

LETTER TO THE EDITOR

Quasiperiodic behaviour in growth oscillations

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Abstract. A very large class of kinetic growth processes manifest oscillatory behaviour in a variety of physical quantities such as the propagation of the growing interface and the density of the resulting cluster. We demonstrate that these oscillations should generically display quasiperiodic behaviour. Using a formalism based on the projection method for quasicrystalline spectra, we elucidate general features such as the dominant frequency and amplitude of the oscillations. We also briefly discuss the effects of interfacial roughening on the spectra of the oscillations.

It has recently been shown [1,2] that kinetic growth processes should generically manifest oscillatory behaviour in a variety of physical quantities. These oscillations are expected in growth processes which satisfy the following two conditions: (i) the growth proceeds by the accretion of discrete particles or lumps of material of finite size and (ii) the growth takes place at a fairly well defined interface. The oscillations are the result of a heating between two length scales. The first is a statically defined length scale characterising the size of the accreting material and the second is a dynamically induced length scale; for example, the distance which the interface moves per time interval.

In this letter we demonstrate that such oscillations are likely to have a quasiperiodic character. This quasiperiodicity is the result of the fact that the static and dynamic length scales characterising the system may be incommensurate. Consequently, the Fourier transform of various physical quantities will consist of a complex set of delta-function peaks, as we expect for a quasiperiodic system. This quasiperiodicity is most easily seen using a version of the projection method developed for the study of quasicrystalline spectra [3]. In addition to elucidating the general quasiperiodic nature of growth oscillations, this formalism allows us to understand the dominant frequency of the oscillations, estimate its magnitude and gain insight into the effects of interfacial roughening.

We will first show that the spectra of the density and of the average position of the interface as a function of time generically display quasiperiodic behaviour. Then we will present a brief discussion of the limitations and implications of our results.

Let us consider a process in which growth proceeds by the addition of discrete particles of material of fixed size. For simplicity we assume also that growth events occur at well defined discrete times. This condition can be considerably relaxed without destroying the oscillations. In a realistic system, growth events occur at well defined moments in continuous time. If the distribution in times between growth events is relatively narrow, or if the observation time is coarsened, growth oscillations will still

be observed. For our present purposes, it is convenient simply to suppose that time is discrete. Similarly, the important effects of the finite size of the aggregating particles can be most simply captured by supposing that the particles can only occupy the sites of a regular lattice. The lattice spacing then plays the role of the particle size. Complications arising from growth of finite-size particles in continuous space are discussed in [1, 2].

The growth of our cluster can be expressed in terms of a quantity, $P_n(x)$, which is the probability for a particle to be deposited at position x at time n . Depending on the problem, x may be a vector or a scalar. For our purposes it is easiest to think of x as the height above some substrate. Generalisations to more complicated geometries will be clear.

Although $P_n(x)$ is more intimately linked with the underlying microscopic growth law, it is not a prominent observable associated with a macroscopic growing cluster, such as a moving interface. Of course we could try to start with a rigorous definition of the interface (a controversial task even in statics) and relate its profile at time n to $P_n(x)$. Here we argue that, heuristically, $P_n(x)$ is the probability for finding the interface at height x at time n and defer rigour to a later publication. Thus, we consider two 'macroscopic' quantities associated with the cluster: the density, $\rho(x)$, and the average position of the interface at time n , $\bar{x}(n)$:

$$\rho(x) \equiv \sum_n P_n(x) \quad (1)$$

$$\bar{x}(n) \equiv \sum_x x P_n(x). \quad (2)$$

Note that, being a probability distribution in x , $\sum_x P_n(x)$ is normalised to 1, for all n . On the other hand, 'holes' deep inside a cluster can never be filled so that $\rho(x)$ is not unity. The rest of this letter is concerned with the quasiperiodic behaviour in the two functions, $\rho(x)$ and $\bar{x}(n)$.

