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COURSE 3

FRACTAL AGGREGATES AND OTHER TENUOUS STRUCTURES

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* Much of the material in these notes comes from the following previously published reviews:
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1. Introduction

Figure 1 shows an electron micrograph of a typical colloidal aggregate. The aggregate is made in aqueous solution starting from a dilute mist of individual gold particles of 100 Å size. The chemical reaction which forms these particles also leaves them with a net electric charge, so that they repel one another and make a stable dispersion. When the reagent pyridine is added, it removes this charge and the particles then attract each other strongly via short-ranged van der Waals forces. Thus the particles stick irreversibly on contact. After a few minutes or hours, one finds that the particles have flocculated to form large, wispy-looking aggregates like the one shown here. (The micrograph was made by putting a drop of the solution on a microscope slide and letting it dry.)

Aggregates like this occur widely in nature and in technology. An example is soot (carbon black), which is formed as a byproduct of combustion and is found in diesel exhausts [1]. Aggregated carbon black, when incorporated into a rubber matrix, forms a tough elastic compound that is used to make vehicle tires [2]. Similar aggregates made of silica particles are used as fluid additives [3]; a small weight percentage of such material added to a liquid such as paint can thicken it dramatically, and give it controllable flow behavior. The flocculation of colloids, leading to precipitation of low-density, finely divided solids, provides an important means of chemical separation (for example of mineral ores) and purification (for example of drinking water) [4]. In nature, the properties of tenuous aggregated materials may determine the mechanical properties of a snow pack or the efficiency of a blood clot.

Another disorderly growth process is shown in fig. 2. This “viscous fingering” pattern is produced when a highly viscous fluid (epoxy) is displaced from a porous material by pumping in a less viscous fluid each other strongly via short-ranged van der Waals forces. Thus the particles stick irreversibly on contact. After a few minutes or hours, one finds that the particles have flocculated to form large, wispy-looking aggregates like the one shown here. (The micrograph was made by putting a drop of the solution on a microscope slide and letting it dry.)

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The pushing fluid tends to flow along thread-like fingers, instead of displacing the viscous fluid uniformly. This tendency poses a major obstacle to schemes that aim to push oil out of the ground by displacing it with water. One way of understanding viscous fingering is in terms of an equivalent aggregation process. Further examples are the closely related patterns that arise by dendritic solidification and electrodeposition [9–12] (such as the familiar mossy deposits that form on the terminals of a car battery) and by the dielectric breakdown of an insulator in a high electric field [13].

The structure of random aggregates has fascinated researchers for many years [14–17]. In these lectures I want to give an overview of recent progress in understanding tenuous structures like these. First, their apparent formless complexity reflects a pervasive scale-invariance property: they appear to be fractals characterized by a fractal dimension $D$. Such tenuous structures have a number of distinctive properties, which I want to survey. I will emphasize how a fractal structure scatters waves, and how it influences diffusion and flow in the space around it. I will also note the special features of disturbances (like vibrations) which propagate through the structure. I will describe some experimental problems in demonstrating the special scaling properties of these fractal structures. Next I will describe the special features that distinguish kinetically formed aggregates from “equilibrium” structures like the percolation clusters discussed by Deutscher (Course 1). A rich variety of aggregation phenomena with different scaling properties has been discovered to date. These fall into two broad classes: aggregation cluster by cluster and aggregation particle by particle. I will describe each class, with its distinctive features and variants, in turn.

2. Properties of scale-invariant structures

2.1. Scale invariance and fractals

The aggregate of fig. 1 has a fascinating self-referent property; each small section of the object appears to embody all the complexity of the whole. Mounting evidence suggests that this appearance reflects the statistical scale-invariance properties of a fractal object [18]. To define scale invariance, we may describe our object by its density profile $\rho(r)$. For the aggregate of fig. 1, $\rho(r)$ would be 1 for all points $r$ within a gold particle, and 0 elsewhere. The statistical properties of an enormous aggregate can then be specified by the correlation functions of this profile:

$$\langle \rho(0) \rho(r_1) \cdots \rho(r_n) \rangle,$$

where $r_1$, $r_2$, etc. indicate specific points in space, and the $\langle \cdot \cdot \cdot \rangle$ indicates an average over all positions and orientations of the structure. We take the separations between the $r_i$ to be much smaller than the size of the aggregate. Statistical scale invariance, or dilation symmetry, means that these correlation functions are essentially unaffected by a dilation of space: $r_i \rightarrow \lambda r_i$. Thus for a dilation-symmetric object

$$\langle \rho(0) \rho(\lambda r_1) \cdots \rho(\lambda r_n) \rangle = \lambda^{-\Delta q} \langle \rho(0) \rho(r_1) \cdots \rho(r_n) \rangle. \quad (1)$$

The only effect of the dilation is a multiplicative factor, controlled by the powers $A(q)$. Some classical examples of fractals in statistical physics are random walks, self-avoiding walks [19], random animals, and percolating clusters [20]. These are known to be dilation symmetric in this sense, and moreover, their powers $A(q)$ are related by $A(q) = q A(1) = q A$.

A simple test for a dilation-symmetric or scale-invariant object is to examine the simplest of these correlation functions, $\langle \rho(0) \rho(r) \rangle$, or equivalently, the pair distribution function $C(r) = \langle \rho(0) \rho(r) \rangle / \rho$. This is the average density of points on the object at a distance $r$ from an arbitrary point. Evidently, in a dilation-symmetric object the correlation function falls off as $r^{-\Delta}$. This power is essentially Mandelbrot’s [18] fractal dimension $D$. This $D$ may be defined by considering the average mass $\langle M(R) \rangle$ within a distance $R$ of an arbitrary point on the object. One may obtain this mass from $C(r)$: $\langle M \rangle = \int_R^\infty \pi \cdot r \cdot C(r) \cdot r \cdot dr$. Evidently, the mass scales as $R^{D-\Delta}$ in $d$-dimensional space. This power also defines the fractal dimension $D$. Measurements of $C(r)$ and related quantities [21] suggest that colloidal aggregation and other random growth processes are fractals in this sense. More detailed studies [22] give indirect evidence that the higher correlation functions of some of these objects are also consistent with the dilation symmetry defined above.

The fractal dimension of these structures is useful for describing their properties in large part because their density profiles are not too heterogeneous. The matter near most points of the structure is similarly arranged. Thus the mass within distance $R$ of an arbitrary point of the fractal does not fluctuate too far from its average $\langle M(R) \rangle$, whose scaling is given by $D$. That is, the statistical distribution $P(M)$ of masses $M(R)$ sampled at various points of the structure is not too broad. For example, a simple random walk ($D = 2$) on a three-dimensional lattice has a peaked

$$\langle \rho(0) \rho(r_1) \cdots \rho(r_n) \rangle.$$
probability distribution. The probability of masses $M \ll \langle M \rangle$ falls off faster than any power, since stretched-out segments of walk are exponentially rare. The probability of masses $M \gg \langle M \rangle$ also falls off faster than any power because compressed segments are exponentially rare [23]. Thus all positive and negative moments $\langle M^n \rangle$ of the mass distribution scale with the average mass: $\langle M^n \rangle = \langle M \rangle^n$. The distribution of masses is narrow enough that $D$ gives the correct scaling behavior for many purposes. Such density profiles are in this sense uniform, or "isometric". Note that any isometric fractal must satisfy $A(q) = q^d$, since one may obtain $\langle M^n \rangle$ by integrating the order-$q$ correlation function.

A contrasting case is the Levy flight [18], a random flight with a broad, power-law distribution of jumps. Levy flights with $D < 2$ (in two or more dimensions) are not "isometric". The average mass is of order $R^D$, but the probability of small masses is not exponentially small; it is of the same order as the probability of the average mass. Many of the scaling arguments mentioned below assume implicitly that aggregates are isometric. This assumption is widely believed, though to my knowledge it has not been tested explicitly. Later we will see that some important properties of fractals other than their density are not isometrically distributed. The special language of "fractal measures" is needed to describe these [24].

2.2. Measuring scale invariance by scattering

A set of particles with fractal dimension $D_f$ scatters waves [25] with a characteristic scattering law. If $q$ is the scattered wavenumber, then the fractal gives a scattered intensity varying as $q^{-D_f}$. A solid bounded by a fractal surface also has a characteristic scattering signature [26–28]. In three-dimensional space, a surface with dimension $D_1$ gives a $q^{D_1}$ scattering law. Surprisingly, the fractal dimension enters with the opposite sign as compared to a fractal set of particles. These scattering laws can be understood by simple scaling arguments.

We suppose that the system consists of a set of particles, all with the same scattering properties. The density $\rho(x)$ gives the distribution of these particles in space. The scattering structure factor $S(q)$ is then proportional to the squared Fourier transform of $\rho(x)$. A qualitative feature of the Fourier transform is that it is insensitive to structure on length scales smaller than $q^{-1}$. More concretely, $S(q)$ is essentially unaltered if the particles are moved over distances smaller than $q^{-1}$.

The scattering from $N$ particles arranged in a fractal density profile of dimension $D_f$ and radius $R$ can be conveniently inferred by dividing space into cubes of length $q^{-1}$, each enclosing some portion of the particles. The cubes are so placed that all particles are enclosed. There are an average of $M(1/q) \sim q^{-D_f}$ particles in each cube. The number of cubes required is of order $(qR)^{D_1}$. We assume that the contents of distant cubes scatter independently, so that $S(q)$ is the sum of the $S(q')$'s from the individual cubes. This assumption is valid for fractals formed by random processes where the relative positioning of two distant cubes is arbitrary to at least $q^{-1}$. [For some mathematical fractals, like the classical Cantor set, this assumption is not valid, and $S(q)$ becomes a complicated function rather than a simple power law.]

The above reasoning relates the scattering from the fractal to that from a single cube. This one-cube scattering is unaltered if all its particles are bunched in the middle. Thus evidently the one-cube scattering is proportional to the square of the number of particles enclosed: $[N(qR)^{-D_1}]^2$. Multiplying the number of cubes by the scattering from each cube, we find

$$S(q) \sim (qR)^{D_1}[N(qR)^{-D_1}]^2 \sim Nq^{-D_f}$$

where we have used $N \sim R^{D_1}$. [One can get this result even more simply by requiring that $S(q) \sim N^2 f(qR)$, and that for $qR \gg 1$, $S(q)$ is proportional to $N$ and otherwise independent of $R$ [29].]

We now consider a fractal surface of dimension $D_1$ imbedded in a $d$-dimensional space. Again, we may cover the object with cubes of radius $q^{-1}$. Only the cubes crossing the surface can cause scattering: if all of space is filled uniformly there is no scattering except at $q = 0$. The number of surface cubes is again $(qR)^{D_1}$. We again expect the contents of distant cubes to scatter independently, so that again $S(q)$ is the number of cubes times the average scattering from a single cube. Each cube on the surface is now roughly half filled with bulk matter at some particle density $\rho_0$, and half empty, with a fractal boundary in between. The scattering, though, is insensitive to the form of this boundary. The scattering must be unaltered if we rearrange the density to form e.g. a simple, smooth boundary within the cube. The scattering contribution from each cube is the square of the number of particles there, $(\rho_0 q^{-d})^2$. In contrast to the volume fractal, the single-cube scattering is now independent of the fractal dimension. Multiplying this single-cube scattering by the number of cubes, we obtain

$$S(q) \sim (qR)^{D_1}(\rho_0 q^{-d})^2 \sim q^{D_1 + 2d},$$

(3)
In three dimensions this is the $q^{D_1 - b}$ result quoted above. Since the fractal surface must have $2 < D_1 < 3$, the power of $q$ varies between $-3$ and $-4$. The power $-4$ is Porod’s law for scattering from smooth surfaces.

Other irregular surfaces have recently been examined extensively. These are the so-called self-affine surfaces [27, 30]. Self-affinity is a generalization of dilation symmetry. A dilation-symmetric object looks the same when space is dilated or stretched uniformly by a factor $\lambda$. A self-affine object looks the same when stretched by a factor $\lambda$ in the $x$ direction and by a factor $\lambda^b$ in the $y$ direction. (Thus dilation symmetry is self-affinity with $b = 1$.) The track of a directed random walker is self-affine. Consider a walker on a lattice who may walk up, down or right, but not left. Then the height occupied by a section of walk is proportional to the square root of its width. The exponent $b$ is $\frac{1}{2}$. Phase boundaries between two-dimensional solid phases are thought to obey (see, e.g., ref. [31]) self-affine scaling with $b = \frac{1}{2}$. Many three-dimensional solid surfaces are also thought to be self-affine [27], with $b = \frac{1}{3}$. Their height is no greater than their width on any length scale.

