KINETIC SURFACE ROUGHENING AND PATTERN FORMATION IN MOLECULAR BEAM EPITAXY

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1. INTRODUCTION

The theory of kinetic surface roughening has attracted attention due to the interest in growth processes like vapor deposition and molecular beam epitaxy, but also from the perspective of fundamental statistical mechanics. Typically the late stages of these growth processes are characterized by generic scale invariance of the correlation functions that is reflected in power-law behavior in space and time. Since the corresponding exponents do not depend on the microscopic details of the system under investigation it is possible to divide growth processes according to the values of these characteristic exponents into *kinetic universality classes*. The association with one particular class depends only on a few properties of the growth dynamics like conservation laws, the importance of defects in the growing film, etc. The determination of these *relevant* features is one of the important problems that have to be addressed by the theory of kinetic surface roughening and is the purpose of this work. Conversely, as soon as these relations are known the determination of scaling exponents allows conclusions about the physical processes that dominate the growth dynamics.

If the growth kinetics are dominated by vapor deposition and desorption processes the surface evolution can be described by the so-called Kardar-Parisi-Zhang (KPZ) equation [6]. This universality class has been studied extensively. In this overview I will concentrate on surface growth under conditions that are characteristic for molecular beam epitaxy (MBE). In a typical MBE experiment the vapor pressure above the surface is very small. Instead, deposition occurs through a particle beam directed towards the surface. Vapor deposition and desorption are usually negligible under these circumstances. As will be shown in Section 2 this leads to growth laws that are different form those of the KPZ class. In fact, it will become clear that there are at least two different universality classes of MBE growth.

Surface growth as in MBE or vapor deposition is typically characterized by dynamics far away from equilibrium, i.e., the deposition rate is sufficiently large so that the surface does not relax through surface diffusion to a state of thermal equilibrium between successive deposition events. In Section 3.1 it will be shown that equilibrium surface diffusion is an extremely slow process so that this condition is almost always fulfilled. Therefore, the growth process will never reach an equilibrium state although it may reach a steady state. But in contrast to dynamics close to equilibrium the growth process is not restricted by the laws of statistical mechanics such as the fluctuation-dissipation theorem. In this respect this kind of surface growth is in marked contrast to growth processes characteristic for phase ordering like domain growth, spinodal decomposition, and Ostwald ripening. In these ordering phenomena the system evolves towards the equilibrium state although it will never reach that state, at least not within finite time. Consequently, phase ordering dynamics are restricted by the fluctuation-dissipation theorem. This is one of the reasons why more analytical results are known for phase ordering dynamics than for kinetic surface roughening (see Ref. [5]). But there are more fundamental differences: In Section 5 I will explain that kinetic surface roughening is similar to critical dynamics whereas ordering dynamics are governed by the low-temperature properties of the equilibrium state the system evolves into. As a consequence kinetic roughening leads to surfaces that are scale invariant in space *and* time: on length scales smaller than the correlation length the surfaces are self-affine and the correlation length itself increases with a power law in time. In contrast systems that phase order are scale invariant in time exclusively, there is a characteristic length scale: the domain size. It is an important result of this work (see Refs. [4,5]) that in the case of diffusion barriers at step edges the growth of surfaces under MBE conditions is *not* described by the scaling theories of kinetic surface roughening. Instead, instabilities develop that lead to pyramid-like structures on the surface and that evolve in time very similar to domains in a magnetic system with conserved order parameter: The size of the pyramids is analogous to the domain size of the corresponding magnetic system. Thus, to study surfaces grown under conditions typical for MBE requires the knowledge of theories of kinetic roughening *and* of phase ordering. Therefore these kind of growth processes point to an interesting connection between these apparently quite different phenomena.

This overview is organized as follows: In Section 2 the scaling theory that forms the basis of the theory of kinetic roughening is presented. The different methods to model kinetic roughening are described in Section 3. In particular, it is pointed out that Monte-Carlo simulations and the description through Langevin equation are fundamentally different and that in principle both methods are needed to obtain a complete description of the growth phenomena. The subsections 3.1–3.3 are dedicated to the description of thermal surface roughening due to equilibrium surface diffusion, the description of nonequilibrium surface roughening under conditions of molecular beam epitaxy, and the description of pyramidal growth on surfaces due to instabilities caused by diffusion barriers at step edges. The final two sections contain derivations that are usually not included in publications, although the conclusions drawn from these calculations are crucial for the understanding of kinetic roughening. Therefore, I regard it as appropriate to include these derivations in this overview. In Section 4 the usual Kramers-Moyal expansion is used to obtain a Fokker-Planck equation that should describe the results of Monte-Carlo simulations. It is explicitly shown that such an approximation fails in the case of surface roughening. The last section describes the derivation of the so-called Martin-Siggia-Rose functional and explains how this theory can be used to distinguish between *relevant* terms in an equation of motion that are important to describe the asymptotic behavior of surface growth phenomena and *irrelevant* terms that can be dropped from the equation without changing growth exponents.

Refs. [1–5] are appended to this overview. Equations and figures of these publications that are cited in this overview are quoted in the following way: Eq. (III.3.6) refers to the equation (3.6) of Ref. [3] and Fig. V.12 corresponds to figure 12 of Ref. [5].

2. SCALING THEORY OF SURFACE ROUGHENING

Throughout this article we describe the surface position through a single valued function $h(\mathbf{r}, t)$ that may be measured from some plane of reference. Instead, since we are mainly interested in the fluctuations of the surface, we define the local position $h(\mathbf{r}, t)$ of the surface at time t as the deviation from the average surface position. Or, in other words, we measure the height in a comoving frame of reference so that $\sum_{\mathbf{r}} h(\mathbf{r}, t) = 0$ for all times t. The theory of kinetic surface roughening assumes that surface configurations generated in the growth process are scale invariant, i.e., surfaces at different stages of the growth process look similar after a suitable rescaling of space $\mathbf{r}$, time t and height h. More precisely, in the calculation of statistical averages such as correlation functions, etc., the rescaled height

$$\tilde{h}(\mathbf{r}, t) = b^{-\zeta} h(b\mathbf{r}, b^z t) \tag{2.1}$$

will produce the same results as the height $h(\mathbf{r}, t)$ for arbitrary scaling factors b. As an example let us consider the height-height correlation function

$$G(|\mathbf{r}|, t) = \langle (h(\mathbf{r} + \mathbf{x}, t) - h(\mathbf{x}, t))^2 \rangle \ . \tag{2.2}$$

Using (2.1) in (2.2) we obtain the homogeneity relation

$$G(r, t) = b^{-2\zeta} G(br, b^z t) \ . \tag{2.3}$$

Such scaling laws are well known in the field of critical dynamics, e.g., the order parameter correlation function $C(r, t) = \langle \mathbf{M}(\mathbf{r}, t)\mathbf{M}(\mathbf{0}, t) \rangle$ of a magnetic system scales at the critical point as [7,8]

$$C(r, t) = b^{d-2+\eta} C(br, b^z t) \ . \tag{2.4}$$

This similarity will be exploited in Section 5 to justify the scaling assumption (2.1) for growing films.

Choosing $b = 1/r$ in Eq. (2.3) the scaling law

$$G(r, t) = r^{2\zeta} g(r/\xi(t)) \tag{2.5}$$

is obtained, where the correlation length

$$\xi(t) \sim t^{1/z} \tag{2.6}$$

has been introduced. This correlation length can be regarded as the wavelength of a typical fluctuation. For late times so that $r \ll \xi(t)$ we expect that $G(r, t)$ becomes stationary, i.e.,

$$G(r, t \to \infty) = g_0 r^{2\zeta} \ . \tag{2.7}$$

The exponent ζ is called roughness exponent and describes the transverse wandering of the surface over a distance r. On scales much larger than the correlation length the surface appears to be smooth. Consequently the correlation function becomes independent of r in this limit. Thus, the scaling function behaves as $g \sim x^{-2\zeta}$ for $x \gg 1$ and

$$G(r,t) \sim t^{2\zeta/z} \sim \xi^{2\zeta} \ . \tag{2.8}$$

The dynamical exponent z describes via Eq. (2.6) the lateral spread of surface fluctuations. At the same time the amplitude of these fluctuations or, equivalently, the surface roughness increases according to Eq. (2.8). A consistent description of the surface through a single valued function $h(\mathbf{r},t)$ requires a roughness exponent $0 \le \zeta \le 1$. If $\zeta < 1$ the amplitude of surface fluctuations, resp. the width of the surface, increases slower than their typical wavelength ξ. Such surfaces are called self-affine in contrast to self-similar surfaces with $\zeta = 1$.

