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LETTER TO THE EDITOR

Growth oscillations

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Abstract. We describe an up till now unrecognised phenomenon in kinetic growth models which leads to observable oscillations in such quantities as the density and velocity of growth. These oscillations, which can occur on length scales of many lattice spacings, arise because of an induced incommensuration in the growth mechanism. To illustrate the phenomenon, we present results for a particularly simple model, but the phenomenon is expected to be quite general and appear in a wide range of growth processes. The essential ingredients for the existence of the oscillations are that the growth take place at a reasonably well defined interface and that the growth process be discrete (e.g. that the cluster grows by the addition of discrete particles of finite size). The growth process is related to a functional stochastic iterative map so that the growth oscillations play the role of limit cycles. We suggest that the fixed point of this map is related to critical fractal kinetic growth.

Models of non-equilibrium kinetic growth have recently attracted considerable attention (see [1] for a representative sample of work). Many interesting features are exhibited by these processes including a wide range of morphologies as well as other phenomena associated with singular growth behaviour, such as velocity selection. In this letter we describe a new aspect of non-equilibrium growth which we believe applies to a very large class of kinetic growth processes. The property we shall discuss leads to oscillations in a variety of observable properties of growing systems such as the density of the structure and the velocity of the growing interface. Furthermore, our treatment of this phenomenon suggests a scenario for understanding a number of other features of kinetic growth including certain kinds of fractal growth.

To most simply describe our growth oscillations, it is useful to consider a specific, rather simple kinetic growth model which exhibits these features. It should be borne in mind, however, that this phenomenon is expected to apply to a very general set of growth processes. (In fact, the growth oscillations we shall describe have also recently been observed in ballistic aggregation [2].) The model we will treat is defined as follows. Place a seed particle on a site of a two-dimensional square lattice. Occupy each nearest neighbour of the seed particle independently with a probability p . Call the newly occupied sites the second generation. Occupy each nearest neighbour of a second generation particle independently with probability p . Call the newly occupied sites the third generation, and so forth. This and related models have a number of interesting features which are described in detail elsewhere [3]. For our purposes it is only important to note the following. For $1 > p > 0.705$ the model grows a structure which looks like a diamond with rounded corners along the axes of the lattice and intervening straight facets oriented at 45° . The facets have an interfacial width of order 1 as the structure grows, while the rounded sections are rough, with an interfacial width growing like N^δ , where N is the number of generations for which the structure

has grown. For $0.54 < p < 0.705$ the structure grows without facets, albeit with an anisotropic shape. For $p = 0.54$ the growth is marginal and the resulting structure is fractal-like, probably a fractal in the universality class of percolation clusters.

Let us now look in more detail at the behaviour of the growing interface. In particular, consider a ray extending out from the centre of the cluster. Let $P_n(x)$ be the probability that a point x along the chosen ray gets occupied exactly at the n th generation. Because our growth is stochastic we need to average over many sample clusters to compute this quantity. In figure 1 we have plotted $P_n(x)$ along a ray coincident with one of the lattice axes. The growth probability in this case is $p = 0.8$, so that the ray intersects a curved section of the cluster interface.

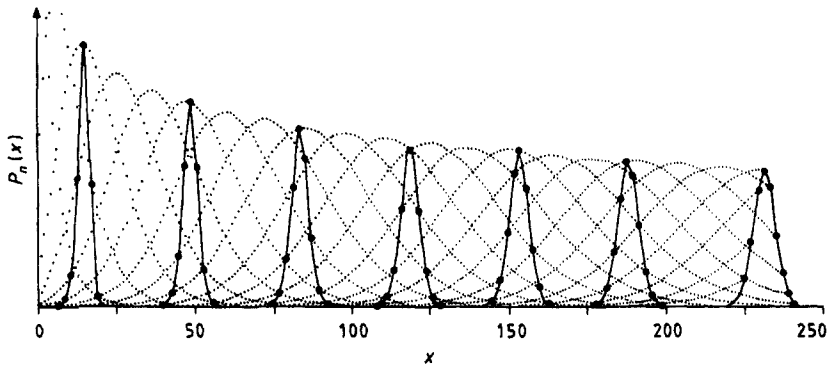


Figure 1. $P_n(x)$ (in arbitrary units) plotted as a function of x , the distance along a lattice axis, averaged over 17 600 samples of the stochastic model grown for 270 generations with $p = 0.8$. The heavy dots, connected for visual clarity by full curves, are those points, $P_n(x)$, as a function of x for one value of n . Curves for $n = 20, 60, 100, 140, 180, 220$ and 270 are shown. The dots are the trails of all values of $P_n(x)$ for all $n \leq 270$ and integer x .