Because we suppose that our growth is on a lattice and time is discrete, x and n are both integers. We are interested in exploring the consequence of this discreteness. To do this we postulate a smooth interpolating function $f(y, t)$ such that

$$P_n(x) = f(y, t)|_{y=x, t=n}. \quad (3)$$

Furthermore, we will suppose for the moment that

$$f(y, t) = g(y - vt) \quad (4)$$

where v is a constant which we identify as the average growth velocity of the interface in units of lattice spacings per time step. Equation (4) embodies our assumption that the growth process reaches a steady state after some initial transience. We suppose a growth process in which the average interfacial shape does not change, and in which the average growth velocity is time independent. Specifically, if v is irrational, $g(y - vt)$ may be considered to be a limiting function representing the superposition of points, $P_n(x)$, for various times. If v is rational then g will be an interpolating function between a finite set of points. The precise form of the interpolation will not affect our results, for rational v . In assumption (4) we have not included any effects of the spreading of the interface due to roughening. Most likely an essential feature in realistic models, it will be addressed below.

For generic aggregation processes, we expect our assumption of simple average steady-state growth as expressed in (4) to be only approximate. Nevertheless, it is

clearly a reasonable assumption. Furthermore, its validity, at least as a semiquantitative guide to the effects of discreteness, is well supported by our study of several stochastic and deterministic models [2, 4].

To explore the consequences of (3) and (4), we first express ρ and x in terms of f and then impose (4). Thus, $\rho(x) = \int dt f(x, t) \sum_n \delta(n - t)$ and $\bar{x}(n) = \int dy y f(y, n) \sum_x \delta(x - y)$. Note that $\bar{x}(n)$ need not be an integer.

This representation can be summarised graphically in figure 1. On a (y, t) plane, the dots indicate the integer values (x, n) of (y, t) . The line is just $y = vt$, and so has a slope of v . Along the y axis we have drawn $f(y, t_0)$ as a function of y for one value of $t = t_0$. If equation (4) is satisfied, the growth process is described by sliding the curve $f(y, t_0) \equiv g(y - vt_0)$ along the line $y = vt$. Each point (x, n) is then assigned the value $P_n(x) \equiv f(y = x, t = n)$, and the appropriate summations are then carried out. (If (4) is not obeyed, then this heuristic picture requires some modifications which involve, in the main, extracting a suitably defined average growth velocity to replace the parameter v in (4). See [4] for an example.)

This construction is just a version of the projection method used in the study of quasicrystals. If v is irrational (and there is no *a priori* reason to suppose that it cannot be) then the quantities deduced from figure 1 will display quasiperiodic behaviour. Since there are many more irrational than rational numbers in $[0, 1]$, we expect that v will usually be irrational unless the dynamics of growth in a given system prevents irrational average velocities. Generically, therefore, we expect to see quasiperiodic oscillatory structure in the time and space dependence of physical quantities associated with kinetic growth.

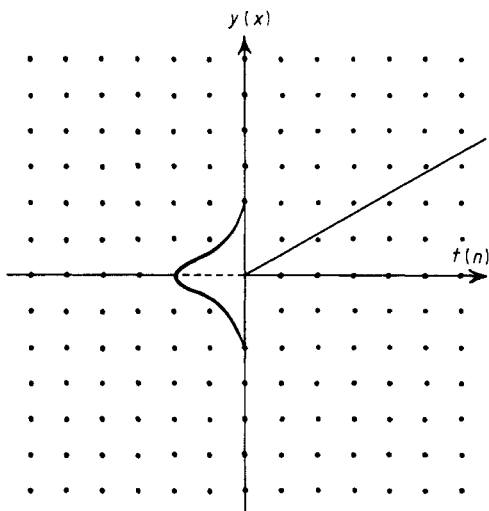


Figure 1. Representation of the projection method applied to growth oscillations.

