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LETTER TO THE EDITOR

Growth oscillations in ballistic aggregation

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Abstract. We demonstrate the existence of growth oscillations in single clusters of particles grown stochastically according to a synchronous (finite density) version of ballistic aggregation, which is a model of relevance to a variety of experimental situations. We find clear evidence for these unexpected dynamically induced oscillations both in the propagation of the interface and in the resulting density of the cluster. We describe the general features of the spectra of the oscillations, and briefly discuss possible experimental systems in which growth oscillations might be observed.

As a result of substantial technological progress in recent years, it has become possible to observe details of kinetic and structural features of materials even at the atomic level. These impressive experimental advances stand as an open challenge to theorists to elucidate and predict the now observable effects in the microscopic and mesoscopic regime. Of particular interest are those effects which are general, transcending the detailed nature of the interparticle interactions.

Such an effect was recently described [1, 2]. It was argued that, contrary to naive expectations, kinetic growth processes should, generally, manifest oscillatory behaviour in a variety of physically observable quantities, including the velocity of the growing interface and the density of the resulting structure. These oscillations can be expected in processes in which the growth proceeds by the accretion of well defined packets of material (so that the process is microscopically discrete) and in which the growth takes place at a moderately well defined interface. The oscillations, which may have wavelengths up to orders of magntidue larger than atomic distances, occur as a result of the beating between two length scales. The first is a static length scale characterising the size of the accreting material, and the second is a suitably defined, dynamically generated length scale, related, for example, to the velocity of the moving interface [1, 2]. While the size of the effects we describe is typically rather small, they should be observable in carefully controlled experiments using recently developed techniques of growth and observation such as molecular beam epitaxy and scanning tunnelling microscopy.

In this letter we present strong evidence for the existence of these growth oscillations in ballistic aggregation, a commonly studied stochastic model of growth, which is applicable to a variety of experimental situations [3]. We will see that oscillations are observable both in the propagation of the moving interface and in the resulting density of large single clusters. Moreover, although the oscillations have a highly multiperiodic structure (in fact, they are expected to be quasiperiodic [4]), we are able to explain some of their main features by simple qualitative arguments.

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The model we have studied is a synchronous version of ballistic aggregation in two dimensions. (We shall comment below on some results in higher dimensions.) Consider a square lattice. To begin, place a line of L seed particles at adjacent sites along the horizontal axis. Particles are added to the cluster at discrete times according to the following algorithm. Let $h(i, n)$ be the height above the substrate (of seed particles) of the uppermost particle in column i at time n. If at time $n+1$ a particle falls in column i, it will stick at a height determined by

$$h(i, n+1) = \max[h(i-1, n), h(i, n)+1, h(i+1, n)]. \tag{1a}$$

If at time $n+1$ no particle falls in column i, then

$$h(i, n+1) = h(i, n). \tag{1b}$$

A particle will fall in a given column at a given time, independently and randomly with a probability p, where p is a fixed control parameter. Thus, at each time step pL new particles, on average, are added to the cluster. To complete the description of the model, we specify periodic boundary conditions for the substrate so that we can think of the L seed particles as being wrapped around the surface of a cylinder. (The asynchronous version, in which one particle at a time is added to the cluster has been studied by a number of groups [5]. To our knowledge, this is the first time the synchronous model has been discussed.)

The cluster grown according to this algorithm is an object with finite density (not a fractal). The interface is rough in the technical sense. For larger values of p, the average density of the cluster and the average velocity of the interface are larger, and the interface is less rough [6].

Now let us suppose that we have grown the cluster for some time, N. There are several different physical quantities we can analyse for the presence of oscillations. First, we can define the average density, ρ, as a function of x, the distance above the substrate. Note that x is an integer in units in which the lattice spacing is equal to one. It is interesting to ask what $\rho(x)$ is well behind the position of the interface. A little thought leads to the expectation that, with the exception of some short-lived initial transients, $\rho(x)$ should be constant. Similarly, if we define the average geometric position of the interface at time n by

$$\langle h(n)\rangle = (1/L)\sum_i h(i, n) \tag{2}$$

and the velocity of the interface by

$$v(n) = \langle h(n)\rangle - \langle h(n-1)\rangle \tag{3}$$

we would expect, again ignoring initial transients, that $v(n)$ would be constant, and thus $\langle h(n)\rangle \sim n$

To see whether this is the case, it is convenient to remove the trivial time dependence from $\langle h(n)\rangle$ by defining

$$H(n) = \langle h(n)\rangle - v_0 n \tag{4}$$

where v_0 is a constant, time-averaged velocity,

$$v_0 = (1/M)\sum_n v(n) \tag{5}$$

with the sum over n running from $n = t$ to $n = t+M$. If we choose a range in n over which $v(n)$ is sensibly constant, then over that range, $H(n)$ should be independent of

n. Since $\langle h(n)\rangle$ is a stochastic quantity, our naive expectation is that the power spectrum of (4) should just exhibit the spectrum associated with white noise.