Because the lattice is discrete, $P_n(x)$ is defined only for integer x . Each dot (heavy or light) represents a value of $P_n(x)$ for some x and n , for all $n \leq N = 270$. The heavy dots, connected for visual clarity by full curves, are those points $P_n(x)$ as a function of x for one value of n and thus represent the shape of the growing interface at time n . Curves for several different values of n are indicated. Notice that while the growing interface is relatively narrow (i.e. $P_n(x)$ extends over ten or so lattice points for a given n), the superposition of all the $P_n(x)$ lie on much broader curves. If we focus our attention on the uppermost points in this figure, we see that the maximum value of $P_n(x)$ undergoes rather regular oscillations as a function of n (or of x), apparently with a period of about 13 lattice spacings. (As we shall see, the actual nature of the periodicity is rather more complicated.) If we think about the meaning of $P_n(x)$, we are led to the conclusion that these oscillations should show up in physically measurable quantities, such as the velocity of the interface and the density of the cluster. As we shall discuss in a moment, this is indeed the case. However, it is useful to first understand qualitatively the origin of the oscillations.

The oscillations exist because of a hidden built-in incommensuration or beating in the problem. Having chosen a value of $p < 1$, the average growth velocity along an axis of the lattice is less than one lattice spacing per generation. (In the case of figure 1, for example, the average growth velocity is very close to $\frac{13}{15}$.) To understand what this implies, imagine that there is a 'meta' interface profile, $Q(x)$, which is a continuous

function of a continuous variable, x , and which merely translates along the x axis with n , without changing shape†. The values of $P_n(x)$ for integer x are the values of $Q_n(x)$ for integer x . Since $Q_n(x)$ moves a fraction of a lattice spacing each generation, the integer values of x will migrate along $Q_n(x)$ as n increases, and the trail of these points will be as shown in figure 1, resulting in an oscillation of the maximum of $P_n(x)$ with n .

There are several important comments to make. Firstly, it is clear that this beating can result in oscillations over periods of many lattice spacings. Thus a microscopic effect can result in oscillations on mesoscopic or macroscopic scales. Secondly, if we study $P_n(x)$ along a direction intersecting a facet, we expect, on the basis of the argument above, no growth oscillations since the facet moves one lattice spacing per generation. This is what is observed and is discussed in detail in [4]‡. Thirdly, we expect these growth oscillations to be quite ubiquitous: a little thought reveals that what is generically necessary for the oscillations to occur is (i) that the growth proceeds by a discrete process (such as the addition of particles of some size to a cluster) and (ii) that the growth takes place at a fairly well defined interface. These kinds of conditions should occur in a wide variety of kinetic processes and in these processes we may expect to see growth oscillations. (The case of off-lattice growth by the addition of discrete particles of finite size as well as asynchronous growth is discussed further in [4].)

We have recently undertaken a study of ballistic aggregation and have observed growth oscillations in this process also, qualitatively similar to the data plotted in figure 1. Details of this calculation will be reported elsewhere [2]. It should also be emphasised that the occurrence of growth oscillations is to be understood as a generic statement: not all processes obeying these conditions will necessarily show growth oscillations (e.g. the facet problem described above) but growth oscillations should be common in such processes. To demonstrate that these oscillations produce physically observable effects, we have studied the density of the cluster along a lattice axis with $p = 0.8$. In terms of the $P_n(x)$ plotted in figure 1, the density, $\rho(x)$, averaged over a number of simulations may be defined as

$$\rho(x) = \sum_n P_n(x) = \bar{\rho} + \eta(x) \quad (1)$$

where $\bar{\rho}$ is a suitably determined average (constant) value of the density and $\eta(x)$ represents oscillations on top of this constant. We have computed the Fourier transform of $\eta(x)$ determined from an ensemble of 17 600 simulations grown for 250 generations. We find a strong Fourier component in the spectrum of $\eta(x)$ corresponding to oscillations in one density with a period of 13 lattice spacings and an amplitude of about 0.5% of $\bar{\rho}$. In addition, we find several other significant amplitudes corresponding to somewhat less pronounced longer wavelength oscillations. Thus the density oscillations in this model appear to have a rich multiperiodic structure dominated by a fundamental periodicity. This spectrum will be described in more detail elsewhere [4].