Wong [27] showed that scattering from a self-affine surface gives a structure factor $S(q) \sim Aq^{D_1 - b} + Bq^{-4}$. But for macroscopically smooth surfaces like those just mentioned, the coefficient $A$ must be small. Our cube construction shows this. Consider covering a horizontally directed random walk by a row of very large cubes. The section of walk within each cube is practically horizontal; the walk only occupies a wide, flat rectangle within the cube. How many cubes are required to cover this walk if their length is halved? It clearly suffices to fit two small cubes side by side within each large cube. There is no danger of the walk escaping out the top of the smaller cubes, since the walk is so flat. This means that the number of cubes required to cover the walk is inversely proportional to the cube size, $D_1 = 1$. Similarly for a self-affine surface in three dimensions with $b < 1$, $D_1 = 2$. By our reasoning above, these self-affine surfaces should have $S(q) \sim q^{-4}$, like smooth surfaces.

If a surface is oriented perpendicular to the scattering vector $q$, then $S(q)$ is sensitive to the height–height correlation function, and thus to the exponent $b$.

2.3. True and apparent power laws

We have shown above that if you scatter from a fractal, $S(q)$ has a power-law form that reflects the fractal dimension $D_1$. But what about the converse question? If a measured $S(q)$ looks like a power law, does that mean the system is a fractal? Often, such an apparent power law can be more naturally explained without supposing any fractal structure. And often, observed power laws prove to be mere “crossover” effects, valid in a limited range and not arising from any fundamental dilation invariance property. I would like to spend a little time discussing when an observed power law gives strong evidence for fractal structure and when it does not.

For this discussion I made up some sample data, shown in fig. 3. The triangles are fictitious scattering data, which suggest the power law $S(q) \sim q^{-3.1}$. (I reveal the real functional form of the data below.) The data closely follows a straight line with slope $-3.1$ over two orders of magnitude in $q^2$. As noted above, this is the scattering law to be expected from a surface fractal with dimension $D = 2.9$. This implies that the data approximate a function which would show the $q^{-3.1}$ law over an arbitrarily wide range of $q$, and with greater and greater precision. That appears to be the case with these data, but it is not. Instead, the asymptotic power for large wavevector $q$ is shown by the broken line, which follows the

![Fig. 3. Fictitious scattering data to illustrate deceptive power-law behavior, shown in a log-log plot of $S(q)$ versus $q^2$. Dashed line shows asymptote which data approach at high $q$. The line through the data has the form $S(q) = \text{const.} \times q^{-3.1}$](image-url)
ordinary Porod law for a smooth surface: $S(q) \sim q^{-4}$. These data have an especially deceptive way of “crossing over” from this $q^{-4}$ behavior at large $q$ to a constant value at small $q$.

It seems clear that with a limited range of the variable $q$ and with limited precision in the measurement, one can never completely rule out this kind of deceptive crossover. How then does one discuss the possibility that the observed power arises from a crossover and is not asymptotic? I would like to describe a general language for discussing this question, which was developed for analysing power laws seen near phase transitions. Guenter Ahlers developed much of this language, and has discussed this subject authoritatively [32].

How consistent are the data of fig. 3 with Porod scattering from a smooth surface? The Porod law holds in the limit of wavevectors large on the scale of the typical radius of curvature $R$: $qR \gg 1$. If $qR$ is not infinite, the scattering must have the form

$$\log S(q) = -4 \log(q) + f(qR),$$

where the correction $f$ tends to a constant as $qR \to \infty$. The nature of $f$ clearly depends on the specific system, and often we have no information about it. But it often happens that it goes to its asymptotic value as a weak power of its argument: $f \sim A + B(qR)^{-c}$.

If the correction power $c$ is small, the measured function $S(q)$ is difficult to distinguish from a fractional power law. If the measured range of $qR$ lies between $x_0$ and $x$, we may rewrite the correction factor as

$$f(x) = f(x_0) + Bx^{-c} - Bx_0^{-c}$$

$$= Bx^{-c} \log(x/x_0) \left(1 + O(c \log(x/x_0))\right).$$

Thus,

$$\log S(q) \approx \text{const.} + \left(-4 - Bc_0^{-c}\right) \log q.$$

The apparent power is no longer 4, but the whole quantity in parentheses. And the approximation is valid as long as $c \log(x/x_0) < 1$.

This shows that any fractional power law is consistent with a nearby integer power law with a correction of the above type. Given the scattering data of fig. 3 one cannot rule out the possibility that the system shows ordinary Porod scattering which has not reached its asymptotic form. This is illustrated in fig. 4, another log-log plot, with the $q^{-4}$ divided out. (This plot is more revealing, since it shows better the slight curvature in the data. It is also more satisfying, since it shows directly how far the data differ from the expected behavior.) We suppose that the data, with their experimental uncertainties, are consistent with the straight line shown. But they are also consistent with the other curve, with $c = 0.8$. Evidently, any $c$ less than 0.8 explains the data as well as the fractal power law does.

If the experimental uncertainties were reduced, or the range of the data were increased, it would be progressively easier to distinguish between an asymptotic power law and nonasymptotic corrections. Conversely, if such data remain consistent with a power law, smaller and smaller $c$ values must be used to give an equally good fit. Thus one measure of the goodness of a power-law fit is how small a value of $c$ would be required to give an equally good non-power-law fit. The smaller the $c$, the more compelling is the evidence of a true power law. This type of fitting, and the determination of $c$, seem to be a good way to evaluate power-law data. The number $c$ suggests what alternative explanations of the data might be possible, and it reflects both the precision and the range of the data. The procedure is not restricted to scattering data. It can be used with any data which seem to show a departure from an expected power law.
Corrections like these are not just mathematical nightmares, nor are they special to esoteric phase transitions. The fictitious $S(q)$ above was a simple function, analytic in $q^2$ for $qR \ll 1$:

$$S(q) = \left[ 1 + \left( \frac{q^2}{c} \right)^{\gamma - \frac{1}{2}} \right]^{-1},$$

(7)

with $c = \frac{1}{2}$. The only thing out of the ordinary is that for large $q$, $S(q) \sim q^{-\left( \frac{1}{2} + \frac{1}{1 + O(q^{-2\gamma})} \right)}$.

Such fractional corrections arise naturally in many physical situations. I will give one example. This is a possible mechanism for the anomalous surface scattering discussed above. Consider a smooth-surfaced object of radius $R$ and mass $M \sim R^3$ with scattering

$$S_0(q, M) = M^2 \left[ 1 + \frac{q^2}{(qR)^2} \right]^{-1}.$$  

(8)

A distribution of such objects with various masses $M$ arranged randomly in space would then have a scattering law

$$S(q) = \int dM N(M) S_0(q, M),$$

(9)

where $N(M) dM$ is the number of objects with mass between $M$ and $M + dM$. It is easy to show that if $N(M) \sim M^{\gamma - 1}$ with $\gamma < \frac{3}{2}$ between limits $M_0$ and $M_1$,

$$S(q) \rightarrow q^{-\gamma} \left[ 1 - O(qR_i)^{-\frac{1}{2} + 1} \right].$$

(10)

If $\gamma$ were close to $\frac{3}{2}$, $S(q)$ would show the same deceptive behavior as our fictitious example. Other examples [33, 34] of this behavior also involve a measured quantity which is an average over a broad distribution of sizes.

2.4. Distinctive properties: internal phenomena

Aggregates are different from ordinary bulk matter in many qualitative respects. A major difference concerns phenomena confined to the structure, such as electric conduction, diffusion along the structure, or mechanical vibration. Alexander and Orbach [35] have suggested that many of these properties may be described in terms of a single new exponent called the spectral or fractal dimension $d$. Deutscher discussed these properties in the context of percolation clusters in Course 1.

Such properties may be discussed even more simply for aggregates than for other fractals, because aggregates are topologically simple: they appear to have no loops except at the smallest length scales. We may illustrate how this simplifies these properties by considering the conductance (inverse resistance) $G$ between an arbitrary point on a loopless fractal and a surrounding conducting sphere of some large radius $R$. As in percolation, most of the mass belongs to dead ends, which carry no current. The remaining backbone structure has relatively long branches at the center connecting to progressively shorter branches near the bounding sphere. The conductance $G$ from center to sphere is evidently larger than that of any single path from center to sphere. This conductance is dominated by the few long branches near the center. For branched structures generally, one may show, with mild additional assumptions, that the conductance of the whole structure is of the same order as that of one path to the boundary. Thus, the conductance scales with the average length of such a path [36–38], which we denote as $L$. Equivalently, $L$ is the average number of particles in the path connecting any two points at distance $R$. Thus the scaling of conductivity in branched structures can be expressed in terms of a purely geometric scaling property. One finds $d = 2/(1 + \delta/D)$. In the computer models we consider later [39], the path length $L$ scales as the distance $R$ to a power $\delta$ between 1 and 2.

An important internal response of fractal aggregates is their elastic response. Large aggregates become progressively less rigid [40], and they ultimately attain a “floppy” state in which the branches are completely flexible. To quantify the elastic response, we imagine anchoring some point of the fractal of fig. 1 in a fixed position and orientation. Then we pull on some point at distance $R$, displacing it by an amount $u$, and storing an elastic energy $U = \frac{1}{2}uK(R)u$. This energy is all stored along the (single) connecting path between the origin and the displaced point. Each point $i$ along this path is bent through an angle $\theta_i$ and gains an energy $\kappa \theta_i^2$. Each of these local bends produces a displacement $u_i = r_i \theta_i$, where $r_i$ is the vector from the point $i$ to the displaced point, projected perpendicular to $u$. Thus the total energy $U$ may be expressed as $\sum_i (\kappa / r_i^2) u_i^2$, where the vector sum of the $u_i$ must be the total displacement $u$. Our problem is now equivalent to a line of simple springs in series. The inverse spring constants add to give $K = \kappa / (r_i^2 L)$, where $L$ is the number of segments $i$.

Knowing the exponent $\delta$ which relates $L$ to $R$, one may readily estimate when a given type of aggregate, made of particles of size $a$, loses its
rigidity in a given situation. Thermal vibrations at temperature $T$ lead to loss of rigidity for sizes $R \approx a(Ta^d)^{1/5}$. Beyond this size, thermal fluctuations produce displacements $u \approx R$. Gravity makes an aggregate sag substantially under its own weight when $R \approx a(ga)^{-\psi}$, under a gravitational acceleration $g$, where $\psi = (1 + D + \delta)^{-1}$. Considering both gravitational and thermal distortions, there is an optimal $a$ that maximizes $R/a$. Under normal gravity and room temperature the optimum diameter $a$ is $10-1000$ Å. This is comparable to colloidal aggregates normally seen. (If the particles were of molecular size, only a few could aggregate before floppiness set in.) The corresponding maximum size is $10^3$ to $10^4$ particle diameters. The scaling properties of floppy aggregates remain unexplored. Do the floppy branches collide, stick, and give rise to a limiting nonfractal density for large aggregates? Or do the resulting arches make "girders" which strengthen the structure sufficiently to produce a new type of fractal?

2.5. Distinctive properties: external phenomena

Complementary to these internal phenomena, confined to the structure, are external phenomena in the space around it. In this category are such effects as the screening of electric fields around a conducting fractal, or the distortion of hydrodynamic flow around a fractal in solution. The most basic scaling properties governing these phenomena, unlike the internal phenomena, can be expressed solely in terms of the fractal dimension $D$. As a simple example of this, we consider the interaction of a fractal with a ray of light. We imagine a tiny light source somewhere in the middle of the fractal, and ask what is the probability that it will be visible from a given point outside. This is simply the probability that the (arbitrary) light ray from the source to our eye intersects the fractal. A parallel problem is the escape of a random walker from an absorbing fractal. The random walker is released from somewhere in the interior. Its probability of escape is one minus the probability of intersection between the fractal and an arbitrary infinitely long random walk.

