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## Fluctuations and Scaling in Aggregation Phenomena

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We introduce a method which enables us to isolate different sources of fluctuations during a typical aggregation process. As an example, we focus on the evolution of islands during irreversible submonolayer epitaxy and show that only spatial fluctuations in their nucleation are required to produce the scaling of the island-size distribution as determined by Monte Carlo simulations. In particular, once the islands are seeded, their growth can be described in a purely deterministic manner.

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Many physical phenomena are governed by the aggregation of material. Such processes have been studied in earnest since the early parts of this century [1] and continue to attract considerable interest within areas that range from astrophysics [2] to biology [3,4]. The most powerful methods for analyzing these phenomena are based on general scaling arguments [5] applied to the number and size distribution of the accreting objects. This scaling theory is supported by analytic solutions of particular mean-field rate equations [6-8] and kinetic Monte Carlo (KMC) simulations [9,10].

However, these methods do not provide a way of systematically addressing the physical *origin* of scaling behavior. The reason for this can be traced to the complex nature of aggregation phenomena, which typically consist of a nucleation regime, when new objects are created, followed by an aggregation regime as these objects grow by accreting matter from their surroundings. Since both nucleation and growth are due to microscopic processes which are inherently statistical in nature, it is difficult to ascertain *a priori* which aspects of either are intrinsically stochastic and which are essentially deterministic. This has proven to be a major obstacle in the quest for a simple, yet physically realistic, model for aggregation phenomena, where it is essential to understand which aspects of nucleation and growth can be described only by including the appropriate fluctuations.

In this Letter, we introduce a model which allows us to address the foregoing issues by examining the effects of individual sources of fluctuations for a prototypical aggregation phenomenon, irreversible submonolayer epitaxy. Our model is based on the level-set method [11,12], which is a general technique for simulating the motion of moving boundaries. As applied to epitaxial growth [13], this method solves the Stefan problem for the motion of island boundaries in the presence of the adatom diffusion field while accommodating the topological changes caused by nucleation and any coalescence. The analytical framework of our model is ideally suited to the task

at hand and, moreover, allows us to ask new questions about the roles of various fluctuations in different regimes of growth.

Submonolayer epitaxial growth is a problem of considerable practical importance in its own right because the surface morphology and, specifically, the size distribution of islands in this regime forms a template for the multi-layer surface morphology [14]. The basic scenario of epitaxial growth by, e.g. molecular-beam epitaxy, involves the deposition of atoms onto a substrate with flux  $F$  and the subsequent diffusion of these atoms with diffusion coefficient  $D$  until they either participate in the nucleation of new islands or attach to existing islands. For irreversible aggregation at fixed coverage, a central quantity which characterizes the morphology of the surface is the ratio  $D/F$ . In particular, in the limit  $D/F \rightarrow \infty$  (the scaling limit), the distribution of island sizes can be expressed in terms of a universal function [5,9]. One purpose of this Letter is to identify the fluctuations which are relevant in this limit. We find that only spatial fluctuations in the seeding of new islands, suitably weighted by the adatom density, are required to produce the correct scaling form of the island-size distribution, as determined by KMC simulations. No other sources of fluctuations (deposition, nucleation times, the motion of island boundaries) are required to produce this scaling form, but away from the scaling regime, the effects of these fluctuations are not altogether negligible.

Because deposition and migration are stochastic processes, there are a number of possible sources of fluctuations in a more "coarse-grained" description of epitaxy based on the nucleation and growth of islands. Of course, a model that correctly describes the spatial and temporal fluctuations of the adatom concentration implicitly captures all fluctuations. Such microscopic models, usually based on KMC methods, have reproduced many aspects of submonolayer epitaxy for a variety of experimental scenarios, especially island size distributions [10,15]. However, in order to build a conceptually simpler macroscopic

model, it is desirable to focus on the fluctuations in nucleation and growth apart from those in the adatom concentration. These fluctuations are difficult, if not impossible, to isolate within a typical KMC simulation because island nucleation and growth are merely consequences of the underlying fluctuations in deposition and diffusion.

