

# Fractal growth processes

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*The methods of fractal geometry allow the classification of non-equilibrium growth processes according to their scaling properties. This classification and computer simulations give insight into a great variety of complex structures.*

ALMOST every theoretical tool of the condensed-matter scientist uses the assumption that the system considered is of high symmetry and is in equilibrium. These assumptions have led to enormous progress; however, to much, if not most, of the natural world such tools cannot be applied. Many systems that we would like to understand are very far indeed from perfectly ordered symmetry and are not even in local equilibrium. Perhaps the most extreme example is disorderly irreversible growth. We mean by this the sort of process which is very familiar in the formation of dust, soot, colloids, cell colonies and many other examples; roughly speaking, things often stick together and do not become unstuck. For example, a particle of soot grows by adding bits of carbon and coagulating with other particles in a random way. A possible result is shown in Fig. 1. We are thinking about cases which are, in some sense, as far from equilibrium as possible, and which have no obvious order.

It is remarkable that the introduction of simplified models has led to quite a good understanding of the morphology of such growth, despite the inapplicability of our usual modes of thinking. Here I will discuss this progress, drawing examples mostly from subjects which have traditionally interested physicists and chemists. However, disorderly growth is ubiquitous in the world around us, and is certainly not limited to inanimate matter. For example, some of the ideas which I will discuss, such as anomalous scaling in kinetic processes, will be useful to biologists. The purpose of the review is to introduce ideas from the area which may be of general use.

The key to our recent progress is the recognition that the most 'interesting' non-equilibrium structures (say, from a visual point of view) are not merely amorphous blobs; they still have a symmetry, despite their random growth habit, albeit a different one than they might have had, had they grown near equilibrium. For example, consider the soot of Fig. 1, or the electrolytic deposit of zinc shown in Fig. 2. Many people will be familiar with branched deposits such as this, and with similar looking objects which form on automobile windshields on cold mornings. In all these cases the structure is disordered, but it is not random. A manifestation of this is that each section of the picture contains holes in the structure comparable in size with that of the section itself. This can only occur if there are long-range correlations in the pattern; particles 'know' about each other over distances far in excess of the range of the forces between them. A truly random pattern, such as that of salt scattered on a table top, shows no such scaling of holes, and correlations are of short range only.

Studies of fractal growth have focused on two questions: how can we characterize and quantify the hidden order in complex patterns of this type, and when and how do such correlations arise? The answer to the first question is now relatively clear, and lies in an application of the fractal geometry of Mandelbrot<sup>1</sup>. The next section gives a brief review of relevant aspects of this subject. The second question has received a partial answer in the formulation and analysis of models suitable for computer

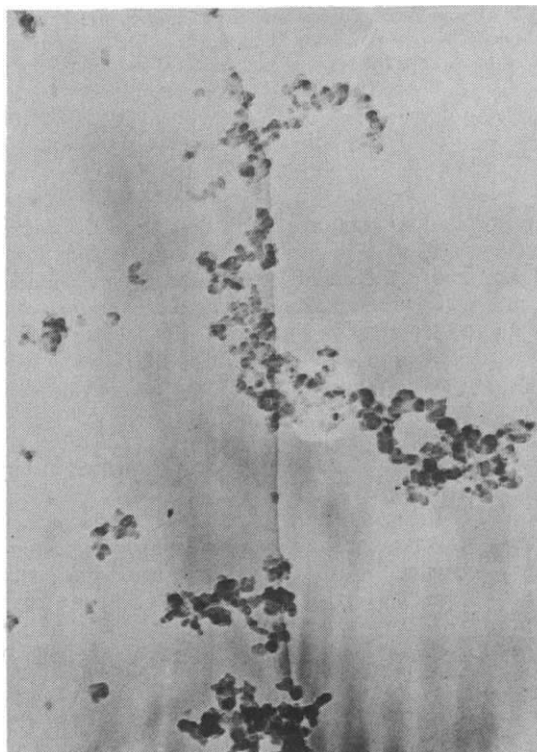


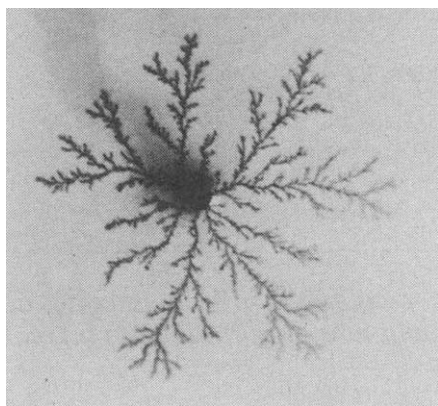
Fig. 1 Electron micrograph of soot. (Supplied by G. Smith, General Motors.)

simulation, which will also be reviewed. For more extensive treatments see refs 2-4.