It is instructive to compare the scaling laws (2.5,2.6-2.8) with the scaling laws valid for phase ordering dynamics. This will be done in Section 3.3.

3. MODELS OF KINETIC ROUGHENING

There are two methods that have been used extensively to model molecular beam epitaxy: Monte-Carlo simulations of microscopic models and the study of Langevin equations. The kind of microscopic models that are used in the simulations have been described in Refs. [1–3]. The Langevin equations that are expected to describe the late time regime of these microscopic models are discussed in [3] as well. The two methods are quite different: The former models the dynamics processes on a microscopic level and measures the resulting macroscopic properties such as correlation functions, surface width, and exponents. Since crossover effects can be quite pronounced (see, e.g., Ref. [2]) it can be difficult to associate the model with a distinct universality class, resp. a corresponding Langevin equation. This has led to incorrect classification of growth models in the past [9,10]. Furthermore, it is almost impossible to decide without additional knowledge, which properties of the model are important or *relevant* for the observed asymptotic behavior and which properties can be changed without affecting the exponents. No analytic approaches are know that could predict the exponents for a given model and therefore the physical insight in the mechanism that determines the asymptotic behavior is rather limited.

These deficiencies require the comparison of the results with those of Langevin equations. The description of growth processes through Langevin equations renounces a microscopic description and therefore cannot explain phenomena for which the discreteness of the lattice is important such as reflection high-energy electron diffraction (RHEED) oscillations. However, in the asymptotic regime, i.e., after the deposition of many monolayers, such effects are no longer important and a continuum description is well suited to study the scaling behavior described in the previous section. The parameters in Langevin equations are macroscopic quantities such as the surface stiffness, the mobility, etc. In principle it would be desirable to calculate these parameters from a microscopic model. Unfortunately no analytic methods, not even approximate, are known, which allow the derivation of the Fokker-Planck equation from a master equation that describes a given microscopic model used in Monte-Carlo simulations. This was already discussed in Ref. [11] and will be explained in more detail in Section 4. Since it is not possible to find the corresponding Langevin equation analytically one usually studies the most general Langevin equation that is compatible with the symmetries of the discrete model. Because it is always possible that some symmetry is either missed or erroneously incorporated this approach can lead to an incorrect association of a discrete model with a Langevin equation. E.g., a term $\sim \nabla^2 h$ was omitted in equations that describe the relaxation of surfaces through surface diffusion since such a term cannot appear under equilibrium conditions when detailed balance is obeyed. I will come back to this point in Section 3.2.

Although only linear Langevin equations can be solved exactly, several analytic methods are known to calculate the scaling exponents ζ and z for a given Langevin equation. First of all, by simple power counting it is possible to determine which terms in the Langevin equation are relevant and which terms can be dropped without changing the asymptotic behavior. This will be demonstrated in Section 5. In that section it is also shown how a Langevin equation for a d-dimensional surface can be transformed into a $d+1$-dimensional static field theory, which in turn can then be investigated by renormalization group methods.

3.1 Equilibrium Surface Diffusion

In this section the relaxation of surfaces into the equilibrium state and the fluctuations around this state are discussed. The dynamical evolution is governed by the fluctuation-dissipation theorem and therefore, as we will see below, the relevant Langevin equation can be derived from the free energy of the surface, given by the product of surface tension σ and area of the surface,

$$\mathcal{F} = \int d^d x \, \sigma \sqrt{1 + (\nabla h)^2} \, . \tag{3.1}$$

For crystalline surfaces the surface tension is anisotropic, $\sigma = \sigma(\mathbf{n})$, $\mathbf{n}$ being the surface normal. Furthermore, a pinning potential $V \cos(2\pi h/a)$ should be added to the integrand of (3.1), a being the lattice constant, to incorporate the effects of the discreteness of the lattice. This pinning potential is responsible for the roughening transition [12]. Since this Kosterlitz-Thouless phase transition does not exist for growing films the pinning term will be neglected in the following discussion. But if, e.g., RHEED oscillations are to be described in a continuum theory the discreteness of the lattice becomes important and the pinning potential cannot be omitted.

In the following I restrict myself to the description of surface diffusion within a temperature range, where desorption from the surface, overhangs and other defects can be neglected. In that case mass conservation inherent of the diffusion process translates into height conservation,

$$\partial_t h(\mathbf{x}, t) = -\nabla \cdot \mathbf{j}[h(\mathbf{x}, t)] + \nabla \cdot \boldsymbol{\eta}_c(\mathbf{x}, t) \, . \tag{3.2}$$

As shown in Ref. [3], the equilibrium surface current $\mathbf{j}$ is obtained from the free energy (3.1) using Fick's law and Herring's equation [13]. The equation of motion takes the form

$$\partial_t h = \Lambda\Delta\frac{\delta\mathcal{F}}{\delta h} + \nabla\cdot\boldsymbol{\eta}\ , \tag{3.3}$$

and because of the fluctuation-dissipation theorem the kinetic coefficient Λ also appears in the noise correlation

$$\langle\eta_{c,i}(\mathbf{x},t)\eta_{c,j}(\mathbf{x}',t')\rangle = 2k_BT\Lambda\delta_{ij}\delta(\mathbf{x}-\mathbf{x}')\delta(t-t')\ . \tag{3.4}$$

In Sec. 5 it is shown that all nonlinearities in Eq. (3.3) are irrelevant, and one concludes that equilibrium surfaces diffusion can be described by the linear equation

$$\partial_t h = -\Lambda\tilde{\sigma}\Delta\Delta h + \nabla\cdot\boldsymbol{\eta}_c\ , \tag{3.5}$$

where $\tilde{\sigma}$ is the surface stiffness [3,12]. Thus, the relaxations towards the equilibrium state is characterized by the *double* Laplacian on the right-hand side of the equation of motion. The corresponding exponents are $z = 4$ and $\zeta = (d-2)/2$ [see Eqs. (5.8a,5.8b)].

The results of this continuum analysis are convincingly confirmed by Monte-Carlo simulations [3]. Regardless of the microscopic surface energy used in the simulation and regardless of the type of the hopping rates (Glauber-type hopping rates or Arrhenius rates) the results are basically indistinguishable from those obtained from Eq. (3.5). For instance, the equilibrium structure factor $S(\mathbf{k}, t\to\infty)$ can be perfectly well fitted to a form $A/(1-\cos k)$ (see Figs. III.2, III.14) that is also obtained when Eq. (3.5) is solved on a discrete lattice. The dynamical exponent $z = 4$ is confirmed by the Monte-Carlo simulations (see Fig. III.15) as well. These agreements are an excellent confirmation of the concepts of kinetic universality classes that are based on the assumption that microscopic details are unimportant for the large-scale behavior of fluctuating surfaces.

A dynamical exponent of $z = 4$ means that correlations between different parts of the surface build up very slowly and fluctuations of size ξ relax within a time $t \simeq \xi^4/(\Lambda\tilde{\sigma})$. Since fluctuations in the deposition rate in a MBE experiment are uncorrelated in space and time these fluctuations increase proportional to $(Ft)^{1/2}$. Thus, any deposition of particles, as small as the flux may be, drives the system out of equilibrium. Nevertheless, it takes some time until the nonequilibrium nature of the growth process becomes apparent. To get an estimate of the crossover time t^* we may solve Eq. (3.5) with an additional nonconserved noise term (see Eq. (3.9), next section) corresponding to the beam fluctuations. Similar arguments have been used by Tang and Nattermann [14]. Since the resulting equation is still linear it can be solved exactly and we obtain the following expression for the structure factor [for the derivation see Ref. [3], section III, Eq. (III.3.6)]

$$S(\mathbf{k},t) \equiv \langle h(\mathbf{k},t)h(-\mathbf{k},t)\rangle = \left(\frac{k_BT}{\tilde{\sigma}k^2} + \frac{D}{\Lambda\tilde{\sigma}k^4}\right)\left(1 - e^{-2\Lambda\tilde{\sigma}k^4t}\right)\ . \tag{3.6}$$

The coefficient D is proportional to the particle flux F. For k-modes with $k > k^* = [D/(k_BT\Lambda)]^{1/2}$ the structure factor (3.6) is dominated by the equilibrium contributions.