In a similar way, we may define an 'active' height, $d(i, n)$, as opposed to the geometric height, $h(i, n)$. $d(i, n)$ is defined by the right-hand side of (1a):

$$d(i, n) = \max[h(i-1, n), h(i, n)+1, h(i+1, n)] \tag{6}$$

and represents the zone of potential growth at the next time step. We may also define $\langle d(n)\rangle$ in analogy with equation (2). It is not difficult to show that $\langle h(n)\rangle$ and $\langle d(n)\rangle$ are related by the following simple expression:

$$\langle h(n+1)\rangle = p\langle d(n)\rangle + (1-p)\langle h(n)\rangle. \tag{7}$$

Finally, we may define for $\langle d(n)\rangle$ a quantity analogous to $H(n)$,

$$D(n) = \langle d(n)\rangle - u_0 n \tag{8}$$

where u_0 is defined, in analogy to v_0, as

$$u_0 = (1/M)\sum_n [\langle d(n)\rangle - \langle d(n-1)\rangle] \tag{9}$$

with the sum in (9) running from $n = t$ to $n = t + M$.

In order to study the behaviour of $\rho(x)$, $H(n)$ and $D(n)$, we have generated four clusters using the algorithm of equation (1). Three were grown with $p = 0.8$ for 1200 generations each, and with three different sizes of substrate: $L = 10^7$, 5×10^6 and 6×10^5. To avoid possible spurious correlations, a different random number generator was used to grow each of the samples†. In addition, we have grown one cluster for 1200 generations with $p = 0.9$ and $L = 10^7$. To analyse $H(n)$ or $D(n)$ for the presence of oscillations, one could perform a simple discrete Fourier transform (FT) and look at the frequency spectrum. We did this and, as we shall soon demonstrate, the results of those calculations are consistent with the conclusions we shall describe below. We leave the detailed description of the simple Fourier analysis to another paper [7], because there is a more efficient method for wresting the signal from the noise in a case like ours. This method is a variant of the simple FT and is called the maximum entropy method (MEM) [8]. The MEM is widely used in signal processing applications, but appears to be relatively unknown among physicists. MEM differs from a simple FT in that the latter makes the implicit assumption that the function to be transformed is zero outside the range for which data are given. MEM, on the other hand, makes no explicit assumption about the data outside the range for which it is known, but rather maximises a certain measure of ignorance of the data where they are not given. In particular, if $R(\nu)$ is the power at frequency ν, then the MEM produces Fourier amplitudes (the absolute squares of which are $R(\nu)$) which maximise $S = \int \log[R(\nu)]\,\mathrm{d}\nu$ consistent with the constraints that these Fourier amplitudes reproduce correctly the values of certain correlation functions of the data. In general, for a process with stationary noise, if we compare the power spectrum produced by a simple FT and by the MEM from a finite set of data, we will find that the power spectrum produced by the MEM is much closer to the power spectrum of the infinite set of data of which the finite data form a subset. In practice, the MEM involves a calculation similar to Padé approximants in the complex frequency plane. As with Padé approximants, the MEM comprises a sequence of approximations. The approximant with N poles maximises

† With an efficient random number algorithm, the largest clusters took approximately 4 h of Cray 2 CPU time.

S, consistent with the constraints that all the time correlation functions of the data from zero to N time steps are correctly reproduced. As with ordinary Padé approximants, the particulars of each situation dictate which order of approximant is best suited to the problem. A fuller description of the method can be found in [7, 8]. For those uninterested in the details of the calculation, the MEM power spectra we discuss here can be thought of as a kind of smoothed ordinary FT of the data. As we stated earlier, for all the spectra we have studied, the ordinary FT always exhibit the same general structure as the spectra produced by the MEM, although the former are somewhat noisier.

Let us first discuss the time evolution of the interface. In figure 1 we have plotted the power spectrum for $D(n)$ estimated by the MEM and, for comparison, obtained by a straightforward discrete FT for our simulation with $p = 0.9$. These curves represent the transform of $D(n)$ over the range $588 \leqslant n < 1100$. Because of our initial conditions, there are some short-lived transients in both the velocity of the interface and the density of the cluster. Since we are not interested in these transients, we choose our data to avoid the initial transient region. Over this range the initial transients have largely died away, and $u(n)$ has a secular variation of less than 1%. u_0 is calculated using equation (9) with $t = 588$ and $M = 512$.