## Fractals and scale invariance

In pure mathematics, it has long been common to study certain 'pathological' geometric shapes that elude ordinary notions such as those of length and area. Figure 3 shows a famous example, which has, in some sense, infinite length, but zero area. It falls between our usual notions of line and solid. Mandelbrot<sup>1</sup> systematized and organized mathematical ideas concerning such objects due to Hausdorff, Besicovitch and others. But, more importantly, he pointed out that such patterns share a central property with complex natural objects such as trees, coastlines, patterns of stars and (as was later discovered) the non-equilibrium growths of Figs 1 and 2. This property is a symmetry which may be called scale invariance. These objects are invariant under a transformation which replaces a small part by a bigger part, that is, under a change in scale of the picture. Scale-invariant structures are called fractals.

There are a number of related properties which follow from the assumption of scale invariance. Consider, for example, the



**Fig. 2** Zinc electrodeposit produced in a thin cell under conditions of low  $\text{ZnSO}_4$  concentration ( $0.01 \text{ mol l}^{-1}$ ). The outer electrode (not shown) is in the form of a ring 6.3 cm in radius. (Supplied by D. Grier, University of Michigan.)

density correlation function  $c(r)$ , of a fractal. This is defined as the average density of the object at distance  $r$  from a point on the object, and is a measure of the average environment of a particle. Clearly,  $c(r)$  must reflect the scale invariance. It is easy to show that the only way that  $c$  may vary is as a power law in  $r$ ; any other function would have an intrinsic scale. It is convenient to write  $c$  in the following form:

$$c(r) = kr^{-(d-D)} \quad (1)$$

Here,  $k$  is a constant, and the exponent is written in terms of the dimension of space,  $d$ , and a new quantity,  $D$ , the fractal dimension. The reason for this terminology will become evident in a moment. As the objects we are dealing with are tenuous,  $c(r)$  is a decreasing function of  $r$ : the average density decreases as the object becomes larger. Now consider how the total mass of the object,  $M$ , scales with the mean radius,  $R$ . We can estimate this by multiplying a typical density, from above, by the volume:

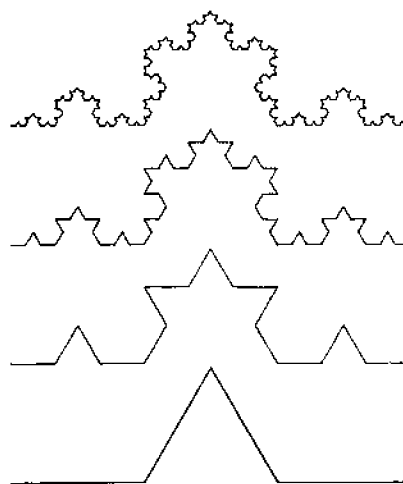
$$M(R) = KR^{D-d}R^d = KR^D \quad (2)$$

Here,  $K$  is another constant. We can now see why  $D$  is called a dimension. For an ordinary curve,  $D=1$ : twice the length gives twice the mass. For a disk,  $D=2$ . For simple objects  $D$  coincides with the usual notion of dimension. But in the cases we are discussing  $D$  is not an integer; it has been measured to be  $\sim 1.7$  for the deposit in Fig. 2, and is 1.26 for the fractal of Fig. 3.

This anomalous scaling with radius, measured by  $D$ , is a very useful means of characterization because the fractal dimension is a 'robust' quantity. Like the famous scaling exponents of phase-transition physics, it has to do with long-range properties, indeed, with the relationship between properties at different scales. Thus we can expect it to be universal in the sense that it should be independent of the details of the interactions between the objects which stick together during the growth, of their detailed composition, and so forth. But, as we will see, the mechanism of growth does affect  $D$ .