Only when the correlation length ξ becomes larger than the crossover length scale $1/k^*$, will the nonequilibrium nature of the growth process be visible. We can define the crossover time t^* by setting $\xi(t^*) = (2\Lambda\tilde{\sigma}t^*)^{1/4} = 1/k^*$. From the result

$$t^* = \frac{\Lambda}{2\tilde{\sigma}} \left(\frac{k_B T}{D} \right)^2 \tag{3.7}$$

we see that the crossover time increases quadratically with the inverse of the flux F.

3.2 Surface Growth in Molecular Beam Epitaxy

As soon as the deposition due to the particle beam is non-zero the surface starts to move and the description of the surface dynamics becomes a truly nonequilibrium problem. Under the conditions of the previous section, i.e., no overhangs, no defects, and no desorption, the mean velocity of the surface must be equal to the incoming flux F of particles. Therefore, the deviations from this mean velocity must again obey a conservation law,

$$\partial_t h(\mathbf{x}, t) = F - \nabla \cdot \mathbf{j}[h(\mathbf{x}, t)] + \eta(\mathbf{x}, t) \ . \tag{3.8}$$

The noise η is associated with the fluctuations of the particle beam and its first and second moments are given by

$$\langle \eta(\mathbf{x}, t) \rangle = 0 \quad \text{and} \quad \langle \eta(\mathbf{x}, t)\eta(\mathbf{x}', t') \rangle = 2D\delta(\mathbf{x} - \mathbf{x}')\delta(t - t') \ . \tag{3.9}$$

In the following the constant F will be dropped from the equations, i.e., we will describe the dynamics in a comoving frame of reference, where the mean height $\bar{h}(t) = L^{-d} \sum_{\mathbf{x}} h(\mathbf{x}, t)$ vanishes. The conservation law associated with Eq. (3.8) leads to a relation between dynamic and roughening exponent [15]. The scale transformation $\tilde{\mathbf{x}} = b\mathbf{x}$, $\tilde{t} = b^z t$ requires the introduction of the rescaled noise $\tilde{\eta}(\mathbf{x}, t) = b^{-(d+z)/2}\eta(\tilde{\mathbf{x}}, \tilde{t})$ in order to keep the noise correlator (3.9) invariant. Therefore Eq. (3.8) transforms as $\partial_t h(\mathbf{x}, t) = b^{z-\zeta-1}\nabla \cdot \mathbf{j}[b^\zeta h(\mathbf{x}, t)] + b^{(z-2\zeta-d)/2}\tilde{\eta}(\mathbf{x}, t)$. After integrating over the system size the term $\nabla \cdot \mathbf{j}$ drops out and we find that $\partial_t \bar{h}(t) = b^{(z-2\zeta-d)/2}\bar{\eta}(t)$ with $\bar{\eta}(t) = L^{-d} \sum_{\mathbf{x}} \tilde{\eta}(\mathbf{x}, t)$. Scale invariance requires

$$z = \gamma \quad \text{with} \quad \gamma = 2\zeta + d \ . \tag{3.10}$$

This exponent relation is obeyed in all models that can be described by equations of the form (3.8) and reflects the fact that the fluctuations in the mean velocity depend exclusively on the beam fluctuations.

The most important problem in the theory of kinetic roughening is the question of how the surface current $\mathbf{j}$ depends on the surface morphology. After the discussion of the previous section it is suggestive to incorporate the deposition of particles in MBE by adding the flux F and its fluctuations η to the right-hand side of Eq. (3.3). The resulting equation of motion reads

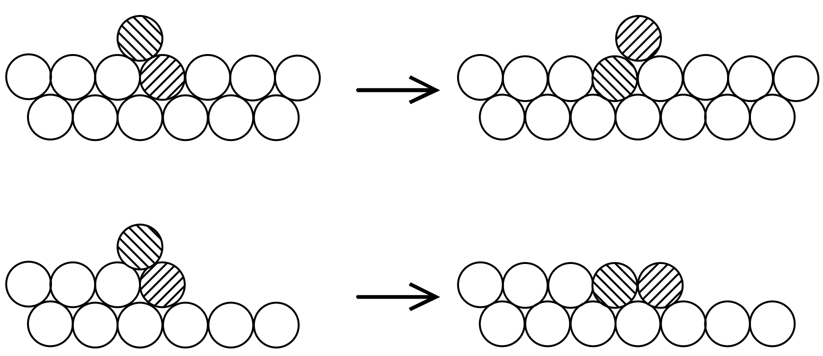
$$\partial_t h = \nu_2 \Delta h - \nu_4 \Delta\Delta h + F + \eta + \nabla \cdot \boldsymbol{\eta}_c + \text{higher order terms} \tag{3.11}$$

with $\nu_2 = 0$. It is one of the most important results of our Monte-Carlo simulations [1–3,16] that this conclusion is incorrect. *Generically, the dependence of the diffusion current on the surface morphology is different in the nonequilibrium case.* The results of the simulations show that the growth can only be described by an equation *with* an additional term $\sim \nabla^2 h$, i.e., $\nu_2 \neq 0$. Furthermore, this coefficient can have either sign, depending on the form of lattice potential for adatoms on the surface. In this way, the results for the $n = 1$ model [2,3,16] can be explained if a coefficient $\nu_2 > 0$ appears in the Langevin equation. In this case all nonlinearities are irrelevant (see Sec. 5) and the dynamical evolution can be described by the Edwards-Wilkinson [17] equation,

$$\partial_t h = \nu_2 \Delta h + F + \eta \ . \tag{3.12}$$

This result, first found in simulations of the $n = 1$ model [2], can be traced back to a slope-dependent surface current $\mathbf{j}(\nabla h)$ [16]. In the $n = 1$ model this current is in the downhill direction and therefore *stabilizes* the growth, i.e., the coefficient ν_2 is positive. Consequently the surface roughness is small. In fact, for two-dimensional surfaces the height-height correlation function increases only logarithmically with distance r and time t corresponding to a roughness exponent $\zeta = 0$ and a dynamical exponent $z = 2$. These results have recently also been observed in a light scattering experiment in the laboratory of T. Tiedge [18]. A microscopic derivation of this result is still missing: The usual Kramers-Moyal expansion for the master equation of the $n = 1$ model (see Sec. 4) yields a coefficient $\nu_2 < 0$ in total disagreement with the simulations. Physically, a coefficient $\nu_2 > 0$ means that the hopping rate on a flat terrace is smaller than the rate across a step edge onto the lower terrace. This is probably realized in materials where surface diffusion occurs via an exchange mechanism: As illustrated in Fig. 1, a diffusing adatom replaces another atom in the lower layer by pushing it onto the upper layer. Since at step edges the second atom is only pushed along the lower terrace, the hopping probability for this process can be larger, at least for some lattice symmetries. Another effect that leads to a positive ν_2 is the knock-out effect described in Ref. [5] (see Fig. V.10).

a)



b)

Figure 1. Illustration of adatom diffusion by an exchange mechanism: a) diffusion on a flat terrace, b) diffusion across a step edge. At least for some lattice symmetries, the diffusion rate for the process b) can be larger than the rate for process a). This leads to an inverted Ehrlich-Schwoebel effect.

The opposite effect, i.e. $\nu_2 < 0$, was in fact discovered earlier in Monte-Carlo simulations [1]. A Laplacian term with a negative coefficient was predicted by Villain [19] for the case when there are diffusion barriers at step edges. These barriers are also called Ehrlich-Schwoebel barriers [20]. They inhibit the diffusion of an adatom across a downward step edge. The adatoms therefore diffuse preferably towards the upper step edge as shown in Fig. 2. This generates a surface current in the uphill direction

a)

b)

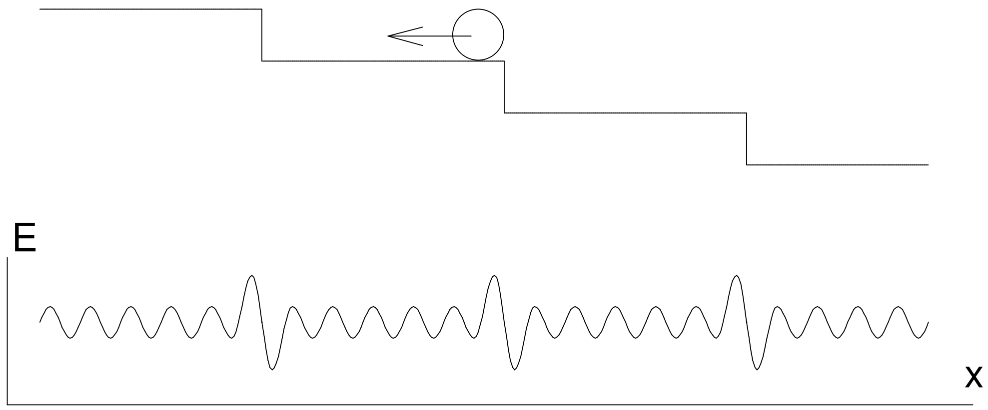


Figure 2. Schematic picture of the Ehrlich-Schwoebel effect: a) a diffusing adatom on a vicinal surface is repelled from a step edge; b) corresponding lattice potential showing the Ehrlich-Schwoebel barriers at step edges.