† For purposes of explanation we assume that the interface profile does not change shape as n increases. However, it is clear that there is some widening in $P_n(x)$ of figure 1 due to stochastic roughening. This does not affect the main argument.

‡ The detailed mechanism with which the system deals with the growth incommensuration is elucidated in reference [4] in the context of certain deterministic models of growth. There it appears that the average growth velocity is a smooth function of p and that small changes in the growth velocity result generally in modulations of, for example, the density by small amplitude long wavelength contributions.

This Fourier transform represents density variations in an ensemble of small clusters. We also expect to see density oscillations in a single larger cluster, which is a more common experimental situation. To understand this in the context of our present model, we need to note first that the interface of our cluster is rough with an interfacial width growing like n^δ where [3] $0 \leq \delta \leq 0.5$ for $p \geq 0.7$ and $\delta < 1$ for $p > p_c$. This means that the dispersion of the peak of a density oscillation will grow in the model like n^δ . On the other hand, the period of the oscillations are independent of n since they are controlled by the average growth velocity which is independent of n [3, 4]. Thus in a cluster of linear dimension L , the Fourier amplitude associated with the fundamental density oscillation will grow like $L^{1-\delta}$. Assuming a background of white noise, we see that for $\delta < \frac{1}{2}$, the signal-to-noise ratio in a diffraction experiment (or Fourier transform) will improve as the linear size of a sample increases, and so density oscillations will in principle be observable in single large samples. For cases in which $\delta > \frac{1}{2}$ there may be an optimum single sample size for observing density oscillations. This is a more detailed experimental question which we cannot discuss here.

These density oscillations, as well as oscillations in the growth velocity, will be analysed in more detail elsewhere [4]. Now, however, we want to turn to a brief discussion of a deterministic model which, in a certain sense, mimics the average behaviour of the stochastic model described above. It is considerably simpler to analyse numerically, but shares enough features with the stochastic model to illustrate a number of important points.

Consider the iterative equation

$$P_{n+1}(x) = p \left(1 - \prod_y (1 - P_n(y)) \right) \left(\prod_{m=1}^n (1 - P_m(x)) \right). \quad (2)$$

The first factor on the right-hand side is the growth probability, p . In the second factor the product over y is a product over all nearest neighbours of the site x . This factor measures the probability that at least one nearest-neighbour site was occupied exactly at time n . The last factor measures the probability that the site x has not been occupied at any previous time. The dynamics of this equation clearly mimics that of the stochastic process although there is no noise in equation (2) and the probabilities are treated as independent. If we choose the initial condition

$$P_1(x) = \delta(x) \quad (3)$$

which just places a single seed particle at the origin, then equation (2) generates a structure with a morphology similar to that of the original stochastic model†.

In figure 2 we have plotted $P_n(x)$ for equations (2) and (3) with $p = 0.8$ along a ray coincident with one of the lattice axes. As in figure 1, we have shown the trail of points of $P_n(x)$ for $1 < n < 270$ as well as the function $P_n(x)$ for a few values of n . This figure shows the same general oscillatory structure as figure 1. Unlike figure 1, we see no broadening of $P_n(x)$ with n , which is to be expected because there is no stochastic noise in equation (2). (Indeed, the width of the interface of the cluster grown according to equations (2) and (3) is always $O(1)$, independent of n .)

We have also studied the density of this deterministic model, defined as in equation (1). Apart from some initial transients, we find that $\eta(x)$ has a multiperiodic structure with a fundamental period of 37 lattice spacings modulated by longer wavelength

† In fact, several related deterministic models all generate structures with similar morphologies. The connection between these deterministic models and stochastic models will be explained in more detail in [4].

