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# Roughening of growing surfaces: Kinetic models and continuum theories

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#### Abstract

The use of scaling concepts in understanding growth by molecular beam epitaxy (MBE) is increasingly important these days. Here we present a critical discussion on the advantages and disadvantages of kinetic theories and continuum models, two main methods frequently used to study the roughening and scaling of surfaces grown by MBE. Finally, some open problems faced by these approaches are also discussed.

## 1. Introduction

Fueled by MBE's potential to produce novel electronic materials, one can observe an increasing activity for understanding and modeling the basic processes taking place at the surface of crystals. Joining forces with various disciplines of physics and chemistry, this lead to the introduction of new techniques and concepts for understanding growth [1]. In particular, approaches based on modern ideas of statistical mechanics resulted in the development of new ideas from which benefitted not only the MBE community, but also the field of irreversible and far from equilibrium statistical mechanics [2-5]. The motivation of these approaches lies in the observation that many processes taking place at the surface during growth are inherently stochastic, and a successful theoretical approach has to incorporate this randomness.

A prominent question regards the roughening of

surfaces grown by atom deposition. One can take a sarcastic approach and affirm that all surfaces in Nature are rough, and that by itself is enough motivation for studying roughening. While this is certainly correct, for completeness one needs to add the following sentence: The roughness depends on the length scale on which one views the system. This has a number of theoretical and practical consequences. For understanding the motivation behind the continuum theories and kinetic models, one has to have a clear picture of what roughening and roughness means. For this in Section 2 we discuss certain issues that one needs to understand in order to grasp the motivation of the following sections. Section 3 discusses briefly the main ideas behind kinetic modeling, focusing on their advantages and disadvantages. In Section 4 the continuum theories are discussed briefly, with their advantages and disadvantages. Recent advances in ion sputtering, reviewed in some detail, support the hope that continuum theories can play an essential role in understanding the experimentally observed morphologies Finally, in Section 5 we discuss some open problems.

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#### 2. Roughening and scaling

We become accustomed to the shape of the interfaces we encounter, so it can be surprising that their morphologies can appear to be quite different depending on the scale with which we observe them. For example, an astronaut in space sees Earth as a smooth ball. However, Earth appears to be anything but smooth when climbing a mountain, as we encounter a seemingly endless hierarchy of ups and downs along our way.

A second example, closer to our focus of study, is the surface of a crystal. There we can also encounter the same scale dependence as for the Earth: While a crystal may seem and feel completely smooth when one observes it unaided with any magnifying tool, it may look rather rough under the microscope. Similarly, a semiconductor surface may seem rather smooth, but the STM reveals its rugged surface, that is unacceptable for many electronic devices.

We can already draw one conclusion: Surfaces can be smooth, such as the Himalayas viewed from space, but the same surface can also be rough, such as the same mountains viewed from earth. In general the *morphology depends on the length scale of observation*!

The *interface width*, which characterizes the *roughness* of the interface, is defined by the rms fluctuation in the height h(x, y),

$$w(L) \equiv \sqrt{\frac{1}{L^2} \sum_{x, y=1, L} \left[ h(x, y) - \bar{h} \right]^2}, \qquad (1)$$

where L is the linear size of the sample, and the *mean height* of the surface,  $\overline{h}$ , is defined by

$$\vec{h} = \frac{1}{L^2} \sum_{x,y=1,L} h(x, y).$$
(2)

Instead of measuring the roughness of a surface over the whole sample size  $L \times L$ , we can choose a window of size  $l \times l$ , and measure w(l). As we mentioned above, a general property of many rough surfaces is that the roughness depends on the length scale of observation. This can be quantified by plotting w(l) as a function of l. A typical plot is shown in Fig. 1. There are two characteristic regimes one can distinguish.

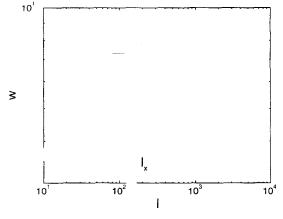


Fig. 1. Typical scaling behavior of the local width, w(l), as a function of the length scale of observation, l.

