Universality issues in surface kinetic roughening of thin solid films

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Abstract

Since publication of the main contributions on the theory of kinetic roughening more than fifteen years ago, many works have been reported on surface growth or erosion that employ the framework of dynamic scaling. This interest was mainly due to the predicted existence of just a few universality classes to describe the statistical properties of the morphology of growing surfaces and interfaces that appear in a wide range of physical systems. Nowadays, this prediction has proved to be inaccurate. This situation has caused a clear detriment of these studies in spite of the undeniable existence of kinetic roughening in many different real systems, and without a clear understanding of the reasons behind the mismatch between theoretical expectations and experimental observations. In this chapter we aim to explore existing problems and shortcomings of both the theoretical and experimental approaches, focusing mainly on growth of thin solid films. Our analysis suggests that the theoretical framework as yet is not complete, while more systematic and consistent experiments need to be performed. Once these issues are taken into account, a more consistent and useful theory of kinetic roughening might develop.

1 Introduction

During the last few years the research community has devoted a great effort to the understanding of a wide variety of interface growth phenomena under the framework of dynamic scaling concepts. These phenomena range from thin solid film growth and erosion processes to the growth dynamics of systems that were beyond the realm of traditional Statistical Mechanics or Condensed Matter Physics, such as tumors, bacterial colonies, etc. [1, 2, 3]. The appeal to describe the interface fluctuations observed in such diverse experimental domains is due to the simplicity of the original framework within which the various observations were described, namely the scaling Ansatz due to Family and Vicsek [4]. Thus, the *surface or interface width* or *roughness*, $W(t)$, defined as the rms deviation of the height field $h(\mathbf{r},t)$ around its mean value $\bar{h}(t)$, is seen to increase with time as a power law $W(t) \sim t^{\beta}$ for as long as the lateral correlation length $\xi(t) \sim t^{1/z}$ is smaller than the system size, L. For longer times the roughness becomes a constant $W \sim L^{\alpha}$. The scaling exponents α, β, and $1/z$ quantify thus, respectively, the spatial correlations of interface features, the temporal evolution of the interface roughness, and the coarsening process of the characteristic lateral correlation length of the interface, for a system displaying scale invariance in time and space. This behavior is a natural extension to a growing surface or interface, of the *dynamic* scaling hypothesis for equilibrium dynamics of critical systems [5]. Actually, the high temperature phase of the discrete Gaussian model, intensively studied in the context of the equilibrium roughening transition of a metallic surface (see e.g. [6, 7, 8]), already provided a well known example of a surface whose equilibrium dynamics fits completely with the FV Ansatz.[1] From

[1] In the kinetic roughening context, the corresponding universality class is termed Edwards-Wilkinson (EW) [9], making reference to the description by the corresponding Langevin equation.

the theoretical point of view, this fact encouraged the modeling of surface/interface growth by the use of discrete models of the Monte Carlo type, or by stochastic differential equations similar, as in the discrete Gaussian model, to, e.g. models A and B of critical dynamics, in the classification of Ref. [5]. These approaches were simplified by the implied existence of *universality* and universality classes, identified by the values of the critical exponents α, and z.[2] Moreover, the main source of phenomenology supporting the FV Ansatz was in the field of *fractal growth* [10]. Indeed the roughness exponent α coincides with the so-called Hurst exponent of the surface, seen as a *self-affine* fractal, and is thus related to the, say box-counting fractal dimension D_B, as [3, 10] $\alpha = d_E - D_B$, where d_E is the embedding Euclidean dimension.[3] Remarkable examples of non-equilibrium surfaces fulfilling the FV Ansatz were found to be the active zones of the *diffusion-limited aggregation* (DLA) or Eden models, much studied in the wider context of *pattern formation* outside of equilibrium [11]. Thus, by the mid-1980's enough theoretical (analytical and simulational) and some experimental evidence strongly conveyed the vision that the large scale properties of rough surfaces grown in equilibrium and far from equilibrium processes should fall into a few universality classes. One of these would be provided by the asymptotic properties of the celebrated Kardar-Parisi-Zhang (KPZ) equation [12], itself directly related to other non-equilibrium systems, such as fluid turbulence or directed polymers in random media [13, 14]. This appealing perspective triggered the interest of the scientific community. Thus, scaling analysis was applied to a large variety of theoretical and experimental systems (see in particular the compilation of experiments reviewed in [1, 3, 15]). Note that, given the interpretation of the scaling exponents, a natural procedure in an experiment is to, first, evaluate these and then, within the universality assumption, to try contrast them with those theoretically predicted in order to fully understand the mechanisms ruling the growth process. This deeper knowledge of the physical mechanisms governing the interface growth dynamics can have implications in many fields. For instance, in the growth of thin solid films, it can contribute to a better control of the deposition conditions in order to produce films with technological applications (electronic, electro-optical, catalytic, etc.) including fabrication of nanostructures [16]. Also for instance, in the growth of tumors [17], this type of identification has led to the conclusion that actual cell duplication (growth) takes place on the surface, rather than, as traditionally assumed, in the bulk, which might lead to appropriate techniques in order to, say, halt tumor growth. However, when trying to apply the above framework to understand the fluctuation properties of actual surfaces, in particular in the context of thin solid film production, the experimental evidence was far less compelling than implied by such a simple, unifying theoretical picture. Unfortunately, as noted in [18], "while many experiments on surface roughening during film growth have been interpreted in the context of the dynamic scaling hypothesis, no consensus has emerged concerning the relationship between roughness and growth exponents appropriate to a given growth process". This situation led M. Kardar to state in [19], that "despite its ubiquitous occurrence in theory and simulations, experimental confirmation of dynamic scaling has been [relatively, this is ours] scarce. In some cases where such scaling has been observed, the exponents are different from those expected". In fact, there do exist many experiments in which the surface fluctuations exhibit scale invariance. However, the spread in the values of the ensuing exponents (see e.g. Table 2 in [15]) questions the very existence of universality in these phenomena, not to mention the difficulty quoted in attributing a given set of experimental exponents to a specific physical mechanism.

Nowadays, seven years after publication of the above quotations, the situation seems to be

[2] Substitution of $L \to \xi(t) \sim t^{1/z}$ in the saturation value of the roughness leads to the scaling relation $\beta = \alpha/z$, which indeed occurs for a flat initial condition.

[3] We will employ d for the dimension of the substrate "on top" of which growth occurs along an additional dimension, thus $d_E = d + 1$. E.g. a one-dimensional random walk is a rough "surface" for which $d_E = d + 1 = 2$.

quite similar to that described in the reviews cited above. It is indeed true that some systems have been analyzed, and fully and consistently assigned to some of the main theoretical universality classes. Examples are found in experiments on paper burning [20] for the KPZ class in $d_E = 2$, sputtered Fe/Au multilayers [21], silica films grown by chemical vapor deposition [22], or growth/etching of epitaxial GaAs films [23], for the KPZ class in $d_E = 3$, or sedimentation of silica nanospheres [24] for the EW class in $d_E = 3$. However, many other systems have not been explained until now, and basically remain just phenomenologically described by a set of experimental exponents. Even in the study of highly idealized models of surface growth by Molecular Beam Epitaxy (MBE), such as those of Wolf-Villain (WV) [25] and Das Sarma-Tamborenea [26] (DT) —see below—, the very notion of universality can be questioned [27]. Thus we are forced into the question whether a consistent physical theory of kinetic roughening can be formulated, and with what use. Given the undeniable experimental evidence on surface kinetic roughening, we believe the answer is positive, but the development of the theoretical framework and tools are as yet incomplete. Moreover, the approach has to be inverted to some extent: rather than postulating universality and trying to fit observations to "idealized" universality classes, the generalization to these need come after detailed study of well defined systems. On the way, this process will also provide new developments in the field of generic models of non-equilibrium systems.

