great technological importance.

4. Ordering Phenomena on Growing Films

In many growth processes particles are highly mobile in an active layer at the surface, but are relatively immobile once incorporated in the bulk. We study models in which atoms are allowed to interact, equilibrate, and order on the surface, but are frozen in the bulk. Order parameter correlations in the resulting bulk material are highly anisotropic, reflecting its growth history. In a flat (layer by layer) growth mode, correlations perpendicular to the growth direction are similar to a two-dimensional system in equilibrium, while parallel correlations reflect the dynamics of such a system. When the growing film is rough, various couplings between height and order parameter fluctuations are possible. Such couplings modify the dynamic scaling properties of surface roughness, and may also change the critical behavior of the order parameter. Even the deterministic growth of the surface profile can result in interesting textures for the order parameter.

4.1. Introduction

For many technological applications, high quality films are grown by the process of vapor deposition. The properties of such films can be quite different from the same material produced in bulk equilibrium,^{55,56} reflecting their

preparation history. For example, during the growth of some binary alloys, the deposited atoms are highly mobile on the surface, but relatively immobile in the bulk.⁵⁶ Consequently, the surface fluctuations occurring during the growth process are frozen into the bulk. A characteristic feature of such (metastable) phases is *anisotropic* correlations related to the growth direction which are absent in bulk equilibrium.

A number of models for composite film growth have been introduced in the past^{57–63} Generally in these models, the probability that an incoming atom sticks to a given surface site depends on the state of neighboring sites in the layer below. Once a site is occupied, its state does not change any more, and thus the surface configuration becomes frozen in the bulk. Such growth rules are equivalent to (stochastic) cellular automata, where each site is updated in parallel as a function of the states of its neighbors. Subsequent states of the cellular automaton correspond to successive layers in the crystal.

It is in general not possible to calculate exact correlation functions for such (nonequilibrium) growth processes. The exception occurs in special cases where the growth rules satisfy a detailed balance condition, relating their stationary behavior to an equilibrium system of one lower dimension.⁶⁴ However, it can be shown that if d -dimensional probabilistic cellular automata with two states, and up-down symmetry, undergo a symmetry breaking, their critical behavior is identical to the corresponding Ising model in equilibrium.⁶⁴ Correlations in time are then equivalent to those generated by Glauber dynamics of the Ising system. $(d + 1)$ -dimensional crystals grown according to the rules of these cellular automata therefore have an order–disorder phase transition with correlations perpendicular to the growth direction characterized by the critical exponent ν , and those parallel to the growth direction by the exponent νz of the d -dimensional Ising model (z being the appropriate dynamical critical exponent).

In the next section, I will introduce a model for *layer by layer* growth of binary films. The atoms on the top layer are assumed to equilibrate completely (by surface diffusion *or* desorption–resorption mechanisms) before another layer is added.⁶⁵ Such an assumption is realistic only if the growth rate is much slower than characteristic equilibration times of the surface layer. The model satisfies detailed balance, and can therefore be analyzed with methods from equilibrium statistical physics. This discrete model is then used to justify a continuum formulation to the problem which is identical to the time dependent Landau–Ginzburg equation for model A dynamics.³

In general, a layer by layer growth mode is unstable, and growing surfaces are rough, as described in the previous chapters. The Kardar, Parisi, Zhang (KPZ) equation describes the dynamic fluctuations in the height of an amorphous surface.²¹ The interplay between roughness and ordering phenomena is then considered by introducing simple equations that couple fluctuations in height and the order parameter. Long-range correlations occur at the critical point for the onset of ordering in the surface binary mixture. This in turn leads to greater roughness fluctuations, whose scaling can be explored perturbatively around $d = 4$ dimensions.⁶⁶

While the order parameter for binary deposition is a scalar, we can more generally examine the case of a continuous order parameter formed on the surface layer. There are soft modes associated with such continuous symmetry breaking whose coupling to height fluctuations are explored at the end. In particular, the deterministic relaxation of the order parameter on an initially rough surface can in fact be described exactly through a generalized Cole-Hopf transformation. Interestingly, the relaxation process is super-diffusive and occurs through coarsening of domains (separated by sharp domain walls) on surface mounds.