Let us now look in more detail at the Fourier spectra of $\rho(x)$ and $\bar{x}(n)$. Using (1, 3), we first study

$$\tilde{\rho}(k) = \sum_x \exp(i2\pi kx) \rho(x) = \sum_{x,n} \int dy dt f(y, t) \exp(i2\pi ky) \delta(y - x) \delta(t - n). \tag{5}$$

Note that we define the Fourier transform with a factor of 2π in the exponent. Defining a square lattice of delta functions, $L(y, t) = \sum_{x,n} \delta(y-x)\delta(t-n)$, we find its Fourier transform to be $\tilde{L}(k, \omega) = \sum_{j,l} \delta(k-j)\delta(\omega-l)$. Writing the Fourier transform of f as \tilde{f} , we obtain

$$\tilde{\rho}(k) = \int d\alpha d\beta \tilde{f}(\alpha, \beta) \tilde{L}(k-\alpha, -\beta). \tag{6}$$

If $f(y, t)$ is of the form of (4), then $\tilde{f}(\alpha, \beta) = \tilde{g}(\alpha)\delta(\alpha v + \beta)$ and $\tilde{\rho}$ becomes

$$\tilde{\rho}(k) = \int d\gamma \tilde{g}(\gamma) \tilde{L}(k-\gamma, -\gamma v) = \sum_{j,l} \tilde{g}(l/v) \delta(k-j-l/v). \tag{7}$$

For irrational v , (7) shows a dense point spectrum with varying weights, which is qualitatively the feature of the spectra obtained in [4]. This is the quasiperiodic behaviour we seek.

Similarly we can examine the Fourier transform of $\bar{x}(n)$ which carries information about the time dependence of the interface. Aside from its oscillatory structure, $\bar{x}(n)$ will generally have a term proportional to n , which merely expresses the average growth of the interface. To simplify the analysis, it is helpful to subtract this term. If $f(y, t)$ is of the form of (4), then we may define

$$H(n) = \bar{x}(n) - vn. \tag{8}$$

To find its spectrum we follow the procedure used for $\rho(x)$ and obtain

$$\tilde{H}(\omega) = \int d\gamma \partial_\gamma \tilde{g}(\gamma) \tilde{L}(-\gamma, \omega - v\gamma). \tag{9}$$

Clearly, (9) is a dense point spectrum as (7) is.

To better understand the implications of these expressions for the spectra, it is worthwhile examining one of them in a little more detail. Consider, for specificity, $\tilde{\rho}(k)$ given by (7). Assuming that $g(x)$ in (4) is smooth, then $\tilde{g}(\gamma)$ should be a rapidly decreasing even function of γ so that the largest single contribution to $\tilde{\rho}(k)$ will come from $|l|=1$. Moreover, since $\rho(x)$ is real and the x are integers, all the information about the spectrum is contained in $\frac{1}{2}(\rho(k) + \rho(-k))$ for $0 \leq k < \frac{1}{2}$. In this interval, the largest delta-function peak occurs (for $j = -l = \pm 1$) at $k = (1-v)/v$ which corresponds to a wavelength of $v/(1-v)$ lattice spacings. This is just the fundamental period of the density oscillations deduced elsewhere by a more intuitive, physical argument [1, 2].

So far most of our discussion has concerned processes in which $f(y, t) = g(y - vt)$. But in real growth processes, there is typically some spreading of the interface due to roughness. To understand, qualitatively, what effect this has on the power spectra, we can consider a situation in which the interfacial width grows slowly with time. One way to introduce such a small dispersion is to let \tilde{f} be of the form $\tilde{f}(\alpha, \beta) = \tilde{g}(\alpha)\delta(\alpha v + \beta + \epsilon\alpha^2 v^2)$, where ϵ is a small parameter.

