# Growing interface in diffusion-limited aggregation

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When an aggregate grows by the irreversible attachment of diffusing particles, only a small fraction of the $N$ aggregated particles will collect new particles. These constitute the growing interface. We study the properties of this interface by computer simulation and analytical estimates. The mass $N_i$ of the interface grows as $N$ and to the 0.6th power in two dimensions and as $N$ to the 0.74th power in three dimensions. Several other masses characteristic of the interface are found to scale as the same power of $N$. We argue that these powers represent a critical exponent $\delta$ independent of the Hausdorff dimension.

## I. INTRODUCTION

In diffusion-limited aggregation[1–5] (DLA) particles aggregate to form a large cluster by diffusing to the cluster one at a time and sticking there. This model is thought to describe aggregation phenomena in particle dispersoids[6,7] such as soot or dust. A striking feature of DLA is that the new growth is confined to the periphery of the cluster.[4] The interior is screened from the incoming diffusing particles. Without this screening effect the aggregates could not have the tenuous, scale-invariant form which one observes. Any attempt to understand the scale invariance must thus take account of the screening. In particular, one must know how much of the cluster is in the interfacial region where growth is occurring. We find that the number of particles in the interface $N_i$ scales as the total number of particles $N$ to a power which depends on the dimension of space. The measured power is in clear disagreement with the simplest mean-field prediction, but is consistent with a naive geometrical argument.

The interfacial region investigated here should occur generally when a fractal object absorbs a diffusing field.[8] This is the situation when a long flexible polymer in solution reacts with a chemical substrate, or in electroplating of ions onto a random conductor describable as a percolating cluster. The same question arises in the electrostatic field distribution around a conducting fractal or the scattering of a Schrödinger wave from a fractal object. Thus, the problem of the diffusive interface is one of considerable practical importance.

If the Hausdorff codimension[9] $d-D$ of a fractal in spatial dimension $d$ is greater than 2, the object should be transparent to a diffusing field[4]; the screening effect is small and the whole object is exposed to the field, so that $N_i \sim N \propto R^D$, where $R$ is the radius of the object. Clearly,[4] diffusion-limited aggregates cannot have $D$ in this range. In the opposite limit of a condensed object, $D=d$, the diffusing field would be absorbed within a distance $\lambda$ of the surface where $\lambda$ is independent of $N$. Then $N_i/N \sim \lambda/R$ and thus $N_i \sim R^{d-1}$. For intermediate values of $D$ we also expect $N_i$ to vary as a power of $N$, which we denote as $\delta$. It is not clear how $\delta$ must vary, or even whether it can be expressed in terms of $D$ alone. $D$ values in this intermediate regime occur in the important cases of percolation clusters and flexible macromolecules.

To define $N_i$ precisely we consider the growth of a cluster initially of "mass" $N$ particles. An $(N+1)$th particle is placed at a large distance from the cluster and it walks randomly in space until it touches the cluster. It then becomes part of the cluster. Particles $N+k$ added subsequently to the cluster have a decreasing probability $P_N(k)$ of touching the first $N$. The number of particles which touch the first $N$ as $k \to \infty$ attains some finite limit $N_i(N)$. Evidently the average $N_i$ can be expressed in terms of the $P_N(k)$,

$$\langle N_i \rangle = \sum_{k=1}^{\infty} P_N(k) \ . \tag{1}$$

We call $\langle N_i \rangle$ the average mass of the interface. The mass attains this asymptote over some characteristic range of $k$. We may define this range as the mass $k_{1/2}$ required to half saturate the interface,

$$\frac{\langle N_i \rangle}{2} \equiv \sum_{k=1}^{k_{1/2}} P_N(k) \ . \tag{2}$$

Another way to define a characteristic range $k_0$ of $k$ is

$$P_N(k_0) = e^{-1} \ . \tag{3}$$

We expect these three measures of the interfacial "mass"—$N_i$, $k_{1/2}$, and $k_0$—to increase indefinitely with $N$. Thus $P_N(k)$ must scale with $N$ for large $n$:

$$P_N(k) = P_N(1) p(k/k_0(N)) \ , \tag{4}$$

where $k_0$ is some function of $N$. The scaling function $p(x)$ is independent of $N$. Evidently, $p(0)=1$. Further, $P_N(1)=1$, since the first particle added is certain to touch one of the first $N$. Then, using Eq. (3) to define $k_0$, $p(1)=e^{-1}$. In view of this scaling behavior, $\langle N_i \rangle$, $k_{1/2}$, and $k_0$ must become proportional for large $N$. From Eq. (1)