In this chapter we investigate the reasons for the mismatch between theory and experiment. Our aim is to assess the current status of kinetic roughening as a consistent physical theory, through the comparison between the "ideal" universality classes, as represented e.g. by the continuum stochastic equations of EW, KPZ, etc., and experimental observations. We will thus evaluate the practical relevance of the universality concept —including its very definition in some instances—, from the point of view of its pertinence to and of the the physical information it provides on the experimental systems. We will not dwell on more fundamental questions such as the origin of scale invariance properties in these systems, see e.g. [28], neither on the generic properties of the representatives of the "ideal" universality classes as instances of non-equilibrium systems with strong fluctuations. As mentioned in the previous paragraph, our approach is that these questions might obtain some answers by generalizing results obtained after close comparison between theory and experiments in well delimited physical instances. Interestingly, we will see that progress can be made here by strengthening and exploiting the connection with those fields in which the subject originated in the first place, namely, those of *pattern formation* and *fractal geometry*. We will restrict ourselves almost exclusively to the relatively reduced domain of thin solid film growth, and those immediately linked to it, while we expect that much of our discussion will carry over to other domains of applicability of the framework provided by dynamic scaling Ansatz. Most notably, we will *not* consider the specific case of kinetic roughening in media with *quenched disorder*. These are reviewed elsewhere for the case of fluid imbibition, see Ref. [29], in a spirit that is somehow akin to that of the present chapter. Moreover, we will restrict our attention, both in the discussion of theoretical and experimental approaches, mostly to those issues which we consider that are not properly understood, or that are promising lines of research with the goal of developing a more consistent picture of the field.

2 Status of the theory

2.1 "Ideal" universality classes

In this section, we briefly recall the main universality classes relevant for kinetic roughening. As stated above, we focus on the case in which the surface or interface is subject to time dependent noise [30]. In typical applications, these fluctuations arise in those of a driving flux (of, say, ag-

gregating units, atoms or molecules) acting on the system. This is a convenient way to represent the nature of the noise, but it does not by any means imply that its amplitude is directly the[4] square root of the average external flux. For instance in studies of growth by MBE [31] or by electrochemical or chemical vapor deposition (ECD, CVD, respectively) [32] the noise term appearing in the Langevin equation for the interface is seen to be rather more involved than that. However, and this can never be overemphasized, the universal behavior applies to asymptotic properties, well beyond all existing transients (induced by, e.g. physical instabilities acting on the system) and crossovers (due to competition among various physical mechanisms, each of which is dominant for a different range in time and space). For the type of systems we are considering, the asymptotic properties are adequately described by equations featuring additive noise, which is Gaussian and uncorrelated in time and space.

Below follows a summary of the current consensus [33] on the universality classes for kinetic roughening. For each of these, the representative continuum equation for the surface height is provided. Again, this has to be understood in the renormalization group (RG) sense, namely, the fact that a given experimental system belongs to one of these universality classes does *not necessarily* mean that the surface height —seen at the spatial and temporal scales resolved in the experiment— follows "in detail" the corresponding Langevin equation. Rather, it means that both systems display the same statistical properties.[5] A natural and standard classification of the universality classes considers the existence of conservation laws on the surface height in the representative equation. Thus, we have:

1. Non conserved systems: If the height evolves in time in the absence of a conservation law on the mass of aggregating units at the interface (such is the case for, e.g., ECD and CVD), for instance if bulk vacancies are significative, or conformal growth occurs [22], then the expected asymptotic behavior is that of the KPZ equation

$$\frac{\partial h}{\partial t} = \nu \nabla^2 h + \frac{\lambda}{2}(\nabla h)^2 + \eta. \tag{1}$$

   In this equation, the first two terms on the right hand side are, to lowest order, *representing* respectively surface tension effects (ν is a positive constant) and growth along the local normal direction with a constant average rate, λ. In this and subsequent interface equations, $\eta(\mathbf{r}, t)$ is the noise term alluded to in the previous paragraph. The scaling exponents characterizing this universality class are exact in $d_E = 2$, $\alpha = 1/2$, and $z = 3/2$ ($\beta = 1/3$), and approximately equal to $\alpha = 0.39$ and $z = 1.61$ ($\beta = 0.24$) in $d_E = 3$ [34]. This universality class is exceedingly important in the theory of non-equilibrium processes.[6] However, as mentioned above there are very few experimental realizations of this scaling behavior.

2. Conserved systems: The other three main universality classes in kinetic roughening refer to systems in which, apart from the noise term, the representative Langevin equation has the shape of a continuity equation, reflecting a conservation law on the mass of aggregating units *at* the surface. Such kind of symmetry may arise dynamically in an effective way, or be explicitly manifest in the physical mechanisms leading to growth. Thus we have:

   (a) EW universality class: There are surfaces whose dynamics can be asymptotically described by an evaporation-condensation effect (combined with fluctuations) of the

[4] E.g., under the assumption of Poisson statistics (*shot noise*).

[5] In that sense, the continuum equation can be thought of as an *effective* description of the physical system at appropriate *coarse-grained* space and time scales.

[6] See e.g. the chapter by M. A. Muñoz in this book.

Gibbs-Thompson type. This is the case, for instance, in CVD experiments with non-negligible vapor pressure. Note that in this case, apart from the external flux, the surface evolves as around an equilibrium state. The EW Langevin equation is thus a linearization of (1):

$$\frac{\partial h}{\partial t} = \nu \nabla^2 h + \eta. \tag{2}$$

For any value of $d_E = d + 1$, the critical exponents characterizing this universality class are $\alpha = (2-d)/2$ and $z = 2$ [$\beta = (2-d)/4$]. These were already familiar from the studies of the equilibrium roughening transition, as mentioned in the Introduction, Eq. (2) being simply model A for the Ginzburg-Landau functional in the Gaussian approximation.

(b) Linear MBE universality class: In the same way as Eq. (2) can be thought of, in the simplest representation, as the equilibrium dynamics of a surface minimizing its area [6, 7], an important class of rough surfaces can be described effectively as minimizing their mean curvature.[7] The Langevin equation representing this universality class reads

$$\frac{\partial h}{\partial t} = -\nu \nabla^4 h + \eta, \tag{3}$$

Again (as reflected in the previous equation being linear) the scaling exponents are exact in this case for any value of d_E, namely, $\alpha = (4-d)/2$ and $z = 4$ [$\beta = (4-d)/8$]. Although the thermodynamic limit of a system like (3) is *not* well defined, this does not prevent real (and finite) systems to display the referred scaling behavior [36].

