RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 30, NUMBER 7 1 OCTOBER 1984

Space-filling constraint on transport in random aggregates

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(Received 11 May 1984)

Transport properties such as conductivity on scale-invariant structures such as random aggregates are governed by a spectral dimension exponent $\tilde{d}$. We investigate the behavior of $\tilde{d}$ for branched structures without loops using geometric constraints on their current-carrying backbone. The requirement that the structure fit into d-dimensional space imposes a new constraint on $\tilde{d}$. The distance Δ between branches of the backbone must be of the order of the span N measured along the backbone. Under mild scaling assumptions this distance Δ controls the conductance. We express $\tilde{d}$ in terms of the fractal dimension D and the scaling power δ relating N to the geometric size L ($N \sim L^{\delta}$): $\delta = D(2/\tilde{d}-1)$. This implies that $\tilde{d} < \frac{4}{3}$ for such branched structures with $D < 2$.

Transport on tenuous random structures such as percolating clusters or diffusion-limited aggregates appears to have remarkably general scaling properties. Alexander and Orbach[1] have argued that conductivity, diffusion, and scalar waves on such structures may be described by a single new exponent called the fracton or spectral dimension $\tilde{d}$. This exponent describes the number of distinct sites $S(t)$ visited by a t-step random walk on the structure: $S \sim t^{\tilde{d}/2}$ (for[2,3] $\tilde{d} < 2$). Remarkably, the spectral dimension appears to have nearly the same value $\frac{4}{3}$ for a wide class of structures, including percolation clusters at length scales smaller than the correlation length[1] and diffusion-limited aggregates.[4] Rammal and Toulouse[2] and Leyvraz and Stanley[5] have argued that this value may arise from some qualitative "homogeneous" characteristic of these structures.

In this Rapid Communication we consider transport on random *branched* structures, such as diffusion-limited aggregates[6] or cluster aggregates.[7,8] These aggregates are defined by irreversible growth processes, and have a negligible number of loops at large scales, as illustrated in Fig. 1. The absence of loops puts constraints on the transport properties, as we show below. Our argument requires certain scaling properties of the sort normally assumed for these scale-invariant structures.[1,9] Given these properties, we may relate the spectral dimension to purely geometrical scaling properties. Further, the spectral dimension and the fractal dimension[10] D relating mass M to geometric size L ($M \sim L^D$) are constrained by an inequality $D \geq 1/(2/\tilde{d}-1)$. A branched structure which violated this inequality would be expected to overfill space of any finite dimension. To our knowledge, such a relation between transport and the filling of space has not previously been realized. In view of our inequality $\tilde{d}$ must be smaller than $\frac{4}{3}$ at least when $D < 2$. Since $D < 2$ for both diffusion-limited aggregates and cluster aggregates in sufficiently small dimensions[7,8,11,12] the arguments leading to $\tilde{d} = \frac{4}{3}$ apparently do not apply to these.

Other authors[13,14] have identified relations between structural and transport properties for branched structures. One such property is the point-to-point conductance. This property is sufficient to determine the spectral dimension in a branched structure with a finite order of ramification.[10] But in the random structures of interest here the order of ramification is not known.

In a uniform medium the point-to-point conductance determines the conductivity of the medium and thence the spectral dimension. But for more general structures, the point-to-point conductance does not suffice to describe transport in other geometries. In a Cayley tree, for example, the point-to-point conductance is inversely proportional to the number of bonds N in the path connecting them.