that in turn triggers the instability. A detailed description of the effect has been given in Refs. [4] and [5]. However, early models for MBE [15,21] failed to show this effect because of an oversimplification of the diffusion processes in these models. Only after the diffusion was modeled by hopping rates that obey detailed balance did the effects of this purely nonequilibrium contribution become apparent [1,3]. If $\nu_2 < 0$ Eq. (3.11) is linearly unstable and the higher order terms have to be taken into account. The instability is reflected in the formation of pyramidal structures, the height of which can increase extremely fast with system size in simulations (see Fig. I.4). The structure factor $S(\mathbf{k}, t)$ (III.2.4) is found to violate the scaling laws (III.2.5) of kinetic roughening (see Figs. I.3, III.10). The steady-state surface profile of the $n = 4$ model consists of one single pyramid (Fig. III.9). The sides of this pyramid have slopes much larger than one and therefore this final profile is physically meaningless in a solid-on-solid model since overhangs would naturally occur under such circumstances. This basically unbounded increase of slopes in the $n = 4$ model was subsequently explained [4] as a deficiency of this model: Generically the surface current $\mathbf{j}(\mathbf{m})$ as a function of the slope $\mathbf{m} = \nabla h$ has a zero at some finite value m_0. This leads to a selection mechanism that will be discussed in the following section. The currents $\mathbf{j}(\mathbf{m})$ can be measured directly in a Monte-Carlo simulation on tilted substrates [16]. It is found that there is no zero in the surface current of the $n = 4$ model other than at $\mathbf{m} = 0$ and consequently the instability does not saturate.

As explained above the generic case of surface growth under conditions typical for MBE is characterized by a nonequilibrium contribution $\nu_2 \nabla^2 h$ in the equation of motion. The special case $\nu_2 = 0$ corresponds to the situation that the diffusion rate for an adatom on a flat terrace is exactly the same as the diffusion rate across a step edge. It is unlikely that this will be found in nature, but if the difference in the diffusion rates is small it is possible that within a certain time interval a behavior can be observed that corresponds to the theory with $\nu_2 = 0$. In that regime the growth is characterized by large roughness exponents; the $n = 2$ model [1,3] provides an example of this case. More general cases are discussed in Ref. [3].

3.3 Pattern Formation in the Unstable Growth Regime

In this section the temporal evolution of the instability in the case $\nu_2 < 0$ will be reviewed. In this case the nonlinearities in the surface current $\mathbf{j}(\mathbf{m})$ have to be taken into account. The analysis of adatom diffusion on vicinal surfaces [19,22,4,5] shows that for tilts m smaller than the inverse of the diffusion length l_d the current $\mathbf{j}$ increases $\sim m$, whereas for $m > 1/l_d$ it decreases as $\sim 1/m$. If this latter regime is extended to all $m > l_d$ it can be shown [4,3] that the slope of the surface profile increases without bound. This form of the surface current is found in the $n = 4$ model [16] and is the reason for the steady-state profiles observed in simulations of this model (Figs. I.4,III.9). But this behavior results from untypical lattice symmetries [4]. As argued in Refs. [4,5] in the generic case there will be at least one zero of the surface current $j(m)$ at some finite $m = m_0 > 0$ with $j'(m_0) < 0$. Whereas at $m = 0$ the derivative of the current is positive and therefore causes the instability, the zero at m_0 corresponds to a stable solution of the equation of motion and leads to an evolution that is similar to that of systems that phase order.

Since the differences between theories of kinetic roughening and theories of ordering phenomena like domain growth, Ostwald ripening and spinodal decomposition will play an important role in the following discussion it is helpful first to recall the scaling theory of ordering phenomena. Whereas kinetic roughening of surfaces can be characterized by *two* length scales, the correlation length and the width of the surface, ordering phenomena are characterized by a single length scale, the domain size $R(t)$*. Consequently, the order parameter correlation function has the simple scaling form

$$C(\mathbf{r}, t) \equiv \langle \mathbf{M}(\mathbf{r}, t) \mathbf{M}(\mathbf{0}, t) \rangle = \Phi_0^2 f(r/R(t)) \ . \tag{3.13}$$

The typical form of the scaling function f is shown in Figs. V.5b and V.6a for conserved scalar and vector order parameters. This form has to be contrasted with the scaling form of the slope-slope correlation function of kinetic roughening that can be deduced from Eq. (2.5)

$$g_s(\mathbf{r}, t) = \langle \mathbf{m}(\mathbf{r}, t) \mathbf{m}(\mathbf{0}, t) \rangle = r^{2\zeta - 2} g_s(r/\xi(t)) \ . \tag{3.14}$$

*For vector order parameters domains in the strict sense do not exist. Nevertheless, there is no problem to define the characteristic length scale with the help of the correlation functions, see Ref. [5], chapter 3.

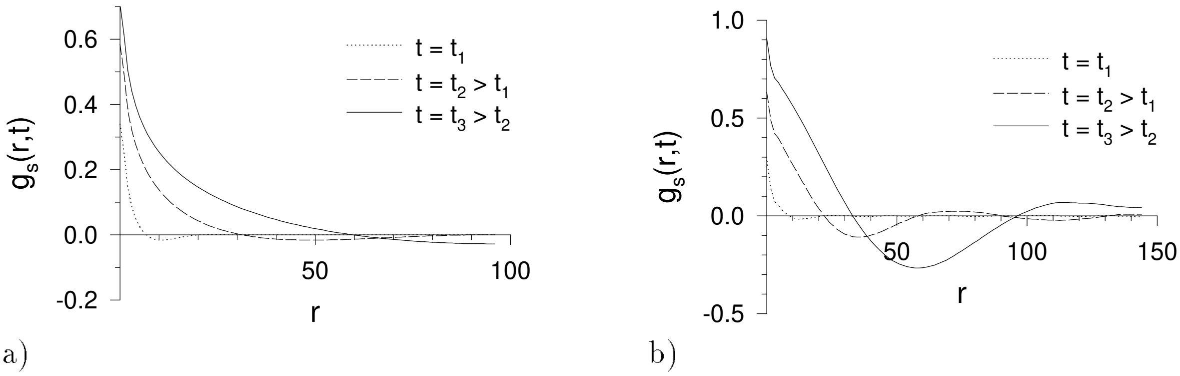


Figure 3. Comparison of the slope-slope correlation functions $g_s(\mathbf{r},t) = \langle \mathbf{m}(\mathbf{r},t)\mathbf{m}(\mathbf{0},t)\rangle$ for kinetic roughening (a) and pyramid formation (b) for three different times.

Thus, the scaling forms (3.13) and (3.14) are formally equivalent if $\zeta = 1$. But the scaling functions are different in the two cases: Whereas the order parameter correlation function $C(\mathbf{r},t)$ (3.13) has an oscillatory behavior with a period of the domain size, the slope-slope correlation function remains constant for $r \gtrsim \xi(t)$. The different behavior is shown in Fig. 3. In Fig. 3a the slope-slope correlation function resulting from the equation $\partial h = -D\Delta\Delta h + \eta$ is plotted. For two dimensional surfaces this corresponds to the case of kinetic roughening with a roughness exponent $\zeta = 1$. In Fig. 3b the slope-slope correlation function that is obtained from a numerical integration of Eq. IV.5 is shown. Whereas for early times it is hard to notice a difference between the two curves, the oscillatory behavior of the slope-slope correlation function in the case of pyramid formation (Fig. 3b) becomes apparent. Thus, the oscillatory behavior seen in Fig. III.13 is already an indication that the unstable growth with $\nu_2 < 0$ cannot be described by theories of kinetic roughening.