$$\langle N_i \rangle = k_0 \int_0^{\infty} dx\, p(x) \ ,$$

and

 

$$\langle N_i \rangle /2 = k_0 \int_0^{k_{1/2}/k_0} p(x)dx \; .$$

Some of the qualitative behavior of $p(x)$ can be anticipated. If the interface consisted of $N_i$ sites each being independently filled with equal probability, then $p(x)$ would be a simple exponential.[10] The ratio $\langle N_i \rangle / k_{1/2}$ would approach 1. In a given cluster the actual mass of the interface will show statistical variations from its average $\langle N_i \rangle$. In the simple model above, where the placement of each particle $N+k$ is an independent event, $\langle (N_i - \langle N_i \rangle)^2 \rangle$ is of order $N_i$.[11]

The spatial dilation invariance of DLA clusters shows up chiefly in the density correlation function $\langle \rho(x)\rho(y) \rangle$, which appears[1,3] to vary as $(x-y)^{D-d}$. It is natural to ask whether the interface has any special dilation invariance of its own. We are thus led to consider the spatial distribution of the $N_i$ particles in a typical interface of a cluster of mass $N$. The density $\rho_i(x)$ of interface particles has a correlation function $\langle \rho_i(x)\rho_i(x+r) \rangle$ which can readily be measured. Since we are interested in the dependence of the separation $r$, we may simply measure the average density of interface particles $C_i(r)$ at distance $r$ from an arbitrary interface particle. The simplest behavior of $\rho_i$ would be that it has no correlations beyond that of $\rho$ itself. If we treat $\rho_i$ as merely that subset of $\rho$ lying within some smooth surface representing the interface region, we would find a local interface density $C_i(r)$ equal to the total density $C(r) \simeq r^{D-d}$ times the fraction of sites at $r$ lying on the interfacial surface. This fraction falls as $r^{-1}$. Thus we expect $C_i(r) \simeq r^{D-d-1}$. If the interfacial surface has a width $\lambda$, then $C_i$ should become equal to $C(r)$ for $r \ll \lambda$.

## II. SIMULATION

We measured the interface exponent $\delta$ in two and three dimensions using the lattice simulation of DLA developed previously.[1,3] The random walker makes nearest-neighbor steps on a cubic lattice and becomes part of the cluster when it is adjacent to another cluster site. In the two-dimensional study nine large clusters of about 9400 particles per cluster were generated. The Hausdorff dimension $D$ of these clusters, measured by $N \simeq R^D$, was[12] $1.72 \pm 0.06$, taking $R$ to be the radius of gyration and using the last 50% of the intermediate clusters obtained during the growth of our final clusters. This result is in good agreement with our earlier[3] value for $D$ of $1.73 \pm 0.06$. The mass $N_i$ of the interface was determined for $N$ ranging in increments of 20 from 200 particles up to 70% of the final mass of each cluster. The lower limit was selected to avoid large statistical fluctuations in the smaller clusters, and the upper limit was selected to ensure that the interface was always saturated, so that the $k \rightarrow \infty$ limit had been substantially attained. The log-log plot of Fig. 1 shows that $N_i(N)$ is well described by a power law. The $\ln(N_i)$ vs $\ln(N)$ data from each of the nine clusters were least-squares fitted to a straight line to obtain the exponent. The nine values of $\delta$ averaged to $0.603 \pm 0.021$. The power is thus in good agreement with that expected

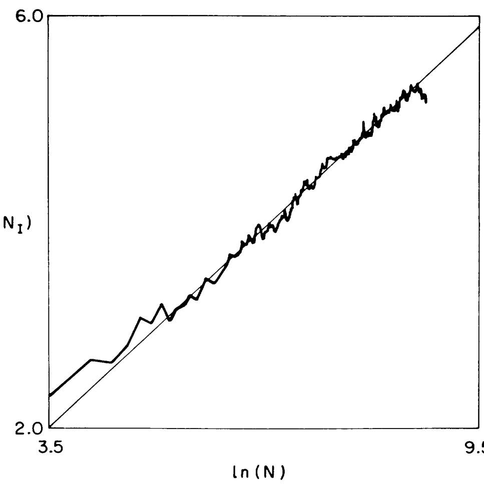


FIG. 1. Average mass of the interface $N_i$ vs the cluster mass $N$ for two-dimensional clusters. Each $N_i$ is the average over six clusters.