(c) Non-linear MBE universality class: To some extent in analogy with the relationship between the KPZ and EW equations, much work has been devoted to elucidating the appropriate non-linear extension of (3) leading to a well-defined universality class. It is represented by the Langevin equation

$$\frac{\partial h}{\partial t} = -\nu \nabla^4 h + \lambda \nabla^2 (\nabla h)^2 + \eta, \tag{4}$$

In this case the critical exponents are close to [37] $\alpha = 1$, $z = 3$ [$\beta = 1/3$] in $d_E = 2$, and $\alpha = 2/3$ and $z = 10/3$ [$\beta = 1/5$] in $d_E = 3$.

Although the above universality classes are those which have focused most work in the field of kinetic roughening, the above list is possibly far from complete. Actually, in a certain way there is a *continuum of universality classes*! Some not covered in the previous list arise in physical mechanisms which cannot be ruled out *a priori* when considering applications to real data:

- Noise properties: It is well known theoretically that the introduction of particular features in the noise distribution can modify the scaling exponents, see e.g. [1, 2, 3]. A somewhat trivial one is considering conserved noise (as in model B [5] for conserved dynamics of an order parameter), instead of non-conserved noise, implicit above. More far-reaching modifications are introducing *(i)* power-law correlations in the noise [38], or *(ii)* a noise amplitude which is itself power-law distributed [39]. This type of properties lead to scaling exponents that depend *continuously* on the parameters that characterize the noise properties.

[7] In analogy with the discrete Gaussian model, there is also a roughening transition for these systems, described through the discrete *Laplacian* model [35].

- Non-locality: Another way to obtain scaling exponents depending continuously on model parameters is by considering long range effects on the surface, as represented in the dynamic equation by integral kernels that couple the height field values at all points on the surface [40]. The parameters mentioned characterize in this case the spatial decay (typically as a power law) of such integral kernels. If non-locality appears as a consequence of a diffusing field (e.g. concentration of depositing species in CVD) being coupled to the height field, then frequently scaling exponents are also different from those of the four universality classes summarized above [32, 41, 42].

- Quenched vs time dependent noise: Another issue obscuring the identification of universality classes is the fact that two surfaces may share the same values of the exponents α and z, but still be physically rather different, the noise distribution being quenched in one case and time dependent in the other. Such a situation occurs e.g. when comparing (in $d_E = 2$) an EW type equation with a diffusion coefficient which is a random variable with a quenched columnar distribution [43], with the linear MBE equation.[8] Incidentally, for surfaces growing in a (quenched) disordered medium, like a Hele-Shaw cell with randomly varying gap spacing, scaling exponents can also depend continuously on parameters characterizing the disorder [44].

As can be concluded from the previous paragraph, the values of the critical exponents α and z may not suffice in order to identify the universality class a given system belongs to. Moreover, as will be seen below (Sec. 2.3), the most general form of the dynamic scaling Ansatz known to date does depend on at least an additional independent "roughness" exponent. There may be even accidental or coincidental equalities among exponent sets, as occurs [45] between those characterizing the KPZ equation in $d_E = 1$ —a far from equilibrium system— and the ones obtained for the *equilibrium* roughening of the interface between two immiscible fluids. The existence of a continuum of exponent sets naturally questions the use of the concept of universality classes, in particular from the practical point of view. Another inadequacy of the universality concept arises when interpreting the scaling properties of some theoretical discrete models of MBE growth, such as the WV or DT models, which seem to belong to different universality classes by changing the value of d_E [27]. For instance the WV model for instantaneous surface diffusion is in the EW class for $d_E = 2$, whereas for higher dimensions it leads to unstable growth.[9] The existence of universality has even been questioned on theoretical grounds in the context of the very same KPZ class [46]. Although the numerical instabilities adduced have been overcome and shown to be due to inappropriate schemes [47], the strong coupling properties of the KPZ class still undoubtedly remain to be properly understood [48].

2.2 The rôle of instabilities

An issue that has already arisen in the previous section is the existence of instabilities in the context of models, or experiments, of kinetic roughening. Already since the first systematic studies of kinetic roughening, physical instabilities have been well known to take place in the systems studied. The paradigmatic example in this context is the *Mullins-Sekerka* instability arising in crystal growth from an undercooled melt and in other growth systems (see reviews in e.g. [49]). More

[8] As will be seen in Secs. 2.3 and 3.2, in this example the value of the roughness exponent measured by the height-difference correlation function is different for the first ($\alpha_{\rm loc} = 1/2$) and second ($\alpha_{\rm loc} = 1$) examples mentioned.

[9] Actually, these phenomena point out the increased complexity of non-equilibrium discrete models as compared to equilibrium ones, in the sense that slight modification of the *dynamical* rules lead to dramatic changes in the asymptotic properties.

recently they have been seen to play a key rôle in the dynamics e.g. of crystal growth by atomic or molecular beams [50]. In these systems, the *Ehrlich-Schwoebel* effect hindering the crossing of steps by adatoms leads —in the case of growth onto a high symmetry surface— to development of mounds or, when growth is on a stepped surface, to step meandering possibly also leading to mound formation. However, in spite of their ubiquity, the effect of physical instabilities on the properties of the surface roughness has been overlooked to a large extent. Recently, however, instabilities have been put forward [32] as a possible explanation for the difficulty in observing experimentally the "ideal" universality classes discussed in Sec. 2.1. A paradigmatic example is provided by the (noisy) Kuramoto-Sivashinsky (KS) equation

$$\frac{\partial h}{\partial t} = -|\nu|\nabla^2 h - |B|\nabla^4 h + \frac{\lambda}{2}(\nabla h)^2 + \eta. \tag{5}$$

This equation has been actually *derived* from constitutive equations —not merely based on scaling and/or symmetry arguments— as a description of kinetically roughened surfaces arising in a number of physical contexts. To name a few of them, we can mention directional solidification of dilute binary alloys [51], solidification of a pure substance at large undercoolings including interface kinetics (see [52] and references therein), erosion (sputtering) by ion bombardment (IBS) [53], dynamics of steps on vicinal surfaces under MBE conditions [31], or growth by CVD or ECD [32]. In our context, a distinguishing feature of this continuum model [54] is the *negative*, and therefore linearly unstable, sign of the first term on the right hand side of Eq. (5). Competition between this term and the *stable* biharmonic one leads to the existence of a finite band of linearly unstable Fourier modes in the surface. The Fourier mode with the fastest increasing amplitude leads to the production of a pattern whose appearance in principle breaks scale invariance. The highly non-linear dynamics of the system eventually leads to a disordered and rough morphology whose statistics are provided, at least for $d_E = 2$, by the KPZ universality class. Note, incidentally, that Eqs. (1) and (5) are *indistinguishable* from the point of view of symmetries, hence "derivations" based on the latter typically end up by proposing the KPZ equation as a continuum description even in systems where the KS equation is a better description, due to the existence of *physical* instabilities, see e.g. [55] for the case of erosion by IBS. Note that the crossover from the pattern-formation transient to the asymptotic kinetic roughening can be exceedingly large in the KS system [56]. Hence, it is conceivable that when studying experimentally systems described by the KS equation and whose asymptotic state is effectively unaccessible, fits to some effective (and improper) exponents are attempted that lead to incorrect conclusions on the scaling properties of the system. At any rate, the long crossover can even hinder the actual experimental observation of the asymptotic KPZ scaling in systems described by Eq. (5). Analogous conclusions can be drawn for systems in which physical instabilities are expected, even if the relevant dispersion relation[10] differs from the KS one, e.g. if it is, rather, provided by the Mullins-Sekerka dispersion relation, ubiquitous for systems with diffusional instabilities and fast interface kinetics [32]. An open avenue for research in this connection is undoubtedly the interplay of noise and instabilities and its implications for the interface morphology, and the possible formulation of generic continuum descriptions incorporating these phenomena, see Sec. 2.5.