These intersection questions are readily answered in terms of the fractal dimension. Indeed, the mean number of intersections [18] $M_\psi(\lambda)$ between any two fractals 1 and 2 of radius $R$ scales as $R^{D_\psi-D_{21}}$. This law applies to fractals placed independently of each other in the same volume of space, like the light ray or the random walk above. Evidently, if the power is negative, the probability of intersection decreases indefinitely as the size of the structure increases. Thus in three dimensions the ray ($D_\psi = 1$) almost always emerges from a sufficiently large fractal provided $D_\psi < 2$. We may say that the two fractals are transparent to each other.

In the complementary case where the number of intersections grows with $R$, we expect the two fractals to avoid each other with a probability approaching zero. The two are opaque to each other. If the absorption probability $b$ per contact is small, the walker or ray may travel some large distance $\lambda$ through the fractal before $M_\psi(\lambda) = b^{-1}$ and the net probability of absorption becomes appreciable. Still, if the fractal is much larger than $\lambda$ we expect that the probability of escape goes to zero. Eventually the opacity becomes virtually as complete as though $b$ had been 1. This means e.g. that random walkers entering a large enough fractal with $D > 1$ are almost always absorbed; an ideal gas of such walkers would be strongly depleted in the interior of the fractal.

Restating this result in more common language leads us to several conclusions about the interaction of fractals with $D > d - 2$. The local concentration $u(r)$ of our gas of walkers obeys a steady-state diffusion equation $\nabla^2 u = 0$, i.e. Laplace's equation. Since the walkers are absorbed, $u = 0$ on the fractal. The distant walkers are unperturbed by the fractal; $u$ is some constant at infinity. Since $u$ is strongly depleted in the whole interior, it must be strongly reduced just outside, as well. The distant $u$ field is thus the result of a perfect absorber with radius $R_u$ of order $R$; specifically, $u(r) = u_0[1 - (R_u/r)^{d-2} + O(R_u/r)^{d-4}]$. Evidently, the net flux of material from infinity is the same as for a perfectly absorbing sphere of radius $R_u$. The electric potential around a conducting fractal evidently obeys the same equation as the diffusing field $u$. Thus a conducting fractal has the same capacitance to ground as a sphere of radius $R_u$.

The fractal behaves similarly with respect to hydrodynamic interactions. These interactions control, for example, how fast the fractal sediments in an incompressible fluid. The velocity $v(r)$ is governed by the Stokes equation, which describes the incompressibility and the local conservation of the fluid's momentum. In a quiescent fluid the momentum, proportional to $v(r)$, moves through the fluid by diffusion. A fractal sedimenting at speed $v_0$ absorbs momentum from the surrounding fluid: $v(x) = v_0$ for points $x$ on the fractal. The absorbed momentum diffuses from infinity, as in the random walker problem above. And as above, the fractal absorbs momentum at the same rate as if it were a perfectly absorbing sphere of radius $R_u$, of the order of $R$. A fractal opaque to diffusers is also opaque to flow. The fluid is obliged to flow around the fractal, and $v(r) \approx v_0$ throughout the interior. For the case where the
sedi
ting fractal is a polymer, this opacity is well known [41 (section VI.2.1)]. Another consequence of hydrodynamic opacity is that fractals dispersed in a fluid increase its viscosity as though they were hard spheres of radius about \( R \).

The notion of opacity may be used also to treat the thermodynamics of mutually interacting fractals. These properties are well known in the context of polymers, an important special case [41 (Ch. III), 42, 43]. We consider two fractals whose constituent particles cannot interpenetrate. In calculating the partition function of the two, confined to some volume \( V \), we must eliminate all configurations in which the two intersect. If the two fractals have size \( R \) and are mutually opaque, then almost all the configurations in which the two occupy the same volume must be eliminated. Each will be excluded from a volume of order \( R^d \) on account of the other.

The opacity of a fractal is easily compromised; it must not behave like a solid object in all respects. Thus the mutual excluded volume of a fractal and one of its constituent particles is much smaller than \( R^d \); instead, it clearly scales as the volume actually occupied by the constituent particles, i.e., it scales as the mass \( M \) of the fractal. The exclusion of a small fractal by a large one gives a means of measuring the latter's fractal dimension. This may be derived formally by a scaling hypothesis; it may also be seen pictorially by imagining enclosing the large fractal with balls the size of the small fractal. We suppose that the large fractal has dimension \( D_2 \) and radius \( R_2 \). Evidently the small fractal is excluded from the interior of each ball, but not from the spaces between them; the excluded volume is the sum of the volumes of the balls. Noting that the mass of the large fractal is the sum of the masses in the balls, we deduce the excluded volume \( V_e \):

\[
V_e = R_2^3 (R/R_2)^D_2. \tag{11}
\]

The case where the small fractal is a short random walk was treated by de Gennes [44]. He pointed out that if a fractal is made to absorb diffusing particles, such as an excited molecular species, for a limited time \( t \), then the amount absorbed should scale as \( V_e \) with \( R_e \sim (\xi t)^{1/2} \), where \( \xi \) is the diffusion constant. The counterpart for flow is the time-dependent friction factor [45] for a fractal in a fluid of kinematic viscosity \( \nu \). If the fractal is given an impulse, uniformly on all of its particles, at time zero, the applied momentum occupies the volume \( V_e \) after a time \( t = R_e^2 / \nu \).

A semidilute polymer solution with correlation length \( \xi \) would be excluded from a fractal in the same way; the region where monomers are depleted is essentially the region excluded to a small polymer of size \( \xi \). For fractal objects like colloidal aggregates, one may infer [45] the change of volume fraction \( \phi \) of a polymer solution upon adding a volume fraction \( \phi_t \) of fractal. The solution must be semidilute, so that the polymer chains overlap strongly, but not so strongly that the correlation length \( \xi \) becomes smaller than the constituent particles of the fractals. When the fractals are added, each monomer is excluded from a volume \( V_e \approx V_b / (\xi b) \). Here \( b \) is the size of the constituent particles. In such a semidilute solution the length \( \xi \) scales with volume fraction by the usual fractal law for the polymers: \((\xi/a)^{D_2-d} = \phi_t \). Combining these, we find a relation for the change of \( \phi_t \):

\[
\Delta \phi = \phi_t (D_2/D_1)^{1/(D_2-d)}. \tag{12}
\]

If \( D_2 \) becomes equal to \( D_1 \), then the change of \( \phi_t \) becomes equal to \( \phi_t \). Adding fractals to the solution becomes equivalent to adding the same volume fraction of polymer.

Thus far we have treated properties equivalent to fractals absorbing random walkers or rays. Now we consider the scattering of rays by a fractal. [This section arose from discussions with E. Ackermans and D. Sornette during the school.] A ray of light emitted from a fractal, if scattered without absorption, must eventually escape, even if the \( D \) indicates opacity. To find the nature of this multiple scattering, we consider a classical ray leaving the fractal at some point. To find the probability \( P(r) \) that it travels a distance \( r \) without scattering, we note that \( dP/dr = -\rho(t) \), where \( \rho(t) \) is the average density of fractal particles encountered by an arbitrary such ray. Assuming that this \( \rho \) is approximately the average density at \( r \), viz. \((r/a)^{D-d} \), we find \( P(r) \sim \exp[-(r/a)^{D-d}] \). For a transparent fractal, \( P(r) \) goes to a constant for large \( r \) at every scattering the ray has a probability of order \( R^d \) to escape. For an opaque fractal, \( P(r) \) falls off faster than any power, and thus the mean free path is of the order of the particle size \( a \). The multiply scattered ray follows the fractal, and diffuses like a random walker confined to it. Its escape time \( t \) is thus controlled by the spectral dimension \( d' \):

\[
t \sim R^{2d'/3}. \tag{13}
\]

Multiple diffraction by a fractal may lead to localization of a wave emitted from the fractal. To estimate when localization might occur, we
may use the Ioffe-Regel criterion introduced by Ramakrishnan and Souillard in their lectures (Courses 4 and 5): localization is indicated when the mean free path becomes smaller than the wavelength $1/\lambda$. If $qa = 1$, the fate of the wave is controlled by the local structure rather than the fractal scaling properties. For Schrödinger waves with $qa \ll 1$, the mean free path $l$ is proportional to the density of scatterers, and becomes independent of $q$. Using the density $\rho(r) \sim r^{D-4}$, we find $ql \sim q^{D-d+1}$. Again, our opacity criterion indicates strong wave scattering and the potential for localized waves.

2.6. Distribution of diffusing flux: fractal measures

The above discussion shows that many aspects of a fractal’s interaction with its environment can be inferred from its fractal dimension. Now I want to note another aspect, for which new and little-studied scaling properties of the fractal are needed. We have seen that an opaque fractal screens out most of an external field from its interior. The question naturally arises how much of the fractal is then exposed to this field. For example, where does the charge reside on a conducting fractal at nonzero electric potential? This question and related ones have been considered by several authors [46, 24, 47]. Such questions are clearly important for understanding the electrical breakdown around a fractal, the distribution of forces on a fractal in flow, or the amount of some chemical reaction occurring at different places on an absorbing fractal. When a fractal grows by absorbing a diffusing substance, it owes its very structure to this distribution of absorption rates.

To make the problem explicit, we imagine a fractal which absorbs a diffusing particle from infinity. Each site $i$ of the fractal has a probability $p_i$ of absorbing the particle. For a solid sphere, only the particles on the surface can absorb, and the absorbing mass scales as the total mass $M$ to the power $(d-1)/d$. A simple way to estimate the absorbing mass in a fractal is to approximate it as a cloud of density $\rho = M/R^d$ spread uniformly within the fractal’s radius $R$. Then the absorption rate at a point $r$ is proportional to the concentration $u$ of diffusers there, and to the density $\rho(r)$. Thus the diffusing field satisfies $\nabla^2 u = up(r)$. One easily checks that $u$ falls off exponentially in a small distance $\lambda$ from the outer boundary:

$$u(r) \sim e^{-(r-R)/\lambda}, \quad \lambda/R \sim R^{(D+2-d)/2}.$$  \hspace{1cm} (14)

The absorbing mass scales as $MA/R \sim M^x$, where $x = (D-2+d)/(2D)$.

Remarkably, this picture seems to account reasonably for the number of absorbing sites. Meakin et al. [47] defined a measure of the absorbing mass $M_c$ as essentially the average number of random walkers that must be absorbed on the fractal before some site absorbs two: $M_c = (1/p_i)^{-1}$. They found with some exceptions that for two-dimensional fractals $M_c$ appeared to scale with the power $x = \frac{1}{2}$ predicted above.

One exception clearly occurs when the absorber is a line or row of particles in two dimensions. Such a line is clearly opaque to a diffusing field in the sense defined above. The “hit probabilities” $p_i$ become larger at the tips; still the $M_c$ defined above is proportional to the total mass. The flux is qualitatively uniform; most of the absorbers get a “fair share” of the total flux, and there is no screening of the flux from the interior. This leads us to ask what is necessary for the interior of a fractal to be screened, and how this screening is to be described. More precisely, what is the form of the mass distribution $n(p) = \sum_i \delta(p-p_i)$? Clearly $n(p) dp$ is the number of sites absorbing with probability between $p$ and $p + dp$. It is useful to define the moments of this distribution $\langle p^n \rangle = \int dp \, p^n n(p)$. Since the number of absorbers is $M$, $\langle p^n \rangle = M$, and since the total probability of absorption is defined to be $1$, $\langle p \rangle = 1$. The $M_c$ defined above is $\langle p^2 \rangle^{-1}$.

To illustrate why these moments are important, we consider a hypothetical problem in catalysis [48]. Suppose that a fractal absorbs a diffusing molecular species A from the space around it. This reactive species becomes inactive and leaves the fractal after a short time on it. But if three such molecules find themselves together on the same site, they may combine to yield a desired product B. Similarly, when two A’s are together, they can combine to make an undesired product C. The desired reaction rate is proportional to $\langle p^n \rangle$, while the undesired rate is proportional to $\langle p^{n-1} \rangle$. One wants to maximize the former and minimize the latter. In the hydrodynamic counterpart of this problem the role of $p_i$ is played by the fluid velocity past the site $i$. Thus, the velocity distribution analogous to $n(p)$ is of interest for questions of velocity dispersion raised by Guyon in his lectures (Course 2).