In contrast to KMC simulations, mean-field rate equations [16] are completely deterministic and spatially homogeneous evolution equations for the adatom and island densities. Rate equations can produce the adatom and island densities determined by KMC simulations [17], but not the island size distribution, unless spatial information is included, e.g., from simulations [7] or experiment [18]. Thus, it is apparent that rate equations do not provide a self-contained framework for describing aggregation phenomena.

The central constructs of our model are a spatially-varying adatom density and the creation and motion of island boundaries by the level set method. The main idea behind this method [11,12,19] is that a (zero thickness) boundary curve, such as the boundary of an island, can be represented by the set  $\varphi = 0$ , called the *level set*, of a smooth function  $\varphi$ , the level set function. For a given boundary, this function evolves according to

$$\frac{\partial \varphi}{\partial t} + \mathbf{v} \cdot \nabla \varphi = 0 \quad , \quad (1)$$

where  $\mathbf{v}$  is the boundary velocity. All physical information about the boundary motion is contained in the normal component  $v_n = \mathbf{n} \cdot \mathbf{v}$ , where  $\mathbf{n}$  is the outward normal of the moving boundary and  $\mathbf{v} \cdot \nabla \varphi = v_n |\nabla \varphi|$ . For the case of irreversible aggregation,  $v_n$  is computed as [20]

$$v_n = a^2 D (\mathbf{n} \cdot \nabla \rho^- - \mathbf{n} \cdot \nabla \rho^+) \quad , \quad (2)$$

where  $a$  is the lattice constant,  $\rho$  is the adatom density, and the superscripts label the contributions from above (+) and below (-) the boundary. The adatom density is obtained by numerically integrating the diffusion equation

$$\frac{\partial \rho}{\partial t} = F + D \nabla^2 \rho - 2 \frac{dN}{dt} \quad , \quad (3)$$

with the condition that  $\rho = 0$  at all island boundaries and where  $N$  is the island density. The last term accounts for the loss of adatoms due to the nucleation of dimers, which are the smallest stable islands,

$$\frac{dN}{dt} = D \sigma_1 \langle \rho^2 \rangle \quad , \quad (4)$$

where  $\langle \cdot \rangle$  denotes the spatial average of  $\rho(\mathbf{x}, t)^2$  and  $\sigma_1$  is the adatom capture number [17]. The square of the adatom density is used because two adatoms are needed at the same position to form a dimer.

The model, as described by Eqs. (1)-(4), is not yet complete because we have not specified whether the

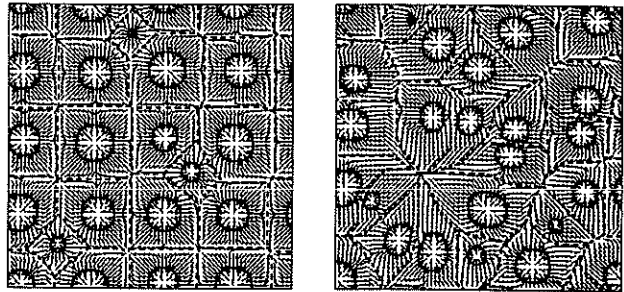


FIG. 1. Typical surface morphologies (solid lines) at a coverage of 0.2 monolayers (ML) using the deterministic (left panel) and probabilistic (right panel) seeding style. The results were obtained on a square lattice of size  $L/a = 90$ , and with  $D/F = 10^6$ . The capture zone boundaries (dashed lines) were obtained by separating the domains of attraction, where the arrows point in the direction of the gradient of the diffusion field.

