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Patterns and Disorder in Fractal Growth Processes

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Many naturally occurring objects are random fractals, that is, scale-invariant random structures. In recent years a series of simple models have been developed which show how fractal structure can occur when rapid (non-equilibrium) growth is involved. The simplest of these models, which is called diffusion-limited aggregation, also seems to describe certain types of orderly growth, e.g., those involved in the formation of snow-flakes. The relation between these two types of pattern will be discussed.

INTRODUCTION

The coexistence of order with disorder, chaos with patterns, simplicity with complexity, pervades nature. In this report I will try to give a theoretical physicist's view of how some of these ubiquitious features of the world arise, and, in particular, review some recent work on the generation of both patterns and disorder in simple physical systems. Some of the elementary features of the model systems we study may turn out to be generic for the production of form; we can hope that some contribution to the emerging science of form will result.

Our focus will be on random-growth processes. It turns out that the most interesting cases correspond to growth far from equilibrium. We will try to discover when there is order, and when disorder, and how complexity arises. For the disordered case, we will find that there sometimes occurs a new and unsuspected symmetry called fractal symmetry (Mandelbrot: 1982). And we will say a bit about transitions between the various growth regimes.

GROWTH PROCESSES AND REGIMES OF GROWTH

The simplest type of growth process which we can imagine involves the production of a large cluster (an aggregate) by the irreversible addition of subunits from outside. In what follows we will suppose that the units are tiny spheres (particles) which stick together by short-range forces.

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One such model was introduced in the context of cell colony growth (Eden: 1961). In this process particles are added one at a time, at random, to any unoccupied surface site of the cluster. The resulting aggregate is a featureless blob, as one might expect. The remarkable thing is that the next two processes which we will discuss which do not seem very different give an entirely different result, and may thus give some clues about the genesis of order and pattern in nature.

A very interesting model called ballistic aggregation is intended to represent processes that contribute to the growth of thin films from a low-density vapor (Leamy, et al.: 1980). In this case particles rain in a parallel stream onto a surface where they stick to the surface or to each other. If the rain is perpendicular to the surface, we get an amorphous deposit, but for large angles of incidence a persistent pattern of parallel streaks (called the columnar microstructure) appears which greatly influence the properties of the film. A theory for this phenomenon has been given by Ramanlal and Sander (1985).

If the particles "wander", i.e., randomly diffuse, to the surface (or to a nucleation center) where they stick to each other on contact we have a process which has become known as diffusion-limited aggregation (DLA). This process describes diffusion-limited crystal growth such in the electrodeposition of a metal (Brady and Ball: 1984); Matsushita, et al.: 1984). In Figure 1 we compare a computer simulation of the DLA model (left) with an electrodeposit of zinc produced in my laboratory by D. Grier (middle. See Grier, et al. (1985).

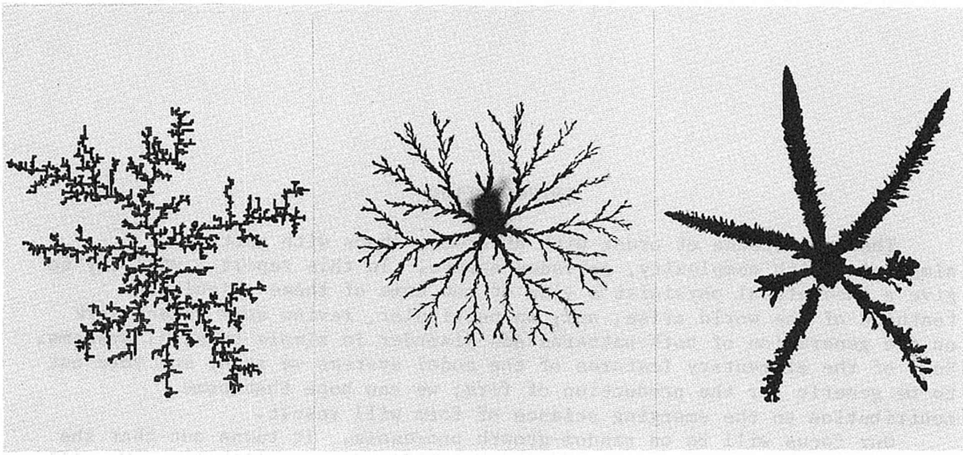

FIG. 1.

