Scaling Structure of the Surface Layer of Diffusion-Limited Aggregates

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We analyze numerical studies of the harmonic measure of diffusion-limited aggregates in two dimensions by modeling this measure by a fractal set of singularities. We discuss the relationship between the dimension of this fractal set and the strength of the singularities, and the implications of these ideas both for the dimension of the entire aggregate and also for a more complete theory of diffusion-limited aggregation.

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The nature of the surfaces of growing diffusion-limited aggregates must play an important role in determining the long-time structure of these aggregating clusters.[1-5] The harmonic measure affords a method of quantitatively characterizing the relevant properties of the surfaces of such clusters. This measure is defined (with respect to a particular cluster) as the probability $\rho(\mathbf{r})d\mathbf{r}$ of a random walker approaching the cluster from infinity first striking the cluster between the points $\mathbf{r}$ and $\mathbf{r}+d\mathbf{r}$ along the boundary of the cluster.[6] We have generated harmonic measures for typical two-dimensional diffusion-limited aggregation (DLA) clusters via numerical simulation. The primary result of this Letter is that this measure has itself a nontrivial scaling structure, and consists of a fractal set of power-law singularities. Furthermore, we are able to relate the dimension of the fractal set of singularities to the power law of the singularities, and both to the geometry and to the radius-of-gyration exponent of a growing cluster.

Consider the probability p_i for a walker to land within an interval ϵ about the point r_i on the boundary $\partial\Omega$ of a cluster Ω. From these $\{p_i(\epsilon)\}$ we can construct an infinite hierarchy of generalized dimensions D_q which describe the harmonic measure,[7,8]

$$D_q = \lim_{\epsilon\to 0}(q-1)^{-1}\log\left[\sum_{i=1}^{N(\epsilon)} p_i^q(\epsilon)\right]/\log(\epsilon). \quad (1)$$

Here $N(\epsilon)$ is the number of intervals of size ϵ needed to cover the boundary. The Hausdorff dimension of $\partial\Omega$ is given by $\lim_{q\to 0}D_q$, and the information dimension of the harmonic measure by[7]

$$\lim_{q\to 1+} D_q = \lim_{\epsilon\to 0}\left[\sum_{i=1}^{N(\epsilon)} p_i\log(p_i)\right]/\log(\epsilon). \quad (2)$$

D_2 is related to the scaling of the two-point correlation function on the harmonic measure, and the higher D_q are related to higher correlations on the measure.[9] It can easily be shown that for[7] $q>q'$, $D_q \le D_{q'}$. It has been shown recently by Makarov that D_1, the information dimension, is exactly 1 for the harmonic measure of any connected object in two dimensions.[10]

The probabilities p_i are related to the Green's function $g(\mathbf{r})$ for the Laplace equation with the boundary conditions that $g(\infty)$ is a constant and $g(\mathbf{r})=0$ along the boundary of the cluster. The probability to land in the ith interval of size ϵ will be proportional to the normal derivative of $g(\mathbf{r}_i)$, for some r_i in that interval, so that $p_i(\epsilon)=\partial_n g(r_i)\epsilon$.

We can quite simply obtain a value for $D_\infty = \lim_{q\to\infty}D_q$. From the definition (1) of the D_q we have

$$D_\infty = \lim_{q\to\infty}\lim_{\epsilon\to 0}\log\left\{\sum_{i=1}^{N(\epsilon)}[g'(z_i)\epsilon]^q\right\} \times[(q-1)\log\epsilon]^{-1}. \quad (3a)$$

For very large q we assume that the sum will be dominated by its largest term, so that

$$D_\infty = \lim_{\epsilon\to 0}\log\{\sup[g'_\epsilon(z)\epsilon]\}/\log(\epsilon), \quad (3b)$$

