Spectral Dimension for the Diffusion-Limited Aggregation Model of Colloid Growth

Paul Meakin
Experimental Station, Central Research and Development Department, E. I. DuPont de Nemours and Company, Inc., Wilmington, Delaware 19898

and

H. Eugene Stanley
Center for Polymer Studies and Department of Physics, Boston University, Boston, Massachusetts 02215
(Received 4 August 1983)

The spectral dimension $d_s = 2d_f/d_w$ is calculated for the diffusion-limited aggregation model of colloids and dendritic growth; here d_f is the fractal dimension of the aggregate, and d_w the fractal dimension of a random walk on the cluster substrate. $d_s = 1.2-1.4$ is found for $d=2$ and 3, to within the accuracy of the present Monte Carlo calculations. Thus Witten-Sander aggregates may possess the same remarkable "superuniversality" discovered for percolation clusters and argued to possibly hold for all homogeneous fractals.

PACS numbers: 64.60.Cn, 05.70.Jk, 66.10.Cb, 82.70.-y

A wide range of natural phenomena occur in spaces of noninteger dimension.[1-6] Such "fractal phenomena" are of tremendous current interest. Most of the attention thus far has focused on characterizing the *geometrical* properties of fractals. However, it is of greater importance to seek to discover how the fundamental laws of nature are modified for fractal objects. For example, the laws of diffusion in Euclidean space involve only a single length scale, the rms displacement ξ_w, which is related to the number of steps in a random walk N_w through the fractal dimension of the walk,[5]

$$N_w \sim (\xi_w)^{d_w}. \tag{1a}$$

When diffusion occurs on a fractal, there is a *second* length scale, the "radius of gyration" ξ_f, which is related to the "mass" N_f through the fractal dimension

$$N_f \sim (\xi_f)^{d_f}. \tag{1b}$$

For Euclidean spaces $d_w = 2$ for *all* values of ξ_w, while we must distinguish two ranges of ξ_w.[5] If $\xi_w \gg \xi_f$, then $d_w = 2$, while if $1 \ll \xi_w \ll \xi_f$, then ξ_w may in general be expected to take on a value that depends on the system under consideration. Moreover, interesting parallels exist between classical diffusion and quantum localization, as can be seen[6] by translating results from Euclidean lattices[7] to fractals.

On what specific features of the substrate fractal does d_w depend? For percolation clusters, an example of *homogeneous* fractals,[4] Alexander and Orbach made a remarkable numerical discovery.[8] While d_w indeed depends strongly on d, the spectral dimension $d_s = 2d_f/d_w$ appears to be independent of d (for $d \geq 2$); this discovery has been confirmed by careful studies using Monte Carlo simulations[9] and exact enumeration procedures[10]; to the available numerical accuracy, $d_s \simeq \frac{4}{3}$. In contrast, for the Sierpinski sponge, an example of a *nonhomogeneous* fractal, d_s depends on d $\{d_f = [\ln(d+1)]/\ln 2$, and $d_w = [\ln(d+3)]/\ln 2\}$.[6] Could a general feature of fractals be that d_s is independent of d for *all* homogeneous fractals? *If so, the homogeneous fractals would possess a marvelous "superuniversality" that would distinguish them in a fundamental way from nonhomogeneous fractals.*

Here we address this intriguing question by considering Witten-Sander aggregates,[11-15] which are *homogeneous* fractals distinctly different from percolation clusters (and also different from lattice animals,[12] another type of homogeneous fractal). Witten-Sander fractals have been used to describe irreversible aggregation phenomena ranging from soot particles to colloid growth.[11,13] By obtaining good statistics from extensive direct Monte Carlo simulations of very large fractals, we find that d_s is approximately the same for $d = 2$ and $d = 3$, suggesting that d_s is independent of d (unlike percolation, however, there may be no upper critical dimension).

Our calculation consists of two steps, the first being the generation of a fractal and the second being the generation of a random walk on the fractal. Step one has been described elsewhere.[14] One places a seed particle on a lattice site at time $t = 1$. At $t = 2$ a second particle is released from a random point on a hypersphere surrounding the seed particle and allowed to undergo a random walk until it reaches a perimeter site of the seed.[16] At $t=3$, a third particle is released, and this process continues until typically at times of order 10^4 fractals with a large number ($N_f = t$) of particles have been formed. The results of computer simulations suggest that $d_f \cong \frac{5}{6}d$ for small d,[14] and this finding has been interpreted theoretically.[15] For our purpose here, nine large clusters (averaging 9568 particles per cluster) were generated for $d=2$, and eleven clusters (averaging 7662 sites) were obtained for $d=3$. In addition, five *very* large (25 000-site) $d=3$ clusters were generated.