(i) For length scales smaller than  $l_{\times}$ , the local width increases as

$$w(l) = Al^{\alpha}, \tag{3}$$

where  $\alpha$  is the *roughness exponent* and A is a proportionality constant. If we are interested in surface phenomena that take place at length scales shorter than  $l_{\times}$ , then we can not neglect the roughness of the surface. In this regime the roughness is not simply a number, but it depends on the length scale available to the method probing the surface.

(ii) For  $l \gg l_{\times}$ , w(l) is independent of *l*. For most processes that take place at length scales larger than  $l_{\times}$ , the surface is *smooth*, i.e. we can neglect the roughness. In this regime we can characterize the surface roughness with a single number, namely the saturation value  $w_{sat}(l)$ .

In general reporting a *number* for characterizing the surface roughness, as it is frequently done, is a misleading and unsatisfactory procedure. The concept of roughness, for many application, has to be replaced with the length scale dependent roughness, requiring the determination of the full w(l) curve.

Regarding the dynamics of the roughening process, at early times the total width increases as  $w(L, t) \sim t^{\beta}$ , where  $\beta$  is the growth exponent.

Next we need to understand what determines the w(l) curve. Can we develop models and theories that can provide/predict A,  $\alpha$ ,  $\beta$ ,  $l_{\times}$  and  $w_{sat}$ ?

Studying scaling relations, such as Eq. (3), allows us to define *universality* classes. The universality class concept is a product of modern statistical mechanics, and codifies the fact that there are but a few essential factors that determine the exponents characterizing the scaling behavior. Thus different systems, which at first sight may appear to have no connection between them, behave in a remarkably similar fashion.

The values of the exponents  $\alpha$  and  $\beta$  are independent of many 'details' of the system. They are universal, i.e. they do not depend on the details of the crystal lattice or on the implementation of the model, as long as the mechanism generating the roughening does not change. In contrast, other quantities, such as A,  $l_{\times}$ , or  $w_{sat}$ , are non-universal, i.e. they depend on almost every detail of the system. It is the goal of the next two sections to show to which degree these numbers can be calculated using various approaches, and how reliable these predictions are.

# 3. Kinetic (atomistic) models

The elementary processes taking place at the crystal surfaces suggest that numerical simulations on discrete models might be helpful in understanding the collective behavior of the atoms during growth. In recent years a number of models have been proposed to describe the roughening of the crystals.

The most widely used models can be classified into three main categories.

(i) The simplest set of models is motivated by the desire to understand the scaling behavior and the corresponding universality classes. These are intrinsically nonequilibrium models, and encompass random deposition, local relaxation and sticking rules [6-15]. They capture the essential properties of the roughening process, but do not include the effects of thermal activation of the atoms on the surface. The deposited atoms usually take a few steps on the surface, after which they stick irreversibly. For this reason we call them irreversible growth models. Due to the simplicity of the relaxation rules, these models are the most efficient from numerical point of view, allowing large scale simulations with long running times, and thus the possibility of reaching the asymptotic scaling behavior characteristic of the underlying relaxation mechanism.

(ii) A second, more realistic, approach regarding the resemblance to the typical processes taking place during MBE is to study an activated type of model for which all surface atoms are mobile, with a hopping probability given by the Arrhenius law [7,16– 20]. These are intrinsically finite-temperature models, with temperature as a tunable parameter. Due to the time-consuming activation process, these models are limited to small system sizes and short simulation times. In addition it is common to find strong finite size effects, again hampering efforts to determine the scaling exponents. However, combined with the experience gained from the study of irreversible models, models with reversible sticking and diffusion are extremely useful in tracing the effect of activated diffusion on the scaling behavior.

(iii) Third, there are a number of phenomenological models, which do not start from the elementary processes observed during MBE, but rather use activated processes driven by a Hamiltonian [21,22]. Despite rather little resemblance to MBE, these models can capture some essential properties of the scaling behavior generated by surface diffusion.

Models (i) and (ii) are used to model a wide variety of processes observed during atom deposition, ranging from surface roughening [1] to submonolayer epitaxy [1,23–25]. We shall not discuss molecular dynamics simulations, which consider the interatom potentials acting between different atoms. Molecular dynamics is extremely useful in understanding the local effects taking place on the surface of the crystals, but is limited by very small sizes and short simulation times.