Even for simple objects, $n(p)$ may be a broad distribution. For the line of absorbers mentioned above, $n(p)$ varies as $p^{-2}$ between some $p_{\text{min}}$ and $p_{\text{max}}$. Other smooth absorbers with singular points have other power laws. In this situation different moments $\langle p^n \rangle$ may scale differently with $M$. High moments are controlled by $p_{\text{max}}$: $\langle p^{n+1} \rangle / \langle p^n \rangle \sim p_{\text{max}}$. Low moments (including negative $q$) are controlled by $p_{\text{min}}$. Clearly, ratios of moments can be constructed which scale with various powers of the
This is a partition function for $X$ independent "spins" $s$ in a magnetic field $h$:

$$
\langle p^h \rangle = \left[ \int ds_1 e^{-U(s_1)+hs_1} \right]^X.
$$

(16)

It is natural to define the quantity in square brackets as $e^{f(h)}$, where $f$ is a free energy per spin. Expressing $X$ as $\log M/\log 2$, this means

$$
\langle p^h \rangle = M^{f(h)/\log 2} = M^{f(h)}.
$$

(17)

The various moments $h$ scale as independent powers of the mass. A measure $\mu$ distributed in space whose moments scale in this way is known as a fractal measure or multifractal [24]. These powers $F(h)$ reflect the details of the original random distribution $U(s)$, so there is no general way to express the moments in terms of a small subset. This is in contrast to simple singular absorbers like the line segment treated above.

For any fractal measure, where $\langle p^h \rangle = M^{f(h)}$, the distribution $n(p)$ must obey a particular scaling with $M$. To find $n(p)$ itself, or equivalently $\tilde{n}(S) = n(p) dp/dS$, we recall that

$$
\langle p^h \rangle = \int dS \tilde{n}(S) e^{hs} = e^{\log M \cdot f(h)}.
$$

(18)

Now, the right-hand side must go to zero or infinity as $M \to \infty$, so that $\tilde{n}(S)$ must do likewise. Expressing $\tilde{n}(S)$ as $e^{g(S)}$, the integral must be dominated by a saddle point $S^*$ such that $d(G + hS)/dS = 0$ when $S = S^*$.

Thermodynamically, $G(S)$ is the Gibbs free energy. It is the Legendre transform of the total free energy $\log(M) F$. Evidently, $G$ must be extensive: $G(S) = \log(M) F(\sigma)$, where $\sigma = S/\log(M)$. The function $F$ is called the effective potential [51]. A single spin $\sigma$ in the potential $F$ would respond the same to a field $h$ as does the average $S/\log(M)$ of $\log(M)$ spins in individual potentials $U$. Since $F(\sigma)$ is independent of system size, we may infer how $n(p)$ scales with system size [52-54]:

$$
\log(pn(p))/\log(M) = F(\log p/\log(M)).
$$

(19)

The scaling function $F$ is essentially the "spectrum of singularities" $f(\alpha)$ defined by Halsey et al. [55]; specifically their $\alpha = -\alpha D$ and their $f(\alpha) = -Df'(\alpha)$. The present derivation of it does not require any singularities, but merely that moments of the distribution scale as independent powers of the mass. To interpret $F$ we first recall our normalization conditions on $n(p)$. Since $\langle p^h \rangle = M$, we have $F(h = 0) = 1$. The minimum value of $F$ is thus $-1$. Since $\langle p \rangle = 1$, $F(h = 1) = 0$, so that the curve $F(\sigma)$
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is tangent to the line of unit slope through the origin, as shown in fig. 5. The minimizing value \( \sigma_0 \) and the point of tangency \( \sigma_1 \) have a special meaning. (Note that \( \sigma_0 < -1 < \sigma_1 \).) Near the minimum, \( \Gamma = \frac{1}{2}(\sigma - \sigma_0)^2/(\Delta \sigma)^2 = 1 \). The distribution \( \tilde{N}(S) \), where \( S = \log \rho \), is essentially a Gaussian with its peak at \( S = \sigma_0 \log(M) \) and with standard deviation \( \Delta \sigma \log(M)^{1/2} \). The median value of \( S \) is clearly \( \sigma_0 \log(M) \); so the median value of \( \rho = e^S \) for all the sites is \( M^{\sigma_0} \).

The quantity \( \sigma_1 \) has a similar interpretation. If the fluxes \( \rho_i \) are sampled by random walkers instead of indiscriminately over all sites \( i \), then a given site \( i \) is represented \( \rho_i \) times as often. The distribution \( N(\rho) \) sampled in this way is thus given by \( N(\rho) = \rho n(\rho) \). We call this the flux-weighted distribution. Its scaling function \( \Gamma' \) is evidently \( \Gamma - \sigma \). Thus its minimum is at \( \sigma = \sigma_1 \). By our previous reasoning, the median flux-weighted flux scales as \( M^{\sigma_1} \). The flux-weighted average of \( \log \rho \):

\[
\int N(\rho) \log \rho \int N(\rho) = \frac{\int \tilde{N}(S)S}{\int \tilde{N}(S)},
\]

is evidently also \( \sigma_1 \log M \). This average is \( \langle \rho \log \rho \rangle \), and its scaling is conventionally expressed in terms of the “information dimension” \( D_i \):

\[
\langle \rho \log \rho \rangle = D_i \log R.
\]

(The scaling with length is usually expressed in terms of a local averaging length \( \varepsilon \) rather than the overall size \( R \). In any case, the moments only depend on \( \varepsilon \) or \( R \) through the ratio \( R/\varepsilon \), so that the two types of scaling are equivalent.) Thus \( D_i = \sigma_1 D \). This shows that the information dimension gives the scaling of the flux-weighted median flux.

Two other salient features of \( \Gamma \) are the points \( \sigma_{\min} \) and \( \sigma_{\max} \), for which \( \tilde{n}(S) = 1 \). The highest flux encountered on the object scales as \( M^{\sigma_{\min}} \). On average there is only one point that has a flux within a factor \( e \) of this highest flux. In the same sense the lowest flux on the object scales as \( M^{\sigma_{\max}} \). The exponent \( \sigma_{\max} \) is important for fractals that grow from the diffusing flux as we will discuss later.

The scaling behavior of the flux distribution \( n(p) \) on a fractal contrasts sharply with that of the mass distribution. We saw above that the distribution of mass values in a neighborhood, \( P(M) \), is typically “isometric” for connected, statistical fractals. That is, the distribution falls off faster than any power on either side of the mean \( \langle M \rangle \), so that all positive or negative moments of the distribution scale with \( \langle M \rangle \). For fractal measures this, by our definition, is no longer true. The \( n(p) \) looks qualitatively log-normal, with apparent \( 1/p \) behavior at small \( p \). But the distribution of \( \log p \) is quite homogeneous—indeed, its relative width \( \Delta \rho / \rho \langle \log M \rangle^{1/2} \) goes to zero for large systems. This fractal-measure scaling is applicable to the distribution of currents in a random resistor network [56], the density of electronic states in an incommensurate solid [57], and to the occupied points in phase space of a chaotic dynamical system [55, 58]. It appears from our random-product approximation that the distribution of flux onto a nonuniform fractal behaves similarly. I now describe a case where this fractal-measure scaling of the flux has been quantitatively confirmed.

Recently, Cates and I [59] calculated the moments of diffusing flux in a particular case. The absorber was taken as a random walk \( (D = 2) \) in \( d \)-dimensional space. This problem can be recast as a self-avoiding polymer problem, and the moments \( p^n \) can be expressed in terms of the critical exponents of that problem.

To see this we imagine our absorber as a walk of \( M \) steps. The flux \( p_i \) onto a given site \( i \) is proportional to the number of possible random walks of some very long length \( N \) which terminate at \( i \) and which do not meet the absorber elsewhere. We denote this number as \( Z_n \). With our normalization, \( p_i \propto M^{12-d}; Z_n; \sigma_i Z_n \), where \( Z_n \) is the number of random walks without the avoidance constraint. Evidently, \( p_i = Z_i^q \) is the number of “star random walks” with \( q \) arms, all of which arrive at \( i \) without touching the absorber. We calculate the average of \( p_i^n \) over all sites of the absorber, and further, over all configurations of the absorber. This

Fig. 5: Schematic plot of the effective potential \( \Gamma(\sigma) \) describing the scaling of the flux distribution. The special values \( \sigma_{\min}, \sigma_0, \sigma_{\max} \) and \( \sigma_{\max} \) are marked. The \( f(\sigma) \) function of ref. [55] is obtained by multiplying both axes by a factor \( D \), and reflecting them through the origin.
average is evidently a sum of $Z^q$, times a normalization factor. Specifically, it is the sum over all $(q + 2)$-arm star random walks in which (i) the $q$ arms each have $N$ steps, (ii) the remaining two arms have a total of $M$ steps, and (iii) the $q$ arms all avoid the two arms.

This sum is the partition function of a star random walk with selective self-avoidance. The scaling of similar partition functions is well understood in the context of polymers [60, 61]. The corresponding behavior for the present case is

$$Z_{q+2} = Z_q M^{-\gamma(q)}.$$  \hspace{1cm} (20)

Thus, $(p^q) \sim M^{q^2 - 4/3} D^{-q}$. These $\gamma$ exponents can be calculated systematically by standard methods [62, 42, 51] in spatial dimensions $d$ near 4: $d = 4 - \epsilon$. And from their $q$-dependence the behavior of $\Gamma(\sigma)$ can be found [54]. Nontrivial fractal-measure scaling appears only in order $\epsilon^2$. At this order $\Gamma(\sigma)$ is parabolic, with a width $\Delta \sigma^2 = \epsilon^2/4$. Thus at this order the flux distribution is log-normal. (For $\sigma$ far from $\sigma_0$, $\Gamma$ must depart from the parabolic form [54].) From this width, we may determine $\sigma_0 = -1 - \epsilon^2/8$, and $\sigma_1 = 1 + 2\epsilon^2/8$. More concretely, in three dimensions the median flux-weighted flux is larger than the median site-weighted flux by a factor of order $M^{1.25}$. If instead the absorber is a self-avoiding walk, then the median flux-weighted flux is larger than its site-weighted counterpart by $\sim M^{\nu - 2/3} D \sim M^{1.17}$. The self-avoiding absorber is more nearly transparent, and thus the distribution of the flux is narrower.

Other calculations and measurements [63, 64] of diffusing flux onto fractals are underway. The implications of the special scaling properties of this flux have only begun to be explored, and I expect that many important consequences are waiting to be discovered.

3. Aggregation cluster by cluster

The preceding section dealt in a general way with various properties expected for fractal structures. In this section we consider how such structures may be produced by kinetic aggregation processes. These kinetic processes produce objects that are qualitatively different from classical “equilibrium” structures such as self-avoiding walks and random animals. I want to illustrate this difference, and then describe the simplest class of aggregation phenomena: aggregation cluster by cluster. Most colloidal aggregation seems to be of this type. I want to sketch some varieties of cluster-by-cluster aggregation and note the main effects that influence the fractal dimension $D$.

3.1. Kinetic versus equilibrium clusters

In a naive sense the ensemble of aggregates is identical to the well-known ensemble of lattice animals. These are defined as all connected clusters that can be formed on e.g. a square lattice. A typical large “animal” has a fractal dimension of about 1.56 in two dimensions [65] and 2 in three dimensions [66]. Even though aggregates are the same objects as random animals, their statistical properties are much different. We may illustrate this point using a simple Eden growth model [67]. In this model a cluster is produced kinetically by randomly adding sites next to the cluster [68]. The statistical weight of a given cluster is markedly different in this Eden model than in the random animal ensemble. To illustrate this, we consider the relative weights of two clusters grown from the origin: a line of four particles extending to the right, and a square with the origin in the upper left corner. In the lattice animal ensemble, each distinct structure is represented exactly once, so that these two have equal weights. In the Eden model, each structure has a weight proportional to the number of ways it would have grown. For the line of sites, the particles must have been added in sequence: 1, 2, 3, 4 reading left to right. But for the square, there are four possible growth histories: 1, 2, 3, 4; 1, 2, 4, 3; 1, 3, 2, 4; and 1, 4, 2, 3. The compact square configuration is evidently strongly favored. This effect increases progressively with the cluster—so much so that a typical large cluster is not fractal at all, but instead fills a region of space nearly completely [69]. Thus we are faced with a puzzle: in view of the nonfractal behavior of the simplest aggregation models, how can aggregation nevertheless lead to fractal structure?