[10] Defined as the wave-vector dependent rate $\omega(k)$ characterizing, within a linear stability analysis, the exponential growth/decay of the k-th Fourier mode of the height field, $h_k(t) = \text{const.}\, e^{\omega(k)\, t}$.

2.3 Dynamic scaling *Ansätze*

As mentioned in the Introduction, one of the main problems with the large variety of exponents found experimentally is that, in the absence of a detailed study of the system considered,[11] which might not necessarily be available, there is no *a priori* argument allowing to identify a clear physical mechanism responsible for the experimental data. Moreover, many of the experiments seemed to contradict the assumed generality of KPZ scaling, the scaling behavior actually observed being termed *anomalous*. In retrospect, this induced a large advance in the field, at least from the theoretical point of view, in the search for potential origins for such violation of the generic expectation. Thus, for instance quenched disorder was advocated as a source for anomalous scaling, as were various other properties of the interface noise distribution [1, 2, 3]. The problem with these was that, again, for given experiments, no *a priori* arguments were provided supporting those kinds of properties in the appropriate noise distribution.

A potentially more fundamental reason for the spread in the values of the scaling exponents was put forward in [43], generalizing results mainly obtained in the context of discrete models of surface growth by MBE [57], namely, the FV Ansatz is not the most general scaling behavior for a surface displaying kinetic roughening. Thus, it might be the case that, at least in some systems (see an explicit example in Sec. 3.2.1), the values attributed to the scaling exponents were not correct but, rather, originated in trying to fit the data using the wrong scaling Ansatz. This type of scaling has also been termed *anomalous*. At least in many cases studied, it originates in slow dynamics of the surface *slopes*, which do *not* reach a stationary state simultaneously with the surface height [58]. In its most general form to date [59], the dynamic scaling Ansatz is formulated in terms of the power spectral density (PSD) or surface structure factor $S(k,t) = \langle \hat{h}(k,t)\hat{h}(-k,t)\rangle$, with $\hat{h}(k,t)$ being the k-th Fourier mode of the surface height deviation around its spatial average for a given time t. Thus, for many rough interfaces one has

$$S(k,t) = k^{-(2\alpha+d)}\, s(kt^{1/z}), \quad \text{where} \quad s(x) \sim \begin{cases} x^{2(\alpha-\alpha_{\rm s})} & \text{for} \quad x \gg 1 \\ x^{2\alpha+d} & \text{for} \quad x \ll 1 \end{cases} \tag{6}$$

Here, $\alpha_{\rm s}$ is an additional exponent, whose values induce different behaviors. Namely, for $\alpha_{\rm s} < 1$ one may have FV scaling [i.e., that expected naïvely for $S(k,t)$ directly from the FV Ansatz for $W(t)$, and which occurs for the "ideal" classes (1) and (2a,c) of Sec. 2.1] or *intrinsic* anomalous scaling, if $\alpha_{\rm s}$ equals α or not, respectively. On the other hand, if $\alpha_{\rm s} > 1$ anomalous scaling ensues which can be either trivial if $\alpha_{\rm s} = \alpha$, in the sense that it is induced by the super-roughness of the interface (this is the case for the linear MBE equation mentioned in Sec. 2.1), or non-trivial if $\alpha_{\rm s} \neq \alpha$. This latter case corresponds also to faceted surfaces featuring large values of the local slopes [59]. In general, signatures of anomalous scaling are different effective roughness exponents for small and large length-scales —see an explicit example in Sec. 3 below—, and finite-size effects in $S(k,t)$.[12] To date, there is no general argument as to what kind of scaling field controls the value of the exponent $\alpha_{\rm s}$ in those cases in which it is actually independent from α, z and geometrical constants. Note that in these cases the number of exponents characterizing the growth process increases as compared with the traditional assumption reviewed in Sec. 2.1. There has been progress, nevertheless, in relating intrinsic anomalous scaling with Lévy statistics for the local height difference distribution [60]. Moreover, in some examples such as interfaces generated in invasion percolation models, the value of $\alpha_{\rm s}$ can be related with the fractal properties of the underlying cluster. This may provide a physical interpretation for the anomalous scaling properties

[11] Say, of a derivation of the interface equation from constitutive equations, as available e.g. in the examples mentioned in Sec. 2.2.

[12] Inexistent within the standard FV scaling.

of the interfaces, when these are seen to occur. Examples of experimental reports on anomalous scaling (see also Sec. 3 below) include wood fracture [61], polymer film growth by vapor deposition [42], and electrochemical or electroless production of Cu films [62]. However, note these include realizations of all types of anomalous scaling *but* for the faceted surfaces characterized by $1 < \alpha_{\rm s} \neq \alpha$.

2.4 Fractal properties

As mentioned above, kinetically roughened surfaces are but a specific example of statistical *self-affine fractals*. A phenomenon which has not been completely addressed, either from the theoretical, or from the experimental points of view is the full relationship between the geometrical fractal properties of the interface and the form of dynamic scaling Ansatz it follows, and the scaling relations among the critical exponents characterizing both.

2.4.1 Multiscaling

In the case of the simple FV Ansatz, there is a direct connection between the unique roughness exponent characterizing the rough surface and the fractal dimension. However, it becomes less evident for the case of anomalous scaling. As a relevant example, for the discrete DT model [26] of MBE growth, anomalous scaling has been seen to occur accompanied by *multiscaling* properties [63]. These relate to the fact that different moments of the height-difference correlation function $G_q(r,t) = \langle |h(\mathbf{r}_1,t) - h(\mathbf{r}_2,t)|^q \rangle$, where $r = |\mathbf{r}_1 - \mathbf{r}_2|$, scale with moment-dependent exponents,[13] $G_q^{1/q}(r,t) \sim \xi(t)^{\gamma_q}\, r^{\alpha_q} f_q(r/\xi(t))$, with $f_q(x)$ appropriate scaling functions, and the exponents γ_q being related to the local height difference distribution properties. These results for the DT model have been recently generalized for any surface with a local height difference distribution of the Lévy type [60]. However, the general interplay between anomalous scaling and multiscaling properties of surfaces is not known. It would be highly desirable to have available sufficient and/or necessary conditions for the occurrence of one of these phenomena as a function of the other.[14] This would allow for a consistent analysis of experimental data by, say, evaluating different moments of the height difference correlation function in real space, and cross-checking such information with the scaling behavior of the PSD in the form (6).

2.4.2 Other geometries

There are two other important aspects —of at least a methodological nature— which have not been addressed in full in a systematic way, and whose study would undoubtedly clarify the interpretation of many experimental observations, as well as their comparison with theoretical models. Both relate to interface growth phenomena in geometries which differ from the strip or slab geometry.