## Growth models

How might one visualize the growth of an object such as the electrolytic deposit in Fig. 2? As we are interested in long-range properties we can ignore the complications of electrochemistry and simply imagine that ions wander randomly in solution (in many cases the electric field is screened out so that this is a good approximation) and stick to the deposit when they happen to get near it.



**Fig. 3** Four stages in the growth of an exact fractal, the Koch curve. This and many other examples are discussed in ref. 1. The fractal dimension may be deduced by thinking of each picture as a part of the picture above, with a change of scale. For each scale change by three, we need four such parts. Thus, according to equation (2),  $D = \log 4 / \log 3 = 1.26$ .

To make a computer model which is a literal translation of this process we start with a centre. Then we liberate a diffusing particle, a 'random walker', and let it wander freely until it is within a fixed distance of the centre, where it sticks. Then we liberate another particle and let it walk until it sticks to the centre or the first particle, and so on. We may, for our purposes here, idealize the process of formation as being completely irreversible: we ignore the possibility that the particles rearrange after sticking to find a more energetically favourable location. This is the diffusion-limited aggregation (DLA) model of Witten and Sander<sup>5,6</sup>. The application of DLA to electrodeposition is due to Brady and Ball<sup>7</sup> and Matsushita *et al.*<sup>8</sup>.

Figure 4 shows the result of an extensive simulation according to the DLA rules; its resemblance to Fig. 2 is evident. Measurements of DLA clusters have shown them to scale according to the relations quoted above, with  $D=1.7$  for  $d=2$ , and  $D=2.4$  for  $d=3$ . Note that the structure is tenuous and open because holes are formed and not filled up. Filling up the holes would require wandering down one of the channels in the cluster without getting stuck on the sides; a random walker cannot do this.

There are several features of the DLA model which should be mentioned. Although it is simple to describe, no progress has been made towards 'solving' it. That is, although we suspect, on the basis of simulations, that DLA clusters are fractals, we cannot prove it. And we have no method of calculating  $D$  (or any other property): we must measure it. There are several reasons for this (I will mention a rather technical one below), the primary one being that DLA presents us with a situation in which our experience in equilibrium systems doesn't seem to help. Note that  $D$ , along with other scaling properties, arises in a non-trivial way from the kinetics of growth: there is no simple geometric argument with which to predict them.

The DLA model can be generalized in various ways, for example, to describe deposition on a surface<sup>9</sup> rather than a point. A more profound generalization is to use the model to describe systems which apparently have nothing to do with particle aggregation, but which share the same universal properties. We may see how one is led to do this by observing<sup>5,10</sup> that the probability,  $u$ , of finding a random walker at some point on its way to the aggregate has the following well-known properties: the flux of walkers,  $v$ , is proportional to the gradient of  $u$ , and, because walkers are absorbed only on the aggregate, this flux

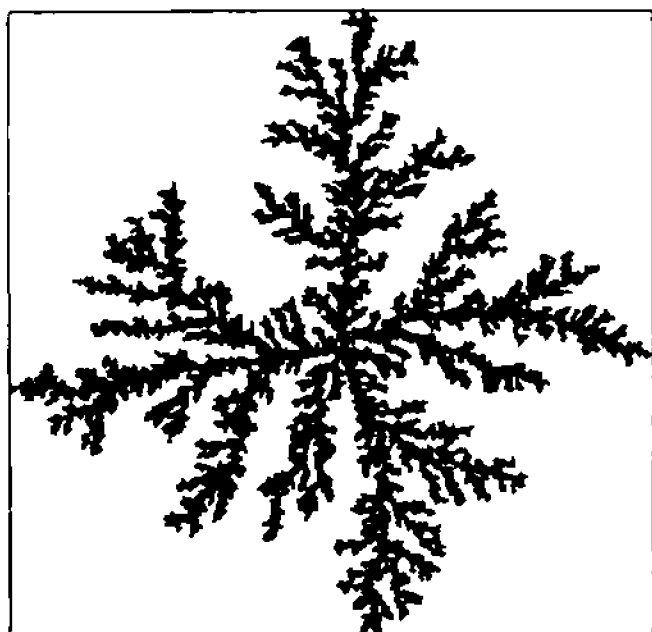


Fig. 4 A large DLA cluster ( $\sim 50,000$  particles) grown on a square lattice. Note the resemblance to Fig. 2, and the beginning of distortion towards a dendritic outline, as discussed in the text. (Supplied by P. Meakin, Dupont.)

has no divergence:

$$v \propto \nabla u \quad (3)$$

$$\nabla \cdot v = \nabla^2 u = 0 \quad (4)$$

As walkers are not allowed to escape from the aggregate, we set  $u = 0$  on the surface. The growth of the aggregate is given by the flux at its surface, that is, by  $\nabla u$ .