Although a kinetic process may produce an ensemble with a biased weighting as seen above, it can also produce an equilibrium ensemble with equal weighting. Monte Carlo algorithms [70] are kinetic processes designed to sample an equilibrium ensemble without bias. To decide whether a given aggregation process gives a bias, one considers an arbitrary cluster, and asks in how many ways the process could have constructed it. If the number of ways is the same for any object, the sampling is unbiased.

3.2. Aggregation mechanisms

To construct a kinetic growth model appropriate for colloidal aggregates like those of the Introduction, we first consider the microscopic phenomena controlling an individual aggregation event: a cluster A fusing
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with a cluster B. The rate $S_A(B)$ at which two such clusters combine depends on how the cluster B moves to the vicinity of A from its initial (typically distant) location. It also depends on the rate at which particles fuse together once they are nearby. The simplest problem of this form [23] is the rate of absorption of spherical particles at concentration $c_n$ onto a partially absorbing sphere of radius $R_A$. The relative separation of any two spheres diffuses with a diffusion constant equal to the sum of those of the spheres, $\zeta_A + \zeta_n$. Since each sphere is opaque to the fluid flow around it, each diffuses according to the Stokes formula: $\zeta \sim R^{2-d}$. If the absorption begins at time $t=0$, the absorption rate attains a steady state in a time of order $R^2/\zeta$. After that, if the sphere is much larger than the absorption length $\lambda$ defined in section 2.5 above, the sphere absorbs nearly as a perfectly absorbing sphere. The absorption occurs when the distance between the centers of the spheres A and B is less than $R_A + R_n$. A volume of order $(R_A + R_n)^d$ is depleted of diffusers in a time $(R_A + R_n)^{d-2}/\zeta$. Thus the absorption rate [23] is proportional to the relative diffusion constant times this radius to the power $d-2$:

$$S_A(B) \sim (R_A^{2-d} + R_n^{2-d})(R_A + R_n)^{d-2} c_n.$$  

Since the same power, $d-2$, occurs in both factors, the overall $R$ dependence of the reaction rate cancels out, and only a weak dependence on $R_A/R_n$ remains. The cancellation is no accident; the two factors reflect the rate of absorption of diffusing momentum and of diffusing particles. This law should describe the reaction rate not only of spheres, but of fractals as well, provided they are opaque to flow and diffusing fields: $D > d-2$. Our picture of the relative reaction rates is almost unmodified by the fact that they are fractals. Thus we may adopt the classical discussion of Smoluchowski [71] to see how the cluster size distribution develops. We discuss this further below. What emerges from this treatment is that throughout the reaction, clusters fuse with other clusters of comparable size.

The reaction rates $S_A(B)$ are different from the form treated above under different conditions. One such case of interest is when the clusters move in straight-line trajectories. This is what happens in the "Knudsen regime" of long mean free paths. The reaction rate is proportional to the velocity of approach times the cross section:

$$S_A(B) \sim |v_A - v_n|(R_A + R_n)^{d+1} c_n.$$  

Sedimenting clusters, such as composite snowflakes, should follow this law, with $v \sim MR^{2-d}$. In thermal equilibrium, the velocities would be mean thermal velocities, proportional to $M^{-1/2}$.

Another situation of importance is the reaction-limited regime, in which the absorption length $\lambda$ is much larger than the aggregates. In this regime the aggregates touch many times before they react and the probability of two particles fusing is a fixed fraction of the probability that these would touch in the absence of reaction. The reaction rate then takes the form

$$S_A(B) = w Q_{AB} c_n.$$  

Here $w$ is the reaction rate for two touching particles, $Q_{AB}$ is the number of ways in which the two clusters can touch (e.g. on a lattice), and the concentration $c_n$ may be thought of as the probability that a given "origin" site of the B cluster occupies any given (lattice) point. For the counting of $Q_{AB}$ we imagine that A and B are in fixed orientations, and treat other orientations as being different clusters. Unlike the previous cases, the scaling of $Q_{AB}$ is strongly influenced by the fractal nature of A and B. Evidently, if B is a single particle, then $Q_{AB}$ is the number of perimeter sites of A, which scales as $M_A$. When A and B are of comparable mass, $Q_{AB}$ appears also to scale [72, 73] roughly as $M$. It may be [74] that the scaling of $Q_{AB}$ influences the dimension of the aggregates, which in turn influences $Q_{AB}$. We consider this point in more detail below.

3.3. Aggregation models

For most colloidal aggregation processes it is natural to suppose that typical aggregates are formed by the combination of clusters of comparable size. From this observation, one may gain a qualitative understanding for why these aggregates are fractal. A simple, solvable model embodying this idea is the "Sutherland's Ghost" model of Robin Ball [75]. In this model, pairs of single particles are linked to form dimers oriented in all directions (e.g. on a lattice). Then a pair of these dimers is selected at random and a particle of each is linked at random. In this way an ensemble of tetramers is made. Then arbitrary pairs of these are linked to make an ensemble of octomers, and so forth. Each linking is done by selecting a monomer of each cluster at random and linking these together (e.g. on adjacent sites of the lattice) in an arbitrary direction. The particles are allowed to interpenetrate freely.
To calculate the scaling of the mass of such an aggregate with its radius is a straightforward exercise [75]. The average number of bonds $q(2N)$ separating one particle from another on a $2N$-site cluster may be written in terms of the same average for the constituent $N$-site clusters, A and B. ($q$ denotes “chemical” distance, along the links explicitly included; it ignores incidental intersections.) If two particles are chosen at random on a $2N$ cluster, there is a probability $\frac{1}{4}$ that they both belong to the A cluster and $\frac{3}{4}$ that they both belong to the B cluster. In either case, the average distance is $q(N)$. There is a probability $\frac{1}{4}$ that one of the chosen particles belongs to A and the other to B. In this case the distance between them is the distance from the chosen particle on A to the particle on A linked to B, plus the distance from this to the chosen particle on B. This distance averages $2q(N)$. Taking the four cases together, one finds $q(2N) = \frac{5}{2}q(N)$. Thus $q(N) \sim N^{\log 5/2}/\log 3/2$. Since the paths connecting the points are random walks, the mean square distance between them is proportional to the $q$-distance. Combining, one finds $N \sim R^{D_0}$, where $D_0 = 2\log(2)/\log(3/2) \approx 3.4$. Like ideal branched polymers [76], these structures have a diverging density in three dimensions, but above four dimensions they are well-behaved fractals with power-law density correlations.

3.4. Effect of self-avoidance

The main flaw in this model is that it permits the particles to interpenetrate freely. The particles do not avoid each other. This self-avoidance constraint has the same qualitative effect on the aggregates as on familiar equilibrium structures like self-avoiding walks. To account for the self-avoidance in a simple way, we may modify the Sutherland’s Ghost model. The two subclusters are linked as before, but then the combined cluster is examined for self-intersections. (For concreteness, this can be implemented on a lattice.) If there are any self-intersections, the cluster is discarded. This hierarchical model [72] describes reaction-limited aggregation of equal-mass clusters. The importance of the discarding feature can be understood using the “opacity principle” described in section 2. The principle says that two such clusters will not intersect appreciably if the sum of their dimensions is smaller than that of space. Even when a particle of one cluster is adjacent to the other as in the present model, the two mutually transparent clusters intersect with only finite probability. Thus the ensemble with intersections discarded has only a finite fraction removed, and the clusters which remain must have the same scaling properties as in the original Sutherland’s ghost. Thus the self-avoidance constraint is not crucial in spatial dimensions above $2D_0 \approx 6.8$; that is, this aggregation model has an upper critical dimension of $2D_0$, in the same sense as the standard equilibrium models [19, 20].

The hierarchical model resembles these standard models in a further respect: it is itself an equilibrium model. The various clusters in the ensemble appear with equal probability. This is because there is only one way each cluster can be constructed. A given cluster can only be made from one specific A and B subcluster. There is only one way in which the connecting particle of each can be chosen and one direction in which the connecting link can be made. (The structure must be specified by its bonds, not just its sites.) The ensemble thus produced is not the ensemble of animals, but only that subset that is “binary decomposable”: each cluster can be divided exactly in half by cutting one bond. The effect of the self-avoidance constraint is to straighten out the branches and to favor attachment near the ends. We expect the heuristic Flory model [77, 78] to give a reasonable estimate for the fractal dimension below the upper critical dimension. This gives $D = (d+2)/[2(d+1)/D_0]$. The Flory estimate for random animals (or randomly branched polymers [76]) is given by setting $D_0 = 4$. Thus, reaction-limited equal-mass aggregation is expected to have a dimension slightly lower than that of random animals. Since it is an equilibrium ensemble, its fractal dimension is not affected if the branches are allowed to be flexible; in any case the clusters form an unbiased sample of the allowed configurations.

Other variants of the hierarchical model describe growth by diffusing [16] or ballistically [17, 79] moving clusters. In the diffusive version, the two clusters are chosen, placed at a large distance, and allowed to diffuse until they touch [16]. Equivalently, one may choose a pair of particles on each subcluster and form the aggregate as in the Sutherland’s ghost model. But then, one of the subclusters is made to execute a random walk to infinity. If the two subclusters intersect during this walk, the combined cluster is discarded. Now the opacity principle applies to the one cluster and the path swept out by the other: this path is an object constructed by replacing each site of the cluster by the random walk of sites. Its fractal dimension is the dimension of the cluster plus 2. For this process then, the upper critical dimension is $2D_0 + 2 = 8.8$. For dimensions between 6.8 and 8.8, these clusters must have a lower dimension than their reaction-limited counterparts. We expect this to be true as well in two and three dimensions. Since each combined cluster could have been made with various choices of the random walk, this
process does not make an unbiased equilibrium ensemble like the reaction-limited case. The same is true for the ballistic case, which has an upper critical dimension $2D_b + 1 = 7.8$.

3.5. Effect of mass distribution of reacting clusters

The ghost model has another important defect. It assumes that the reacting masses are exactly equal. Actually, the reacting masses are taken from a statistical distribution, controlled by the reaction rates $S_A(B)$ above.

To see the effect of the unequal mass on the aggregates, we generalize the ghost model above by allowing clusters of unequal mass to react. We suppose that the two reacting clusters have masses $i$ and $j = ai$, with the ratio $a$ fixed throughout the process. We may again express the chemical distance $q(i+j)$ as a weighted average of the $q(i)$ and $q(j)$ of its constituents:

$$q(i+j) = [(i/j)(i+j)]^q(i) + [(j/i)(i+j)]^q(j)$$

$$+ 2ij/(i+j)q(i) + q(j)).$$  \hfill (24)

Assuming that $q(n) \sim n^{-D}$, we have an implicit equation for $D$ in terms of the ratio $a$. If $a$ is small or large, one readily checks that $D \to \infty$ as $|\log(a)|$. Thus the disparity of the reacting masses has the potential to ruin the fractal structure seen in the simple ghost model. To see that this does not occur requires a further level of detail, which takes account of the distribution of mass ratios.

Smoluchowski's theory [71], analyzed extensively in recent years [80], shows how the reaction rates $S_A(B)$ influence the distribution of cluster masses. The theory involves two assumptions. The first is that the reaction rate $S_A(B)$ for two particular cluster configurations $A$ and $B$ is about the same for any clusters having the same masses as $A$ and $B$. Thus we may use a common reaction rate $S_{i,j}$ for all pairs of clusters with masses $i$ and $j$. From this it follows that the concentrations $c_A$ are about the same for all clusters $A$ with the same mass, so that we need only consider the total concentration $c(i)$ of clusters with mass $i$. The second assumption is that these concentrations may be represented by their spatial averages. Thus, we exclude the possibility that, say, large clusters are preferentially surrounded by small ones. Further, the Smoluchowski theory assumes that the reaction rate between two types of cluster is uninfluenced by the presence of other clusters; it is a dilute theory.