Thus, as a *first* instance we have growth on circular ($d_E = 2$) or spherical geometries ($d_E = 3$). Although the earliest systematic studies of kinetic roughening actually originate in the dynamics of the active zone in "circular" DLA or Eden clusters, the vast majority of analytical studies have focused on the strip ($d_E = 2$) or "slab" ($d_E = 3$) geometries. Some exceptions are partial studies of the KPZ equation in radial geometry [64]. Although, admittedly, these geometries are infrequent in applications to thin solid film production, the clarification on the correct scaling Ansatz to be performed in circular geometries would undoubtedly complete the theoretical framework. For

[13] In order to compare with other common notations, one has to replace $(\alpha_q, \gamma_q) \to (\chi_q, \alpha_q)$ [60], or $(\alpha_q, \gamma_q) \to (\zeta_q, \alpha_q)$ [63].

[14] For instance, examples are known of models which display intrinsic anomalous scaling, but not multiscaling [43].

instance, in these systems an essential feature is the fact that the system size is *not fixed* to a constant value. One can account for the experimental data by the Ansatz (6), provided the replacement $k \to k/f(t)$ is performed, where $f(t) \sim t^a$ (a being a constant) is a dilation factor taking into account the increase of the system size with growth time [65]. However, differing values of a are needed for different systems, such as e.g. tumor [17] or plant calli growth [65], being unrelated, moreover, with the value of z in the various systems. Note a possibly related non-stationarity phenomenon in the behavior of the local slope for the case of anomalous scaling. It would be interesting to provide an estimation for a based on first principles, as well as deriving its value for the main interface growth equations, say, the EW and KPZ equations in non-conserved growth, and the linear and non-linear MBE equations for conserved growth, see Sec. 2.1.

The *second* issue mentioned is the deduction of scaling properties of fully two-dimensional surfaces ($d_E = 3$) from data for one-dimensional cuts ($d_E = 2$) across them. This is a rather frequent procedure, specially for Transmission Electron Microscopy (TEM) analysis of the growth of multilayer films. In this case very thin cross-sections (i.e. one-dimensional profiles) of the multilayers are analyzed under the framework of dynamic scaling. Thus, values for the scaling exponents are obtained. However, discrepancies arise in the literature when these values are compared to models in $d_E = 2$ [66] or in $d_E = 3$ [67]. Although in the isotropic case the correct relationship between roughness exponents is clear, it becomes less immediate [68] when there exists physical anisotropies on the substrate plane. This issue can be important because it also affects the correct procedure to analyze *anisotropic* growth or erosion processes. Physical realizations of such anisotropies are provided e.g. by vapor deposition of gold films [69] or IBS of highly pyrolytic graphite [70], both under oblique incidence conditions (for attaching or bombarding particles, respectively).

2.5 Universal (continuum) descriptions

As implicit from the previous sections, the physics of surface/interface dynamics is naturally more involved than the mere intermediate or asymptotic scaling laws and, even if one is interested in these (as encoding the kinetic roughening properties of the system), one needs take into account the various physical mechanisms acting on the surface morphology, which actually may have an impact on the behavior of, say, the surface roughness. In practice, this requires resorting to an underlying physical model of the phenomenon at hand in order to derive the pertinent interface equation[15] in a consistent fashion. Technically, such physical models usually have the shape of a free or moving boundary system[16] into which fluctuations are incorporated. This is the case in the examples quoted in Sec. 2.2 [31, 32, 52, 53, 71]. This procedure, moreover, allows one to relate the coefficients appearing in the Langevin equation for the interface —say, ν, B, and λ in (5)— to phenomenological parameters (diffusivities, temperature, mass-transfer coefficients, etc.). Admittedly, the moving boundary problem couples effects at the interface which are strictly kinetic, with others of a different nature, such as e.g. mechanisms for the transport of matter, latent heat, or interfacial free energies. Our point here is that these have to be taken into account if one wishes to have a physical theory of kinetically roughened interfaces which is relevant to experiments. Interestingly, one can still put forward *generic* —rather than *universal*, borrowing terminology from the field of *pattern formation* and non-linear science [54]— descriptions of kinetically roughened surfaces that strongly follow those available for standard[17] pattern forming systems. For instance, the deterministic stabilized KS equation has been shown [72] to be generic

[15] Typically through a weakly non-linear analysis.

[16] See e.g. the chapter by R. González-Cinca *et al.* in this book.

[17] In the sense of *deterministic* partial differential equations.

for systems featuring bifurcations with a vanishing wave number, provided certain symmetries occur. The advantage of this approach is that one can provide continuum models that feature both instabilities leading to pattern formation, together with fluctuation effects that tend to disorder the interface and induce scale invariance properties. The behavior observed will be then a matter of a delicate balance between these opposing effects. Continuing with the same example, within this approach the noisy KS equation is more generic than the KPZ equation, in the sense that it can account for more complex dynamics that applies to *both* asymptotic and pre-asymptotic features. It will be of great interest if, proceeding along these lines, one could provide generic continuum descriptions that feature for instance anomalous scaling, known e.g. in the context of ECD to be a preasymptotic feature [73].

3 Status of the experiments

As mentioned above, regarding the experimental issues we will mainly focus our attention on processes related to the growth or erosion of the surfaces of thin solid films. These processes take place usually under far-from-equilibrium conditions. Several techniques have been employed to characterize the growth morphologies appearing; among them Scanning Tunneling Microscopy (STM), Atomic Force Microscopy (AFM), X-ray reflectivity, Reflection High-Energy Electron Diffraction (RHEED) and Transmission Electron Microscopy (TEM). The suitability of each of these techniques depends on the specific sample to be studied, the length scale to be sampled, the interface roughness, and the character of the study to be made, since not all of them yield values for all the scaling exponents [15]. Besides, for instance X-ray reflectivity measurements can be carried out *in situ*, whereas Scanning Probe Microscopy (SPM) has to be employed *ex situ*. Nevertheless, in later years SPM techniques seem to be predominant for analyzing surfaces of thin solid films, since they allow a wide length window for analysis (from nanometers up to several dozens of microns) and they do not require any special sample preparation (in particular with AFM). They allow to measure the surface morphology for different growth (erosion) times and thus directly obtain e.g. the value of exponent β. Moreover, from the analysis of the topography one can obtain different observables, such as the power spectral density (PSD), the height-height correlation function, the height autocovariance, etc., from which the values of other critical exponents, such as α and z can be determined. However, it has been pointed out that SPM techniques can overestimate the value of α when this is relatively small [74, 75], although consistent values have been also found when the AFM values have been compared to those obtained by different techniques for the same system [76, 77, 78, 79, 80, 81]. In the following sections we consider additional difficulties that can be met in experimental works.

3.1 Scaling ranges

In principle, in order to characterize appropriately the growth process under study,[18] one should determine all independent exponents by separate means. Ideally, not even employing scaling relations of the type $\beta = \alpha/z$, that can fail due e.g. to specific initial conditions employed [82]. In this sense it is very useful to identify the characteristic lateral correlation length with some morphological film surface features (for instance, with the grain structure). Also, a relevant issue concerns the magnitude of the window (either spatial or temporal) over which the value of the exponent is being determined. This is important since the theoretical models usually deal with windows of several decades, whose extent can be very hard to achieve in experimental systems [83]. As a

[18] Without *a priori* assumptions on the universality class.

consequence of this limitation it is very convenient to carry out deposition (erosion) experiments for very long times in order to be able to analyze the truly asymptotic growth (erosion) scaling behavior [22, 84]. Admittedly, the relevant time scales may lie beyond the physical constraints of the experimental setup. Finally, another experimental problem found in many systems [22] is related to the existence of *crossovers* between different growth regimes. This limitation is aggravated by the fact that the crossovers are in practice not sharp but, rather, even more gradual than as seen in many theoretical models. Thus, reliable identification of the extent of the different growth regimes becomes hampered, their temporal or spatial extension being reduced.