As Niemeyer *et al.*<sup>10</sup> pointed out, a set of equations of identical form govern dielectric breakdown of a solid if we ignore many short-range details. As we are looking for universal features, making such simple, indeed, crude approximations is justified. If we think of  $u$  as the electrostatic potential in a solid about to be destroyed by a discharge, its negative gradient is, of course, the electric field. But  $u$  then obeys the Laplace equation of electrostatics, which is of the same form as the steady-state diffusion equation, equation (4), above. The breakdown channel will grow in a way determined by the electric field, that is, the gradient of  $u$ , on its surface. If the growth rate is linear in the field, we expect to have exactly the same situation as in DLA, and indeed, direct solutions of the equations, as well as measurements of photographs of real discharges, give the same fractal dimension as DLA. Non-linear breakdowns (lightning in the atmosphere is probably an example) give rise to patterns with different values of  $D$ .

Paterson<sup>11</sup> noticed an even more remarkable manifestation of the wide applicability of the model. When a fluid flows under conditions of large friction, inertial effects are negligible and the flow rate can be taken to be proportional to the hydrostatic force, that is, to the gradient of the pressure: this is known as D'Arcy's law. The situation is commonly realized in the laboratory by letting fluid flow between thinly spaced plates, a so-called Hele-Shaw cell. In nature, the flow of crude oil through the porous rock in which it is found is an example of quite serious interest. Suppose we try to force such flow by blowing a bubble of air or another low-viscosity substance into the cell (or by pumping water into an oil field—a scheme known as enhanced recovery). It has long been known that the air will not uniformly displace the fluid; instead it will break up into a complex

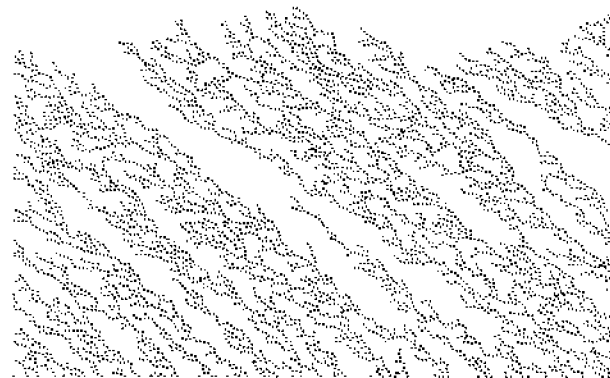


Fig. 5 Columnar microstructure in ballistic aggregation. Particles stick to the substrate and to each other after raining onto the structure in parallel trajectories at an angle to the vertical somewhat smaller than that of the columns. The fluctuations of the upper surface scale with the height for small height, and with the total width for large height. (Simulation performed by P. Ramantlal, University of Michigan.)

structure with many arms<sup>12</sup>, which are called 'viscous fingers'. This phenomenon has an obvious detrimental effect on enhanced recovery.

Paterson's<sup>11</sup> speculation was that the pattern of the viscous fingering would scale like DLA. His reasoning was as above: the pressure in an incompressible fluid obeys equation (4), with  $u$  now standing for pressure, because fluid, like particles, is conserved. D'Arcy's law is of the same form as equation (3). Once more, many details have been ignored. In particular, the role of surface tension in this and similar situations will be discussed below.

The reasoning has been verified most directly by Chen and Wilkinson<sup>13</sup>, who introduced discrete randomness into a Hele-Shaw cell—the effect should be that of the random arrivals of particles. Their patterns look almost exactly like Figs 2 and 4. Another experiment, by Nittmann *et al.*<sup>14</sup>, used the clever trick of eliminating surface effects by taking for the two fluids water and an aqueous polymer solution; the fluids are miscible but mix slowly. Once more the pattern of fingering resembled the simulations. There seems to be a source of randomness in this experiment, probably arising from the non-newtonian flow characteristics of the polymer solution; such shear thinning could amplify noise. Even more startling is the experiment of Ben-Jacob *et al.*<sup>15</sup>, who used a smooth Hele-Shaw cell, and one with a periodic pattern, with newtonian fluids. In some conditions they observed DLA-like scaling without an evident source of randomness, and without discrete 'particles'.