where g'_ϵ represents the average of g' over an interval

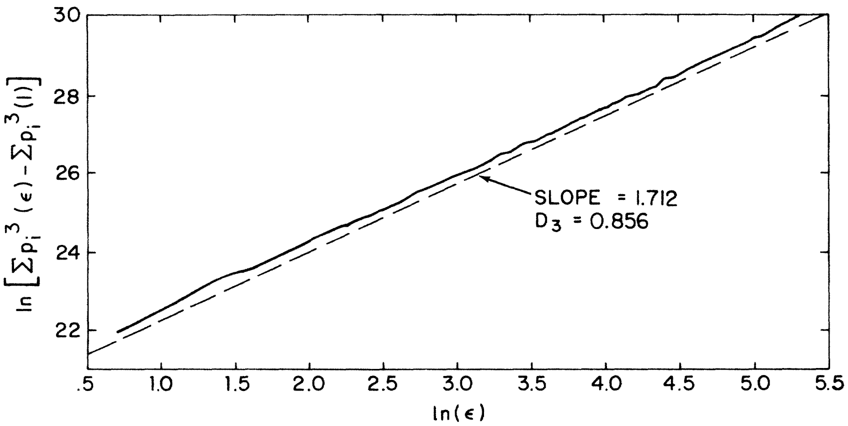

FIG. 1. A typical determination of a D_q. Here $\log[\sum p_i^3(\epsilon)]$ is plotted vs $\log(\epsilon)$.

ϵ. If we imagine that the strongest singularity in the harmonic measure along the boundary has the power-law form $(s-s_0)^{\tilde{\alpha}-1}$, then $D_\infty = \tilde{\alpha}$.

The quantities D_q are thus bounded for $q \geqslant 1$ by $\tilde{\alpha} \leqslant D_q \leqslant 1$. If we assume that there is only one type of singularity on the measure, then we can calculate all of the D_q. Of course, such an assumption must be justified *a posteriori,* which we do by examining numerical calculations of the D_q. Suppose that there are power-law singularities of the type $(s-s_0)^{\alpha-1}$ sitting on a set of fractal dimension f, and that the nonsingular portion of the harmonic measure sits on a one-dimensional set. Since the measure must be integrable, $\alpha \leqslant 1$ and by definition $f \leqslant 1$. It is now straightforward to calculate the D_q. Two types of terms contribute to the sum in (2): ϵ^{-1} terms of strength ϵ^q from the nonsingular background, and ϵ^{-f} terms of strength $\epsilon^{q\alpha}$ from the singularities. Thus,

$$\log \sum_{i=1}^{N(\epsilon)} p_i^q(\epsilon) \approx \log(\epsilon^{q-1} + \epsilon^{q\alpha - f}).$$

Since $\sum_i p_i \approx 1 + \epsilon^{\alpha - f}$, normalizability of the measure requires that $\alpha > f$. Taking the limit $\epsilon \to 0$, we have

$$D_q = \frac{1}{q-1} \min(q-1, q\alpha - f) = \min\left[1, \frac{(q\alpha - f)}{(q-1)}\right]. \tag{4}$$

This formula is in agreement with the above results for D_1 and D_∞.

We have studied the D_q numerically for the harmonic measure for large off-lattice DLA clusters. One hundred clusters were grown to a size of 50 000 particles by use of recently developed fast off-lattice algorithms.[11] The probabilities to land at various sites on these clusters were obtained by the successive launching of 100 000 random walkers at each cluster and the recording of the point of first contact (while not allowing the cluster to grow further). The values of the D_q were obtained by examination of the scaling behavior of $\sum_i p_i^q(\epsilon)$ vs ϵ. In this way we obtained values for D_q for $q = 2$ through 8. A log-log plot illustrating the determination of a typical D_q is displayed in Fig. 1. Unfortunately, the direct determination of D_1 by this method is cumbersome for numerical reasons. However, all D_q determined are less than $D_1 = 1$, which we expect from the general properties of the D_q. In Table I we show the determined values of the D_q with their estimated errors. We also show the best fit by a formula of the form of (4), for values of $\alpha = 0.705$ and $f = 0.42$. This fit is quite good; we are able to fit the determined dimensions almost within the statistical errors (see Table I). We should note that the deviations of the numerical values from the best fit, although small and generally within the statistical error bars, are systematic. We expect that the representation of the harmonic measure in terms of singularities of one type superimposed on a nonsingular background, although a good approximation, will not be exact.[12] The dimensions satisfy the inequalities $D_q \geqslant D_{q'}$ for $q < q'$ and $D_q > \alpha$. We thus conclude that the harmonic measure

TABLE I. Values of D_q obtained from the numerical simulations, and the results of the theoretical fit, Eq. (4), with the best values $\alpha = 0.705$ and $f = 0.42$. The uncertainties in the simulation column are only statistical.

q	D_q (simulation)	D_q (theory)
2	0.980 ± 0.010	0.990
3	0.856 ± 0.005	0.848
4	0.810 ± 0.006	0.800
5	0.782 ± 0.006	0.776
6	0.763 ± 0.008	0.762
7	0.748 ± 0.010	0.753
8	0.735 ± 0.012	0.746

is well described in terms of singularities of the form $(s-s_i)^{-0.295}$, with the positions of the singularities $\{s_i\}$ forming a set of dimension 0.42, superposed on a nonsingular background.