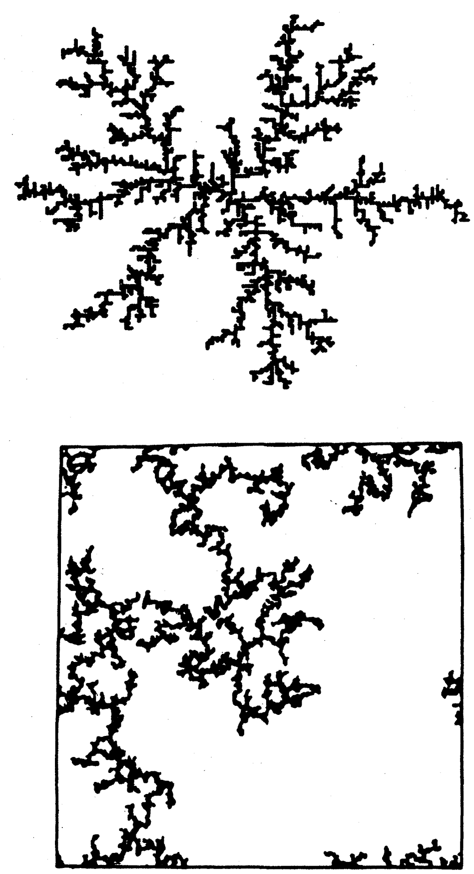

FIG. 1. Diffusion-limited aggregate (top) and cluster aggregate (Ref. 7) (bottom) in two dimensions. The absence of loops on all but smallest scales is evident.

But the conductance between a point and a surrounding sphere is independent of the size of the sphere. It is this latter conductance which is related to diffusion and to the spectral dimension. This point-to-sphere conductance is the quantity considered here. Our space filling constraint implies that the point-to-point and point-to-sphere conductances should scale the same for physical structures, in contrast to the Cayley tree. Thus, our result justifies the widely made assumption that various conductances may be treated as equivalent in practice.

To describe transport on a branched structure, we choose a point on it at random, and draw a large spherical boundary of radius L around it. The entire structure is assumed to be much larger than the bounding sphere. Then we may investigate transport, e.g., by measuring the conductance $\Sigma(L)$ between the center and the sphere. The conductance[1] scales with L according to

$$\Sigma(L) \sim L^{D(1-2/\tilde{d})} \quad . \tag{1}$$

Most of the original structure consists of dead ends, with no path to the boundary. Thus, in our conductance measurement, current flows only through a small subset of the structure called the backbone. An example of such a backbone in a diffusion-limited aggregate is depicted in Fig. 2. The backbone is itself a branched structure. But unlike the original object, each new branch on the backbone means a new path to the boundary , each with its own branches and subpaths. Unless the growth of the number of branches with increasing L is limited, the density of the backbone becomes impossibly large. To avoid this, the distance between branches must be large. We define $N(L)$ as the average chemical distance along the backbone from the center to the boundary. Equivalently, $N(L)$ is the average distance along the full branched structure between two points at geometric separation L. We suppose that $N(L)$ scales as L^δ. This power δ governs the point-to-point resistance in a branched structure. This exponent has been extensively investigated for percolation clusters: the "spreading dimension" of Ref. 15 equals D/δ, while the exponent $\tilde{\nu}$ of Ref. 16 is the inverse of δ.

First we consider a regular nonrandom structure in which all branches have the same length Δ, and the length of any path from the origin to the boundary is N. Then any path has $k=N/\Delta$ branches. This structure is a Cayley tree of coordination number three. The number of branches B_j at j steps from the origin doubles at every step, so that $B_j=2^j$. Thus, the mass $M(N)$ of this tree $\Delta\sum_{j=1}^{k} B_j$ grows exponentially with k. In order to fit into space of finite dimension, M must grow no faster than a power of N. Thus, k can grow no faster than $\ln N$ and Δ must be of order $N/\ln N$ or larger.

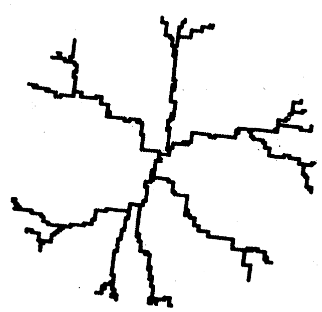

FIG. 2. Backbone of the diffusion-limited aggregate depicted in Fig. 1. Note that only a small number of original branches survive in the backbone.