for a condensed object, viz., $N_i \simeq R$. The mass $k_{1/2}$ required to half saturate the interface was found (Fig. 2) to be proportional to $N_i$, as anticipated. The data in Fig. 2 are on a 9045-particle cluster with $N$ ranging from 20 to 7000 in steps of 20. The ratio $N_i/k_{1/2}$ was about 0.9, in near agreement with the hypothesis that $P_N(k)$ is a simple exponential. Our preliminary measurements of $P_N(k)$ itself show a slower-than-exponential falloff for large $k$. The statistical fluctuations of $N_i$ and $k_{1/2}$ are also evident in Fig. 2. The variance of either quantity is roughly proportional to the quantity itself. Thus, e.g., $\langle (N_i - \langle N_i \rangle)^2 \rangle \simeq 3N_i$. This is in agreement with our hypothesis of statistical independence.

The bunching of the points in Fig. 2 is noteworthy. It indicates significant memory effects in the growth of the cluster. We believe this is related to the instabilities present in dendritic growth,[13] a continuum counterpart of DLA.

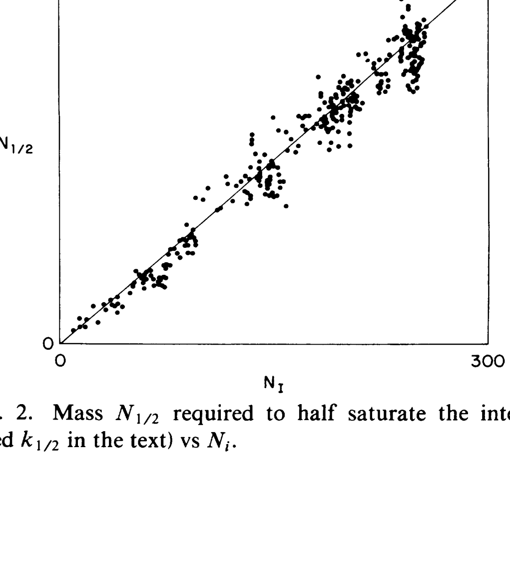


FIG. 2. Mass $N_{1/2}$ required to half saturate the interface (denoted $k_{1/2}$ in the text) vs $N_i$.

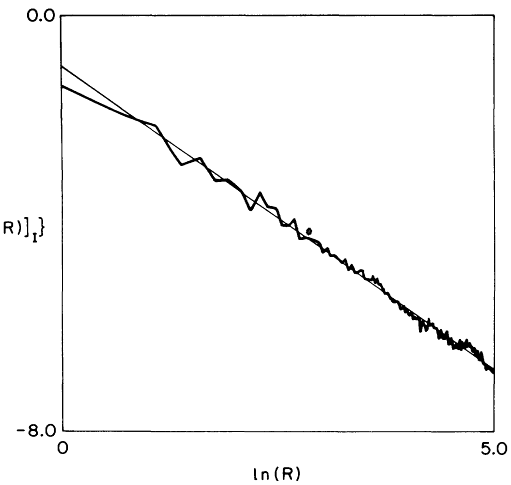


FIG. 3. Interface correlation function $C_i(r)$ defined in the text plotted vs the distance $r$ for two-dimensional aggregates.

The function $C_i(r)$ giving the correlations of the interface density $\rho_i$ is plotted in Fig. 3. For this plot the mass $N$ ranged from 35% to 70% of the final mass of the cluster. For $r<50$ lattice sites, the $C_i(r)$ had a form independent of $N$ and followed a power law: $C_i(r) \sim r^{-B}$. The average power for the six clusters was $B=1.13\pm0.07$ for $5 \leq r \leq 50$. The expected power for distances $r$ much greater than the interface thickness is $B=4/3$. For distances $r<5$ the $C_i(r)$ tended to fall below the power-law line as we would expect for $r \lesssim \lambda$, but no clear short-distance power-law behavior was evident.

Nine clusters averaging 7600 particles each were used for our three-dimensional study. These had $D=2.52 \pm 0.08$ as determined by the radius of gyration, in agreement with $D=2.49\pm0.08$ from previous work.[3] Again the mass $N_i$ of the interface varied as $N^{\delta}$, with $\delta=0.744\pm0.018$. Here the fitting was performed over the range $N=100$ up to half the mass of the cluster. For a condensed cluster, with $N_i \sim R^2$, one would have $\delta=0.74$–$0.82$. This prediction is not inconsistent with the data. In three dimensions the ratio of $N_i$ to $k_{1/2}$ was about 0.8; the plots of $N_i$ vs $k_{1/2}$ show the same scatter and the same bunching of points seen in two dimensions. We did not study the correlations of the interface in three dimensions.