These values of α and f have some immediate implications for the geometry of the surface layer of the cluster. It is a simple exercise in conformal mapping to find the Green's function around the tip of a wedge of included angle β. If the tip is at s_0, the singularity in the harmonic measure near the tip is of the form $(s-s_0)^{\alpha-1}$, with α given by $\alpha=\pi/(2\pi-\beta)$. The value of β corresponding to $\alpha=0.705$ is $\beta=105°$. Of course, we do not see in the DLA cluster actual solid wedges including this angle; we instead expect to see ramified, low-density wedges at the largest length scale including this angle, with smaller wedges arranged on this wedge, yet smaller wedges on those wedges, and so forth in a self-similar manner, so that the fractal dimension of the set of wedge tips is $f=0.42$. Wedge models of the surface of DLA aggregates have also been proposed recently by other authors.[13,14]

If we require that these wedges now grow in such a way that their internal angles are conserved by the growth process, we can obtain a dynamical relation between α and f. In Fig. 2 we display a wedge on a large length scale, whose sides are occupied by a succession of wedges on smaller length scales. Consider the growth of the wedge shown into the larger wedge shown by the dashed lines. Clearly the smaller wedges at the front of the large wedge must grow more quickly than the side wedges if the angle is to be preserved. In fact, the ratio of the tip growth rate to the side growth rate is

$$G_t/G_s=1/\sin(\tfrac{1}{2}\beta). \tag{5}$$

Since the large wedge only grows when particles entering its area strike one of its subtips, the rate of growth in the tip region (between $-E$ and E) can be estimated as $G_t=f_tp^s/n_t$, where f_t is the total flux arriving in the tip region, p^s is the probability to hit one of the subtips in the large-tip region, and n_t is the total number of subtips in the large-tip region. The total flux arriving is proportional to $2\int_0^E x^{\alpha-1}\,dx=2E^\alpha$. The density ρ_ω of smaller wedges falls off as one moves away from the tip as $\rho_\omega(x)\approx(x)^{f-1}$. Thus the total number of subtips is $2E^f$, and the average distance between them $2E/n_t$. A particle approaching a wedge of size l has a probability to land on the tip of that wedge, which is given by

$$p_t=\int_0^a x^{\alpha-1}dx\Big/\int_0^l x^{\alpha-1}dx=(a/l)^\alpha, \tag{6}$$

where a is the size of the particles, which here serves as the ultraviolet cutoff. Thus we have that

$$G_t=2E^\alpha(2E^fa/2E)^\alpha(1/2E^f), \tag{7}$$

and by exactly the same arguments, we obtain for the growth rate of the side region (between E and $3E$, or between $-E$ and $-3E$),

$$G_s=[(3E)^\alpha-E^\alpha]\left(\frac{[(3E)^f-E^f]a}{2E}\right)^\alpha\frac{1}{(3E)^f-E^f}.$$

Since $\sin(\beta/2)=\sin(\pi/2\alpha)$, we have finally that

$$\frac{1}{\sin(\pi/2\alpha)}=\left[\frac{2}{3^\alpha-1}\right]\left[\frac{3^f-1}{2}\right]^{1-\alpha}. \tag{8}$$

This determines α as a function of f, given the requirement of self-consistent growth on all length scales. The assumption that the tip grows straight forward, while the side regions grow perpendicularly to the wedge, is rather crude. In reality, there will be a gradual modulation of the growth direction over the entire large wedge. However, we do believe that (8) offers a reasonable approximation to the true relationship between α and f, and this approximate relation is certainly physically informative. In particular, observe that three length scales were necessary in formulating this argument: the ultraviolet length scale a, the large-wedge length scale E, and the intermediate length scale of the subwedges l, which was allowed to vary.