There are several remarkable aspects of these patterns. One is their open sprawling structure. They are not merely amorphous, but <u>fractal</u> (see below). Also, the metal does not have the overall structure of a typical non-equilibrium dendritic crystal, of which snowflakes are a well-known, beautiful example. On the right side of the figure we show a dendrite which was produced in the <u>same</u> cell as the other deposit by slightly raising the voltage. We believe that this change enhances the effect of crystalline anisotropy. An qualitative change in morphology results. The lesson to be learned from this example (which we will discuss further, below) is that diffusion-limited growth plus weak external anisotropy can yield an astonishing variety of complex patterns.

This example falls into a general framework of discussion of non-equilibrium growth. Very often we find three regimes: near-equilibrium; disorderly, which may be merely amorphous, but which can also be fractal; and patterned, with complex shapes reflecting some underlying symmetry.

FRACTAL SUMMARY

Fractals have received a large amount of attention in recent years because it seems that many natural objects are of this type (Mandelbrot: 1982). They are relevant to our subject because DLA and many similar processes produces fractal clusters; this may account for some of the observations, as we will see below.

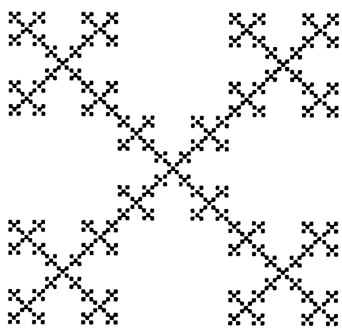

FIG. 2

Briefly, we may define a fractal as an object with an anomalous kind of scaling symmetry. Figure 2 (a Vicsek snowflake) is an example. Note that every growth stage for the figure (by a factor of 3 in radius) multiplies the number of units by 5. This is more than we would expect for a linear pattern (we would expect 3) and less than for a filled area (we would get 3x3=9). This relationship is expressed in the fractal dimension (this version is called the mass dimension):

$$N = cR^D \ . \qquad (1)$$

For the Vicsek snowflake $D=\ln(5)/\ln(3)=1.46$. The fractal dimension need not be integer. It is a very useful characterization of the overall scaling symmetry of an object. DLA in two dimensions of space has $D=1.71$; for three dimensions D is 2.4.

DIFFUSION-LIMITED PROCESSES IN NATURE

In this section we will describe how complex patterns and fractals are generated in diffusion-limited growth, and give some physical examples to demonstrate how a common mechanism can operate in a wide range of systems.

The essential feature of these systems seems to be that growth instabilities underlie the formation of complexity. This is the mechanism for amplifying either initially asymmetric form or external noise in order to generate complex shapes. For the case of diffusion this instability (known as the Mullins-Sekerka instability, Mullins and Sekerka (1963)) amounts to pointing out that a wandering particle is more likely to encounter a protruding tip of the aggregate and to stick there than it is to explore the rest of the surface. Thus, tips grow even larger as the particles accrete. It is remarkable that the same effect operates in other, very different systems in a way that may be shown to be mathematically identical.

For example, when an inviscid fluid is pumped into a viscous fluid (the viscous fingering problem, see Paterson (1984)) it is easier for the viscous fluid to flow away from protruding tips than from flat surfaces.

Thus tips grow and a complex interfacial pattern results which looks like the patterns pictured above.

Latent heat flow away from a solidifying crystal also has the property of being more efficiently dissipated by tips. The tips grow sharper until controlled by surface tension. Thus there is a direct link between DLA and this kind of solidification (Witten and Sander: 1981).

Even more remarkable is the fact that electrostatic systems have the same sort of property. The tip of a grounded conductor in an electric field is more accessible to field lines than a hole or a flat surface. Thus field lines concentrate at tips; this is how a lightning rod works. If the growth of a portion of surface is proportional to the field there, then an open branched structure will result. Lightning is the most familiar example. Dielectric breakdowns in solids have been analyzed in this fashion; they seem to have the fractal dimension of DLA (Niemeyer, et al.: 1984). In fact, if the rate-limiting step in electrochemical deposition is not diffusion but inhomogenities in the conduction current flow, this is the important instability. The patterns of Figure 1 probably result from this effect and not from diffusion (Grier, et al.: 1985).