There is another important difference between phase ordering dynamics and kinetic roughening: Whereas kinetic roughening of surfaces is driven by the noise into a state far from equilibrium, phase ordering dynamics are governed by the so-called $T = 0$ fixed point, i.e., the noise is unimportant [23]. For the unstable growth due to the Ehrlich-Schwoebel effect this means that beam fluctuations force the system out of equilibrium and generate the nonequilibrium dependence of the surface current on the local tilt, but in the resulting equation of motion (IV.4) the noise can be set to zero; it does not influence the formation of pyramidal structures on the surface.

As explained in Refs. [4,5] the Langevin equation that describes the evolution of the instability can be cast into the form

$$\partial_t \mathbf{m} = \Lambda \nabla\nabla \cdot \frac{\delta \mathcal{F}}{\delta \mathbf{m}} + \nabla \eta \tag{3.15}$$

with a surface current $j_\alpha = \partial V / \partial m_\alpha$, $\alpha = 1, 2$, that can be derived from a pseudo free energy

$$\mathcal{F} = \int d^d x \left[\tfrac{1}{2}(\nabla m_x)^2 + \tfrac{1}{2}(\nabla m_y)^2 + V(\mathbf{m}) \right] \ . \tag{3.16}$$

Of course, there is no guarantee that the surface current can be written in this form, but first results for surface currents with additional terms that cannot be derived from a potential $V(\mathbf{m})$ indicate that these additional terms are irrelevant and do not change the results [24]. The minima of the potential V correspond to the zeros of the surface current. In this formulation (3.15) it is immediately clear that the surface evolution will develop towards a state in which the surface is preferably oriented in directions that correspond to the minima of V. In fact, these are the only stable solutions of Eq. (3.15).

It is impossible to derive the locations of the zeros of the surface current in a continuum theory. The symmetry argument used in Refs. [4,5] guarantees the existence of at least one zero in the surface current, but the orientations of the sides of the growing pyramids may not coincide with the zeros that are obtained by this symmetry argument. As shown in Ref. [5] the knock-out effect can lead to additional zeros in the surface current that are not related to the crystalline symmetries of the growing film. There are several other, usually material dependent, mechanisms that can have similar effects. In any case the selected slopes correspond to the zeros of the surface current that are closest to $\mathbf{m} = 0$.

The typical form of the surface current is shown in Fig. 4. The functional dependence

$$j_\alpha(\mathbf{m}) = m_\alpha f_\alpha(\mathbf{m}) \ , \quad \alpha = 1, 2 \tag{3.17}$$

may be fairly complicated, but it turns out that the form of the growing pyramids and the results for the growth exponent are basically independent of these functions as long as the following conditions are fulfilled:

$$\begin{array}{l} f_\alpha(\mathbf{0}) = -\nu_2 > 0 \ , \quad f_\alpha(\mathbf{m}_0) = 0 \ , \quad \alpha = 1, 2 \\ \text{and, for cubic symmetries,} \quad f_\alpha(-\mathbf{m}) = f_\alpha(\mathbf{m}) \ , \quad f_1(m_1, m_2) = f_2(m_2, m_1) \ . \end{array} \tag{3.18}$$

For other lattice symmetries similar conditions must be obeyed. The solutions $\mathbf{m} = \mathbf{m}_0$ with $f_1(\mathbf{m}_0) = f_2(\mathbf{m}_0) = 0$ determine the orientations of the sides of the growing

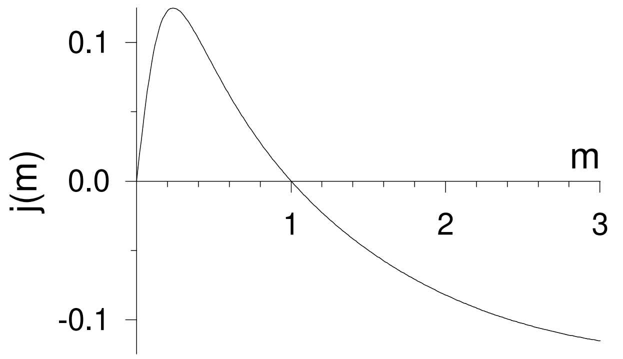


Figure 4. Surface current $|\mathbf{j}|$ as a function of the tilt $m = |\mathbf{m}|$. The current plotted here corresponds to Eq. IV.3 with $D_s = 1$ and $l_d = 4$.

pyramids. Both functions, (IV.3) and (IV.5), used for the surface current in Ref. [4] are in conformity with the conditions (3.18). But, the symmetry of these two functions is different: Whereas the current corresponding to Eq. (IV.3) is zero for all tilts $\mathbf{m} = \mathbf{m}_0$ with $|\mathbf{m}_0| = 1$, the current Eq. (IV.5) has only a discrete set of zeros, $\mathbf{m}_0 = (\pm 1, 0)$ and $\mathbf{m}_0 = (0, \pm 1)$. Therefore, the current (IV.3) leads to cone-like structures and only (IV.5) leads to true pyramids. In the language of phase ordering dynamics Eq. (IV.3) corresponds to a two-component vector order parameter with a potential $V(\mathbf{m}) = V(|\mathbf{m}|)$ in the free energy (3.16) similar to an XY model [5,25], whereas Eq. (IV.5) has the same symmetry as a four-state clock model. The differences in the potentials $V(\mathbf{m})$ are illustrated in Fig. V.12.

The evolution of the order parameter in systems that phase order has been reviewed in Ref. [5]. For a conserved order parameter the relevant Langevin equation is the Cahn-Hilliard equation [26]

$$\partial_t M_\alpha = \lambda \nabla^2 \frac{\delta \mathcal{F}}{\delta M_\alpha} + \nabla \cdot \boldsymbol{\eta}_\alpha \ , \quad \alpha = 1, \ldots, n, \tag{3.19}$$

with a free energy of the form (3.16). For a scalar order parameter ($n = 1$) this equation describes phenomena like spinodal decomposition and Ostwald ripening. In that case the average domain size† $R(t)$ grows proportional to $t^{1/3}$, the well-known Lifshitz-Slyozov-Wagner growth law [27]. In the case of a vector order parameter, $n \geq 2$, $V(\mathbf{m}) = V(|\mathbf{m}|)$, the characteristic length scale grows more slowly, $R(t) \sim t^{1/4}$, with additional logarithmic corrections for the XY model, $n = 2$ [28,25,5]. If the analogy between surface growth and phase ordering holds we would expect that the size of the cones generated by the current (IV.3) grows proportional to $t^{1/4}$, and the size of the pyramids generated by the current (IV.5) grows as $t^{1/3}$. In the case of the current (IV.5) this turns out to be correct, but surprisingly in the case of pyramidal growth the numerical integrations of Eq. (3.15) still show a growth law $\sim t^{1/4}$ [4,5].

The differences between phase ordering and surface growth become most apparent if we choose a surface current (3.17) with functions $f_1(\mathbf{m}) = 1 - m_1^2$, $f_2(\mathbf{m}) = 1 - m_2^2$. The free energy (3.16) then becomes the familiar φ^4 free energy with

$$V(\mathbf{m}) = \sum_{\alpha=1,2} \left[-\tfrac{1}{2} m_\alpha^2 + \tfrac{1}{4} m_\alpha^4 \right] \tag{3.20}$$

that is commonly used in the theory of phase ordering [5]. For this free energy the equations for the components of the order parameter (3.19) are completely decoupled so that this case is equivalent to the case of a scalar order parameter $n = 1$, i.e., the domains grow as $R(t) \sim t^{1/3}$. When the same free energy is used in Eq. (3.16) the equations for the components of the local slope of the surface profile turn out to be coupled,

$$\partial_t m_\alpha = -\Lambda \partial_\alpha \sum_{\beta=1,2} \partial_\beta \left[-\Delta m_\beta - m_\beta + m_\beta^3 \right] \ , \quad \alpha = 1, 2 \ , \quad \partial_\alpha \equiv \frac{\partial}{\partial x_\alpha} \ , \tag{3.21}$$

†As in Ref. [5] I will continue to use magnetic language, although, e.g., in the case of spinodal decomposition in fluids this is in fact the average droplet size.

although there is no coupling in the free energy. Rather, the coupling is due to the dynamics and a consequence of the constraint $\nabla \times \mathbf{m} = 0$ that is not present in domain growth phenomena. As shown in Fig. V.15 this difference has a profound effect on the growth law: The size of the pyramids grows proportional to $t^{1/4}$ in contrast to the Lifshitz-Slyozov-Wagner growth law.