Clusters grown using an Eden[14] model were also analyzed. In the Eden growth process, particles are added with equal probability to any vacant site adjacent to the cluster (including interior vacant sites). The radius-of-gyration exponent $D$ is shown for various sizes of clusters grown by the Eden mechanism in Table I. From these data we conclude that the limiting ($N \rightarrow \infty$) value of $D$ is 2, i.e., equal to the Euclidean dimensionality $d$. In this sense the Eden clusters are "classical." This table also shows the results obtained for the interface exponent $\delta$. In the case of the Eden clusters, our measure of the interface size is the number of unoccupied sites next to cluster sites. The results shown in Table I indicate that there is a significant finite size effect for the exponent $\delta$ for Eden clusters. However, the results indicate that the limiting $N \rightarrow \infty$ value for $\delta$ is the classical value of 0.5.

TABLE I. Growth exponents in the Eden model.

| Mass $N$ (thousands) | Number of clusters | Hausdorff dimension $D$ | Interface exponent $\delta$ |
|---|---|---|---|
| 50–100 | 6 | 2.0064±0.003 | 0.529±0.05 |
| 25–50 | 14 | 2.0096±0.002 | 0.521±0.022 |
| 12.5–25 | 21 | 2.0173±0.003 | 0.542±0.035 |
| 5–10 | 21 | 2.0305±0.008 | 0.552±0.04 |

## III. DISCUSSION

Our various measurements of the mass of the DLA interface are for the most part consistent with our simple estimates made in the Introduction. But two of our findings require further discussion. The first is the behavior of the correlation function $C_i(r)$. The observed behavior—$r^{-1.1}$—appears inconsistent with the prediction of $r^{-1.33}$ assuming no special interface correlations. The predicted power requires $r$ to be much larger than any skin depth $\lambda$. If this condition were not fulfilled, one would see an effective exponent closer to 0, as we did. In view of this effect, we see no evidence for special interface correlations.

A more serious problem is the interpretation of the measured exponents $\delta$. In two dimensions the measured exponent is in good agreement with the condensed-object behavior $N_i \sim R^{d-1}$. In three dimensions the measurements are also consistent with this law. We have not been able to understand this behavior in a simple way, in view of the known properties of fractal density profiles. The problem becomes immediately apparent if we use mean-field methods to predict $\delta$. To this end we replace the density $\rho(r)$ at distance $r$ from the origin of an $N$-particle aggregate by its ensemble average. This density falls off as $r^{D-d}$ out to the cluster radius $R$, and falls quickly to zero beyond. A steady-state diffusing field $u(r)$ gives the probability that the random walker arrives at $r$. The diffusing field is absorbed by the cluster, the local absorption rate being proportional to the probability $\rho(r)$ that a cluster particle is present. Thus $u$ obeys

$$0=\nabla^2 u - \text{const} \times \rho(r) u \ . \tag{5}$$

For distances less than $R$, $u$ falls off exponentially in a distance $\lambda$. The scaling of $\lambda$ with $\rho$ may be found by using $d^2/dr^2$ for $\nabla^2$ and $\rho(R)$ for $\rho(r)$. Then $u(r) \sim u(R) e^{-|r-R|/\lambda}$ with $\lambda^{-2} \propto \rho(R)$. The interface particles are thus added over this depth. We expect the mass of the interface $N_i$ to be proportional to the total number of particles within the skin depth $\lambda$:

$$N_i \propto \frac{\lambda}{R} N \sim [\rho(R)]^{-1/2} \frac{N}{R} \ . \tag{6}$$

Since $\rho(R) \sim N/R^d$, this gives

$$N_i \sim N^{1/2} R^{d/2-1} \sim N^{1/2+(d/2-1)/D} \ . \tag{7}$$

In this mean-field approximation the power $\delta$ is predicted to be 0.5 in two dimensions (cf. 0.6) and 0.7 in three dimensions (cf. 0.74). The mean-field model neglects correlations: It treats the cluster as a cloud of independent particles. Each particle at distance $r$ has an equal chance of absorbing the diffusing field. The real aggregate has clustering at all length scales. This clustering should make a particle at distance $r$ less effective in absorbing the diffusing field than it would otherwise be. Thus the total absorption rate should be reduced.[15] If the clustering were only local, so that mean-field theory could still be used for the aggregate as a whole, a reduced growth rate would mean an increased skin depth and, hence, an increased interfacial mass. We expect the same to be true in the actual case, where clustering occurs on all length scales, and the skin depth $\lambda$ becomes difficult to define. Thus the mean-field prediction of Eq. (6) should *under*estimate $N_i$, i.e.,

$$\delta \geq 1/2+(d/2-1)/D \equiv \delta_{\rm MF} . \tag{8}$$

This agrees with our empirical observations. Along with this lower bound on $N_i$, there is the upper bound $N_i < \text{const} \times N \sim R^D$, i.e., $\delta \leq 1$.

