

The equilibrium behavior is unchanged, and

$$\lim_{t \rightarrow \infty} \langle |h(\mathbf{q}, t)|^2 \rangle = \frac{Dq^2}{\mu q^2(\rho g + \sigma q^2)} = \frac{D}{\mu(\rho g + \sigma q^2)}, \quad (61)$$

as before. Thus the same static behavior can be achieved by different dynamics. The static exponents (e.g.,  $\chi$ ) are determined by the equilibrium (stationary) state and are unchanged, while the dynamic exponents may be different. As a result, dynamical critical phenomena involve many more universality classes than the corresponding static ones.

## 2. Dynamic Scaling in Growing Films

Inhomogeneities in a deposition process may lead to formation of rough surfaces. Fluctuations in the height  $h(\mathbf{x}, t)$  of the surface (at location  $\mathbf{x}$  and time  $t$ ) can be probed directly by scanning microscopy, or indirectly by scattering. Analytical or numerical treatments of simple growth models suggest that, quite generally, the height fluctuations have a self-similar character; their average correlations exhibiting a dynamic scaling form,  $\langle [h(\mathbf{x}, t) - h(\mathbf{x}', t')]^2 \rangle = |\mathbf{x} - \mathbf{x}'|^{2\alpha} f(|\mathbf{x} - \mathbf{x}'|^z / |t - t'|)$ . The *roughness* and *dynamic* exponents,  $\alpha$  and  $z$ , are expected to be *universal*; depending only on the underlying mechanism that generates self-similar scaling. Despite its ubiquitous occurrence in theory and simulations, experimental confirmation of dynamic scaling has been scarce. In some cases where such scaling has been observed, the exponents are different from those expected on the basis of analysis or numerics. I shall briefly review the theoretical foundations of dynamic scaling, and suggest possible reasons for discrepancies with experimental results. Note that in Eq. (47) we used the symbol  $\chi$  to indicate the roughness exponent. Following Family and Vicsek,<sup>8</sup>  $\alpha$  is the symbol used in the context of growing surfaces. The symbols  $\zeta$  and  $H$  are also used to describe the same exponent.

### 2.1. Dynamic scaling

The growth of films by deposition is clearly of great technological interest. It has recently been recognized that such growth processes also pose important issues in the physics of nonlinear and complex systems. Rather crudely, we can distinguish between three types of growth morphologies:

- (i) *Layer by layer* growth is naturally most desired from the technological stand-point. From a theoretical perspective, this type of growth is not

stable in the presence of inhomogeneities in the deposition beam. However, this is only a statement about asymptotic behavior, as it is clearly possible to grow many layers in this mode.

- (ii) *Unstable* growth occurs when the selected orientation of the substrate cannot be maintained. The instability is usually manifested by the formation of mounds, or other macroscopic features on the surface. Another type of instability results from growth that is controlled by a diffusive field. This instability may lead to formation of fractal aggregates. This is probably unrelated to growth by deposition and will not be discussed here.
- (iii) *Self-affine* surfaces appear in a growth mode that is intermediate between the above. The average orientation of the surface is maintained, but it becomes rough; the amount of roughness grows with time and/or scale of observation in a self-similar fashion.

The most economical way to characterize self-affine roughness is by appealing to a dynamic scaling form<sup>11</sup>: If the height of the surface at location  $\mathbf{x}$ , at time  $t$ , is described by a function  $h(\mathbf{x}, t)$ , its average correlations may satisfy the scaling form

$$g(\mathbf{r}, t) \equiv \langle [h(\mathbf{x}, \tau) - h(\mathbf{x} + \mathbf{r}, \tau + t)]^2 \rangle = |\mathbf{r}|^{2\alpha} f\left(\frac{t}{|\mathbf{r}|^z}\right). \quad (62)$$

Family and Vicsek<sup>11</sup> first proposed such scaling in the context of interface growth based on numerical results. There is now a firm theoretical basis for such behavior. The *dynamic exponent*  $z$  describes the evolution of correlated regions with time: initially different parts of the surface are independent; but regions of correlated roughness form over time, their size growing as  $\xi(t) \propto t^{1/z}$ . In each correlated region, the width of surface grows as the observation scale raised to the *roughness exponent*  $\alpha$ . Thus, the overall width of the surface initially grows as  $t^\beta$  with  $\beta = \alpha/z$ , until it saturates at  $L^\alpha$  where  $L$  is the sample size.