There are many analytical theories that obtain the $t^{1/3}$ growth law in the case of spinodal decomposition. In the case of surface growth such a theory is still missing. The problem is not so much to find a theory that gives a $t^{1/4}$ growth law, but to explain why no $t^{1/3}$ behavior is observed. There were many theories proposed for spinodal decomposition that resulted in a $t^{1/4}$ behavior (see, e.g., Ref. [29]). Thus, a convincing theory must be applicable in both cases, domain and surface growth, and must yield a $t^{1/3}$ law in the case of domain growth and a $t^{1/4}$ law in the case of of surface growth.

The agreement between theoretical and experimental results in the field of kinetic surface roughening has so far been quite poor. However, in the case of the pyramidal instabilities recent experiments [30,22,31,24] seem to be at least in partial accord with the theory presented in Refs. [4,5]. In Ref. [30] the slope selection was observed in the epitaxial growth of Cu on Cu(100). Furthermore, at low temperatures the size of the pyramids was found to grow proportional to t^β with $\beta \simeq 0.26$. Similar values have been obtained by Palasantzas and Krim [31], whereas Stroscio *et al.* measure for Fe on Fe(100) an exponent that is slightly smaller, $\beta \simeq 0.16$. Again the slope selection was clearly observed. In the case of GaAs [22] pyramidal structures were found with a very small angle of about 3°. In this case it is not clear whether the slopes have saturated or not.

4. FROM MASTER TO FOKKER-PLANCK EQUATIONS

The models used in Monte-Carlo simulations of kinetic roughening [1–3] incorporate two different mechanisms: deposition and surface diffusion. In each time step a particle is added to the growing film with probability f, and a diffusive move is attempted with probability $(1-f)w_{i\to j}$, where i and j are nearest-neighbor sites. The corresponding master equation for the probability distribution $P\{h(t)\}$ of the height variables $h_i(t)$ reads

$$\begin{aligned} P(\{h(t+1)\};t+1) = {} & P(\{h(t)\};t) \\ & + \sum_{\{h'(t)\}} w\{h'(t),h(t)\}P(\{h'(t)\};t) - w\{h(t),h'(t)\}P(\{h(t)\};t) \end{aligned} \tag{4.1}$$

The sum runs over all possible sets $\{h'(t)\}$ of height variables. The transition rates $w\{h,h'\}$ from the set of variables $\{h\}$ to the set $\{h'\}$ is a sum of two parts: $w = w_{\rm dep} + w_{\rm dif}$. The deposition rates $w_{\rm dep}$ are nonvanishing only for sets $\{h'\}$ that differ from the set $\{h\}$ only at one site i by $h'_i = h_i + a$ with a being the lattice constant,

$$w_{\rm dep}\{h,h'\} = fL^{-d}\sum_i \left[\delta_{h_i+a,h_i'}\prod_{j\neq i}\delta_{h_j,h_j'}\right] . \tag{4.2}$$

The diffusion rates $w_{\rm dif}$ are non-zero for transitions between sets that differ at two nearest-neighbor sites i, j:

$$w_{\rm dif}\{h,h'\} = \frac{1}{n_b}(1-f)L^{-d}\sum_{<ij>} w_{i\to j}\{h\}\left[\delta_{h_i+a,h_i'}\delta_{h_j-a,h_j'}\prod_{l\neq i,j}\delta_{h_l,h_l'}\right] , \tag{4.3}$$

where n_b is the number of nearest neighbors, e.g., $n_b = 4$ for a square lattice and the sum runs over all nearest-neighbor pairs $<ij>$. The hopping rates $w_{i\to j}$ depend on the the local surface morphology and on the chosen dynamics, e.g., Glauber-type hopping rates or Arrhenius rates. Based on the work of Kurtz [32], Fox and Keizer [33] claimed that under quite general preconditions the master equation (4.1) can be approximated by the Fokker-Planck equation that is obtained through the usual Kramers-Moyal expansion [34]

$$\partial_t P(\{h\};t) = -\sum_i \frac{\partial}{\partial h_i}\left[K_i^{(1)}\{h\}P(\{h\};t)\right] + \frac{1}{2}\sum_{ij}\frac{\partial^2}{\partial h_i\partial h_j}\left[K_{ij}^{(2)}\{h\}P(\{h\};t)\right] . \tag{4.4}$$

The first and second moments $K_i^{(1)}$ and $K_{ij}^{(2)}$ are defined as

$$K_i^{(1)}\{h\} = \sum_{\{h'\}}(h_i'-h_i)w\{h,h'\} \tag{4.5}$$

and

$$K_{ij}^{(2)}\{h\} = \sum_{\{h'\}}(h_i'-h_i)(h_j'-h_j)w\{h,h'\} . \tag{4.6}$$

There are several difficulties with this approach, especially in the case of equilibrium surface diffusion [11]. Nevertheless, this approximation has been widely used to derive the Langevin equation that describes the evolution of surfaces in a particular Monte-Carlo simulation [35].

Introducing the flux $F = afL^{-d}$ and the attempt frequency $D = \frac{a}{n}(1-f)L^{-d}$ the moments (4.5,4.6) can be rewritten as

$$K_i^{(1)} = F + D\sum_{<j>_i} w_{j\to i} - w_{i\to j} \tag{4.7}$$

and

$$K_{ij}^{(2)} = aF\delta_{ij} + aD\sum_{<l>_i}\left[\delta_{ij}\Big(w_{l\to i}+w_{i\to l}\Big) - \delta_{jl}\Big(w_{i\to j}+w_{j\to i}\Big)\right] , \tag{4.8}$$

where $<l>_i$ denotes the nearest-neighbor sites of site i, and a is again the lattice constant. These moments constitute the diffusive part and the noise correlator of the Langevin equation that corresponds to the Fokker-Planck equation (4.4):

$$\partial_t h_i(t) = K_i^{(1)}\{h(t)\} + \eta_i(t) \tag{4.9}$$

with

$$\langle \eta_i(t) \rangle = 0 \quad \text{and} \quad \langle \eta_i(t)\eta_j(t') \rangle = \langle K_{ij}^{(2)}\{h(t)\} \rangle \delta(t-t') \; . \tag{4.10}$$

The noise η can be decomposed into a nonconserved part η_F describing the fluctuations in the deposition,

$$\langle \eta_{F,i}(t) \rangle = 0 \quad \text{and} \quad \langle \eta_{F,i}(t)\eta_{F,j}(t') \rangle = aF\delta_{ij}\delta(t-t') \; , \tag{4.11}$$

and a conserved part associated with the fluctuations of the surface current,

$$\eta_{c,i}(t) = \sqrt{aD} \sum_{x=1}^{d} \Big[w_{i_{x+} \to i} + w_{i \to i_{x+}} \Big]^{1/2} \nu_{i_{x+}}^{(x)}(t) - \Big[w_{i \to i_{x-}} + w_{i_{x-} \to i} \Big]^{1/2} \nu_i^{(x)}(t) \tag{4.12}$$

with

$$\langle \nu_i^{(x)}(t) \rangle = 0 \quad \text{and} \quad \langle \nu_i^{(x)}(t) \nu_j^{(y)}(t') \rangle = \delta_{ij}\delta_{xy}\delta(t-t') \; ,$$

and $i_{x\pm}$ denote the nearest-neighbor sites of i in the positive and negative x direction.

In the remainder of this section this approximation will be discussed in the context of models that use Glauber-type hopping rates

$$w_{i \to j} = \Big[1 + \exp\Big(\tfrac{1}{k_B T} \Delta\mathcal{H}_{i \to j} \Big) \Big]^{-1} = \tfrac{1}{2} - \tfrac{1}{2} \tanh\Big(\tfrac{1}{2k_B T} \Delta\mathcal{H}_{i \to j} \Big) \; , \tag{4.13}$$

where

$$\begin{aligned} \Delta\mathcal{H}_{i \to j} = {} & J \sum_{<l>_i} \Big[|h_i - h_l - a|^n - |h_i - h_l|^n \Big] + J \sum_{<l>_j} \Big[|h_j - h_l + a|^n - |h_j - h_l|^n \Big] \\ & + J \Big[|h_i - h_j - 2a|^n - |h_i - h_j|^n - 2\Big(|h_i - h_j - a|^n - |h_i - h_j|^n \Big) \Big] \end{aligned} \tag{4.14}$$

is the change in the surface energy $\mathcal{H} = J \sum_{<ij>} |h_i - h_j|^n$ associated with the move. These hopping rates are in many respects less realistic than Arrhenius-type hopping rates, but they show the difficulties connected with the approximation (4.4) more clearly.