3.2 Morphological analysis of experimental data

Any rationalization of experimental data draws unavoidably upon some theoretical model, and can thus be biased by the limitations of the latter. This fact also occurs in our context, where perhaps the best example is the existence of the anomalous scaling. Note that the theoretical understanding of the difference it implies with respect to the simplest FV scaling Ansatz has been taking place during the last ten years, well after formulation of the latter [4]. Specifically, as was explained above, in principle several different types of dynamic scaling can be distinguished: *(i)* the simplest one is the classical Family-Vicseck (FV) scaling, that is associated with overlapping PSD [the function $S(k,t)$ defined in Sec. 2.3] or height-difference correlation [the function $G_2(r,t)$ introduced in Sec. 2.4.1] functions for different deposition times,[19] and with only two independent exponents, $\alpha_{\rm s} = \alpha$ and z, with $\alpha_{\rm s}$ the spectral exponent defined in Eq. (6). *(ii)* The merely "super rough" case in which, while the PSD graphs overlap for different times, the height-difference graphs do not, but are shifted upwards as deposition (erosion) time increases. In this case one has to distinguish between local ($\alpha_{\rm loc} \equiv 1$) and global ($\alpha > 1$) roughness exponents,[20] since they appear different as measured from $G_2(r,t)$ or from $S(k,t)$, which are sensitive to, respectively, local or global fluctuations. In this case $\alpha_{\rm s} = \alpha > 1$, and the value of the local roughness exponent is fixed to 1 by geometrical constraints [3], hence it is not really an independent exponent. *(iii)* The "intrinsic" anomalous case in which neither the PSD nor the height-difference correlations overlap for different times but are, rather, shifted as deposition (erosion) time increases [for some faceted surfaces $\alpha_{\rm s} > 1$, which induces time shift only in the PSD and not in the $G_2(r,t)$ function]. In this case, again local and global fluctuations are accounted for by different roughness exponents, there existing now *three* really independent exponents.

It is clear from the above that proper analysis of the data requires previous inspection of the two functions, $G_2(r,t)$ and $S(k,t)$, in order to anticipate the scaling behavior at least in terms of the existence of the mentioned non-overlapping[21] features. Once it is identified, the adequate procedure of analysis of the data can be found in the literature. In general, the most elegant method consists in the collapse of the height-difference correlation or/and the PSD experimental curves, according to the theoretical scaling relations [80]. Another possibility requires performing roughness analysis at short and long length scales in order to measure the local and global roughness exponents [62, 85, 86].

Different situations can be found when one examines the existing literature regarding this subject. First, there are many studies in which the authors did not realize that anomalous scaling was present in their systems, or they did but a consistent analysis was not performed because, rather,

[19] For length-scales smaller than the lateral correlation length.

[20] The local roughness exponent $\alpha_{\rm loc}$ coincides with exponent α_2 as defined in Sec. 2.4.1. Equivalently, it characterizes the spatial scaling of the local width $w^2(\ell,t) = (1/\ell^d)\langle\sum(h(\mathbf{r},t) - \bar{h})^2\rangle$ where spatial averages are restricted to boxes of lateral size ℓ. Thus, for $\ell \ll t^{1/z}$ one has $w(\ell,t) \sim \ell^{\alpha_{\rm loc}}$.

[21] As mentioned in Sec. 2.3, for a finite system the non-overlapping features of the PSD amount at saturation to finite size effects, inexistent for FV scaling.

FV scaling was assumed [18, 42, 87, 88, 89]. An example of this can be found in an earlier paper by one of the present authors [90], see details in Sec. 3.2.1 next. A second set of experiments exists in which the authors explicitly identified the anomalous behavior and analyzed it accordingly, mostly through the behavior of the interface width for different length scales [36, 62, 85]. It should be noted that most of these studies in three-dimensional systems are relatively recent, and are mostly related to electrodeposition systems. In general, we can say that the description of anomalous scaling is an example of a situation in which the theoretical developments are ahead from the experimental analysis. There is still, however, an important issue than remains unsettled, related with the physical origin of this type of behavior. While the theoretical papers on the subject address more mathematical aspects, one can already obtain some light on this issue from the (scarce) experimental works on the matter. As we have mentioned, in the context of thin solid film growth, most of these are focused mainly on electrodeposited films, suggesting that non local —in this case bulk diffusion— effects are likely responsible for such behavior. This scenario would be consistent with different local growth rates, which is qualitatively consistent with other explanations suggested for two-dimensional systems of a different nature [44]. It should be stressed, however, that the physical origin of anomalous scaling is an issue that still has to be explicitly addressed. Another important (possibly related) aspect regarding consistency of the analysis of the experimental data, especially when instabilities are found, is that one of multiscaling. It is important to check explicitly for such type of behavior in any chosen system for, in the case it does exist, it does not make sense to refer to a single set of scaling exponents but, rather, one should consider an (in principle, infinite) set of exponents, one for each moment of the height difference distribution.

3.2.1 An explicit example

In order to provide an example that illustrates the various difficulties met in this type of works, we devote this section to reanalyzing an experiment by one of the present authors [90] in the light of the more recent developments on the types of dynamic scaling Ansätze. We will see that even doing so, a complete understanding of the corresponding growth process is still open. The experimental system is growth by CVD of copper films on Si(100) substrates. In Fig. 1 we display three STM images of films with thickness (proportional to time, given the constant growth rate imposed) of 44 nm (a), 334 nm (b) and 2220 nm (c), respectively. The film roughening and coarsening are evident in the figure. As can be observed in Figs. 2 (corresponding to Fig. 3(a) in Ref. [90]) and 3 for the local surface roughness $w(\ell)$ and for the height-difference correlation function $G_2(r,t)$, respectively, the surface morphology displays anomalous scaling in the sense that curves for different times do *not* overlap for small values of r. Specifically Fig. 2 contains data for the thinnest and thickest copper films. However, in Ref. [90] the scaling analysis was made according to the FV Ansatz, with results for the critical exponents $\alpha_{\rm loc} = 0.81 \pm 0.05$, $\beta = 0.62 \pm 0.09$, and $1/z = 0.74$, the last value having a relatively large error bar since it was obtained by averaging the grain distribution functions. In order to improve the scaling analysis, we present now in Fig. 3 the $G_2(r,t)$ curves for various film thicknesses t. The anomalous scaling displayed by $G_2(r,t)$ can be seen even more clearly in the systematic upward shift of the different curves with increasing thickness. From the slope of the curves for small r values (such as was the procedure employed in [90]) we already obtain $\alpha_{\rm loc} = 0.86$. In order to characterize quantitatively the anomalous scaling, we have used the data from Fig. 3(a) and collapsed the curves [see Fig. 3(b)] by dividing the horizontal axis by $t^{1/z}$, and by dividing the vertical axis by the product $r^{\alpha_{\rm loc}}\, t^{\beta_*}$, with $\beta_* = (\alpha - \alpha_{\rm loc})/z$ [43]. The best collapse of the data is obtained for $1/z = 0.63$ and $\beta_* = 0.11$. These results imply that the system is characterized by $\alpha = 1.03$, $\alpha_{\rm loc} = 0.86$,