DISORDER AND PATTERN

Growth instabilities have another surprising effect in the case of diffusion-limited growth. They allow a macroscopic expression of microscopic symmetries. For example, in the case of snowflakes tiny anisotropies in surface energy and small differences in the sticking rate of incoming molecules on different crystal faces are amplified in a complex way. These effects are among the most difficult problems in mathematical physics, but it does seem clear that the sensitivity of unstable growth to small perturbations is essential to the form of such dendrites.

Very recently, some work has been undertaken to see exactly what conditions lead to patterned growth and what to disorderly, fractal patterns. The remarkable result is that there can be a sharp morphological transition in the growth depending on the exact parameters of the system. Qualitatively, as a fractal pattern grows, tips form, split, and reform. The proliferation of splittings and tip-interactions give rise to the fractal. Slightly more anisotropy can stabilize the tip growth direction. Then the dendrite has its major growth in a single direction, though it can shed side-branches as it develops (cf. Figure 1).

A very beautiful demonstration of this effect was given by Ben-Jacob, et.al. (1985) in a modified version of the viscous fingering experiment described above. Glycerine was confined between plates and air was introduced in the center. The interface was a disorderly structure of branches showing tip-splitting, as before. But one of the plates in this version of the apparatus was inscribed with a lattice of shallow grooves. As the pressure increased, the effect of the anisotropy increased and tip-splitting was supressed. A pattern looking very like a snowflake was produced, see Figure 3.

The recent experiment of Grier, et al. (1985) was an attempt to explore the same sort of transition. Once more, as a function of voltage and electrolyte concentration we were able to tune from disordered to ordered patterns (cf. Figure 1).

SUMMARY

This paper reports a quick look at a tiny subset of the processes that produce the complex forms that we see in the world around us.

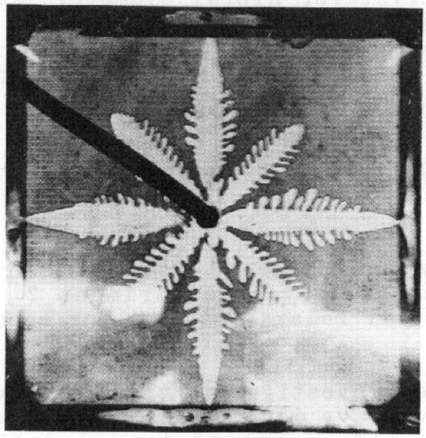

FIG. 3

Perhaps the isolation, in these simple cases, of a few organizing principles will inspire a more comprehensive treatment.

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Q: In growing crystals, global symmetry may be understood to come from microscopic crystaline structure (e.g. the six-fold symmetry of ice crystals). In the Hele-Shaw experiment (fingering in a viscous fluid) there is no such microscopic structure due to an underlying lattice. Why does your experiment show an approximate six-fold symmetry ? (N. Packard)

A: We imposed the symmetry by inscribing grooves on one of the plates of our cell.

Q: Is there any dependence of transition pressure on the speed of changing pressure in your experiment ? My guess is there might be some special relaxation time for each special pattern. (T. Haseda)

A: We have never investigated this question. In fact, in all the experiments we have done, we have taken care to ensure that the changes were slow compared to relaxation times. In the analogous problem of electrodeposition of zinc, we have looked at AC electrical characteristics. The relaxation times are quite short.

Q: You mentioned about 4,000,000 particle aggregation, which is the largest DLA pattern I've ever heard of. What happened, I wonder ? (M. Matsushita)

A: The very large simulations were done by P. Meakin and R. Ball in order to see whether lattice effects become important at long times. The preliminary result is that on a square lattice, anisotropy does begin to dominate and may eventually destroy the fractal for this case.

C: The relevance of the Hele-Shaw experiments reported by Dr. Sander to the current problems of icosahedral growth is very clear. As well as using an anisotropic environment of five-fold symmetry, there are possibilities of changing the metric of the space of the liquid into which air is injected so that the perimeter of the region changes to some other power of the distance than linearity.

Dr. Sander replied that experiments with the Penrose tiling were already being done but that it would also be interesting to use, for example, a hyperbolic film. (A. Mackay)

C: I'd like to make a comment on the snow crystal. In principle, there are two processes relevant to growth of snow crystals. One is the diffusion process and the other is the surface kinetic process for incorporating water molecules into the crystal lattice. (T. Funakubo)