Experts will notice that equations (3) and (4) are of the same form (except for surface effects) as the description of solidification when the limiting factor in growth is diffusion of latent heat away from the surface of the growing crystallite. Why, then, does a snowflake (unlike the crystalline deposit of Fig. 2) not look like DLA, but is instead dominated by the crystal symmetry? I will return to this aspect of growth in the final section.

If particle aggregation doesn't need particles, what does it need? More generally, we can ask what different types of model give rise to scaling objects. For example, it is often the case that aggregates are formed by adding particles with a long mean free path, for example, in the formation of thin films by vapour-phase deposition<sup>16</sup>. In this case we may assume that the paths of the particles are straight lines. This model has become known as ballistic aggregation, and it has a number of very curious features. It is now known that the deposit itself is not a tenuous object but achieves a constant density<sup>17,18</sup>. (In contrast, diffusion-limited growth on a surface<sup>9</sup> yields an open deposit whose average density decreases with height.) It is of great interest to understand the upper surface of the film, which is a

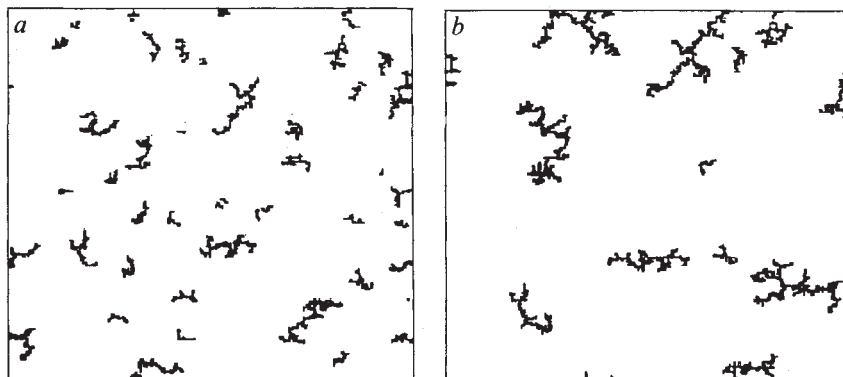


Fig. 6 Two stages in the formation of cluster-cluster aggregates: *a*,  $t=3,669$ ; *b*,  $t=17,409$ . Note the resemblance of the clusters in *b* to the soot particle of Fig. 1. (Supplied by P. Meakin, Dupont.)

model of a random rough surface. It has been shown numerically<sup>19</sup> that for normal incidence of the depositing particles this surface also has scaling properties: for example, the fluctuation of the height scales with a non-integral power of the height, for small height. This surface is probably not an ordinary fractal curve, like Fig. 2, but is probably an example of a self-affine fractal<sup>1,20</sup>. 'Self-affine' means that the scaling in two different directions (width and height in this example) is different.

For non-normal incidence another effect appears, which is well-known in thin-film technology<sup>16</sup>. This is the columnar microstructure: the film spontaneously forms as a set of nearly parallel columns as it grows (see Fig. 5). The beginnings of a theory of this effect exist<sup>21</sup>, but it is not known what, if any, relationship these giant fluctuations have with the scaling fluctuations at normal incidence.

The simplest aggregation process of all was introduced into mathematical biology by Eden<sup>22</sup>. This is a model for the growth of a cell colony: a cluster is grown by adding particles at random to perimeter sites. Once again the object is compact, but the surface has interesting scaling properties which seem to be the same as for ballistic aggregates with normal incidence<sup>23</sup>. Scaling is ubiquitous, and tends to have common features despite widely different details of growth.

We still have not described how soot forms. The structure of Fig. 1 is far more open than a DLA cluster: its fractal dimension is  $\sim 1.8$  (DLA in three dimensions has  $D=2.4$ ). Extensive measurements of soot<sup>24</sup>, colloids<sup>25</sup> and other similar objects leads one to suspect that a different class of clusters is involved. In fact, we have omitted a central feature of the formation process of clusters which can coagulate, namely the aggregation of clusters with each other<sup>26,27</sup>. Figure 6 shows two stages of a simulation of this process in two dimensions. We start with a vapour of freely moving particles which stick together whenever they come into contact, and then allow the clusters to continue to move with, perhaps, a smaller diffusion constant. The large fractals which are eventually formed have  $D=1.4$ . The corresponding simulations in three dimensions give  $D=1.8$  and yield the open structure of real colloids and aerosols. At each stage of the process almost all of the clusters are of roughly the same size.