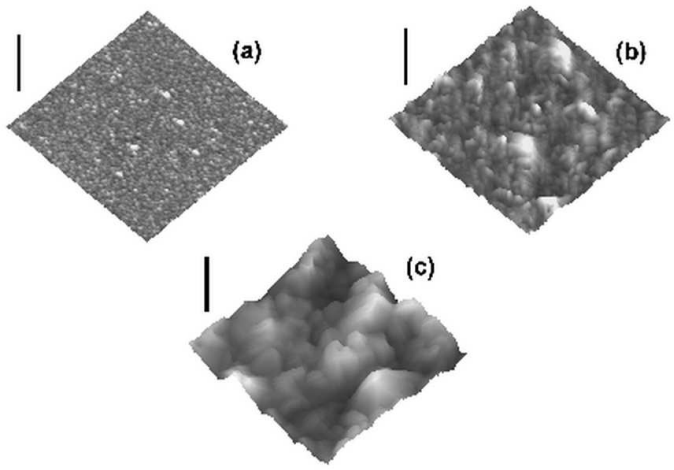

Figure 1: $4 \times 4\ \mu\mathrm{m}^2$ STM images of Cu films grown by low-pressure CVD onto Si(100) substrates, for values of the thickness: 44 nm (a); 334 nm (b), and 2220 nm (c). The vertical bars represent 1000 nm.

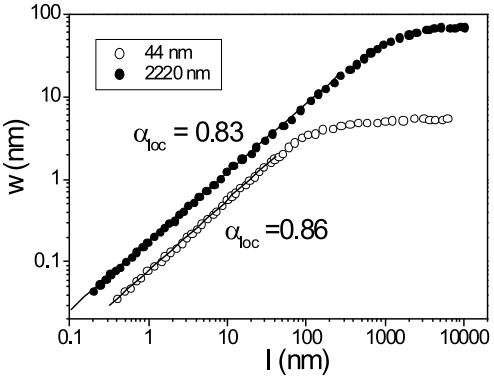

Figure 2: Logarithmic plot of the local roughness, $w(\ell)$, vs window size, ℓ, for the 44 nm thick film (lower curve) and for the 2220 nm thick film (top curve). The values indicated for α_{loc} provide the slopes of the corresponding solid lines. This figure corresponds to Fig. 3b of Ref. [90].

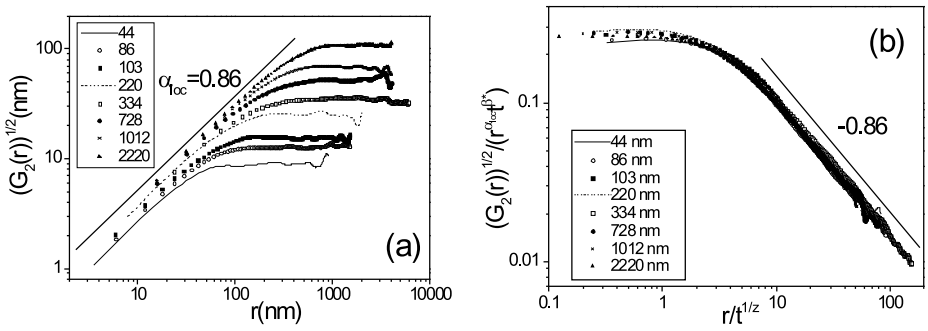

Figure 3: (a) Logarithmic plots of the square root of $G_2(r,t)$ vs r for different thicknesses t (corresponding thickness values and symbols are indicated in the figure). The value indicated for $\alpha_{\rm loc}$ provides the slope of the thick solid line. (b) Collapse of the data provided in panel (a). Vertical coordinates are rescaled by a factor $r^{\alpha_{\rm loc}}\, t^{\beta *}$, using $\alpha_{\rm loc} = 0.86$ and $\beta_* = 0.11$, while horizontal coordinates are divided by a factor $t^{1/z}$ with $z = 1.59$. For the sake of reference, a thick solid line has been drawn whose slope equals -0.86.

$z = 1.59$, $\beta = 0.65$, and $\beta* = 0.11$. Note that the collapse is consistent since the behavior of the scaling function for large arguments should decay as $(kt^{1/z})^{-\alpha_{\rm loc}}$, as indeed occurs in Fig. 3(b).

Although there are some discrepancies between the values of the scaling exponents reported in [90] and those obtained here, the difference lies within the error bars. Yet, conceptually there is a large difference, since the system seems to display intrinsic anomalous scaling, and is thus characterized by three, rather than two, independent exponents. Any theoretical model of the present growth system has to be able to account for this fact. Moreover, from the physical point of view a result that remains to be explained is the large value of $\beta > 0.5$. This *unstable* growth could be due to non local effects such as shadowing, that are likely to occur in CVD, or alternatively to the existence of step-edge barrier effects as recently shown for copper films deposited by sputtering [91]. In summary, the growth system studied in this section certainly exemplifies several (possibly generic) features of experimental studies in the field. Namely, it displays unstable growth reflected in a large growth exponent and in the existence of intrinsic anomalous scaling, that remained unnoted when studying the properties of the roughness under the FV Ansatz. The more complex scaling uncovered might provide clues as to the understaning of the relevant physical mechanisms acting in the system, and remains to be explained theoretically.

3.3 Identification of growth modes

As described above, the experimental works are mainly based on measurements of two or three of the scaling exponents and their further comparison with those predicted by the different existing growth models. In this analysis, one should consider the physical properties of the growth system, such as growth temperature, vapor pressure of the depositing material, kinetic or diffusive control of growth, sticking coefficient, etc. in order to be able to neglect those processes which are irrelevant and should not contribute, say, to the continuum equation for the interface, if such a description is available. For instance, if the growth temperature is relatively high the surface diffusion should be taken into account, or if the vapor pressure of the depositing material is very low the Gibbs-Thompson effect (and thus terms accounting for surface tension) can be neglected. In this sense, the very morphological data obtained for the system can also contribute to including or discarding terms in the equation of motion. This is the case for the skewness of the growing