These bounds show that $N_i$ can vary as $R^{d-1}$ within, at most, a limited domain. Since $\delta \leq 1$, we can only have $N_i \sim R^{d-1}$ as long as $D \geq d-1$. In fact, $D$ must remain strictly greater than $d-1$. Otherwise, if $D=d-1$, then $\delta=1$ and the interface is a substantial fraction of the aggregate. Mean-field theory becomes qualitatively valid, and it can be used to estimate $\delta$. From Eq. (8) this would imply $D=d-2$, contradicting the supposition that $D=d-1$. Thus, if $D=d-1$ for some $d$, the law $N_i \sim R^{d-2}$ cannot hold.

If $D$ merely approached $d-1$ as $d \rightarrow \infty$, as recent work suggests,[16] we still expect $N_i \sim R^{d-1}$ to break down. Otherwise, $N_i/N \sim R^{d-1-D} \rightarrow R^0$, and, again, mean-field theory should become more and more nearly applicable. However, mean-field theory predicts $N_i/N \sim R^{(d-D)/2-1} \rightarrow R^{-1/2}$, again contradicting the supposition.

## IV. CONCLUSION

This work shows that DLA clusters have a characteristic scaling property besides their Hausdorff dimension, namely, the mass of the interface. Various measurements of this mass are consistent with each other. This mass in turn scales with a well-defined power $\delta$ of the total mass $N$. However, we have been unable to account for this power. It seems likely, rather, that this power cannot be expressed in a simple way in terms of the Hausdorff dimension of the aggregate and the dimension of space. Studies of the absorption of diffusive fields by other fractals would clarify this question. Such studies would be of practical interest as well.

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10The probability $P_N(k)$ that the $k$th particle arrives on a remaining interface site is

$$P_N(k)=\frac{\langle N_i \rangle - \sum_{k'=1}^{k-1} P_N(k')}{\langle N_i \rangle} ,$$

so that

$$dP/dk=(-1/N_i)P_N(\kappa) .$$

11We let $q_N(k)$ be the random variable—1 or 0—whose average is $P_N(k)$. Then

$$\langle N_i \rangle = \sum_k \langle q_N(k) \rangle ,$$

$$\begin{aligned} \langle N_i^2 \rangle &= \sum_{k,k'} \langle q_N(k) q_N(k') \rangle \\ &= \sum_{k,k'} \langle q_N(k) \rangle \langle q_N(k') \rangle + \sum_{k,k'} \langle q_N(k)^2 \rangle - \langle q_N(k) \rangle^2 \\ &= \langle N_i \rangle^2 + \sum_k P_N(k) - P_N^2(k) , \end{aligned}$$

where we have used $\langle q^2 \rangle = \langle q \rangle$. Both of the sums here are of order $\langle N_i \rangle$ as claimed.

12The uncertainties given in this paper are 95% confidence limits (1.3859 standard deviations) and include only statistical contributions to the uncertainty.

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15To illustrate the screening effect, we solve Eq. (5) as an expansion in powers of the density $\rho$. This gives

$$u(r)=u_\infty - u_\infty \int d^d r' \frac{\rho(r')}{|r-r'|} + O(\rho^2) .$$

We treat the cluster region of radius $R$ as having an average density $\langle \rho \rangle$, so that $\rho(r)=\langle \rho \rangle + \delta\rho(r)$. Then the average absorption rate $\langle I \rangle$ can be written

$$\langle I\rangle \equiv \int_{|r|<R} d^d r \langle u(r)\rho(r)\rangle$$
$$=\langle \rho\rangle u_\infty R^d - \langle \rho\rangle^2 u_\infty \int_{|r|,|r'|<R} \frac{d^d r\, d^d r'}{|r-r'|}$$
$$-u_\infty \int_{|r|,|r'|<R} d^d r\, d^d r' \frac{\langle \delta\rho(r)\delta\rho(r')\rangle}{|r-r'|} .$$

In the absence of correlations among the particles, the last term vanishes. With clustering, $\langle \delta\rho\,\delta\rho\rangle$ is positive at short distances and the last term reduces the absorption rate.

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