flux and diffusion field are stochastic or deterministic, nor have we specified how new boundaries are created through nucleation and incorporated into the level set function. The accommodation of new boundaries within the level set function is accomplished generically by adding to the right hand side of Eq. (1) a source term of the form  $\sum_n \delta(t - t_n) \delta(\mathbf{x} - \mathbf{x}_n)$  to account for nucleation events which occur at times  $t_n$  and at positions  $\mathbf{x}_n$ . Note that the structure of these equations implies that any fluctuations (spatial or temporal) in either the adatom density or flux produces only temporal fluctuations in the nucleation rate, because a spatial average of  $\rho(\mathbf{x}, t)^2$  is taken in Eq. (4). Spatial fluctuations of nucleation events must be specified independently. A natural choice is to treat deposition and adatom diffusion deterministically, which implies that the  $n$ th island is seeded at time  $t_n$  defined by the number of islands  $NL^2$  passing the integer value  $n$ , where  $L$  is the system size. The justification for this choice will be provided below. For the spatial dependence of nucleation, we consider two schemes: deterministic seeding and probabilistic seeding. By deterministic seeding we mean that new islands are seeded at the point where  $\rho^2$ , as computed from Eq. (3), has its maximum value. In contrast, the position of a new island seeded probabilistically is weighted by a probability that is proportional to the local value of  $\rho^2$ . Note that, according to Eq. (2), the motion of island boundaries is deterministic for *any* seeding scheme if  $F$  and  $\rho$  are deterministic.

Figure 1

shows typical island morphologies obtained with the two different seeding styles. While the number of islands nucleated on the surface is (almost) independent of the seeding style [22], it is apparent that deterministic seeding leads to a much more regular island morphology. This

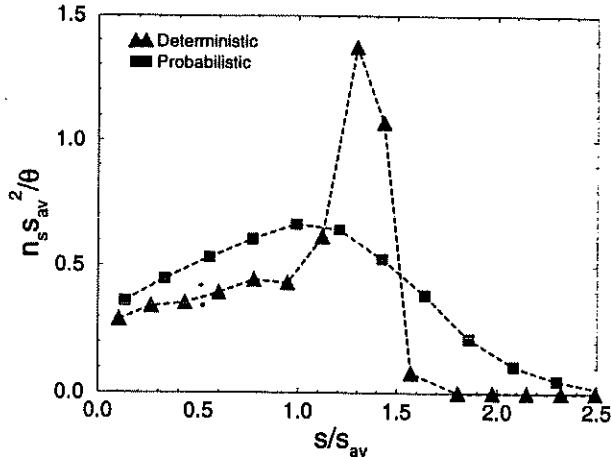


FIG. 2. Scaled island-size distributions obtained with different seeding styles, where  $n_s$  is the number of islands of size  $s$ ,  $s_{av}$  is the average island size, and  $\theta$  is the coverage. All results were obtained with  $L/a = 180$ , a numerical grid of linear size 512, and with  $D/F = 10^6$ . The data for probabilistic seeding represents an average over at least 50 independent lattices.

can also be seen in Fig. 2, where we show scaling plots of the island size distributions [9] for the two different seeding styles. The distribution function produced by the deterministic seeding style is seen to exhibit a pronounced peak with a sharp drop-off for large islands. As is evident in Fig. 1, islands are always seeded as far away as possible from existing islands, because this is where the adatom concentration is highest. This has the effect of diminishing the competition between neighboring islands for adatom capture, so many islands grow to approximately the same size.

The vastly different shapes of the distribution functions invite comparison to data obtained from KMC simulations and from experiment. In Fig. 3 we compare the scaled island-size distribution functions with probabilistic seeding to that obtained from KMC simulations and from experimental data for Fe/Fe(100) [23]. The agreement between the distributions generated by the probabilistic seeding and those from the KMC simulations is quite striking and both give an acceptable account of the experimental data. Moreover, we obtain excellent data collapse of the distribution functions for different coverages and different values of  $D/F$ .