4.2. Layer by layer growth

4.2.1. Discrete model

Binary growth is modeled by two kinds of atoms, A and B , which occupy the sites of a $(d+1)$ -dimensional hypercubic lattice. Let ϵ_{AA} , ϵ_{AB} , and ϵ_{BB} denote the interaction energies between neighboring atoms of types AA , AB , and BB , respectively. When each layer has N sites, there are 2^N possible configurations for a layer. The energy cost for adding a new layer of configuration γ on top of one in configuration α is the sum of the internal energy E_γ of the new layer, and the interaction energy $V_{\alpha\gamma}$ with the previous layer. These energies are the sums of all local bonds ϵ_{ij} between nearest neighbors ij within the new layer, and between the two layers, respectively. In addition, E_γ contains a chemical potential $\mu_A N_A + \mu_B N_B$ related to the partial pressures of A and B atoms in the gas phase.

Assuming that the top layer is in thermal equilibrium, the conditional probability that it is in configuration γ , given configuration α for the layer below, is

$$W_{\gamma\alpha} = \frac{\exp[-\beta(E_\gamma + V_{\alpha\gamma})]}{\sum_\delta \exp[-\beta(E_\delta + V_{\alpha\delta})]}, \quad (118)$$

where $T = (k_B\beta)^{-1}$ is the temperature at which the crystal is grown. After adding many layers, the steady-state probability for finding a configuration γ is determined by the stationarity condition

$$P_\gamma = \sum_\alpha W_{\gamma\alpha} P_\alpha, \quad (119)$$

which has the solution

$$P_\alpha = \frac{\sum_\gamma \exp[-\beta(E_\alpha + E_\gamma + V_{\alpha\gamma})]}{\sum_{\delta,\nu} \exp[-\beta(E_\delta + E_\nu + V_{\delta\nu})]} \equiv \frac{\sum_\gamma \exp[-\beta H_{\alpha\gamma}]}{\sum_{\delta,\nu} \exp[-\beta H_{\delta\nu}]}. \quad (120)$$

The above expression is the equilibrium probability for the top layer of a two-layer system, obtained after summing over the states of the bottom layer. Transverse correlation functions (i.e. perpendicular to the growth direction) are therefore exactly the same as correlation functions in a two-layer system.

From Eqs. (118) and (120) it follows that the system satisfies detailed balance, i.e.

$$W_{\alpha\gamma} P_\gamma = W_{\gamma\alpha} P_\alpha. \quad (121)$$

Thus, beyond a transient thickness, the crystal looks the same along or against the growth direction, and the sequence of layers corresponds to time evolution of thermodynamic equilibrium states. This generalizes previous results for cellular automata, which are obtained by setting the in-plane interactions E_α to zero. As in such cellular automata, the $(d+1)$ -dimensional system has transverse properties like d -dimensional models. In particular, phase transitions occur at the same temperature as for a d -dimensional two-layer system.

Generalizing the model, by allowing several layers at the surface to equilibrate, is straightforward. To mimic the large energy of the impinging particles, as well as their modified environment, we can assign each of the top ℓ layers from the surface a different temperature, through scaled interaction energies depending on its depth. The probability that a layer with configuration γ follows one in configuration α in the bulk is obtained by considering the layer at the moment when it is the ℓ th layer from the top, that is, immediately before its configuration is frozen. Denoting the configuration of the first $\ell-1$ layers by \mathcal{C}_γ and their energy (including the coupling to the ℓ th layer, and different interaction constants in the different layers) by $E(\mathcal{C}_\gamma)$, the conditional probabilities $W_{\gamma\alpha}$ can be written as

$$W_{\gamma\alpha} = \frac{\sum_{\mathcal{C}_\gamma} \exp\{-\beta[E_\gamma + V_{\alpha\gamma} + E(\mathcal{C}_\gamma)]\}}{\sum_{\delta,\mathcal{C}_\delta} \exp\{-\beta[E_\delta + V_{\alpha\delta} + E(\mathcal{C}_\delta)]\}}.$$