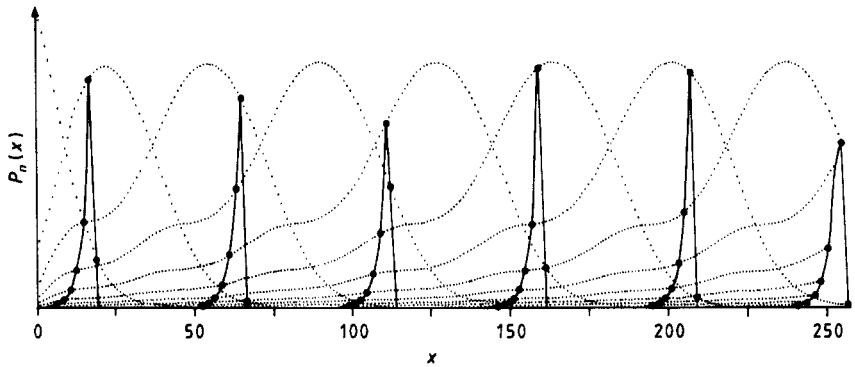


Figure 2. $P_n(x)$ (in arbitrary units) plotted as a function of x , the distance along a lattice axis for the iterative map, equation (3), carried to 270 generations. Each full curve connects the points $P_n(x)$ as a function of x for one value of n . Curves for $n = 70, 120, 170, 220$ and 270 are shown. The dots are the trails of all values of $P_n(x)$ for all $n \leq 270$ and integer x .

oscillations with smaller amplitude. As in the stochastic model, the oscillations of the density here are about 0.5% of the constant, $\bar{\rho}$. The deterministic map of equation (2) therefore has the same qualitative structure as the stochastic model, but is rather simpler†. In fact, we have been able to calculate analytically the dotted curves in figure 2 [4]. The mathematical relation between the stochastic and deterministic models is also quite interesting and will be discussed in detail elsewhere.

The formulation of a deterministic model like that of equation (2) which has the structure of an iterative map suggests that growth oscillations may be thought of as limit cycles of a somewhat complicated non-linear map. Thinking about the problem in this way one is led to ask a host of questions familiar from the study of non-linear dynamical systems, including the dependence of the period of oscillation on p , the absence or presence of mode-locking, as well as questions concerning the more detailed multiperiodic nature of the oscillations and their physical consequences. In reference [4] we have been able to partially answer a number of these questions in the context of deterministic models of the form of equation (2).

One may also be led to inquire about the nature and meaning of a fixed point of this iterative map. In this regard an extremely interesting speculation suggests itself: it is observed, both in this stochastic model and in the deterministic map of equation (2), that as p decreases so does the period of the oscillations associated with $P_n(x)$. On the other hand, we know that in these models growth does not proceed for $p < p_c$, where p_c is a critical value at which the growth is marginal and fractal-like. For $p \leq p_c$, therefore, the growth of the system, thought of as an iterative map, is controlled by the trivial functional fixed point of the map, $P_n(x) = 0$, and the fractal structure of the cluster when $p = p_c$ is determined by the rate at which the trivial fixed point is approached as $n \rightarrow \infty$. This, then, is the fixed point with which a random kinetic fractal is associated. The Hausdorff dimension and other properties of such clusters can therefore be calculated by renormalising the appropriate stochastic iterative map. An analysis of the fixed point for the stochastic model considered above, for example,

† It is also possible to further simplify equation (2) considerably and still maintain the general features of growth oscillations. In fact, even a simplified one-dimensional version of equation (2) exhibits growth oscillations (see [4] for details).

should allow us to calculate explicitly the properties of fractal percolation clusters. (In this context we note that we have been able to determine the behaviour of the model of equation (2) for p near p_c . Details will be reported elsewhere [4].)

We believe that the growth oscillations discussed here are likely to be a quite ubiquitous phenomenon and should be apparent in kinetic processes in which the dynamics builds in some incommensurations or beating in length or timescales. In the models discussed here both space and time were discrete, which made the description of the incommensuration relatively simple. A more common growth process is one in which time is not discrete, in the sense that growth events do not take place at regular intervals. Nevertheless, we may expect to see growth oscillations in certain quantities even in this case. For example, if we imagine a process in which particles are added to a cluster in a way which is affected by the local geometry, then if the local geometry is shielded on a length scale different from the size of the particles, we may expect that, on average, the local geometry will pass through a number of different configurations before returning to its original one, thus giving rise, in general, to an oscillation in the density. If, on the other hand, the width distribution of time intervals between the growth events is very broad, i.e. comparable to or greater than the period of the expected growth oscillations, then the growth oscillations in the velocity may be quite difficult to observe.

In summary, the growth oscillations we have described appear to be a very general fundamental feature of a wide range of kinetic growth processes. They have been observed both in the models studied in this letter and in ballistic aggregation [2]. Our discussion has led us to a picture of the kinetic growth of a cluster in terms of iterative maps. An important implication of this picture is the suggestion of a method for calculating the properties of fractals generated by kinetic growth mechanisms. We have also shown that these growth oscillations lead to physical effects which should be observable in carefully controlled growth experiments.

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