With these assumptions, we may readily express the rate of change of $c(n)$ in terms of all the $c(i)$. Each $i$-cluster reacts with $j$-clusters at a rate $S_i(j) = K(i,j)c(i)c(j)$. (Since $S_i$ is proportional to $c(i)$, the $K$'s are independent of concentration.) The concentration of product clusters increases at a rate $S_i(j)c(i)c(j) = K(i,j)c(c(i)c(j))$. The concentration of $i$- and of $j$-clusters decreases at the same rate. The effect of the $i$-$j$ reaction on the concentration derivatives $\dot{c}(n)$ is

$$\dot{c}(n) = K(i,j)c(i)c(j) \quad \text{if} \quad n = i + j,$$

and

$$\dot{c}(n) = -K(i,j)c(i)c(j) \quad \text{if} \quad n = i \text{ or } n = j.$$  \hfill (25)

Thus

$$\dot{c}(n) = \frac{1}{2} \sum_{i,j} K(i,j)c(i)c(j)(\delta_{i+\lambda n} - \delta_{i-n} - \delta_{j,n}).$$  \hfill (26)

Some features of this "coagulation equation" are easy to understand. The system starts out with all the $c(n) = 0$ except $c(1)$. As time passes, clusters develop with higher and higher $n$; the typical $n$ in the system becomes arbitrarily large. In this regime, where the typical clusters have very large mass $M$, it is natural to expect scaling behavior, viz., the distribution $c(n) = M^{n/nM}/(n/M)$. When such scaling holds, it is easy to infer the basic scaling of typical mass with time. First, the total number of particles cannot vary as $M$ changes:

$$M^n \sim \sum_n nc(n) = \int dn \ t^{nM/2}$$

$$\approx M^{n+2} \int du u^{\lambda/2}.$$  \hfill (27)

This shows that $x = -2$. We now examine how the coagulation equation transforms when the mass $M$ has increased by some factor $\lambda$, and $c(n)$ has become $c(n)/\lambda^n$. Time has also increased from $t$ to $\lambda t$. The equation at the rescaled point reads schematically

$$\frac{d\lambda}{dt} \sim \lambda^2 K(\lambda i, \lambda j)c(i)c(j).$$  \hfill (28)

Now, we suppose that $K$ is homogeneous of degree $h$: $K(\lambda i, \lambda j) = \lambda^h K(i,j)$. Extracting powers of $\lambda$ on the right-hand side, we recover the
original equation for \( \frac{dc_i}{dt_i} \):
\[
\frac{dc_i}{dt_i} = \lambda^{1+b-4} \int dt K(i,j)c_i c_j - \lambda^{1+b-4} \frac{dc_i}{dt_i}.
\]
(29)

In view of the scaling of \( c_i \), this implies \( dt_i \sim \lambda^{1-b} dt_i \) or \( M \sim \lambda^{1/(1-b)} + \) constant.

For reaction kernels \( K \) with \( b < 1 \) (like the diffusion-limited case above with \( b = 0 \)) the mass grows as a power of time. For \( b > 1 \), the power is negative; the larger the typical mass, the shorter its doubling time. The typical mass \( M \) diverges after a finite interval of time. The system is said to "gel". The cluster size distribution \( c(n) \) is often broad, especially when the system gels. Then \([81]\), \( c(n) \) for \( n < M \) goes as \( n^{-\tau} \), with \( \tau = (3+b)/2 \). Such broad distributions have the largest potential effect on the fractal dimension, since these involve the most disparate masses.

To see how the mass distribution affects the cluster sizes \([74]\), we consider the chemical-distance size \( q(n) \) of clusters of mass \( n \) produced in a short time interval \( dt \). The \( i-j \) reaction produces clusters of some average size \( q(i,j) \), which depends on the subcluster sizes \( q(i) \) and \( q(j) \).

In the ghost models discussed above, \( q(i,j) \) was a simple weighted average of \( q(i) \) and \( q(j) \). In more realistic cases it is not clear that the averages \( q(i) \) and \( q(j) \) suffice to determine the combined average \( q(i,j) \). The clusters of mass \( n \) produced in a time \( dt \) have an average size \( q(n) \) of
\[
q(n) = \frac{\sum_{i,j} \delta(n, i+j) K(i,j) c(i)c(j)q(i,j)}{\sum_{i,j} \delta(n, i+j) K(i,j) c(i)c(j)} ,
\]
(30)
or
\[
\sum_{i,j} \delta(n, i+j) K(i,j) c(i)c(j)[q(i,j) - q(n)] = 0.
\]
(31)

(The \( n \)-clusters removed in this \( dt \) are assumed to have the same size as those produced.) To see concretely how this equation determines the fractal dimension of the clusters, we consider Ball's \([82]\) refinement of the ghost models above. This model is again an ensemble of monomers which may react to form larger clusters. But now the reacting clusters are not restricted to having a particular mass ratio: all clusters may react. The aggregation is reaction limited, so that \( K(i,j) = wQ_{ij} \), as in eq. (23) above. The clusters may interpenetrate freely as before, so that any site on the \( i \)-subcluster may bond to any on the \( j \)-subcluster. The number of reaction positions \( Q_{ij} \) is thus proportional to \( ij \). The Smoluchowski equation has been solved in this case \([83]\). The \( c(n) \) have the \( i^n \) form discussed above, with \( \tau = \frac{3}{2} \). The average chemical length \( q(i,j) \) for a reacting pair is the same as for the other ghost models (eq. 24). Thus the \( q(n) \) are determined by the equation
\[
\sum_{i,j} \delta(n, i+j) [q(i,j) - q(n)] = 0.
\]
(32)
Starting from \( q(1) = 1 \), one may construct each higher \( q(n) \) step by step from this equation. If the sum is dominated by \( i < j \) and \( i \gg j \) then the resulting fractal dimension must diverge, in line with our above discussion. But in fact, if \( i < j \), the quantity in brackets is of order \( i/n \), so the integrand is of order \( i^{-1/2} \), and does not dominate the integral. Thus the fractal dimension remains finite. By solving eq. (32), one readily verifies \([82]\) that \( q \sim n^{2/D}(-R^2) \) with \( D = 4/(\sqrt{17} - 3) \).

In real reaction-limited aggregation, the reaction kernel is clearly different from this model. Its homogeneity degree \( b \) must be smaller than 2, the case just treated. For \( b \) somewhat less than 2, the region of \( i < j \) in eq. (32) becomes progressively more important. For \( i < j \) \( K(i,j) \sim j^{-b+1} \), as discussed below eq. (23). The quantity \( c(i) \sim i^{-\alpha(b+2)/2} \). When \( b \) approaches 1 from above the integral is dominated by \( i < j \), and \( D \) goes to infinity \([74]\). Some further effect must intervene to assure that \( D \approx d \).

The scaling of the reacting surface \( Q_{AB} \) clearly has a dramatic influence on \( D \). This \( D \) must in turn influence \( Q_{AB} \); more compact A and B subclusters should have a smaller reacting surface. Larger \( D \) thus makes smaller \( Q \), and presumably smaller \( b \). This mutual constraint may be the dominant effect determining both \( D \) and \( b \).

Experimentally \([84]\) one may infer \( b \) from the scaling of mass with time, as discussed above. Both this scaling and the cluster size distribution \([84]\) suggest that \( b \approx 1 \), adjacent to the singular regime described above. How and when this simple value of \( b \) arises is an important unanswered question about reaction-limited aggregation.

The combined effects of self-avoidance and the distribution of reacting masses can evidently be studied directly by simulation. The fractal nature of cluster-by-cluster aggregation was in fact discovered by such simulations \([85]\). These simulations appear to give a realistic picture of colloidal aggregates in the diffusion-limited regime \([86]\). The \( D \) values found are
scarcely distinguishable from those found in simple hierarchical models like those discussed above [72, 79, 87, 88]. For reaction-limited aggregation the distribution of reacting masses plays a central role, and the hierarchical model is less adequate [73].

4. Aggregation particle by particle

This section deals with a type of aggregation complementary to the cluster-by-cluster processes treated above. Here we consider clusters that are constructed one particle at a time. It is convenient to classify these processes, as above, according to how the particle moves to join the cluster. The case where it does not move, but simply appears at a site adjacent to the existing cluster is the Eden model treated in the preceding section. In ballistic particle aggregation [89], the particles follow straight-line paths to join the cluster. Neither of these models produces a fractal structure [69, 90], though both produce surfaces with interesting scaling properties [68, 89, 91].

If, on the other hand, each particle executes a random walk to join the aggregate, an apparently fractal structure [92, 69, 93] results. This “diffusion-limited aggregation” (DLA) model is the more interesting because of its relationship to several continuum growth phenomena in nature. All these are varieties of growth controlled by Laplacian fields, as I will describe below.

Growth from diffusing or Laplacian fields yields a wide range of patterns. These patterns may be simple convex shapes, regular dendritic branched structures, or irregular tenuous structures. Langer will discuss the regular patterns and the problems of how the system selects them in Course 10. I will concentrate on how the irregular, tenuous structures arise. I will describe the various physical realizations leading to such structures. I will outline some basic variations of the growth process and how these affect the structure. I will discuss the scaling properties of these shapes; these are much different from those of ordinary fractals, and some of them seem paradoxical. Then I will give an introduction to the theory of these structures, with soluble models, an exact “Langevin equation” for the ensemble of structures, and some bounds on the fractal dimension, which show its strong differences from the fractals we have met up to now.

4.1. Realizations

In diffusion-limited aggregation on a lattice, the probability of growth at a perimeter site next to the cluster is the probability that the random walker visits this site before visiting any other perimeter site. The probability \( u \) obeys the steady-state diffusion equation \( \nabla^2 u = 0 \), where \( u \) is some constant at infinity, and zero on the perimeter sites. The probability that a perimeter site is visited is proportional to the flux onto that site—i.e. the sum of \( u \) on neighboring sites. This quantity for a smooth surface of perimeter sites would be the normal gradient of \( u \). The relative rates of growth are thus controlled by a Laplacian field.

The fluid pattern in fig. 2 results from a growth process of this type [5]. The flowing fluid is in intimate contact with a solid which absorbs the fluid’s momentum. Flow in a gel, a porous rock, or between parallel plates has this property. For such flows, the Stokes equation is replaced by the Darcy [94] equation, analogous to Ohm’s law, relating local current or velocity \( j \) to pressure \( p: j = \mu \nabla p \). As Guyon explained in his lectures (Course 2), the mobility \( \mu \) is proportional to the viscosity of the fluid and also depends on the local geometry of the contacting solid. In an incompressible fluid, the divergence of \( j \) must vanish, so that the pressure satisfies Laplace’s equation. When a fluid of negligible viscosity displaces a viscous fluid, the pressure in the pushing fluid is practically constant. The pressure on the whole interface may be taken to be zero. A point on the interface advances at a rate proportional to \( \nabla p \) there. This flow thus corresponds closely to the diffusion-limited aggregation process. The resulting interface shows the familiar viscous fingering instability, leading to parallel fingers of the pushing fluid penetrating into the viscous fluid. When the background solid is locally random as it is in fig. 2, a tenuous pattern like DLA appears. The effects of this randomness were studied by Lenormand and Zarcone [7] and Chen and Wilkinson [7].

Diffusion-limited solidification is another phenomenon of this type [95]. Here the diffusing field may be heat or a chemical species; the local growth rate is proportional to the flux of this field. Usually, the structure forms in a regular, dendritic pattern. But Brady and Ball [10] have shown that such growth can lead to a tenuous structure with fractal scaling behavior consistent with three-dimensional DLA.

Fractal structure seems to occur in a broader range of conditions. One may, for instance, relax the requirement that the local growth rate be proportional to the flux of diffusing field. In dielectric breakdown an insulating medium separates two conductors at different electrical potential. The potential is raised until some point of the dielectric ionizes and becomes conducting. The growth of this ionized region again corresponds to diffusion-limited aggregation, except that the local growth rate is some
nonlinear function of the local electric field [69]. Niemeyer et al. [13] have used simulations to study the effect of nonlinear growth laws by modifying the DLA model to make the local growth probability proportional to a power $\eta$ of the flux. (With $\eta = 1$ the model reduces to DLA.) This power affects the fractal dimension observed: larger $\eta$ makes a smaller fractal dimension $D$.