Following the approach for the case $\ell = 1$, we can show that the set of weights

$$P_\alpha = \frac{\sum_{\gamma, \mathcal{C}_\gamma, \mathcal{C}_\alpha} \exp\{-\beta[E_\alpha + E_\gamma + V_{\alpha\gamma} + E(\mathcal{C}_\gamma) + E(\mathcal{C}_\alpha)]\}}{\sum_{\delta, \nu, \mathcal{C}_\delta, \mathcal{C}_\nu} \exp\{-\beta[E_\delta + E_\nu + V_{\delta\nu} + E(\mathcal{C}_\delta) + E(\mathcal{C}_\nu)]\}},$$

describe a stationary state. It is easy to verify that this stationary solution satisfies detailed balance. The stationary state corresponds to an equilibrium Hamiltonian with 2ℓ layers, with interactions which depend on the distance from the closest surface. The top (or the bottom) layer describes the deposited surface, while the middle (ℓ or $\ell + 1$) layers describe transverse correlations in the bulk. While the correlations parallel to the growth direction are more complicated, the general conclusions for $\ell = 1$ remain valid.

4.2.2. Continuum formulation

In the above discrete model, we can use an Ising variable $\sigma_i = \pm 1$ to indicate if site i is occupied by atom A or B . Close to the critical point, density fluctuations occur over long distances and universal properties are better captured by considering a coarse-grained order parameter $m(\mathbf{x}, t)$. Here \mathbf{x} labels the d directions transverse to growth, while t which indicates time is also proportional to the coordinate parallel to the growth direction. Hence $m(\mathbf{x}, t)$ encodes the time history of the growth process. From the exact solution of the discrete problem, we know that the behavior of these configuration is equivalent to the time evolution of a d -dimensional system at equilibrium. In the continuum limit, the latter is described by the time-dependent Landau–Ginzburg equation³

$$\partial_t m = K \nabla^2 m + r m - u m^3 + \eta_m(\mathbf{x}, t), \quad (122)$$

where $\eta_m(\mathbf{x}, t)$ is a random noise of zero mean, whose variance is proportional to the growth temperature.

Away from the critical point at $r = r_c$, fluctuations in m decay over a transverse correlation length ξ , and a longitudinal correlation “time” ξ^z . At the critical point itself, there is no intrinsic scale, and correlations decay as

$$\langle m(\mathbf{x}, t) m(\mathbf{x}', t') \rangle = \frac{1}{|\mathbf{x} - \mathbf{x}'|^{-2\chi_m}} g_m \left(\frac{|t - t'|}{|\mathbf{x} - \mathbf{x}'|^z} \right). \quad (123)$$

In dimensions $d > 4$, criticality occurs for $r = u = 0$ (the diffusion equation), leading to $z = 2$ and $\chi_m = (2 - d)/2$. On approaching criticality, the correlation length diverges as $\xi \propto |r - r_c|^{-\nu}$, with $\nu = 1/2$. For $d \leq 4$, the nonlinear

term um^3 is relevant, and the exponents can be calculated perturbatively³ in $\varepsilon = 4 - d$.

4.3. Rough growth

4.3.1. Dynamic roughening

The layer by layer growth mode cannot be maintained indefinitely, and the surface eventually becomes rough.⁶⁷ Let us denote the height of the surface at location \mathbf{x} at time t by a function $h(\mathbf{x}, t)$. As discussed in the previous sections, there is considerable evidence from simulations (and some experiments) that the resulting surfaces exhibit self-affine fluctuations, well described by the continuum KPZ equation (71). The self-affine fluctuations in the surface height will be described by dynamic scaling exponents χ_h and z , defined through

$$\langle [h(\mathbf{x}, t) - h(\mathbf{x}', t')]^2 \rangle = |\mathbf{x} - \mathbf{x}'|^{2\chi_h} g_h \left(\frac{|t - t'|}{|\mathbf{x} - \mathbf{x}'|^z} \right). \quad (124)$$

The linear equation for $\lambda = 0$ gives diffusive exponents $\chi_h = (2 - d)/2$ and $z = 2$. Any nonlinearity is relevant in $d \leq 2$, while sufficiently large λ is required in $d > 2$ to produce a rough phase ($\chi_h \geq 0$).