The second step is to simulate random walks on each cluster. To do this, we select at random one of the cluster sites to be the origin.[17] We randomly choose one of the neighboring cluster sites to be the first step of the random walker, and then continue this process until a long random walk has been generated. We started by generating large numbers ($\simeq 10^4$) of short walks (200 steps for $d=3$ and 1000 steps for $d=2$) to obtain good statistics and avoid the outer (not fully developed) regions of the Witten-Sander clusters. However, we found that the effective exponents describing the random walks were dependent on the length of the walks[18] and that longer walks could be taken without significant sampling of the outer regions. Consequently, several thousand walks of up to 2^{13} (8192) steps were generated on each of the $d=2$ clusters and several thousand walks of up to 2000 steps were generated on the $d=3$ clusters; walks of 5000 steps each were used on the very large (25 000-site) $d=3$ clusters. Figure 1 displays a walk of 2500 steps on a very small (1000-site) $d=2$ fractal.

Figure 2 shows double logarithmic plots of ξ_w^2 vs N_w, for typical walks on $d=2$ and $d=3$ Witten-Sander aggregates. From the definition (1a) we expect the data to be linear for $1 \ll \xi_w \ll \xi_f$; a least-squares fit to the linear region for longer walks gives the values $2/d_w = 0.78 \pm 0.03$ for $d=2$ and 0.6 ± 0.05 for $d=3$ corresponding to

$$d_w = \begin{cases} 2.56 \pm 0.10, & d=2, \\ 3.33 \pm 0.25, & d=3. \end{cases} \tag{2}$$

Both statistical uncertainties and our estimates of systematic uncertainties due to finite-size effects, etc., are included in the error limits.

To determine the spectral dimension $d_s = 2d_f/d_w$, we calculated d_f for the *same* aggregates used in this study. Using the definition (1b), we found $1/d_f = 0.580 \pm 0.019$ for the nine $d=2$ clusters and 0.398 ± 0.012 for the eleven $d=3$ clusters. These results are in good agreement with earlier

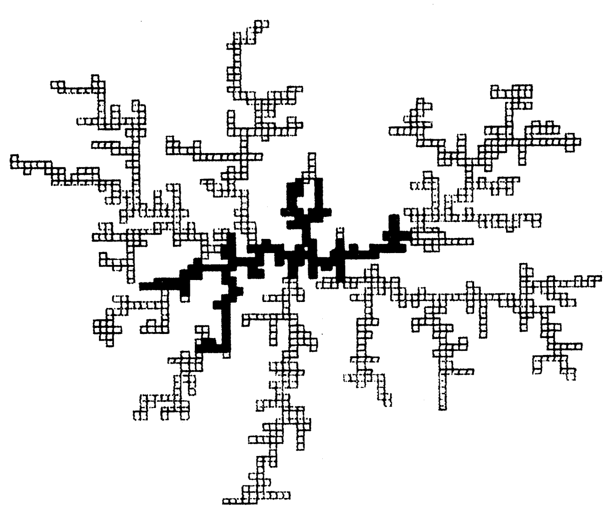

FIG. 1. A random walk of 2500 steps on a fractal with 1000 sites (a small aggregate). The sites visited by the walk are indicated by heavy lines.