One important effect of this dispersion is to split delta-function peaks in the spectra. We can see this, for example, in (the symmetric part) of $\tilde{\rho}$. With the non-dispersive interface the delta-function peaks corresponding to (j, l) and $(-j, -l)$ are degenerate, as in (7). With dispersion, this degeneracy is split. In particular, the fundamental peak in $\rho(k)$ corresponding to $j = -l = \pm 1$ splits into two peaks at k values of

$$k_{\pm} = (1-v)/v \pm \epsilon. \tag{10}$$

Similar splitting will occur for other values of j and l , and will also appear in quantities like \bar{x} and \tilde{H} . Because the interface is generically rough and widening, such splitting

should be a common occurrence in these power spectra. Indeed, this peak doubling has been observed in computer simulations of ballistic aggregation [4].

Finally, we address the question of normalisation of $P_n(x)$:

$$\sum_x P_n(x) = 1 \quad \text{for all } n. \quad (11)$$

This condition places non-trivial constraints on $f(y, t)$. For example, functions of the form (4) are heuristically appealing. But even apart from the lack of dispersion, they are generally too simple since, if v is irrational, no simple g can be chosen which satisfies (11). To see this, we consider the Fourier transform of (11) and follow the procedure outlined for ρ and \bar{x} . The result is $\sum_{j,l} \tilde{g}(j) \delta(\omega + jv - l) = 0$ for $0 < \omega < \frac{1}{2}$. If v is irrational, the δ are linearly independent in that, for distinct (j, l) , the δ have support on distinct ω . So, $\tilde{g}(j) = 0$ for all $j \neq 0$, forcing g to be a constant.

This 'no-go' theorem is, however, easily avoided in a realistic growth process. First, we have indicated the need to include broadening of the interfacial width, leading to dispersion in f . At a more fundamental level, for finite systems evolving in finite times, Fourier transforms of various quantities contain peaks with finite widths rather than delta-function peaks. The no-go argument given above is therefore inapplicable. There is, of course, a constraint analogous to (11) even in finite systems, but its consequences are not as simple (and probably not as severe). A more detailed analysis of finite systems is clearly desirable. Furthermore, with finite space and time, the distinction between rational and irrational v is, strictly speaking, moot, so that the appropriate normalisation constraint can be satisfied with a non-trivial g . At the same time, the power spectra of the other physical quantities will still resemble quasiperiodic ones.

Regardless of the detailed way in which the no-go theorem is evaded, the important point is that the normalisation condition does not rigorously preclude quasiperiodic behaviour in other quantities. As a *mathematical* example, consider $f(y, t) = g(y - vt) + \eta(t)$, where $\eta(t)$ is *chosen* to satisfy the normalisation condition. Since η is independent of x , $\tilde{\rho}(k)$ will have the same quasiperiodic structure as before.

In this letter we have shown that growth oscillations should, generically, exhibit quasiperiodic behaviour. This quasiperiodicity results from an incommensuration between a static and dynamic length scale. Using a projection method developed in the study of quasicrystals [3], we have shown how to relate the shape of the interface and the growth velocity to various features of the spectra of physical quantities, such as the density of the cluster. In particular, we have been able to derive the fundamental frequency of the oscillations, and have shown that the roughening of the growing interface results in split peaks in the spectrum. If the dynamics of a particular growth process prevents the static and dynamic length scales from being incommensurate, the spectrum of growth oscillations will collapse into one characteristic of a normal multiperiodic structure. In either case, the techniques we have presented will be very useful in understanding the spectrum of the oscillations.

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References

- [1] Cheng Z and Savit R 1986 *J. Phys. A: Math. Gen.* **19** L973
- [2] Baiod R, Cheng Z and Savit R 1986 *Phys. Rev. B* **34** 7764
- Cheng Z, Baiod R and Savit R 1987 *Phys. Rev. A* **35** 313

- [3] de Bruijn H G 1981 *Ned. Akad. Wet. Prod. A* **43** 39, 53
Zia R K P and Dallas W 1985 *J. Phys. A: Math. Gen.* **18** L341
Elser V 1986 *Acta Crystallogr. A* **42** 36
Duneau M and Katz A 1986 *J. Physique* **47** 181
- [4] Cheng Z, Jacobs L, Kessler D and Savit R 1987 *Growth Oscillations in Ballistic Aggregation. Preprint*
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