Another nonlinear effect concerns the Darcy-law relation between velocity and pressure. In some fluids the mobility $\mu$ depends on the flow rate $v$. When the mobility is increased at higher velocities, inhomogeneities in the velocity are accentuated, and tenuous structures resembling DLA are observed [6, 96].

4.2. Factors affecting diffusive growth patterns

One expects complex structure to emerge from diffusive growth, because a smooth surface growing by diffusion is intrinsically unstable against wrinkling [97]. Any small perturbation of e.g. a spherical surface, grows exponentially. To see this directly, consider a “snowman” shape made by stacking progressively smaller spheres one upon the next, so that the stack fits into a conical envelope. We know from electrostatics that if these spheres were grounded conductors, the electric field near each would be inversely proportional to its radius. (This becomes strictly true in any dimension $d$ if the spheres are far apart.) Thus if the snowman grows by diffusion, the radius of a smaller sphere will increase faster than that of the larger ones beneath. The initially conical envelope must sharpen into a more singular shape. By contrast, if the snowman grew uniformly, the envelope would keep its conical shape. The patterns emerging from this instability depend on other properties of the system, which we outline in this section.

In a physical system, the spatial scale of the smallest structures cannot be infinitely small. In continuum systems, such as viscous fingering, the smallest scale is set by a “capillary length”, which is controlled by the surface tension of the interface. At scales smaller than this capillary length the surface potential is not constant, but is higher at more exposed points, so that the growth rate there is less than it would be otherwise. The effect is to reverse the instability at small length scales, so that sections of surface smaller than the capillary length are smooth. Langer will discuss this in more detail (Course 10). In aggregation of discrete particles (DLA), the attachment or sticking probability controls the size of the smallest structures. If the sticking is perfect, the smallest structures are the size of the particles. If the sticking probability $b$ is small enough, the aggregate is transparent to the walkers, they penetrate freely and growth occurs uniformly. There is no instability, and a compact structure results. Then the growth is equivalent to the Eden model discussed above. But the transparency achieved by small $b$ is only transitory. When the structure grows to a size $R$ and mass $M$ such that $MR^{2-d}$ is larger than $b^{-1}$, it becomes opaque, as noted in section 2.5 above. Growth at larger scales is unstable, and the fractal structure of DLA reemerges.

Other factors alter the fractal structure predominantly at the largest length scales. If the growth occurs from a gas of random walkers of density $u(\infty)$ rather than from a single walker, then the rate of growth is not vanishingly small relative to the motion of the walkers. The time derivative in the diffusion equation is no longer negligible. The length scale $B$ associated with this time derivative is called the diffusion length. If the growth advances at a speed $v$, then $B \sim \zeta/v$, where $\zeta$ is the diffusion constant. In DLA with a nonzero density of walkers, the radius of the aggregate is of order $B$ when $[98]$ the average density of the aggregate decreases to that of the initial gas of walkers. For much smaller aggregates, the density of walkers is negligible, and the structure is the same as though there had only been one at a time. Aggregates much larger than the length scale $B$ have local spatial correlations like DLA, but are globally of uniform density. Thus $B$ gives a practical upper limit on the range of DLA scaling behavior.

Another factor relevant to the large-scale structure of DLA is sticking anisotropy. Sticking is anisotropic when the sticking of one particle to another is favored for certain directions in space. Twofold anisotropy means that the sticking probability $b$ is larger in, say, the x and $-x$ directions than in the perpendicular directions. Any twofold anisotropy in the growth is amplified, and the result is a needle-like asymptotic shape, with ever increasing ellipticity [99]. Growth on a lattice entails anisotropy, since growth occurs only along nearest-neighbor directions. Meakin [100] has studied three-, four- and sixfold anisotropy in two dimensions by examining the asymptotic shape of the aggregate grown on lattices of various coordination numbers. Threefold and fourfold anisotropy lead to dendritic growth: the aggregate becomes a rosette of spear-like dendrites, one for each favored direction. The regular side-branches of dendritic growth are also observed, and any fractal structure is restricted to small length scales. For sixfold anisotropy, i.e. triangular lattices, the largest simulations to date have not observed the emergence of dendritic growth. Sticking anisotropy of any amplitude or number of
preferred directions appears to be amplified in deterministic diffusive growth \cite{101, 111}. But with the local randomness inherent in discrete DLA, the ultimate effect of the lattice is only apparent in large clusters of tens of thousands of particles. Lenormand and Zarcone, and Chen and Wilkinson \cite{7} have studied the interplay between lattice anisotropy and local randomness. But there seems to be no quantitative understanding of how anisotropy and local randomness compete to control the length scale at which an anisotropic structure appears.

4.3. Scaling properties of DLA correlations

Superficially, DLA resembles the fractals we have encountered above. It has branches of all different sizes, so that small sections of an aggregate resemble larger sections. And its density correlation function $\langle \rho(0) \rho(r) \rangle$ falls off as a power of $r$. The scaling of the mass with size giving $D = 1.7$ for $d = 2$ has been confirmed over at least three orders of magnitude in length scale \cite{102}. In three dimensions $D \approx 2.4-2.5$, so that $d - D$ increases with dimension. This increase \cite{93} appears to continue to at least $d = 6$, where $d - D \approx 1$.

The properties of DLA grown from an extended substrate such as a line or plane also appear consistent with those of simple fractals such as polymers. These \cite{103} have a single length controlling the large-scale behavior. Their local correlations $\langle \rho(0) \rho(r) \rangle$, for points 0 at distance $h$ from the substrate are unaffected by the substrate for $r > h$. And their average density at height $h$, $\langle \rho \rangle_h = \langle \rho(0) \rho(h) \rangle_h \sim h^{D - D}$, for DLA clusters the measured $\langle \rho(h) \rangle$ appears consistent with this prediction \cite{104}.

Another property also may be predicted by assuming that the spatial correlations of DLA are those of a normal fractal. When DLA is grown from an extended substrate, the deposit consists of a set of interconnected trees with a broad distribution of masses $s$. Racz and Vicsek \cite{105} showed that the distribution of these masses is related to the fractal dimension $D$. Their treatment can easily be extended to a general substrate of dimension $D_1$; a linear substrate has $D_1 = 1$; a planar substrate has $D_1 = 2$.

The Racz-Vicsek reasoning uses two basic assumptions. First, an individual tree has the same height-mass scaling as in an entire DLA cluster: $s \sim h^D$. Second, the distance between two neighboring trees of height $h$ or more is of the order of $h$.

Given these assumptions, one may infer the probability $P(h)$ that an arbitrary substrate site has a tree of height at least $h$. If a given site does have such a tree, we draw a sphere of radius $h$ around that substrate site. By our second assumption above, the growth from substrate sites within this sphere is inhibited; the trees within it have height of order $h$ or less. Thus, out of $h^{D_1}$ sites, only about one has height of order $h$ or greater: $P(h) \sim h^{-D_1}$. Using our first assumption, the probability $P(s)$ that the mass of a tree exceeds $s$ goes as $s^{-D_1/D}$. The probability $n(s)$ that the mass is exactly $s$ is $n(s) = dP/ds = s^{-D_1/D - 1}$. Meakin \cite{104} has found distributions $n(s)$ consistent with this prediction.

The properties treated above show the behavior expected for a fractal. But several striking differences between the basic two-dimensional DLA and ordinary fractals have been discovered. These show that DLA has a more regular structure than classical random fractals like percolation clusters. An important structural property is the length $L$ of the path connecting arbitrary points at distance $r$ well within a large cluster. For all the classical fractals $L$ grows faster than linearly with $r$: $\delta > 1$. For DLA in two, three, and four dimensions the length of the connecting path appears to scale linearly with $r$, as Stanley and collaborators discovered \cite{106}. The connecting paths are in a sense maximally stretched out. Because these paths are stretched, we expect them to be both less random and more sensitive to the local connectivity, e.g. lattice structure.

Another unusual property of DLA is the regularity in the overall shape of the cluster. This may be seen, for example, by measuring the moment $\langle r^2 \rangle = S_x^2$ in various directions $x$ and then comparing $S_{\max}$ to $S_{\min}$. For an ordinary fractal such as a random walk, there is anisotropy of order unity: $S_{\max}/S_{\min}$ approaches some definite value greater than 1. But in DLA \cite{107} the ratio may approach unity. In off-lattice simulations, the shape seems to become more and more circular. In on-lattice simulations, the dendrites which emerge may have increasingly equal length.

Other information about the outer boundary concerns the “active zone”—the region where particles are currently being absorbed \cite{108}. The next particle to be added to a large cluster has a probability distribution of distances from the center with a spread of order $R$. This must be true for any fractal structure with open spaces of order $R$ connecting it with the exterior. But the most active sites need not be so broadly distributed. To see this we consider an “asterisk” aggregate made of, say, six radial needles. There the active zone has a width of order $R$. But if each site is weighted with, say, the cube of its absorption probability, then the width of the distribution is of order $R^3$. The width of the active zone depends on the definition of activity used.

Other recent results concern the local homogeneity and isotropy of DLA. The classical fractal structures can be shown to be locally
homogeneous and isotropic [109]. This property concerns correlations
\( p(r_0)p(r_1) \cdots p(r_n) \) for the \( r \), confined within a neighborhood of radius
\( R \) at distance \( T \) from the center of an aggregate of radius \( S \). A definition
of local homogeneity is that such correlation functions become independent
of the position of the neighborhood (thus independent of \( T \)) in the
limit \( R \ll T \) and \( R \ll S \). "Finite-size" corrections to this limit are of the order \( (R/T)^{\text{power}} \) or \( (R/S)^{\text{power}} \). Local anisotropy means that the correlations
above are invariant under an overall rotation of the vectors \( r \), up
to similar finite-size corrections.

DLA clusters grown from a point have a distinct center and in this
sense they lack homogeneity. The average mass \( \langle M(R) \rangle \) measured at the
center is systematically higher [110] than that around an arbitrary point
of the aggregate. In this respect, a DLA cluster resembles a star polymer
[111], a well-understood scale-invariant structure. A structure may have a
distinct center in this sense, yet still be locally homogeneous and
isotropic in the sense just defined.

Meakin [102], Kolb [112] and Meakin and Family [113] have investigated the isotropy of DLA numerically by comparing local two-point correlations in the direction toward the seed and in the perpendicular
direction. The data suggest that the correlations fall off with a steeper
power in the perpendicular direction. It will be important to clarify this
effect, since this picture violates the simple fractal ideas used to under-
stand several properties discussed above. This picture seems to suggest,
for example, that the width of DLA trees grown from a surface becomes
much narrower than their height.

4.4. Theory of particle-by-particle aggregation

Some fundamental features of stochastic growth can be captured in a
solvable model, which might be called "Eden’s ghost". This model is a
variant of the Eden model treated above, but like the "Sutherland’s ghost" model, it ignores the mutual avoidance of the cluster with the
deposited particles. In this model as in the Eden model one adds particles one by one to a particle at the origin of a lattice to form a connected
cluster. The next particle is added at random adjacent to an existing
particle; that is, a cluster particle is chosen at random, a nearest-neighbor
site is then chosen at random and the new particle is put there. In contrast
to the Eden model, the particles freely interpenetrate so that many may
occupy a given site.

The scaling properties may be found by considering a space-dependent probability \( G(r, t+1) \), probability that the \((t+1)\)st particle is at the lattice
site \( r \). Any particle \( t_i \) preceding the \((t+1)\)st may be the one picked to gain a neighbor. The probability that any one is picked is \( 1/t_i \). The probability that this one is next to \( r \) is \( G(r+n, t_i) \), where \( r+n \) is the neighboring site. Finally, the probability that the site \( r \) will be chosen out of the \( z \) possibilities is \( 1/z \). Combining these, we find

\[
G(r, t+1) = \frac{1}{z} t^{-1} \sum_{t_i=1}^t \sum_n G(r+n, t_i). \tag{33}
\]

This can readily be expressed as a differential equation by multiplying through by \( t \) and then subtracting the resulting equation at two adjacent
time steps. One finds

\[
\frac{\partial G}{\partial (\log t)} = (a^2/z)^{1/2} G. \tag{34}
\]

Evidently, the \( G \) function obeys the random-walk equation, but with \( t \)
replaced by \( \log(t) \). Now instead of \( t \sim R^2 \) we have \( t \sim \exp(R) \); the mass
grows faster than any power of the radius. This is not a fractal object in
any usual sense; but it is a useful point of departure for studying many
growth models. This model is closely related to the impenetrable Eden
model on a Cayley tree, treated by Parisi and Zhang [114], and by Bradley
and Stenknis [115].