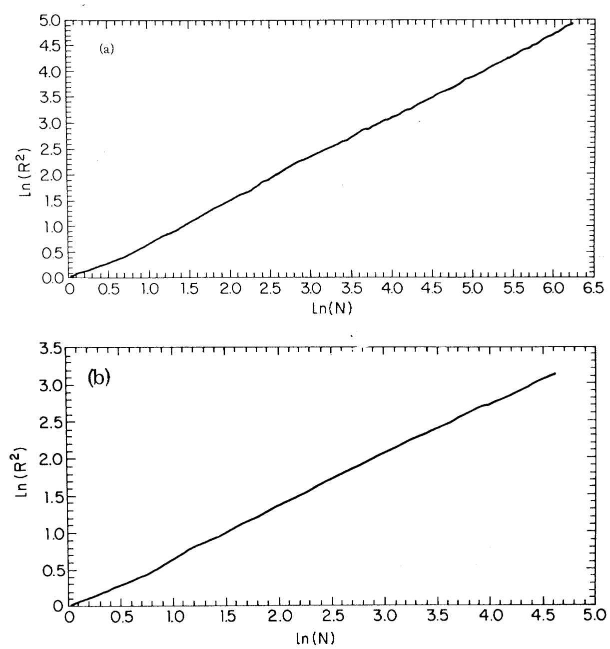

FIG. 2. Double-logarithmic plot of dependence of mean square end-to-end distance ξ_w^2 on the number of steps in the walk, N_w, for walks on (a), a $d=2$ fractal ($d_f \simeq \frac{5}{3}$), and (b), a $d=3$ fractal ($d_f \simeq 2.4$). The slope of the straight line is a measure of $2/d_w$, where d_w is the fractal dimension of the walk. This particular $d=2$ cluster had 6540 sites, while the $d=3$ cluster had 7076 sites.

work.[11,14] A more detailed analysis[19] of the five large (25 000-site) $d=3$ clusters indicates that $d_f \simeq 2.4 \pm 0.1$ ($1/d_f = 0.416 \pm 0.016$) and it is this result that we use to analyze our $d=3$ data. From these results we find

$$d_s = \begin{cases} 1.35 \pm 0.10, & d=2, \\ 1.44 \pm 0.20, & d=3. \end{cases} \tag{3}$$

Thus our results are consistent with the concept that d_s is independent of dimension.

We also calculated the mean number of sites visited by the random walk,[6]

$$\langle s \rangle \simeq (N_w)^{d_s/2}, \tag{4}$$

so that d_s can be obtained directly without the need to measure d_f. The plots for typical $d=2$ and $d=3$ walks are shown in Fig. 3. Averaging over all walks and all clusters studied, we find

$$d_s = \begin{cases} 1.20^{+0.10}_{-0.05}, & d=2, \\ 1.30 \pm 0.06, & d=3. \end{cases} \tag{5}$$

To check for systematic trends for longer walks, we carried out two additional types of calculation. The first of these consisted of binning data by powers of two. The binned data displayed little

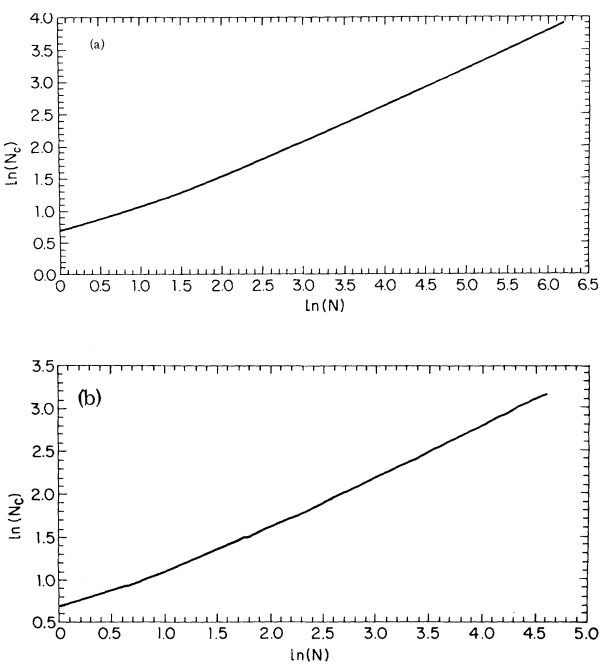

FIG. 3. Double-logarithmic plot of the dependence of the mean number of sites visited $\langle s \rangle$ on N_w for typical walks on the same $d=2$ and $d=3$ fractals shown in Fig. 2. The slope of the straight line is a measure of $d_f/d_w = d_s/2$ where d_s is the spectral dimension (Ref. 8).

scatter, and the exponents showed no significant tendency to increase or decrease for longer walks. The second type of calculation was for "higher moments." For example, Eq. (1a) implies that the mean of the *fourth* power of the displacement scales as $(N_w)^{4/d_w}$ so that measurements of this quantity provide an estimate of d_w that weights the longer walks more than the shorter walks, and is *independent* of the value obtained from Eq. (1a). Similarly, Eq. (4) implies $\langle s^2 \rangle \sim (N_w)^{d_s}$. Our calculations for these higher moments give essentially the same results as in (2)–(4), increasing our confidence that the walks studied were sufficiently long to provide reliable exponent values.