4.3.2. Coupling growth and ordering

There are few studies of the interplay between fluctuations in height and the order parameter. Some numerical simulations have incorporated both elements: as a model for diamond growth, Capraro and Bar-Yam⁶¹ introduced a variant of ballistic deposition which exhibits sublattice ordering. Kotrla and Predota⁶³ have examined binary deposition in 1+1 dimensions, resulting in domains with rough surfaces. In a recent work with Barbara Drossel,⁶⁶ we took an analytical approach to this problem. A different set of equations was given by Léonard and Desai⁶⁸ for the case of phase separation during molecular beam epitaxy. Their equations reflect the situation of MBE where particle deposition is random (in contrast to having sticking probabilities that depend on the local environment), and where the order parameter can only be built up through surface diffusion.

The starting point is the continuum Eqs. (122)–(71), describing the order parameter $m(\mathbf{x}, t)$ and height $h(\mathbf{x}, t)$ fluctuations. To these equations we added all terms consistent with the symmetries of the problem. The lowest order (potentially relevant) terms result in the following pair of coupled differential

equations

$$\begin{cases} \partial_t h = \nu \nabla^2 h + \frac{\lambda}{2} (\nabla h)^2 + \zeta_h - \frac{\alpha}{2} m^2, \\ \partial_t m = K \nabla^2 m + r m - u m^3 + \zeta_m + a \nabla h \cdot \nabla m + b m \nabla^2 h + \frac{c}{2} m (\nabla h)^2. \end{cases} \quad (125)$$

(Note that these equations satisfy the symmetry $m \mapsto -m$.) Fluctuations of the surface are modified by coupling to the order parameter, through the term proportional to αm^2 . There are also three coupling constants a , b , and c , which modify the order parameter fluctuations due to coupling to h .

As long as the binary mixture is disordered ($r > r_c$), fluctuations in m and hence m^2 , are short-ranged, and αm^2 acts as another source of white noise. The surface fluctuations should thus scale with the standard KPZ exponents. However, the range of correlations increases as $r \rightarrow r_c$ and $\xi \sim |r - r_c|^{-\nu} \rightarrow \infty$. This modifies (most likely increases) the overall amplitude of surface roughness, and height fluctuations over a scale L behave as

$$\sqrt{\langle \delta h^2(L, r) \rangle} = \xi^{\chi_h^c - \chi_h} L^{\chi_h} g(L/\xi), \quad (126)$$

where χ_h^c is the roughness exponent at criticality, which is discussed next.

4.3.3. Critical roughness

Under a change of scale $\mathbf{x} \mapsto b\mathbf{x}$, $t \mapsto b^z t$, $h \mapsto b^{\chi_h} h$, and $m \mapsto b^{\chi_m} m$, the nonlinear coefficients in Eq. (125) scales as $x \mapsto b^{y_x} x$, with

$$y_\lambda = y_a = y_b = \chi_h + z - 2, \quad y_c = 2\chi_h + z - 2, \quad y_\alpha = 2\chi_m - \chi_h + z. \quad (127)$$

The critical point in dimensions $d \geq 4$ occurs at $r = u = 0$. The linear diffusion equations at this point result in the bare field dimensions $\chi_h^0 = \chi_m^0 = (2-d)/2$. Taking account of the nonlinearities, we observe the following behaviors:

- $d > 6$: All nonlinearities are (perturbatively) irrelevant; the surface is smooth, and the order parameter goes through a classical phase transition.
- $4 < d < 6$: The leading nonlinearity is the term αm^2 describing the correlated noise acting on the surface height, with ($z = 2$)

$$y_\alpha^0 = 4 - d - \chi_h^c = 3 - d/2. \quad (128)$$

In these dimensions, the correlated noise is more relevant than the white noise from the flux variations.³¹ The correct result can in fact be obtained

simply by setting y_α to zero, leading to critical height fluctuations with

$$\chi_h^c = 4 - d > \frac{2 - d}{2}.$$

Note that while the roughness exponent is larger than its bare value, it is still negative. The scaling of the order parameter is not modified, and $\chi_m = (2 - d)/2$.