## 2.2. Discrete models

A large number of numerical models of growth, of varying levels of sophistication, have been developed.<sup>12</sup> Here, I shall briefly describe some of the simplest versions.

*Random Deposition:* Particles are dropped randomly over deposition sites, and stick to the top of the pre-existing column on that site.<sup>13</sup> The height of each column thus performs an independent random walk, such that

$$\langle h_i(t) \rangle = vt \quad \text{and} \quad \langle (h_i(t) - vt)^2 \rangle = Dt, \quad (63)$$

where  $v$  is the average deposition rate per column, and  $D$  denotes its fluctuations. Since the columns are independent, the correlation length does not grow with time,  $\xi = 1$  and  $1/z = 0$ . This model leads to unrealistically rough surfaces whose overall width grow with the exponent  $\beta = 1/2$  without saturation.

*Sedimentation:* After the dropped particle reaches the top of a column, it rolls down-hill to neighboring columns until it reaches a local height minimum, where it stops. The smoothening effects of the rolling particles generate correlations in height, and lead to more realistic surfaces. Since the motion of rolling particles is more or less diffusive, it is likely that the correlation length grows as  $\xi(t) \propto t^{1/2}$ . This value of  $z = 2$  is supported by numerical simulations,<sup>14</sup> which find in  $1 + 1$  dimensions (i.e. for a one-dimensional substrate) the exponents  $\alpha = 0.48 \pm 0.02$  and  $\beta = 0.24 \pm 0.01$ .

*Ballistic deposition:* Originally introduced for particles moving in a continuous space,<sup>15</sup> several variants with discretized particle locations have since been developed.<sup>11,12</sup> The deposited particles are now attached at the *first point of contact* with the growing aggregate. Since the first contact may be to the side of a high column, this model leaves behind a finite density of voids as it grows.

*Restricted solid on solid models:* There are no voids, and the height is a single-valued function of position. These models impose a restriction on the maximum height difference between neighboring columns.<sup>16</sup> A growth attempt that violates such a restriction is rejected. (This can be regarded as a crude form of desorption.) The last two models are expected to be in the same universality class, but better numerical values for the exponents are found from the latter. The simulations in  $d = 2$  give  $\alpha \approx 1/2$  and  $\beta \approx 1/3$ , while for  $2 + 1$  dimensions they result in  $\alpha \approx 0.38$  and  $\beta \approx 0.24$ .

*Realistic models:* Attempt to more closely mimic the physical processes involved in the growth of surfaces.<sup>17</sup> These include the detailed motion of the deposited atom, its subsequent movement on the surface; incorporation on steps or islands, island nucleation, and desorption. The main difficulty with simulating

all these processes is that they take place over a wide range of time scales, making the analysis of large scale roughness quite difficult.

### 2.3. Continuum equations

Rather than describing the detailed microscopic evolution of the surface, continuum equations focus on the (hopefully universal) macroscopic aspects of its roughness, e.g., the exponents  $\alpha$  and  $\beta$ . The general philosophy is to examine the evolution of coarse-grained (hydrodynamic) variables; in the example at hand, the height function  $h(\mathbf{x}, t)$ . Whereas in near equilibrium situations the evolution equation is obtained from variations of an energy functional, such an approach is not appropriate far away from equilibrium. Here we follow an approach described in Refs. 18 and 19: the equation of motion is decomposed as

$$\frac{\partial h}{\partial t} = \eta(\mathbf{x}, t) + \Phi[h, \nabla h, \nabla^2 h, \dots]. \quad (64)$$

*Particle deposition* is described by the first term. Thus  $\eta(\mathbf{x}, t)$  is a random function whose mean value gives the average particle flux at  $\mathbf{x}$ , and its fluctuations represent the shot noise in deposition. For simplicity, we shall assume that the noise is uncorrelated at different sites and different times, that is, it is a Gaussian process with

$$\begin{aligned} \langle \eta(\mathbf{x}, t) \rangle &= v, \\ \langle \delta\eta(\mathbf{x}, t) \delta\eta(\mathbf{x}', t') \rangle &= D\delta(\mathbf{x} - \mathbf{x}')\delta(t - t'). \end{aligned} \quad (65)$$

*Surface relaxation* subsequent to deposition is described by the functional  $\Phi$ . As we shall see in the following examples, it can depend on various properties of the height, such as its slope  $\nabla h$ , or curvature  $\nabla^2 h$ . We shall assume that the relaxation is *local*, that is, it can be adequately described by the first few terms of an expansion of  $\Phi$  in  $h$  and its gradients. Which terms can be included in such an expansion are then determined by the underlying symmetries and conservation laws appropriate to the dynamics. The basic idea is that any term that is not excluded for fundamental reasons of symmetry or conservation, will be generically present.