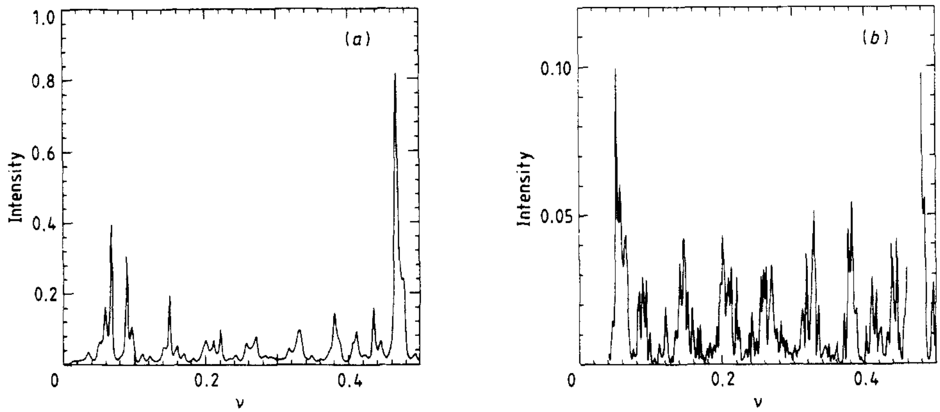

Figure 1. (a) Power spectrum of $D(n)$ for $588 \leqslant n < 1100$ as determined by the maximum entropy method with 100 poles. The horizontal (frequency) axis is normalised so that an oscillation of wavelength λ corresponds to a frequency λ^{-1}. The vertical (intensity) axis is in arbitrary units. Because $D(n)$ is real, the power spectrum for ν in the range $0.5 < \nu \leqslant 1.0$ is just the mirror image of the one shown. (b) Power spectrum of $D(n)$ for $588 \leqslant n < 1100$ obtained by a simple discrete Fourier transform.

In both figures 1(a) and (b), we see a sequence of groups of peaks in moderately well defined regions of ν. These power spectra are clearly not the white-noise spectra one might have expected, and in fact show a definite oscillatory signal. It is tempting to interpret successive groups of peaks in these spectra as successive harmonics. This is likely to be largely correct. However, it should be stressed that were we able to resolve the spectrum with extremely high accuracy, we would probably discover that the signal is really quasiperiodic [4].

These spectra, as well as the spectra for a number of other physical quantities, will be analysed in detail in a subsequent publication [7], but for the moment we wish to make a few semiquantitative comments. First, in the power spectrum of $D(n)$, there is a very large peak at very small ν ($\nu < 0.01$) which we have not indicated. This peak is just due to the fact that, as noted earlier, there is still a very small secular change in $u(n)$ over this range of n. Next, we see a very strong peak at $\nu \approx 0.47$. This peak represents an effect intrinsic to the growth algorithm (1), whereby the interface becomes alternately more smooth and more rough approximately every other time step. This effect is largely independent of p. (It is also seen in our $p = 0.8$ simulations [7].) Aside from these features, the first strong group of peaks occurs in the range $0.05 < \nu < 0.07$. As we shall argue in a moment, we can identify a ν in this range as the 'fundamental' frequency of the dynamic growth oscillations. From the size of these peaks, it is straightforward to extract the amplitude of the growth oscillations. In terms of the growth velocity, for example, if we write $u(n) = c + \delta(n)$, where c is a constant and $\delta(n)$ represents the effects of the oscillations, then $c^{-1}|\delta|$ is of the order of 10^{-4} or 10^{-5}. Effects of a similar size are also seen in the density.

On the basis of a simple intuitive picture of the origin of growth oscillations that was presented in [1], we can show that a frequency in the range $0.05 < \nu < 0.07$ should be the fundamental frequency for this process. In summary the argument is this: imagine that we can represent the interface at which growth takes place by a smooth curve, $P_n(x)$, where for the moment we suppose that x, the distance above the substrate, is a continuous variable. $P_n(x)$ is just the probability to deposit a particle at the position (height) x at time n. For each successive time step, $P_n(x)$ will have moved along the x axis a certain amount given, roughly, by $\Delta x = v_0 \approx u_0$. In units of the lattice spacing, $\Delta x < 1$, so that, relative to the underlying lattice, the curve $P_n(x)$ will now be in a different position. The values of $P_n(x)$ for integer values of x, therefore, will also be different. If we write $\Delta x = 1 - \varepsilon$, then $P_n(x)$ will have to move for approximately ε^{-1} time steps before the curve will be in the same relative position with respect to the underlying lattice. Thus we expect to see an oscillation in time-dependent quantitites such as $H(n)$, $v(n)$, $u(n)$ or $D(n)$ with a wavelength, in units of the time interval, given roughly by