### 3.1. Advantages and disadvantages of kinetic models

One of the main advantages of the kinetic models is their *conceptual* and *technical simplicity*, coming from the simplicity of the elementary processes taking place on the atomic surface. Most of these models include simple processes as atom deposition, surface (activated) diffusion, desorption. Thus the mechanisms acting in these models are easy to understand, and also easy to implement on a computer. As a result, the effect of various processes can be separated, providing an insight into the role played by each of them in determining the morphology of the surface.

Interestingly, the simplicity of the kinetic models can be viewed as a disadvantage as well. In many experimental situations the particularities of the system play a leading role in shaping the surface morphology at the interesting time and length scales. Thus generic models, such as most kinetic models are, have a hard time capturing this behavior. Of course, many system specific properties can be added to these models at the expense of the computation times, limiting the time span and the size of the system.

Another advantage of these models is that the time and length scales one can reach with them is fairly large. In most experimental situations one needs to deal with a large number of atoms, and the growing time can also be long. Currently only kinetic models are able to get close to the experimental probes regarding these two, sometimes rather critical, parameters.

#### 4. Continuum theories

Kinetic models are well suited for *measuring* numerically the scaling exponents. However, this does not necessary tell us much about the mechanism of the growth. Thus we need guidance on what exponents we should expect for the various models. This guidance is even more important when the observed scaling behavior is hampered by various crossover effects, that question the accuracy of the measured values.

Continuum theories represent a further step of abstractization compared to the kinetic models, since many of the 'hands on' aspects of the kinetic models are absent. However, they are rather successful in addressing questions that range from the collective behavior of atoms to the universality of the roughening processes. There are only a few universal numbers, namely the scaling exponents, such as  $\alpha$  or  $\beta$ . These numbers are independent of the particular system under study, and depend only on the elementary processes taking place on the surface. Only in the context of continuum theories can we understand the full strength of the universality concept, and identify the relevance or irrelevance of various physical processes in shaping the surface morphology [1].

The full strength of continuum theories comes from the prediction of the *asymptotic*, or *hydrodynamic* behavior of the growth process, which means that the predictions are valid in the long time limit, and for the large scale limit regarding the morphology of the surface. These limits are often beyond the experimentally or practically interesting time and length scales. However, by providing rather reliable information about the asymptotic limits, they allow us to separate the asymptotic from the transient behavior observed in the growth models and experiments, and thus lead to a much complete understanding of roughening.

Although the prediction of the continuum theories are rarely applicable directly to the experimental situations, there are cases when continuum models can help understand directly the experimental behavior, without being aided by the intermedial role of the kinetic models. To show this, next we present an example in some detail, regarding the roughening and pattern formation observed in ion sputtering [26,27], a common experimental procedure, that almost every MBE experimentalist uses in its laboratory.

#### 4.1. Example: ion sputtering

Much of the attention has focused on the kinetics of interfaces generated in growth processes. However, for a class of technologically important phenomena, such as sputter etching, the surface morphology evolves as a result of erosion processes [28]. Motivated by the advances in understanding growth, recently a number of experimental studies have focused on the scaling properties of surfaces eroded by ion bombardment [29-31]. For graphite bombarded with 5 keV Ar ions, Eklund et al. [29] reported  $\alpha \approx 0.2$ -0.4, and  $z \equiv \alpha/\beta \approx 1.6$ -1.8, values consistent with the predictions of the Kardar-Parisi-Zhang (KPZ) equation in 2 + 1 dimensions [32-37]. Krim et al. [30] observed a self-affine surface generated by 5 keV Ar bombardment of an Fe sample, with a larger exponent,  $\alpha = 0.52$ . On the other hand, there exists ample evidence about the development of a periodic ripple structure in sputter etched surfaces (see e.g. [38]). Chason et al. [31] have recently studied the dynamics of such eroded surfaces for both SiO<sub>2</sub> and Ge bombarded with Xe ions at 1 keV, and found that it differs from the dynamics expected for the self-affine morphologies observed in [29] and [30].

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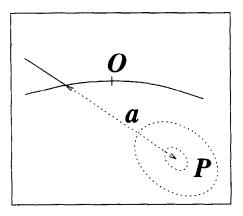


Fig. 2. Following a straight trajectory (solid line) the ion penetrates an average distance a inside the solid (dotted line) after which it completely spreads out its kinetic energy. The dotted curves are equal energy contours. Energy released at point P contributes to erosion at O. (After [26].)