We shall now describe continuum equations for the discrete models introduced in the previous section.

*Random deposition* with no subsequent relaxation corresponds to  $\Phi = 0$ . Integrating  $\partial_t h = \eta(\mathbf{x}, t)$  yields

$$h(\mathbf{x}, t) = \int_0^t dt' \eta(\mathbf{x}, t'), \quad (66)$$

from which we can immediately obtain

$$\langle h(\mathbf{x}, t) \rangle = vt \quad \text{and} \quad \langle \delta h(\mathbf{x}, t) \delta h(\mathbf{x}', t) \rangle = Dt \delta(\mathbf{x} - \mathbf{x}'), \quad (67)$$

corresponding to  $\beta = 1/2$ , and a zero correlation length.

*Sedimentation* was originally analyzed by Edwards and Wilkinson (EW) in Ref. 20. They concluded that the main relaxational process is proportional to local curvature, leading to

$$\partial_t h = \eta(\mathbf{x}, t) + \nu \nabla^2 h. \quad (68)$$

This linear (diffusion) equation is readily solved in Fourier space, leading to

$$\langle |h(\mathbf{k}, \omega)|^2 \rangle = \frac{D}{\omega^2 + \nu^2 k^4}. \quad (69)$$

Recasting the above result in real space then leads to the exponents

$$z = 2, \quad \alpha = \frac{2-d}{2}, \quad \beta = \frac{2-d}{4}, \quad (70)$$

for a  $d$ -dimensional substrate in  $d+1$  dimensional space. In particular, in  $d=2$ ,  $\alpha=1/2$  and  $\beta=1/4$ , while for  $d=3$ , the mean-square width grows logarithmically in both space and time.

*Ballistic deposition* is generically a nonlinear process. There is no *a priori* reason why the relaxation function  $\Phi$  should not depend on the slope  $\nabla h$ . (A direct dependence on  $h$  itself is ruled out by the translational symmetry  $h \rightarrow h + \text{constant}$  of the underlying dynamics.) By symmetry, the relaxation should be the same for slopes  $\pm \nabla h$ , and hence the first term in an expansion in powers of slope starts with  $(\nabla h)^2$  leading to

$$\partial_t h = \eta(\mathbf{x}, t) + \nu \nabla^2 h + \frac{\lambda}{2} (\nabla h)^2 + \dots \quad (71)$$

Higher order terms can also be added, but are irrelevant in that they don't change the scaling properties. There are several excellent reviews of Eq. (71),

known as the KPZ equation.<sup>21</sup> Since the origin and properties of the nonlinear term are discussed in detail in these reviews,<sup>9,10</sup> I shall not elaborate on them any further. In  $d = 1$ , the nonlinear equation can in fact be solved exactly, and leads to  $\alpha = 1/2$  and  $\beta = 1/3$  (as compared to  $\beta = 1/4$  for the EW equation), in excellent agreement with the numerical simulations.<sup>11,12</sup> There are no exact solutions in  $d = 2$ , but based on simulations we can estimate  $\alpha \approx 0.38$  and  $\beta \approx 0.24$ . A characteristic signature of Eq. (71) is the exponent identity

$$\alpha + \frac{\alpha}{\beta} = 2, \quad (72)$$

obeyed in almost all simulations. The KPZ equation appears to describe the asymptotic behavior of most local, random growth processes.