The open structure and low fractal dimension which characterize cluster-cluster aggregation are relatively easy to understand. It is difficult for a random-walking particle to penetrate a significant fraction of the radius of a growing cluster for particle aggregation; it is even more difficult for an aggregate of comparable size to do so. Thus, as aggregation proceeds, open, fluffy structures are produced.

One variant of this model which is worth mentioning is reaction-limited (chemically limited) aggregation<sup>28</sup>. In many cases, because of the details of the growth process, the sticking is inefficient, and many attempts are required to form a new cluster. In the limit of a very large number of attempts, the fractal dimension increases from 1.8 to  $\sim 2$ . Reaction-limited aggregation was probably discovered experimentally<sup>29</sup>, before the simulations were done. Later experiments<sup>30</sup> have carefully

controlled the growth conditions and shown both growth mechanisms, and both types of geometry, in the same system for different growth rates. The encoding of kinetics in the scaling in a form independent of details should be a powerful tool for identifying growth mechanisms.

### Attempts at theory

There is no general theory of irreversible growth. The descriptions given in the previous section must be regarded as a kind of phenomenology, albeit a useful one. We can point to situations in which there is scaling, but we are compelled to do experiments, either in the laboratory or on the computer to calculate anything. We do have a few analytical results, but they give only partial information.

The best understood type of aggregation is the cluster-cluster process. Suppose we assume, as stated above, that the dominant cluster-cluster collision is between clusters of similar mass. If we make the masses strictly equal we have a hierarchical model<sup>31</sup>. It is easy to believe then that we do have a fractal: agglomerating parts in this way is exactly how the artificial fractal of Fig. 2 was made. (Note that particle aggregation is not hierarchical, but it seems to be fractal nonetheless.) The specification of the size distribution of clusters in the vapour, and the verification of the hierarchical assumption, have been the objects of detailed studies<sup>32</sup> which have shown that, indeed, the most common collision is between clusters of nearly equal mass. Some of these investigations use the techniques of colloid chemistry, in particular the Schmoluchowski kinetic equations, as well as computer simulations.

There remains the problem of finding the geometry of the clusters formed. Some progress has been made here because of a detail which allows one of the favourite tricks of the theoretical physicist to be applied. The real difficulty in visualizing the process is excluded volume, that is, the tendency of clusters to get in each other's way because they can attach to each other only on the outside. If they could attach anywhere it would be much simpler to sort out what is going on. It is quite obvious that excluded volume problems become less serious in high spatial dimensions: there are more ways into a three-dimensional cluster than into a two-dimensional one. Often, in equilibrium studies, it is found that for sufficiently large dimension of space,  $d$ , excluded volume is no problem at all: essentially any part of a cluster is accessible from outside. The dimension at which this starts to happen is called the upper critical dimension.

Above the upper critical dimension, calculations are simple, anomalous scaling is independent of  $d$ , and there exist methods (for equilibrium problems) which allow us to extrapolate to the physical world of  $d=3$ . For cluster-cluster processes, this is exactly what happens<sup>33</sup>. The fractal dimension,  $D$ , for a cluster-cluster aggregate cannot grow above  $\sim 3.4$  and it attains this value at about  $d=7$ . This is rather far from the real world, of course, and no one has yet figured out how to extrapolate.

The situation for particle aggregation is very different. Suppose that the entire cluster were to become accessible to added particles for a large enough value of  $d$ . Then the mass in the

interior would grow without adding to the volume. The cluster would quickly become so dense that it would no longer be accessible. Thus, there is no upper critical dimension for DLA. In fact, careful considerations of this sort can be turned into a bound<sup>33</sup> on  $D$ :

$$d - 1 \leq D \leq d \quad (5)$$

The fractal dimension is never independent of the spatial dimension, and the standard technique cannot be applied.