Ion-sputtering is in general determined by atomic processes taking place along a finite penetration depth inside the bombarded material. The incoming ions penetrate the surface and transfer their kinetic energy to the atoms of the substrate by inducing cascades of collisions among the substrate atoms, or through other processes such as electronic excitations. Whereas most of the sputtered atoms are located at the surface, the scattering events that might lead to sputtering take place within a certain layer of average depth  $\alpha$ .

A convenient picture of the ion bombardment process is sketched in Fig. 2. According to it the ions penetrate a distance  $\alpha$  inside the solid before they completely spread out their kinetic energy with some assumed spatial distribution. An ion releasing its energy at point P in the solid contributes an amount of energy to the surface point O, that may induce the atoms in O to break their bonds and leave the surface. Following [39,40], we consider that the average energy deposited at point O due to the ion arriving at P follows the Gaussian distribution. However, the sample is subject to a uniform flux J of bombarding ions. A large number of ions penetrate the solid at different points simultaneously and the velocity of erosion at O depends on the total power  $\varepsilon_0$  contributed by all the ions deposited within the range of the Gaussian distribution. If we ignore shadowing effects among neighboring points, as well as further redeposition of the eroded material, we can calculate the normal velocity of erosion, v, at O. From the expression of v we can obtain the equation of motion for the profile h(x, y, t). The time evolution of h is given by

$$\frac{\partial h}{\partial t} = -v_0 + \gamma \frac{\partial h}{\partial x} + v_x \frac{\partial^2 h}{\partial x^2} + v_y \frac{\partial^2 h}{\partial y^2} + \frac{\lambda_x}{2} \left(\frac{\partial h}{\partial x}\right)^2 + \frac{\lambda_y}{2} \left(\frac{\partial h}{\partial y}\right)^2 - K \nabla^2 (\nabla^2 h) + \eta, \qquad (4)$$

where two additional relevant physical processes were taken into account. First, the bombarding ions reach the surface at random positions and times. We account for the stochastic arrival of ions by adding a Gaussian white noise  $\eta(x, y, t)$  with zero mean and variance proportional to the flux J. Second, at finite temperature atoms diffuse on the surface. To include this surface self-diffusion we allow for a term  $-K\nabla^2(\nabla^2 h)$ , where K is a temperature dependent positive coefficient.

We can also compute the expressions for the coefficients appearing in Eq. (4) in terms of the physical parameters characterizing the sputtering process.

The scaling behavior of the surface described by Eq. (4) depends on the relative signs of  $\nu_x$ ,  $\nu_y$ ,  $\lambda_x$ and  $\lambda_{y}$ . The variations of these coefficients as functions of  $\alpha_{\sigma}$  and  $\theta$  lead to the phase diagram shown in Fig. 3.

To summarize, at short length scales the morphology consists of a periodic structure oriented along the direction determined by the largest in absolute value of the negative surface tension coefficients [31]. Modifying the values of  $\alpha_{\alpha}$  or  $\theta$  changes the orientation of the ripples [38,40]. At large length scales we expect two different scaling regimes. One is characterized by the KPZ exponents, which might be observed in region I in Fig. 3. Indeed, the values of the exponents reported by Eklund et al. [29] are consistent within the experimental errors with the KPZ exponents in 2 + 1 dimensions. The other regions (II and III) are characterized by logarithmic scaling ( $\alpha = 0$ ), which has not been observed experimentally so far. Moreover, by tuning the values of  $\theta$ and/or  $\alpha_{\alpha}$  one may induce transitions among the different scaling behaviors. For example, fixing  $\alpha_{\alpha}$ 

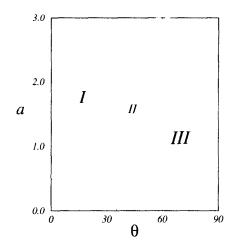


Fig. 3. Phase diagram for the isotropic case  $\sigma = \mu = 1$ . Region I:  $\nu_x < 0$ ,  $\nu_y < 0$ ,  $\lambda_x < 0$ ,  $\lambda_y < 0$ ; Region II:  $\nu_x < 0$ ,  $\nu_y < 0$ ,  $\lambda_x > 0$ ,  $\lambda_y < 0$ ; Region III:  $\nu_x > 0$ ,  $\nu_y < 0$ ,  $\lambda_x > 0$ ,  $\lambda_y < 0$ . Here  $\alpha$  is measured in arbitrary units and  $\theta$  is measured in degrees. (After [26].)

and increasing the value of  $\theta$  would lead from KPZ scaling (region I) to logarithmic scaling (II, III) for large enough angles.