Finally we calculated the probability of return to the origin by measuring the fraction, F_0, of walks that return to the origin after a walk of length N_0. One expects that[6,8]

$$F_0 \sim (N_0)^{-d_s/2}, \tag{6}$$

and we estimate

$$d_s = \begin{cases} 1.20 \pm 0.1, & d=2, \\ 1.30 \pm 0.1, & d=3. \end{cases} \tag{7}$$

In summary, we have introduced the problem of random walks on Witten-Sander aggregates, which is of interest because one can determine the spectral dimension d_s for a random fractal. Our results support the concept that *all* the properties of a random walk on a homogeneous fractal substrate can be described in terms of the fractal dimension (d_f) of the substrate and the spectral dimension ($d_s = 2d_f/d_w$). In addition, our results are consistent with the conjecture that the spectral dimension is independent of the Euclidean dimension (d). However, our data could also be interpreted to indicate a slightly larger spectral dimension for $d=3$ than $d=2$. We are hoping to carry out larger-scale simulations to obtain more accurate estimates.

We thank S. Redner for a useful conversation. This work was supported in part by the National Science Foundation, the U. S. Army Research Office, and the U. S. Office of Naval Research.

[1] See the recent review, C. Mitescu and J. Roussenq, to be published, and references therein [particularly P. G. de Gennes, Recherche 7, 919 (1976)].

[2] A. Kapitulnik and G. Deutscher, Phys. Rev. Lett. 43, 1444 (1982).

[3]R. F. Voss, R. B. Laibowitz, and E. I. Allessandrini, Phys. Rev. Lett. 49, 1441 (1982).
[4]F. Leyvraz and H. E. Stanley, to be published.
[5]Y. Gefen, A. Aharony, and S. Alexander, Phys. Rev. Lett. 50, 77 (1983).
[6]R. Rammal and G. Toulouse, J. Phys. (Paris), Lett. 44, L13 (1983); see also A. Coniglio and H. E. Stanley, to be published.
[7]E. Abrahams, P. W. Anderson, D. Licciardello, and T. V. Ramakrischnan, Phys. Rev. Lett. 42, 673 (1979).
[8]S. Alexander and R. Orbach, J. Phys. (Paris), Lett. 43, L625 (1982).
[9]R. B. Pandey and D. Stauffer, Phys. Rev. Lett. 51, 527 (1983); S. Havlin and D. Ben-Avraham, to be published.
[10]H. E. Stanley, K. Kang, S. Redner, and R. L. Blumberg, Phys. Rev. Lett. 51, 1223 (1983).
[11]T. A. Witten, Jr., and L. M. Sander, Phys. Rev. Lett. 47, 1400 (1981), and Phys. Rev. B 27, 5686 (1983).
[12]H. Gould, F. Family, and H. E. Stanley, Phys. Rev. Lett. 50, 686 (1983); S. Wilke, Y. Gefen, V. Ilkovic, A. Aharony, and D. Stauffer, to be published.
[13]J. Deutch and P. Meakin, J. Chem. Phys. 78, 2093 (1983), and references therein.
[14]P. Meakin, Phys. Rev. A 27, 604, 1495, 2616 (1983).
[15]M. Muthukumar, Phys. Rev. Lett. 50, 839 (1983); R. Ball, M. Nauenberg, and T. A. Witten, to be published; M. Tokuyama and K. Kawasaki, to be published.
[16]If the mobile particle reaches a position that is a great distance from the growing cluster before reaching the cluster, then a new particle is released and the process continued until an aggregate of two particles has been formed.
[17]The origin of each random walk was selected to avoid biasing the walk by reaching either the center of the cluster, which may be atypical, or the outer portions of the fractal, whose growth is not complete. The former "potential systematic error" is the less serious since density correlation measurements show that the center of the cluster is probably not atypical. Typically, for $d=2$, the origin was selected at random from the subset of sites that had become occupied when the cluster was between 10% and 30% complete (the corresponding numbers for $d=3$ are 10% and 25%). Since d_w is so large, the origin and outer portions of the clusters are rarely visited by the overwhelming majority of the walks.
[18]Similar trends have very recently been found by Pandey and Stauffer (Ref. 9) and by Havlin and Ben-Avraham (Ref. 9) for diffusion on percolation clusters.
[19]P. Meakin and Z. R. Wasserman, to be published.