In order to proceed one has to expand the moments $K_i^{(1)}$ and $K_{ij}^{(2)}$ in powers of the height differences. For arbitrary n the absolute value function in (4.14) poses an additional problem for the expansion. For the sake of clarity I therefore will concentrate on the one-dimensional $n = 4$ model [3]. The main difficulties in the approximation of master equations by Fokker-Planck equations will already appear in this model. The energy change for the $n = 4$ model is

$$\Delta\mathcal{H}_{i \to i+1}^{(4)} = Ja \Big\{ -4\partial_x^2 (\partial_x h_i)^3 + 6a \left[6(\partial_x h_i)^2 + \partial_x^2 (\partial_x h_i)^2 \right] + 4a^2 \left[6\partial_x h_i - \partial_x^3 h_i \right] + 18a^3 \Big\} \, . \tag{4.15}$$

The derivatives have to be interpreted in their discretized form, i.e., $\partial_x h_i \equiv h_{i+1} - h_i$, $\partial_x^2 h_i \equiv h_{i+1} - 2h_i + h_{i-1}$, etc. Since the discrepancy between the results of the approximation discussed in this section and the Monte-Carlo simulations will already occur at the linear level, the nonlinear terms will not be written down explicitly in the following. The Glauber-type diffusion rates

$$w^{(4)}_{i\to i+1} = \frac{Ja^3}{k_B T}\left[-6\partial_x h_i + \partial_x^3 h_i - 18a\right] + \mathcal{O}((\partial_x h_i)^2) \tag{4.16}$$

obey the symmetry relation $w^{(4)}_{i\to i+1}\{h\} = w^{(4)}_{i+1\to i}\{-h\}$. Thus, the following result is obtained for the first moment:

$$K_i^{(1)} = F + \frac{DJa^3}{k_B T}\left[6\partial_x^2 h_i - \partial_x^4 h_i\right] + \mathcal{O}((\partial_x h_i)^3)\ . \tag{4.17}$$

Eq. (4.9) can be interpreted as the discretized version of a continuous Langevin equation. But, if we use the result (4.17) this Langevin equation reads

$$\partial_t h(x,t) = F + \nu_2 \partial_x^2 h(x,t) - \nu_4 \partial_x^4 h(x,t) + \mathcal{O}((\partial_x h_i)^3) + \eta_F(x,t) + \eta_c(x,t) \tag{4.18}$$

with a *positive* coefficient ν_2. This is in marked contrast to the results of Monte-Carlo simulations of the $n = 4$ model [1,3] that are consistent with a *negative* value of ν_2. Even more disturbing is the fact that the coefficient ν_2 is non-zero for the equilibrium case $F = 0$. As mentioned in Sec. 3.2 the equilibrium fluctuations are well described by the equation $\partial_t h = -\nu_4 \Delta\Delta h + \eta_c$, in fact a term $\sim \nabla^2 h$ violates the fluctuation-dissipation theorem and must not appear. This failure of the expansion was already noticed in Ref. [11] and points to the main deficiency of Eq. (4.4), resp. Eq. (4.9): The form of the right-hand side of the Langevin equation, i.e., the first moment $K_i^{(1)}$, does not depend on the nature of the noise. Regardless of whether conserved or nonconserved noise is used, the deterministic part of the Langevin equation (4.9) does not change. This cannot be true for the Langevin equation that describes the Monte-Carlo simulations: In the nonequilibrium case ($F \neq 0$) there is a coefficient $\nu_2(F) < 0$ for the $n = 4$ model [1,3,16], whereas $\nu_2(F = 0) = 0$ for all models considered in Refs. [1–3].

For the $n = 2$ model the expansion described above gives a coefficient $\nu_2 = 0$ that is in agreement with the Monte-Carlo results for that model [1,3]. This is due to a special symmetry of the $n = 2$ model [3]. To study models with $0 < n < 2$ the absolute value function on the right-hand side of Eq. (4.14) must be replaced by an analytic function. When this is done a coefficient $\nu_2 < 0$ is obtained, again in contrast to the simulations of the $n = 1$ model, the results of which can be explained by the Edwards-Wilkinson equation [1,3].

The coefficient ν_2 can be measured directly in Monte-Carlo simulations on tilted substrates [16]. In these simulations the surface current j is measured as a function of the tilt m. In the limit $m \to 0$ the coefficient ν_2 is obtained: $\lim_{m\to 0} j(m) = -\nu_2$. The results of these simulations [16] are in agreement with the usual Monte-Carlo simulation [11,1–3] and, again, disagree with the results obtained by the treatment discussed in this section.

It is not completely clear why Kurtz's theorem [32] does not apply to these surface models. But in the usual Kramers-Moyal expansion the Fokker-Planck equation is obtained in the limit of a small parameter ϵ going to zero keeping $K^{(1)}/\epsilon$ and $K^{(2)}/\epsilon^2$ constant [36]. Eqs. (4.7,4.8) suggest the choice $\epsilon = a$. But the result (4.17) shows that the dominant term of the first moment is the trivial constant F that can be scaled away by measuring the height in a comoving frame of reference, $h \to h + Ft$. Thus, the crucial terms are the subdominant terms of the first moment and obviously the expansion fails to give the correct expression for these terms.

5. THE MARTIN-SIGGIA-ROSE FUNCTIONAL

As already mentioned in Section 3, one of the important problems in the theory of kinetic roughening is the determination of those properties of a model that influence the asymptotic behavior. In this section it will be demonstrated how this task can be achieved in the framework of Langevin dynamics, i.e, it will be shown in which way one can distinguish between *relevant* terms of the equation that are important for the calculation of growth exponents and *irrelevant* terms that can be omitted without changing the asymptotic results.

To start with, the Langevin equation is cast into the form of a Lagrangian, the so-called Martin-Siggia-Rose functional [37,38], that can be investigated using field theoretical methods. The derivation of the Lagrangian is quite standard [38–40], nevertheless, it is worth-while to repeat it here in the context of surface dynamics. Let us consider the set of stochastic equations

$$\partial_t h_j(t) = f_j\{h(t)\} + \eta_j(t) \ . \tag{5.1}$$

Here $\{h(t)\}$ denotes the whole set of height variables $h_j(t)$ that describes the position of the surface measured from some plane of reference. The index j labels the sites of this d-dimensional substrate and $\eta_j(t)$ is a stochastic variable representing thermal noise and beam fluctuations, the first and second moments of which obey

$$\langle \eta_j(t) \rangle = 0 \quad \text{and} \quad \langle \eta_j(t)\eta_n(t') \rangle = 2\Gamma_{jn}\delta(t-t') \ . \tag{5.2}$$

In case of equilibrium relaxation, the function f is related to the free energy of the system through

$$f_j\{h\} = -\Gamma_{jn}\frac{\delta \mathcal{F}\{h\}}{\delta h_n} + V_j\{h\} \ . \tag{5.3}$$

Throughout this section summation over repeated indices is implied. The appearance of the kinetic matrix Γ_{jn} in Eqs. (5.2) and (5.3) is a consequence of the fluctuation-dissipation theorem and ensures that the stationary solution of the corresponding Fokker-Planck equation is the equilibrium distribution $P_{\mathrm{eq}}\{h\} = e^{-\mathcal{F}\{h\}}$. The second term on the right-hand side of (5.3) must obey the condition

$$\frac{\delta}{\delta h_j}\left(V_j\{h\}e^{-\mathcal{F}\{h\}}\right) = 0 \ , \tag{5.4}$$

but is arbitrary otherwise. For purely dissipative dynamics, corresponding to discrete models that obey detailed balance, it can be shown that V vanishes identically [41].