Some progress has been made in the study of particle aggregation by exploiting the similarity of the process to the famous 'snowflake' problem, that is, the study of dendritic crystallization<sup>34</sup>. We can see, for example, why tenuous structures are likely to arise in DLA and not in ballistic aggregation by noting that in the DLA case we have a growth instability of exactly the same form as the well-known Mullins-Sekerka<sup>35</sup> instability of crystal growth. The reasoning<sup>5,35</sup> goes as follows: suppose we start with a smooth aggregate and ask why it grows sharp tips. If we start with a tiny bump on the surface it will be magnified into a tip by the fact that the bump will grow faster than the rest of the surface: it will catch random walkers more efficiently than the flat portions of the surface, and certainly much more efficiently than the holes in the aggregate. The analogous dielectric breakdown case will make this even clearer: recall that the growth rate of any point on the surface of the structure is proportional to the electric field there. Sharp tips have large electric fields (the lightning rod effect). They grow ever sharper and dominate the growth. In the viscous fingering problem the same instability arises because it is easy for viscous fluid to flow away from a growing tip. It is even possible to specify a relationship between  $D$  and the characteristic opening angle of the tips<sup>36</sup> by using the mathematical theory of lightning rods. Unfortunately, no one knows how to calculate these angles. In fact, recent work indicates that there is an array of sharp tips on the surface of the fractal DLA cluster whose distribution is itself fractal<sup>37</sup>.

For ballistic aggregates or for the Eden model there is no growth instability: it is easy to see that a bump on the surface neither grows or shrinks, but just adds a uniform skin, and tips do not grow. The bulk of the material remains compact.

## Fractals and snowflakes

In the last section we noted the usefulness of the analogy of DLA with the kind of solidification most familiar (at least to those in cold climates) in the formation of snowflakes, that is, branched (dendritic) crystals. But particle aggregates do not look like snowflakes. To be precise, in a typical dendrite, a growing tip forms by the Mullins-Sekerka instability but then stabilizes. It retains its shape and continues in a definite direction, although it may spawn side-branches as it grows. In DLA (and in, for example, the zinc deposit of Fig. 2) the tips repeatedly split and wander.

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There are three obvious differences between DLA clusters and dendritic crystals: DLA has essentially zero surface tension, it has a significant source of noise in the discrete arrivals of the particles, and it has (at least in some versions of the model) no analogue of crystal anisotropy. Sorting out how these affect the process is a subject of current controversy and great intrinsic interest.

Surface effects can be added in various ways to DLA simulations<sup>5,38,39</sup>; the result is to thicken the branches of the aggregate, but the scaling is unaffected for large sizes. Nor do surface effects, by themselves, make the equations of crystallization give rise to snowflakes<sup>40,41</sup>. Instead, something unexpected happens: a growing tip with surface tension does not stabilize, but undergoes repeated splittings, which are caused by the surface tension itself. This is because surface tension slows the growth of sharply curved surfaces and the end of the tip is the most sharply curved. In order to make real dendrites, anisotropy arising from the crystal structure must be introduced. The relationship of anisotropy to tip-splitting was verified experimentally<sup>15</sup> using fluid flow in a Hele-Shaw cell with a lattice of grooves.

How does this relate to DLA? It is common to do DLA simulations on a lattice (for convenience). Will the same thing happen here as in the noise-free case; that is, will stable tips form because of lattice anisotropy? It seems that the answer is yes<sup>42,43</sup>: sufficiently large clusters on a lattice have the outline of a crystallite, with tip splitting only on a small scale. But why, without surface tension, do we ever get tip splitting? This is because noise due to the discreteness of the arriving particles can split the tips. This can be verified in various ways, for example, by experiments and calculations which vary the noise<sup>13</sup> at fixed anisotropy. In cases where tip splitting is mainly due to surface tension rather than noise, will we get an object which scales? The answer to this question is not yet clear, but there are indications<sup>15,44</sup> that there is scaling, and that it is close to that of DLA.

These considerations are of more than technical interest, because they show how small effects (such as anisotropy) can make qualitative changes in growth habit. A series of recent experiments<sup>45,46</sup> have shown, for example, how changes in growth conditions, such as an increase in voltage in electrodeposition, can change a fractal pattern like Fig. 2 into an ordered dendritic crystal by increasing the effective anisotropy. This is a fascinating example of the competition between scaling symmetry and ordinary spatial symmetry.

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*Note added in proof:* There have been two interesting recent attempts<sup>47,48</sup> to explicate the competition between anisotropy and noise.

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