The strong qualitative differences between the island-size distributions obtained with the different seeding styles implies that spatial fluctuations in the seeding of new islands are an essential ingredient of any model. This is ultimately the reason why standard mean-field rate equations do not reproduce the correct island-size distribution. The spatial information of the local environment of each island is necessary for the correct evolution of the surface morphology and is a direct consequence of

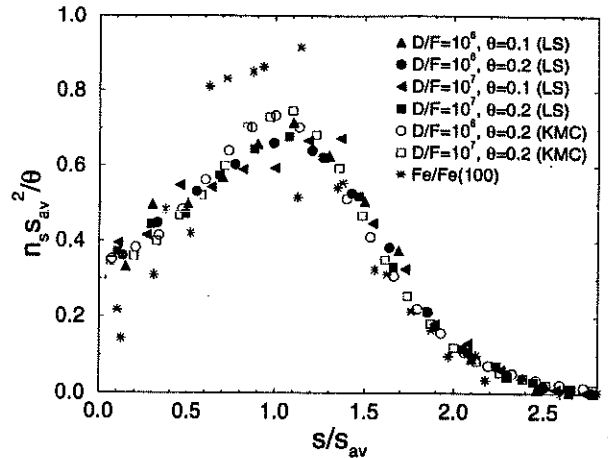


FIG. 3. Scaling of island-size distributions obtained from the level-set method (LS) and KMC simulations. The numerical parameters are the same as those in Fig. 2. The KMC data are the average of 8 runs on a  $2000 \times 2000$  lattice. Edge diffusion was included in the simulation to obtain compact island shapes. Also shown is the scaled island-size distribution for Fe/Fe(100) [23].

the spatial fluctuations in the seeding of islands. This can be illustrated by examining the domains of attraction of the adatom diffusion field associated with the islands (Fig. 1). These “capture zones” provide a direct indication of the growth rate of an island [7,24]. It is immediately apparent from Fig. 1 that the deterministic seeding style leads to a much more regular distribution of capture zones, which is why the islands have a much more regular size distribution. It is interesting to note that this size distribution is qualitatively very similar to that obtained from a model where the velocity is assumed to be spatially constant [13]. The placement of new islands in the deterministic seeding style has the effect of eliminating (almost) all spatial information because all the islands have (almost) identical local environments.

We have chosen the time of nucleation events deterministically, as given by Eq. (4). We can now understand why this is justified. With increasing  $D/F$ , there is a greater tendency for the nucleation of all islands to occur at earlier times. Thus, time interval between successive nucleation events decreases, so islands grow very little between successive nucleation events. Substantial island growth occurs only after nucleation has almost completed. Therefore, any temporal fluctuations in the seeding will have very little influence on the size of an island. All that appears to be relevant is the spatial correlation of island positions, which is accounted for by the spatial fluctuations in the seeding.

Once the islands are seeded the island boundaries can be evolved in a purely deterministic fashion. This means that the fluctuations in the velocity of the island boundaries can be averaged, while the spatial fluctuations in

the seeding cannot. This can be understood as follows: Once a "mistake" is made in the choice of the location of a new island, it cannot be corrected, while a fluctuation in the growth of an island boundary can be corrected over time.

Finally, we discuss the effect of fluctuations on scaling. From the quality of the data collapse in Fig. 3, we conclude that our model has attained its scaling limit. Since we have included only one type of fluctuation, it is quite plausible that only spatial fluctuations in island seeding are relevant in this limit. We cannot exclude the importance of other fluctuations in the pre-asymptotic regime, but the data in Fig. 3 suggest that these effects are small for typical values of  $D/F$  ( $\geq 10^5$ ).

Quite apart from the specific results presented here, we stress that our methodology allows us to ask questions and address issues that were previously difficult, if not impossible, to pursue with established methods. The central issue considered here, namely, the identification of the dominant fluctuations in different regimes of irreversible aggregation, has a natural solution within the framework of our model. Moreover, more general aggregation phenomena which include mobile clusters and coagulation, in addition to the processes we have described, are natural extensions of our methodology. We believe that new avenues of investigation into the relevance of different types of fluctuations in various regimes of aggregation phenomena may be opened by considering simpler models that encapsulate the essential features of this model.

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