$$\lambda_t = [(1 - v_0) - 1] \tag{10}$$

where [ ] denotes the integer part. In the cluster of figure 1, $v_0 \approx 0.95$, so that λ_t is about 20 lattice spacings. The frequency corresponding to this wavelength should be very close to 0.05, in semiquantitative agreement with the first group of peaks in figure 1†. There are a variety of other ways to analyse the data used in figure 1 to see the signal of this fundamental frequency and they will be described in detail elsewhere [7]. It is also worth mentioning that the small size of the oscillations is related to the shape of the interface. In particular, we observe that the oscillatory effect is strongly enhanced if the tails of the function $P_n(x)$ are suppressed. Growth processes with relatively narrow active interfaces should be good candidates for observing growth oscillations. This notion is currently under investigation.

We have performed a similar analysis for the three clusters grown with $p = 0.8$. In those cases we also see clear signals in the power spectrum of $D(n)$, in analogy with figure 1, although the stochastic noise is somewhat more pronounced. Furthermore, the wavelength of the fundamental period of oscillation in those cases is about ten

† Notice that according to equation (10), $\lambda_t \to 0$ as $v_0 \to 0$. But $v_0 \to 0$ as $p \to 0$ in an infinite system, so that the growth oscillations disappear in the usual asynchronous ballistic aggregation. This is indeed the case, and is related to a conjecture concerning the generic relationship between growth oscillations and random fractal growth. See [1] and [2] for more details.

time steps, consistent with the semiquantitative estimate of equation (10), since for $p=0.8$, $v_0 \approx 0.90$.

We have also analysed $H(n)$ and the density $\rho(x)$. Details of these results will be reported elsewhere [7]. Here we only wish to make a few descriptive comments. The results for the power spectra of $H(n)$ are consistent with those we have reported for $D(n)$ and with equation (7). There is a clear sign of oscillatory behaviour with a large fundamental peak occuring near $\nu = 1 - v_0$, as we expect.

The power spectra of $\rho(x)$ (plotted, of course, as a function of k, the variable conjugate to x) are somewhat more complicated than those of $H(n)$ and $D(n)$, in that the harmonics and multiperiodic effects are more pronounced. Nevertheless, three important characteristics of these spectra are clear. First, the three spectra of $\rho(x)$ for the three independent clusters grown with $p=0.8$ are all qualitatively similar with significant peaks occuring for similar values of k. This indicates that the peaks represent real physical effects and are not just artefacts of stochasticity. Second, in all four spectra, the three for clusters grown with $p=0.8$ as well as the one for the cluster grown with $p=0.9$, the first major non-trivial peak occurs at a value of k near $[v_0/(1-v_0)]^{-1}$. Applying the simple arguments presented earlier, based on the picture discussed in [1, 2], this is the value of the fundamental k vector we expect for the oscillations in quantitites which depend on x. The extra factor of v_0 occurs because, in $(1-v_0)^{-1}$ time steps, the interface will have moved about $[v_0/(1-v_0)]$ lattice spacings. Finally, as mentioned earlier, the amplitude of the oscillations in $\rho(x)$ are 10^{-4}–10^{-5} times the background constant value of the density.

In this letter we have demonstrated the existence and observability of growth oscillations in single clusters generated by a stochastic ballistic aggregation process. In particular, we have shown that oscillations in the overall density, as well as in the propagation of the interface, can be observed with an amplitude of the order of 10^{-4} or 10^{-5} of the average constant background. The model we have studied here is defined on a lattice. However, as we have argued elsewhere [1, 2], we do not believe that the lattice is necessary for the existence of growth oscillations. In fact, such oscillations should be observable in a variety of well controlled growth experiments with materials that are amorphous as well as imperfectly crystalline. It may be possible, for example, to observe growth oscillations in carefully controlled molecular beam epitaxy processes on cold substrates, or in the evaporative growth of amorphous silicon. In order to observe the oscillations in the density, it is important that there be relatively little relaxation of the structure after deposition. Thus suggests that amorphous silicon or germanium grown by physical vapour deposition at or below room temperature may be a good candidate for observing the effect†. In this regard, it is worth noting that we have performed some small-scale simulations of ballistic aggregation onto a two-dimensional substrate. While the details differ from one-dimensional systems, growth oscillations are also observed in two dimensions.

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