The fractal behavior of DLA can be obtained by perturbing the solvable
Eden’s ghost model described above. We call this variant “penetrable
DLA” [116]. In the ghost model each particle was added at random next to
existing particles. In penetrable DLA a random walker is launched at a
great distance from the existing cluster as in ordinary DLA. Whenever
the walker arrives next to a cluster particle, it may remain there and join
the cluster. This sticking event occurs with only a small probability \( b \).
The probability that the walker is absorbed upon visiting a site is the
number of particles on adjacent sites times \( b \). If the walker is not absorbed,
it continues to walk through the aggregate. It need not avoid the aggregate.
Evidently, if \( b \) is small enough, the walker may traverse any finite cluster
many times before being absorbed. Then the growth probability at a site
is simply that of the penetrable Eden model. After about [116] \( 1/b \)
particles have grown, a walker touching the cluster has a probability of
order unity of being absorbed. From then on the interior is strongly
screened, and the growth takes on the fractal properties of DLA. This
shows that DLA behavior does not require an explicit limitation of
multiple occupancy. This contrasts with the classical fractals, such as
self-repelling walks, which owe their nontrivial scaling properties to a suppression of multiple occupancy.

We have noted the similarity between DLA growth by the adsorption of random walkers and other forms of growth controlled by diffusing fields. We now make this connection explicit by deriving exact growth equations for DLA in which the random walkers are replaced by a deterministic diffusing field [69, 13, 116]. These growth equations completely specify the ensemble of clusters by giving the statistical weight of any cluster [117, 118].

A particular aggregate may be specified by giving the density $\rho$ for all lattice sites $x$. For ordinary DLA with perfect sticking, $\rho$ is zero or one; for the penetrable DLA introduced above, $\rho$ may span a large range of (nonnegative) values. To characterize the ensemble of aggregates we must somehow give the relative statistical weights of various aggregates ($\rho(x)$). For this it suffices to give the probability $P_{\rho}(y)$ that for a particular aggregate ($\rho(x)$), the next particle is added at a given point $y$. With this, one can in principle compute the probabilities of all $(N+1)$-particle aggregates given the probabilities of all $N$-particle aggregates, and hence determine the absolute probability of a given aggregate.

The probabilities $P_{\rho}(y)$ can be readily computed, because any cluster with a given density profile (no matter how it grew) has the same probability to gain its next particle at $y$. In computing this probability, we are considering the ensemble of all possible ways in which the next walker can be adsorbed onto the given aggregate, i.e., the ensemble of all random walks around that cluster.

The probability that growth occurs at $y$ on a particular time step $t$ is the probability that the walker is at $y$ then, times the conditional probability that a walker there is absorbed. The probability that the walker is present is $u(y, t)$. This probability may be found explicitly by solving a diffusion equation. The absorption effect occurs between steps of the random walk. If the walker is now at $y$, it terminates with a probability $Q(y) = b \sum_n \rho(r + n)$. (If $Q$ is understood [116] to be 1.) Given $u(y, t)$, the probability that the walker survives the random absorption is $u'(y, t) = [1 - Q(y)] u(y, t)$. This $u(y, t)$ is related to $u'$ on the neighboring sites at the last time step:

$$u'(y, t) = [1 - Q(y)] u(y, t)$$

$$= [1 - Q(y)] \sum_n u'(r + n, t - 1).$$

We may rearrange and express this equation in terms of the lattice Laplacian $\Delta^2 u' = \sum_n u'(r + n) - 2u'(y)$:

$$\Delta^2 u' = [1 - Q(y)] \Delta^2 u' + z Q(y) u'.$$

(36)

From $u'$ we may determine the growth probability $P_{\rho}(y)$. The probability of growth at time $t$ is just the probability of absorption $u(y, t) Q(y) = \{Q(y)/[1 - Q(y)]\} u'(y, t)$.

The total probability of growth is a sum over $t$:

$$\{Q(y)/[1 - Q(y)]\} \sum_t u'(y, t);$$

this is $P_{\rho}(y)$. Performing the sum over $t$ on the $u'$ equation yields

$$0 = [1 - Q(y)] \Delta^2 u'(y) + z Q(y) u(y),$$

(37)

where

$$Q(y) = b \sum_n \rho(r + n) = [2z b u(y) + \Delta^2 \rho].$$

(38)

Then, the growth probability is given by

$$P_{\rho}(y) = \Delta^2 u(y)/z.$$  

(39)

We note that the diffusion equation of this problem is in the steady state limit—the time derivative has disappeared. This is because there is strictly no growth of the aggregate in the time before the next walker is adsorbed [13]. The limit of perfect absorption is attained when the sticking parameter $b = 1$. In the limit $b \rightarrow 1$, $u(y)$ goes to zero for all sites adjacent to occupied sites; the $u(y)$ equation may be thought of as a Laplace equation with the boundary condition that $u = 0$ adjacent to the aggregate.

We have seen above that DLA scaling properties occur even when $b < 1$; then both $\rho$ and $u$ vary smoothly in space, and the growth rate $P(y) = \langle \partial \rho/\partial t \rangle$ can be expressed in a quasi-continuum language. Equations (37) and (39) become

$$\Delta^2 u = z b u + z \Delta^2 \rho$$

(40)

and

$$\langle \partial \rho/\partial t \rangle = z^{-1} \Delta^2 u.$$  

(41)

The language suggests further simplifications, which we mention below. This formulation provides a natural way to describe lattice anisotropy. The lattice enters the equations through the use of the lattice $\Delta^2$ instead of the true differential $\nabla^2$. The lattice effects are thought to arise from the diffusing field $\nabla^2 u$, but rather from the confinement of cluster particles
to a lattice, i.e. $\hat{\nabla}^2 \rho$. The finite difference $\hat{\nabla}^2$ may be expanded in true derivatives $\nabla^2$, $\nabla^4$, etc. For sticking confined to an oriented triangle of sites, discussed by Meakin [100], there is a nonzero $\nabla^3$ correction. For a square lattice, the lowest-order correction is $\nabla^4$. For a triangular, six-coordinated lattice, the lowest correction is of order $\nabla^6$.

Beyond these alternative descriptions of DLA, there are a few rigorous limitations on its scaling properties [69]. One is a bound [90] on the rate of growth, and hence on the fractal dimension. It is obtained by considering a large DLA cluster growing in a finite but very small concentration $u(\infty)$ of walkers. The outermost particle in the cluster has a distance $S(t)$ from the seed. This $S$ may increase only if a walker is absorbed on the outermost particle. But the rate of absorption onto such a particle is limited. It can be no greater than the rate of absorption onto the initial seed. The total rate of absorption $M$ is also constrained. The aggregate absorbs the diffusers like a perfect spherical absorber whose radius is of order $S$. As shown above, $M = u(\infty)S^d$. Now $\frac{dS}{dM} = S/\dot{M}$. Since $\dot{S}$ is bounded above, we infer $\frac{dS}{dM} \sim S^{d-1}$. If $M$ is to grow as $S^d$, then $D$ cannot be less than $d - 1$. Thus $D$ must increase steadily as the dimension $d$ of space increases. This property distinguishes DLA from cluster aggregates, for which $D$ becomes simple to calculate and independent of $d$ in high dimensions.

This $\frac{dS}{dM}$ can also be related [119] to the flux distribution $n(p)$ for DLA. It is reasonable that the outer site at $S$ has more flux than any other site on the aggregate; we have denoted this flux as $p_{\text{max}}$. The probability that the next walker increases $S$ is $p_{\text{max}}$; it is also the average change of $S$ per unit mass: $\frac{dS}{dM} = a p_{\text{max}}$, where $a$ is the particle size. If $M \sim S^D$, then $p_{\text{max}} \sim S^{1-D} \sim M^{1-D}/S^D$. This scaling property was defined in section 2.6 above in terms of the upper limit $\sigma_{\text{max}}$ of the flux distribution; evidently, $\sigma_{\text{max}} = (1-D)/D$. This scaling has been checked by simulations [53].

In contrast with these deductive findings about DLA, a number of heuristic models and approximations have been introduced to account for its fractal structure. I list here a representative sampling of these, to serve as an introduction for further reading. The models may be grouped into four classes. First, early attempts were based on the scaling properties of equilibrium structures like random walks. Standard energy-entropy balance arguments [78] for polymer conformations were modified heuristically to take account of the diffusive growth condition [120, 121].

A second class of models may be called "shape hypotheses". They start from the premise that the essential feature needed to explain DLA scaling is some qualitative information about the form of the structure. The earliest models of this type [122] constructed regular, hierarchical structures, which resembled DLA simply in being branched structures. These structures had $D$ values qualitatively like those measured for DLA. Pietronero and Wiesmann [123] introduced a "branch competition" model with the additional feature of random branching. The recent work of Turkевич and Sheker [119] and of Ball et al. [99] likens the distribution of adsorption probabilities of DLA to that of a right-angle wedge. It accounts explicitly for the diffuse feature of the DLA growth. Halsey, Meakin and Procaccia [124] extend this idea to address the question of how the hypothetical shape maintains itself.

There has been a persistent hope that DLA, like other random fractals, should become simple to treat in high-dimensional space. One model of the high-dimensional limit is a Cayley tree. And DLA becomes much simpler in that limit; it becomes equivalent to an Eden model, since there is no screening. The growth from one site is not reduced by absorption by more exposed sites. Many properties [114, 115] can be treated exactly in this limit. A third class of model exploits the Cayley-tree limit to learn about DLA. The behavior of this Cayley-tree DLA seems at odds with numerical evidence about high-dimensional DLA [114]. The numerical data suggest that $d - D = 1$ in high dimensions. For the Cayley tree, $A = d - D$ seems to be 0 up to logarithms. Parisi and Zhang [114] have extended this picture by including screening effects approximately for less than infinite dimensions.

A final approach is based on the similarity of various diffusive growth processes, with widely varying amounts of explicit randomness. This approach could be characterized as perturbing in the randomness. To this class belong studies of the instability of a smooth surface under diffusive growth [9, 125, 126]. Complementary to this work are the continuous-density models motivated by the DLA growth equations presented above. One may take the probability $P_o(y)$ as the actual growth rate $\frac{1}{\ell \partial \eta(x)}$. This yields a mean-field model [127], and gives a scaling of density with size consistent with $d - D = 1$. Perturbing this model with randomness [128, 116] appears as a promising avenue for studying DLA.

5. Conclusion

The tenuous structures resulting from aggregation and growth hold fascination from many points of view, as these lectures have illustrated.
These structures have a controllable penetrability, which distinguishes them from simpler structures. The penetration of random walks into these structures shows the rich scaling of a fractal measure. The mutual penetration of fractal aggregates with each other and with other tenuous objects like polymers, such as in tire rubber, seems to yield unique mechanical and rheological properties, which have yet to be understood. Another unexplored domain is the regime of very large aggregates, in which flexibility is important. The characteristics of the flexible state may prove interesting for fundamental and practical reasons.

Apart from these properties, much remains to be understood about the fractal structures themselves. For structures made by cluster aggregation, the basic mechanisms determining the fractal scaling seem to be identified, but it remains for these to be incorporated into a systematic theory. For those structures made by diffusive growth, however, our understanding is comparatively meager. We are faced with several fundamental unresolvable questions about the nature of DLA. The first is to find a formulation valid at least in some limit which is explicitly scale invariant, analogous to the Sutherland’s ghost model for cluster aggregation. The second is to understand the role of randomness in determining the structure. Our understanding of DLA is in a state of flux. The recently suggested anisotropy properties described above have become obvious only with the largest-scale simulations now practical. These show that our intuitions about what features are important for accounting for DLA are not to be trusted. They cast doubt on previously accepted laws based on the notion that DLA has only one macroscopic length scale. These studies raise the suspicion that the asymptotic behavior of large DLA may be quite different than what has been observed up to now. And they suggest that the scaling of DLA may be qualitatively richer than that of structures we now understand.

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