An arbitrary correlation function $\langle A\rangle$ of the height variables $h_j(t)$ is obtained by averaging over the noise distribution $P\{\eta\}$,

$$\langle A\{h\}\rangle = \int \mathcal{D}\eta P\{\eta\} A\{h_\eta\} \tag{5.5}$$

where $\mathcal{D}\eta$ is a short hand for $\prod_{j,t} d\eta_j(t)$ and h_η is the solution of the Langevin equation (5.1) for one particular realization of the noise variables $\eta_j(t)$. Instead of integrating over the noise variables $\eta_j(t)$, the integral is transformed into an integral over the height variables $h_j(t)$, i.e.,

$$\begin{aligned}
\langle A\{h\}\rangle &= \int \mathcal{D}h A\{h\} P\{\eta\{h\}\} \det(\mathcal{J}\{h\}) \\
&= \int \mathcal{D}h\mathcal{D}\eta A\{h\} P\{\eta\} \det(\mathcal{J}\{h\}) \prod_{j,t} \delta(\eta_j(t) - \partial_t h_j(t) + f_j\{h(t)\}) \\
&= \int \mathcal{D}h\mathcal{D}\tilde{h}\mathcal{D}\eta A\{h\} P\{\eta\} \det(\mathcal{J}\{h\}) \\
&\qquad\qquad \times \exp\left[i\int dt\, \tilde{h}_j\Big(\partial_t h_j(t) - f_j\{h(t)\} - \eta_j(t)\Big)\right] \ ,
\end{aligned}$$

where $\mathcal{J} = \partial\eta/\partial h$ is the Jacobian that is generated by the transformation. The discretized version of Eq. (5.1) in the Ito-interpretation reads

$$h_j(t+1) = g_j\{h(t)\} + \eta_j(t+1)$$

with $g_j\{h(t)\} = h_j(t) + f_j\{h(t)\}$. Thus, starting from $h_j(t=0) = 0$ the evolution of the surface morphology can be generated recursively:

$$h_j(t) = \eta_j(t) + g_j\{\eta(t-1) + g\{\eta(t-2) + \ldots\}\} \ .$$

Since $\eta_j(t)$ appears only in the first term and there are no dependencies on $\eta_j(t+1)$, $\eta_j(t+2)$, etc., the matrix $\mathcal{J} = \partial\eta/\partial h$ is triangular and the diagonal elements are all equal to one and consequently, $\det(\mathcal{J}) = 1$. In the Stratonovitch interpretation the matrix $\mathcal{J}$ is still triangular, but the diagonal elements are no longer equal to one. Nevertheless, the results of the theory remain the same [39,40].

Taking the average over $\eta_j(t)$ with a gaussian probability distribution

$$P\{\eta\} \sim \exp\left[-\frac{1}{4}\int dt\, \eta_j(t)(\Gamma^{-1})_{jn}\eta_n(t)\right]$$

makes the calculations of correlation functions (5.5) formally equivalent to a problem of equilibrium statistical mechanics:

$$\langle A\{h\}\rangle = \int \mathcal{D}h\mathcal{D}\tilde{h} A e^{-J\{h,\tilde{h}\}} \Big/ \int \mathcal{D}h\mathcal{D}\tilde{h} e^{-J\{h,\tilde{h}\}} \ .$$

In the continuum limit Γ_{nm} transforms into a differential operator, and $J[h,\tilde{h}]$ becomes the so-called Martin-Siggia-Rose functional depending on the conjugate fields h and $\tilde{h}$,

$$J\{h,\tilde{h}\} = \int d^d x\, dt\, \Big\{ -\tilde{h}(\mathbf{x},t)\Gamma\tilde{h}(\mathbf{x},t) + i\tilde{h}(\mathbf{x},t)\Big[\partial_t h(\mathbf{x},t) - f\{h(\mathbf{x},t)\}\Big]\Big\} \ . \tag{5.6}$$

If dynamics of, e.g., magnetic systems are investigated one usually uses a free energy of the form

$$\mathcal{F}\{M\} = \tfrac{1}{2}\int d^d x\, \Big[(\nabla M)^2 + \rho M^2 + \tfrac{1}{2}uM^4\Big] \ . \tag{5.7}$$

The parameter ρ is proportional to the deviation $T - T_c$ from the critical temperature; for $\rho > 0$ the system is in the disordered phase, whereas for $\rho < 0$ there exists a finite order parameter M. Only for $\rho = 0$ is the system critical and scale invariant. However, as long as gravitational effects are neglected, the surface free energy does not depend on the position of the surface itself because of translational invariance. This is still true for the function $f\{h\}$ in Eq. (5.1), even for nonequilibrium dynamics, where f is not necessarily related to a free energy as in (5.3). Thus, $f\{h\}$ depends only on derivatives of the height h. In that respect surfaces are always “critical” without fine-tuning of parameters. This is the justification for the scaling hypothesis (2.1) for growing films.

There are a few counter examples, where the scaling hypothesis is not valid: To describe the dynamics of surfaces below the roughening temperature T_R a pinning term $V\cos(2\pi h/a)$ must be included in the free energy. This term leads to a Kosterlitz-Thouless-type phase transition and below T_R the system is no longer critical [12]. But under nonequilibrium conditions, where the incoming flux of particles is nonzero, the pinning potential is averaged to zero, and the roughening transition disappears. The second example is discussed in Section 3.2: the behavior of surfaces that develop instabilities leading to pyramidal structures is in fact similar to ordering dynamics as in spinodal decomposition [4,5].

Eq. (5.6) has the form of a Lagrangian of a $d+1$ dimensional field theory, therefore the same method of power counting can be used here. I will restrict myself here to the two cases of nonconserved and conserved noise, i.e, $\Gamma = D$ and $\Gamma = -D\nabla^2$. Since $J\{h,\tilde{h}\}$ is dimensionless we find that the dimensions of the fields h and $\tilde{h}$ are

$$[h] = [Dt]^{1/2} q^{(d+\alpha)/2} \qquad \text{and} \qquad [\tilde{h}] = [Dt]^{-1/2} q^{(d-\alpha)/2} \ ,$$

where q has the dimension of an inverse length and $\alpha = 0, 2$ for nonconserved and conserved noise, respectively. We now assume that the linear part of the function $f\{h(\mathbf{x},t)\}$ has the form $\nu_p \nabla^p h$. The two cases that are relevant for surface roughening are $p = 2$ and $p = 4$ [1–3]. This relates the dimension of time t to the inverse length scale q and fixes the dimensions of the fields to

$$[t] = [\nu_p]^{-1} q^{-p} \ , \tag{5.8a}$$

$$[h] = [D/\nu_p]^{1/2} q^{(d-p+\alpha)/2} \ , \tag{5.8b}$$

and

$$[\tilde{h}] = [D/\nu_p]^{-1/2} q^{(d+p-\alpha)/2} \ . \tag{5.8c}$$

Eq. (5.8) determines the scaling properties of the Lagrangian $J\{h, \tilde{h}\}$ at the gaussian fixed point. These are equivalent to the scaling properties of the linearized Langevin equation, e.g., Eq. (5.8a) reflects the scaling of the correlation length, $\xi \sim t^{1/z}$ with $z = p$ for linear equations, and $\zeta = (d - p + \alpha)/2$ is the roughness exponent at the gaussian fixed point.

Nonlinearities in $f\{h(\mathbf{x}, t)\}$ may or may not change these exponents. Here, I will discuss nonlinearities that differ from the linear term $\nu_p \nabla^p h$ by factors of $(\nabla h)^{2n}$, e.g., we will consider terms of the form

$$f_n = a_n \nabla \cdot \nabla^{p-2} [\nabla h (\nabla h)^{2n}] \ . \tag{5.9}$$

These are the most important nonlinear terms that can appear in the Langevin equations discussed in Refs. [1–3]. The scaling dimension of these terms can be inferred from the scaling dimension of $\nabla h \sim q^{(d+2-p+\alpha)/2}$. The dimension of the coefficients a_n is therefore

$$[a_n] \sim q^{-n\chi} \quad \text{with} \quad \chi = d + 2 - p + \alpha \ . \tag{5.10}$$

We now apply a coarse graining transformation, $\tilde{q} = bq$ with $b > 1$, to the functional (5.6). If applied successively this operation eliminates short-wavelength modes that are unimportant for the asymptotic scaling behavior. Under the transformation the coefficients a_n increase as $b^{n\chi}$ if $\chi > 0$. Otherwise they scale to zero, in which case these terms are unimportant, or *irrelevant*, in the asymptotic limit. Thus, the nonlinearities are *relevant* only if $\chi > 0$. This is the case for $d = 1$ in the presence of nonconserved noise ($\alpha = 0$) and the absence of a Laplacian term in the Langevin equation ($p = 4$). For $d = 2$ the nonlinear terms are marginal, i.e. $\chi = 0$, and no information about the importance of these terms can be obtained from power counting arguments. In all other cases the nonlinearities are irrelevant, i.e., $\chi < 0$. This includes the case of equilibrium surface diffusion, $p = 4$ and $\alpha = 2$, and cases with an Edwards-Wilkinson term $\nabla^2 h$.

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