- $d \leq 4$: When the roughness exponent is positive, all the couplings λ , a , b , and c become relevant. Also in $d \leq 4$, the critical point of the Landau–Ginzburg model is no longer at $r = u = 0$, and a full renormalization group (RG) study is called for.⁶⁶ Ignoring the feedback from height fluctuations to the order parameter, we find to leading order an RG equation of the form

$$\frac{1}{(\alpha\lambda)} \frac{d(\alpha\lambda)}{d\ell} = \varepsilon - C(\alpha\lambda),$$

where $\varepsilon = 4 - d$ and C is a *positive* constant.

There is a fixed point at $\alpha\lambda = \varepsilon/C$, with roughness exponent $\chi_h^c = 0$. In $d > 4$, this is an unstable fixed point governing a transition between flat ($\chi_h^c = 4 - d < 0$) and rough phases (occurring for $\alpha\lambda < -(d - 4)/C$). For $d < 4$, this fixed point is stable and attracts all points with $\alpha\lambda > 0$. Negative values of $\alpha\lambda$ flow to a rough phase which is not perturbatively accessible. Including all nonlinearities in the equation for m complicates the analysis, but we did not find a fixed point whose critical behavior is different from the ordinary Landau–Ginzburg model (at least to lowest order).

Since the above analytical results are inconclusive, we also undertook numerical simulations. To study the interplay between surface roughening and phase separation we simulated a brick-wall restricted solid on solid model with two species of particles.⁶⁹ Already the $(1 + 1)$ -dimensional system shows a variety of different scaling behaviors, depending on how the two phenomena are coupled. In the most interesting case, which is related to the advection of a passive scalar in a velocity field, nontrivial scaling exponents are found.

4.4. *Continuous order*

4.4.1. *Stochastic evolution*

The situation on the ordered side of the phase transition is more complex. The analogy to the dynamics of the lower-dimensional system suggests that the

leading process is the gradual coarsening of the ordered domains. Such domains would then appear as cone-shaped columns in the bulk film, a reasonably common feature of growth textures. However, more work is necessary to verify and quantify this picture.

Another interesting situation is when the symmetry breaking involves a continuous, rather than a discrete (Ising like), order parameter. For example, we may consider deposition of spins which can realign on the surface but are frozen in the bulk. More interestingly, the growth of crystals involves translational and orientational symmetry breakings in the plane. In the simplest case of a vector order parameter, we can simply generalize Eqs. (125) by replacing the scalar m with an n -component vector $\vec{m}(\mathbf{x}, t)$. While the discussion of critical roughening is not significantly modified from the Ising case ($n = 1$), new issues arise pertaining to the ordered phase.

The most common excitations of the broken symmetry phase are *soft (Goldstone) modes*, which can in principle couple to the surface roughness. The simplest example is provided by the XY model ($n = 2$), where the direction of the vector can be described by an angular field $\theta(\mathbf{x}, t)$. Including the lowest order terms which satisfy rotational symmetry leads to the coupled equations of motion

$$\begin{cases} \partial_t h = \nu \nabla^2 h + \frac{\lambda}{2} (\nabla h)^2 + \zeta_h - \frac{\alpha}{2} (\nabla \theta)^2, \\ \partial_t \theta = K \nabla^2 \theta + \zeta_\theta + a \nabla h \cdot \nabla \theta. \end{cases} \quad (129)$$

Interestingly, these are precisely the equations that Eq. (100) proposed in the contexts of moving flux lines and drifting polymers earlier. In particular, in $d = 1$ the KPZ exponents ($\chi_h = 1/2$ and $z = 3/2$) are recovered for the surface roughness, while the angular fluctuations remove any long-range order. Further analysis is again necessary for the case $d = 2$. Specifically, an important aspect of the field θ not present in the earlier studies is its angular nature. It could thus include vortices which are *topological defects*. Such defects typically play an important role in equilibrium two-dimensional systems, and have been recently considered in a number of related nonequilibrium situation.⁷⁰