We display in Fig. 3 the relation between α and f determined by (8). For $f<0.296$, the only solution is $\alpha=1$, which corresponds to the case of a uniformly growing smooth surface. For $f>0.296$, there is a solution with $\alpha<1$, in which $\alpha(f)$ continuously declines until it reaches $\alpha=0.59$ at $f=1$. Note, however, that we required above that $\alpha>f$ for normalizability of the measure. Thus the part of the curve to the right of $\alpha(f)=f=0.64$ is not physically relevant. We have also indicated the numerically obtained values of

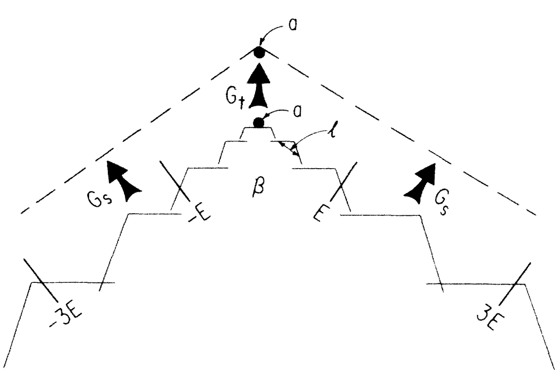

FIG. 2. The growth of a wedge that is formed from a fractal set of subwedges. The requirement that the shape remains unchanged by the growth leads to a dynamical relation between the dimension of the set of singularities and their strength. Shown are three lengths used in the theory: the ultraviolet cutoff a, the large scale E, and the intermediate scale l.

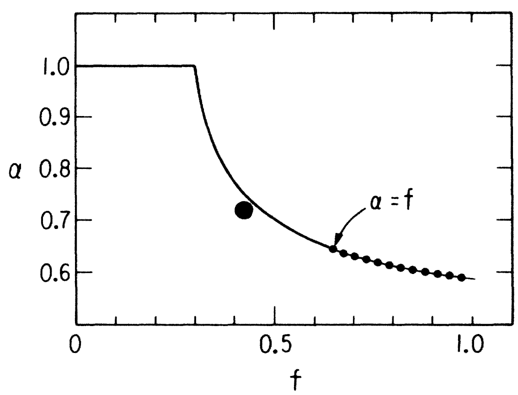

FIG. 3. The strength of the singularities plotted vs their fractal dimension f, as determined by the requirement of self-consistent growth. The dotted portion in the curve corresponds to $\alpha < f$ and is therefore unphysical. From this curve one can bound the dimension of two-dimensional DLA aggregates from below by about 1.64.

α and f on the graph. The relation (8) gives $\alpha(0.42) = 0.75$, which is within about 5% of the numerical result. Given the approximations entering into our relation, we believe that this is a reasonable error.

This model can be used to justify a relation between α and the radius-of-gyration exponent of the aggregate that has been obtained previously by Turkevich and Scher.[13] The arguments above assume that after initial transients have decayed, the cluster grows in a completely self-similiar manner. The number of largest features will be conserved by such a self-similiar growth process, which serves only to decorate these large wedges with wedges of smaller and smaller length scales. Using this fact, we can relate the dimension associated with the radius of gyration of the cluster to the exponent α. If we call the number of largest wedges n_L, then the length scale of these largest wedges will be $E = R/n_L$, where R is the size of the cluster. The rate of growth of the largest wedges is also the rate of growth of the radius of the entire cluster. Using formula (6) above for the probability of a particle hitting the tip of a wedge, we thus have for the derivative of the radius of the cluster with respect to the total number of particles N

$$dR/dN = (a/E)^{\alpha} = (an_L/R)^{\alpha}. \tag{9}$$

Since dR/dN is proportional to R^{1-d_G}, where d_G is the dimension of the cluster as estimated from the radius-of-gyration exponent, we predict $d_G = 1 + \alpha = 1.705$, in excellent agreement with the numerical result of $d_G = 1.71$ for large off-lattice simulations. Given the one-singularity approximation used in the analysis of the D_q, the accuracy of this agreement is probably, however, fortuitous. The same relation between d_G and α has been found in Ref. 13. Note that the result $\alpha(0.64) = 0.64$ effectively bounds d_G from below by 1.64.

The ideas presented in this Letter do not constitute a complete scaling theory of DLA in two dimensions. Given any one of α, f, or d_G, we are able to estimate the other two. However, we still need some numerical or experimental input. It is clear that the understanding of the harmonic measure in terms of its scaling structure does provide an important clue to the growth mechanism of DLA clusters, and may well be the foundation of a complete theory. We have argued that such a theory must deal self-consistently with three length scales, the ultraviolet cutoff, the infrared cutoff, and an intermediate running length scale.

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