#### 2.4. Conservative MBE models

We may ask why the KPZ nonlinearity is not present in the model of sedimentation. The reason is that it is forbidden by a conservation law. If the growth process does not allow the formation of *overhangs* or *voids*, and there is also no *desorption*, then all the incoming flux is incorporated into the growing aggregate. This means that the net mass of the aggregate, proportional to  $H(t) = \int d^d x h(\mathbf{x}, t)$ , can only change due to the random deposition; the relaxation processes should conserve  $H(t)$ . This immediately implies that the relaxation function must be related to a surface current, i.e.

$$\Phi = -\nabla \cdot \mathbf{j}[\nabla h, \nabla^2 h, \dots]. \quad (73)$$

The KPZ nonlinearity is thus ruled out, as it cannot be written as the divergence of another function. Following some observations of Villain,<sup>22</sup> many studies have focused on such conservative models in the context of MBE growth. The basic idea is that aggregates formed by the MBE process are typically free from holes and defects, and that the desorption of the adsorbed particles is negligible. It is thus argued that, at least over some sizeable preasymptotic regime, the relaxation processes should be conservative. Some examples of such conservative models are discussed in the remainder of this section.

*Surface diffusion* currents in equilibrium can be related to variations of a chemical potential via  $\mathbf{j} \propto -\nabla\mu$ . Since in the equilibrium between two phases, the chemical potential is proportional to local curvature ( $\mu \propto -\nabla^2 h$ ), this leads to an equation of motion

$$\partial_t h = \eta(\mathbf{x}, t) - \kappa \nabla^4 h. \quad (74)$$

This equation is again linear, and as for Eq. (69) can be solved by Fourier transformation to yield,

$$\langle |h(\mathbf{k}, \omega)|^2 \rangle = \frac{D}{\omega^2 + \kappa^2 k^8}. \quad (75)$$

The corresponding exponents in real space are,

$$z = 4, \quad \alpha = \frac{4-d}{2}, \quad \beta = \frac{4-d}{8}. \quad (76)$$

This leads to  $\alpha = 3/2$  and  $\beta = 3/8$  in  $d = 1$ , and  $\alpha = 1$  and  $\beta = 1/4$  in  $d = 2$ . Note that a surface remains self-affine, maintaining a well defined orientation, only as long as  $\alpha < 1$ . The above large values of the exponent  $\alpha$  indicate a break down of validity of the above equation for  $d \leq 2$ .

*Nonlinear MBE* models have been proposed partly to remedy the break down of the linear equation for  $\alpha \geq 1$ . One such model starts with a nonlinear chemical potential introduced by Sun, Guo, and Grant,<sup>23</sup> resulting in

$$\partial_t h = \eta(\mathbf{x}, t) - \kappa \nabla^4 h + \frac{\lambda'}{2} \nabla^2 (\nabla h)^2. \quad (77)$$

Despite its nonlinear form, Eq. (77) can in fact be analyzed to yield the exact exponents<sup>24</sup>

$$z = \frac{8+d}{3}, \quad \alpha = \frac{4-d}{3}, \quad \beta = \frac{4-d}{8+d}. \quad (78)$$

In particular  $\alpha = 2/3$  and  $\beta = 1/5$  in  $d = 2$ . However, in nonequilibrium circumstances, there is no reason for the surface current to be derivable from a chemical potential. Removing this restriction allows the inclusion of other nonlinearities,<sup>24,25</sup> such as  $\nabla(\nabla h)^3$ , which are in fact more relevant. More importantly, nonequilibrium currents should generically include a term proportional to  $\nabla h$ , which is the dominant gradient as discussed next.

*Diffusion bias* refers to such generic nonequilibrium currents that are proportional to the local surface slope.<sup>26</sup> One possible origin of such currents is in the Schwoebel barriers<sup>27</sup>: Atoms on a stepped surface are easily incorporated at the step to a higher ledge, but are reflected by a barrier towards jumping to a lower ledge.<sup>28</sup> This sets up a net up-hill current<sup>22</sup>  $\mathbf{j} = \nu' \nabla h$ , leading to an equation of motion

$$\partial_t h = \eta(\mathbf{x}, t) - \nu' \nabla^2 h + \dots. \quad (79)$$

This equation leads to an unstable growth of fluctuations, and therefore higher order terms are necessary to ensure stability. For example, Johnson *et al.*<sup>29</sup> have proposed the following nonlinear equation

$$\frac{\partial h}{\partial t} = \eta(\mathbf{x}, t) - \nabla \left( \frac{\nu' \nabla h}{1 + (\nabla h)^2} \right) - \kappa \nabla^4 h. \quad (80)$$

The instabilities in this equation develop into a complex array of mounds dubbed as SLUGs (Super Large Unstable Growths). Finally, it is also possible for the nonequilibrium currents to be oriented down-hill (as in sedimentation), in which case the behavior is the same as the EW equation (68) discussed earlier.