4.4.2. Deterministic textures

As discussed before, the nonlinear KPZ equation can be recast as a linear diffusion equation through the Cole-Hopf transformation. This transformation can in fact be generalized to describe the coupling of the surface height to a

vector order parameter. Consider a field of unit spins, $|\vec{s}(\mathbf{x}, t)| = 1$, and set

$$\vec{W}(\mathbf{x}, t) = \exp\left[\frac{\lambda h(\mathbf{x}, t)}{2\nu}\right] \vec{s}(\mathbf{x}, t). \quad (130)$$

A diffusive equation of the field $\vec{W}(\mathbf{x}, t)$, as

$$\partial_t \vec{W} = \nu \nabla^2 \vec{W} + \frac{\lambda}{2\nu} \eta_h(\mathbf{x}, t) \vec{W}, \quad (131)$$

can be recast into the pair of coupled differential equations

$$\begin{cases} \partial_t h = \nu \nabla^2 h + \frac{\lambda}{2} (\nabla h)^2 + \left(\frac{2\nu^2}{\lambda}\right) \vec{s} \cdot \nabla^2 \vec{s} + \eta_h(\mathbf{x}, t), \\ \partial_t \vec{s} = \nu [\nabla^2 \vec{s} - (\vec{s} \cdot \nabla^2 \vec{s}) \vec{s}] + \lambda \nabla h \cdot \nabla \vec{s}. \end{cases} \quad (132)$$

Note that the transverse component of $\nabla^2 \vec{s}$ contributes to $\partial_t \vec{s}$, thus ensuring that the magnitude of \vec{s} is not changed in time, while the longitudinal component of this quantity couples to the surface height.

It can be checked easily that for $n = 2$, the parametrization $\vec{s} = (\cos \theta, \sin \theta)$, reduces Eqs. (132) to Eqs. (129) in the special limit of $\alpha\lambda = 4\nu^2$, $K = \nu$, $a = \lambda$, and $\eta_\theta = 0$. It is also possible to construct other Cole-Hopf transformations for cases when $\alpha\lambda < 0$.

Starting from any arbitrary initial condition at $t = 0$, the deterministic limit ($\eta_h = 0$) of Eqs. (131)–(132) is easily solved using the diffusion kernel, as

$$\begin{aligned} \vec{W}(\mathbf{x}, t) &= \exp\left[\frac{\lambda h(\mathbf{x}, t)}{2\nu}\right] \vec{s}(\mathbf{x}, t) \\ &= \int \frac{d^d \mathbf{x}'}{(4\pi\nu t)^{d/2}} \exp\left[-\frac{(\mathbf{x} - \mathbf{x}')^2}{4\nu t} + \frac{\lambda h(\mathbf{x}', 0)}{2\nu}\right] \vec{s}(\mathbf{x}', 0). \end{aligned} \quad (133)$$

The saddle-point evaluation of the above integral (formally exact as $\nu \rightarrow 0$) captures the long-time behavior of the solution. The surface profile

$$h(\mathbf{x}, t) = \min_{\mathbf{x}'} \left[h(\mathbf{x}', 0) - \frac{(\mathbf{x} - \mathbf{x}')^2}{2\lambda t} \right], \quad (134)$$

consists of a set of parabolic mounds centered at locations $\mathbf{x}' = \mathbf{x}_0(\mathbf{x}, t)$ corresponding to high points of the initial surface. Note that the evolution of the

surface profile in this limit is independent of \vec{s} . The evolution of spins on the other hand is completely controlled by the surface height, and given by

$$\vec{s}(\mathbf{x}, t) = \vec{s}(\mathbf{x}_0(\mathbf{x}, t), 0), \quad (135)$$

that is, each of the surface mounds carries the spin of its initial high point! Such behavior is quite different from the diffusive evolution of spins in the absence of coupling to the surface profile. For self-affine initial surface profiles, the relaxation of the height and spins is now both diffusive. Furthermore, the spin textures produced by this process are domains separated by sharp domain walls, very different from the soft modes and vortices that characterize diffusive relaxation. Similar extensions of the Cole-Hopf transformation to matrix order parameters are also possible, and could for example describe relaxation of crystalline substrates.

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