interface, that measures the symmetry of the interface. A negligible value indicates that the surface is quite symmetric with respect to the average height. In contrast, noticeable values of the skewness, whether positive or negative, indicate that the surface presents large protuberances or depressions (namely, an asymmetric morphology), respectively. Under such circumstances, the continuum equation should include terms with odd derivatives of the height field [92, 93]. Moreover, the analysis of the behavior of the surface slope can also contribute to identifying the growth mode; particularly in the case in which step-edge barriers are present in the growth leading e.g. to mounded morphologies [94]. These arguments may be successfully combined with others of the types described in Sec. 2.5 in order to produce continuum descriptions for various thin solid film growth processes. Thus, many systems have been described and understood adequately under the framework of the dynamic scaling theory. Among them we can mention thin solid films grown by physical evaporation, sputtering, sedimentation, electrodeposition and chemical vapor deposition [22, 24, 36, 62, 95, 96]. Among the cases where a clear identification of the growth mode was *not* possible we can distinguish roughly two classes. *First*, those cases in which some exponents are very close to those of an existing model but other fail to agree [92, 97, 98, 99, 100] or where the adscription to a given model is made without determining, at least, two of the exponents [101]. In this group we can find some cases in which the growth mode seems to be close to KPZ, but it cannot be demonstrated. *Second*, systems for which the exponents simply are not explained by any existing model or class [102, 103, 104]. In many of these cases, the exponent that fails to agree is the growth exponent β, which frequently results to be larger than the theoretically expected one. Usually, although most models of the type considered in Sec. 2.1 predict[22] a value of β smaller than 0.3, larger values are reported for many systems [79]. Even more, in many cases $\beta > 0.5$ is found, see Sec. 3.2.1. Given that $\beta = 0.5$ is the value predicted by the random deposition model, which does not include any sort of relaxation mechanism, this situation is referred to as unstable or rapid roughening in the literature [2, 79]. In fact, "rapid roughening has been reported for a number of systems [105, 106, 107, 108, 109] but not a general mechanism has been identified", as pointed out recently by Dürr *et al.* [79]. Usually, the authors of the corresponding works usually try to develop more or less simple models that consider some physical parameter relevant in their system to try to understand and interpret their results. Thus, phenomena like surface heterogeneity [79], "cluster coalescence" [92], mass transport diffusion effects [99], sticking effects [103, 104], grain growth [110], step-edge barrier effects [111], and shadowing effects [112], among others, have been invoked and sometimes modeled to explain the observed behaviors.

3.3.1 Polycrystallinity, step-edges and other issues

In relation to some of the above effects, an important open issue has to do with the polycrystalline character of many of the films studied. In fact, many of the growth systems with wider impact on technological applications lead to polycrystalline films. Already in one of the earlier experimental reports [98], dated 1994, on the growth scaling behavior of gold films, the authors noted that "one significant discrepancy between the experimental conditions and all of the deposition models is that grain boundaries are not included in the modeling". Generally, these models do not include any mechanism to account for the formation and development of the polycrystalline grains. These grains are expected for the majority of vapor-deposited and sputtered-deposited films. Thus, it would be very important for non equilibrium growth models to consider also growth on the surfaces of the crystallites, which are misoriented with respect to one another [98]. This same idea was also considered by Kardar two years later in his short review on dynamic scaling phenomena in growth processes [19]. In that work Kardar insisted that "variations in crystallinity had so far

[22] For the case $d_E = 3$ relevant to thin solid film production.

been left out of the theoretical picture". This statement is still valid. In his work Kardar suggested the suitability to include in the models an additional order parameter $M(\mathbf{r}, t)$ describing the local degree of crystallinity.[23] Moreover, this generalized approach should account for experimental instances which have been nevertheless adequately described according to the existing models for polycrystalline systems [36]. Thus, further research addressing different experimental issues, such as the influence of the growth substrate and other growth parameters, could contribute to obtaining a deeper knowledge of the influence of polycrystallinity on film growth dynamics. Another related issue is the investigation of the possible rôle of step-edge barriers on film growth for polycrystalline materials. The effect of these barriers has been invoked for describing the growth for different metallic and semiconductor systems [2, 50]. However, its rôle in polycrystalline systems is more debated. It is assumed in a polycrystal that step-edge barriers will be improbable if growth starts out with randomly orientated grains and grain sizes much smaller than film thickness [36]. However, recently step-edge barrier effects have been proved to be a key ingredient on the growth of one micron thick polycrystalline copper films by sputtering, when the grains have a size close to 200 nm [91]. Despite these efforts, the above issues exemplify a situation in which theoretical developments are well behind experimental findings, there being in particular a need of further theoretical studies that considering unstable interface growth. From the experimental point of view, more efforts should be addressed to studying the dependence of the unstable growth behavior on growth parameters. Recent examples of such type of studies can be found, for instance, in the report on dependence of anomalous scaling on the current density for copper electrodeposition [62], or that on the dependence of the growth instability on the sticking coefficient in the case of growth of silica films by CVD from silane precursor [22].

Finally, it would be important to check systematically for the presence of multiscaling, as well as for non-trivial noise correlations in unstable experimental growth systems. This would allow for a better insight into the physical mechanisms behind growth instabilities and, thus, would improve theoretical modeling of such type of systems. Regarding comparison with experimental data various improvements would be required in the theoretical descriptions. Thus, most theoretical models work within the small-slope approximation, that is assuming morphologies in which $|\nabla h|$ is small. Obviously, this is not the general case for unstable features. Also, theoretical analysis of growth instabilities requires simulations to be carried out on systems with considerably larger sizes, in order to probe the asymptotic properties. This fact is aggravated by the fact that most of the thin solid film or surface erosion systems are in $d_E = 3$ (i.e. onto fully bidimensional substrates) [41]. Thus, one can find many publications where the experimental system studied is effectively three-dimensional, but its scaling behavior is contrasted to that obtained from two-dimensional models. As the values of the scaling exponents depend on dimensionality of the system, this situation could limit full understanding of the system behavior.

4 Conclusions and outlook

The situation described in the previous pages clearly indicates a mismatch between theoretical predictions and experimental findings in the area of thin film surface growth. Also, it is evident that the different experimental results as they stand can be neither adscribed to nor understood under a reduced group of universality classes. However, most of the reported experimental results indeed show clearly kinetic roughening properties. Even more, several of them have been adequately adscribed to some of the former universality classes. Thus, although this situation has generated some confusion in the scientific community we believe that further research in this

[23] See analogous theoretical work for growth of a *binary* film in [113].

field is still worthy. Our aim in this chapter has been to describe this situation and suggest future lines of research. On the theoretical side, we expect that the search for *generic* descriptions that capture both, the pattern-forming mechanisms and the relevance of strong fluctuations, will provide a more accurate theoretical framework for the field. Progress along these lines will undoubtedly come through addressing specific growth systems, and after a suitable generalization process. Naturally, benefit can be gained from studies of a more general nature such as, e.g. on the origin of anomalous scaling and of multiscaling in fluctuating interfaces. Note that many of the experimental systems whose behavior does not lie within any of the former universality classes do show kinetic roughening with the features of unstable or rapid roughening growth, and/or with anomalous scaling. Thus, we have tried to bring the attention of the reader, either experimentalist or theoretician, to some topics that are not a particular focus in the literature (anomalous scaling, instabilities, polycrystallinity, etc.), with the aim that in future their consideration and study may contribute to reaching a deeper knowledge of the phenomenon of kinetic roughening. In this sense, more systematic experimental studies addressing the influence of the different growth parameters on the kinetic roughening properties would be welcome.

Acknowledgments

The authors wish to acknowledge J. M. Albella, T. Ala-Nissila, J. Asikainen, M. Auger, A.-L. Barabási, M. Castro, R. Gago, I. Koponen, K. B. Lauritsen, J. M. López, H. A. Makse, E. Moro, F. Ojeda, M. A. Rodríguez, M. Rusanen, R. Salvarezza, A. Sánchez, and O. Sánchez for their assistance and collaboration during the last years in many issues related to this work. They also acknowledge partial support by MCYT (Spain) grants Nos. BFM2000-0006 (R.C.) and MAT2000-0375-C02-02 (L.V.).

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