## 2.5. Discussion

An excellent review of the experimental research in this subject is provided by Krim and Palasantzas.<sup>30</sup> The observational methods include diffraction (specular or diffuse x-rays, RHEED, LEED, HR-LEED, and helium scattering), direct imaging (STM, AFM, SEM and TEM), and surface adsorption. A variety of metallic (silver, gold, copper, iron), and other (Si, InP, NbN, polymer) surfaces grown under a host of different conditions have been examined by such probes. Some of these surfaces exhibit unstable growth, while others appear to satisfy self-similar scaling. However, there is usually no clear-cut identification of the exponents with the theoretical models. Some experiments on gold and silver give roughness exponents consistent with the KPZ value, but larger values of  $\beta$ . Other surfaces give larger values of  $\alpha$ , consistent with those of the nonlinear MBE equation (78). The reader is referred to this review article<sup>30</sup> for the details. Perhaps the following statements at the end of the review are most revealing of the experimental situation: “Over 50% of the experimental work reported on here was published in the interval from January 1993 to August 1994. The pace of experimental work is clearly accelerating, and rapid advances in the field can be expected.”

Given the discrepancies between experiment and theory, we can also ask if important elements have been left out of the analysis. The formalism presented so far deals solely with a single coarse-grained variable, the height  $h(\mathbf{x}, t)$ . Other variables may play an important role in the evolution of  $h$ . For example, in many cases the roughness is intimately related to formation of microcrystalline grains. Variations in crystallinity have so far been left out of the theoretical picture. In principle, one could introduce an additional “order parameter”  $M(\mathbf{x}, t)$

describing the local degree of crystallinity. Surface relaxation may then depend on this order parameter, leading to  $\Phi(\nabla h, \nabla^2 h, M, \nabla M, \dots)$ . We should then also include an additional dynamical equation for the evolution of  $M$ . This direction will be explored further in the final section.

Equation (64) can be regarded as representing a complex filter, converting the white input noise  $\eta(\mathbf{x}, t)$  to the correlated random function  $h(\mathbf{x}, t)$  through the action of  $\Phi$ . So far, the focus has been on the relaxation function  $\Phi$ , assuming that the input noise is uncorrelated. Not surprisingly, correlations in the input noise lead to more correlated surface roughness with larger values of the exponents  $\alpha$  and  $\beta$ .<sup>31</sup> Maybe, in view of the rather large exponents observed experimentally, this point should be further investigated. Starting with  $D(\mathbf{k}, \omega) \equiv \langle |\eta(\mathbf{k}, \omega)|^2 \rangle \propto |\mathbf{k}|^{-2\rho}$ , roughness exponents may be tuned continuously by changing the parameter  $\rho$ . This is not satisfactory, as it relies on a rather arbitrarily chosen exponent for noise correlations. Here I shall propose another possibility which is less arbitrary, and may be relevant to some experiments. My choice of noise correlations is motivated by the scaling phenomena observed in turbulence, another interesting problem in nonlinear physics.

In the case of turbulence, the fluid is stirred at long length scales, setting up a *Kolmogorov energy cascade*<sup>32</sup> by which energy is transferred to shorter wave-length modes, ultimately dissipating at a microscopic scale. In the intermediate (inertial) regime, the energy density follows simple power laws. We may similarly assume that the deposition noise is correlated over long distances  $\ell$ , such that  $D(\mathbf{k})$  is large only for  $|\mathbf{k}| < 1/\ell$ . Distances intermediate between the atomic scale and  $\ell$  are then analogous to the inertial regime in turbulence. Adapting the arguments of Kolmogorov to the KPZ equation leads to

$$z \approx \frac{4}{3}, \quad \alpha \approx \frac{2}{3}, \quad \beta \approx \frac{1}{2}, \quad (81)$$

in all dimensions. (Because there is no conservation of a corresponding energy for the KPZ equation in  $d \neq 1$ , the arguments leading to Eq. (81) are considerably less compelling than those in turbulence. Hence, the above exponents should be regarded as a first approximation.) The exponent  $\beta = 1/2$  is similar to that of random deposition. Indeed, an early experiment on recrystallization of amorphous GaAs films,<sup>33</sup> obtained an exponent of  $\beta \approx 0.50$  for highly correlated surfaces.