# 4.2. Advantages and disadvantages of continuum theories

One of the clear advantage of continuum theories is that they provide *analytic* predictions regarding the scaling properties of growth models. These predictions help in (i) understanding the nature of the basic processes that lead to certain universality classes; (ii) understanding the mechanism by which these universality classes are generated; and (iii) guide us in interpreting the properties of kinetic models and experiments.

The robustness of the continuum theories against additional (irrelevant) effects can be quoted as both advantage and disadvantage. We can add a number of terms that affect the short scale morphology of the surface but do not influence the scaling exponents. This is closely related to the robustness of the kinetic models against additional microscopic effects. But this robustness also means that one can not ask system specific questions, since the results are not affected by irrelevant terms. Thus the continuum theories can be used to describe only generic systems, and additional methods have to be used to include system specific effects.

Similarly, these theories provide a description of the system at a coarse grained length scale, thus any attempt to use them to provide microscopic information is bound to fail.

# 5. Open problems

In the previous sections we discussed the main role of the kinetic models and continuum theories. They work rather successfully within their limits of applicability. In this section to mention some of the problems that one faces when one tries to apply these methods to experimental systems. It is difficult to give a complete list, so the following can be regarded only as a rather personal collection of problems that need to be addressed in the near future so that the applicability of the continuum theories become more widespread.

#### 5.1. From discrete models to continuum theories

One of the major unsolved problems is to find the continuum theory that describes a particular kinetic model. Even if we limit ourselves to simple models, such as ballistic deposition, we do not have a method for *deriving* the continuum theory starting from the microscopic rules defining the model. This does not mean that there are no attempts in this direction. In fact, for a number of growth models very encouraging advances have been achieved [41,42], however, they are rather exceptions than prove the rule that no general methods exist to connect rigorously these two methods of study. On the other hand, it is rather unfair to call this a specific problem of MBE, but it is rather a problem that is characteristic of statistical mechanics in general.

The link between the kinetic models and continuum theories is provided by phenomenological methods, that relay on symmetry and conservation law arguments [1]. These methods provide continuum theories that predict successfully the asymptotic scaling of the discrete models, but are not capable of predicting more specific connections, such as the one we have seen in the case of ion sputtering, where the continuum equation has been derived starting from a model of the physical processes that take place on the surface.

#### 5.2. Stress

Currently the applicability of the stochastic continuum theories is limited to homoepitaxial systems, for which stress plays no or negligible role. However, due to the growing application of strained layers in microelectronics, much of the current experimental and theoretical efforts in MBE have focused on understanding the effect of stress on the morphology of epitaxial surfaces. Due to the nonlocal character of the stress so far it was not possible to incorporate stress in the continuum stochastic equations. Coupling the local properties of the growth process to the nonlocal stress field deprives us from the very nature of the continuum stochastic equations that makes them attractive and useful from theoretical point of view, their locality.

This does not mean that there is no continuum theory for describing stress, for it is rather straightforward to formulate the continuum theory of stress [43,44]. The problem lies in the nonlocal nature of the stress field, that depends not only on the local morphology of the surface, but also on the stress on the surface and in the bulk. Introducing such nonlocality in the continuum theories studied in the context of MBE, that are all *local*, strips us from the advantages of the continuum theories, namely that one can predict analytically their behavior.

Thus the dynamics of a growing interface, coupled to a dynamically varying stress seems to be a hopelessly complicated open problem. However, advances can be obtained if one tries to understand specific effects, such as nucleation affected by the stress [45]. Hopefully, the solution of such specific problems will guide us in understanding the general problem of stress dominated roughening as well.

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