This result may be extended to structures with branches of arbitrary lengths. For any given L we can define an average length Δ_j of jth generation branches. This average is obtained by dividing the sum of lengths of all jth generation branches by B_j; thus, if some of the branches are absent they contribute zero length to the average. The mass M of this structure is $\sum_{j=1}^{\infty} B_j\Delta_j$, and the average distance $\overline{N}$ to the boundary is $\sum_{j=1}^{\infty}\Delta_j$. This $\overline{N}$ is a different average than the $N(L)$ used above. For arbitrary structures as for Cayley trees the Δ_j's must increase with $\overline{N}$ to avoid overfilling space. To see this we denote the largest of the Δ_j by Δ and find the minimum mass M for the same Δ and $\overline{N}$. Evidently the smallest mass is attained when all the branches have the length Δ. (Increasing one Δ_j at the expense of a later one decreases the mass without changing $\overline{N}$.) Thus, the largest of the average lengths Δ_j in any branched structure must be of order $\overline{N}/\ln\overline{N}$ or larger,[17] as in a Cayley tree. (Clearly the Δ_j are also bounded above, since each Δ_j must be smaller than $\overline{N}$.)

Given this space-filling limitation on the lengths, we may estimate the conductance in actual random structures. To make this estimate, we must assume that the structure is sufficiently homogeneous. That is the initial Δ_j's — Δ_1, Δ_2, etc. must be of the same order of magnitude as the maximum Δ_j. Then the space-filling limitation implies that all these branches have the characteristic length Δ between order $\overline{N}/\ln\overline{N}$ and order $\overline{N}$. We presume that the point-to-point average $N(L)$ is of the same order as $\overline{N}(L)$. This is true as long as the number of branches along different paths to the boundary does not vary too much. Now for $L\rightarrow\infty$ the resistance of the backbone must increase at least as fast as this Δ but not faster than N,[18] and since both these quantities scale the same (up to logarithmic corrections) we conclude that $\Sigma\sim N^{-1}\sim L^{-\delta}$. Using Eq. (1), we may relate the scaling of Σ to the D and $\tilde{d}$ of the entire structure:

$$\delta = D(2/\tilde{d}-1) \quad . \tag{2}$$

Thus, $\tilde{d}$ may be determined by purely geometric measurements, viz., measurement of $N(L)$ and $M(L)$. This relation has been obtained for finitely ramified structures.[13,14] The formula is valid for the infinite percolating cluster in high dimensions,[19] where $D=4$, $\delta=2$, and $\tilde{d}=\frac{4}{3}$. We have shown that the formula is valid generally for branched structures.

The mass of any connected path must grow at least as fast as the radius L, but not faster than L^D; thus $1\leq\delta\leq D$. This sets bounds on $\tilde{d}$:

$$1\leq\tilde{d}\leq 2D/(D+1) \quad . \tag{3}$$

Thus, a branched structure with $D<2$ must have $\tilde{d}<\frac{4}{3}$. Aharony and Stauffer[20] have suggested that the upper bound in Eq. (3) is the actual expression for $\tilde{d}$ whenever $D\leq 2$. They used an heuristic argument which applied to any fractal structure with $D<2$ (although several counterexamples exist), while our bound is restricted to only branched structures but is not restricted to $D<2$. In view of our Eq. (2) this would imply that $\delta=1$, i.e., $N\sim L$, whenever $D\leq 2$; the branches would be qualitatively equivalent to straight lines. For diffusion-limited aggregates in two dimensions[11] $D \cong 1.7$, so that Eq. (3) implies $\tilde{d} \leqslant 1.26$. For cluster aggregation in three dimensions (Meakin 1984), the limit is about the same. For two-dimensional cluster aggregation with[7] $D \cong 1.5$, the limit is stronger: $\tilde{d} \leqslant 1.2$.

These bounds on $\tilde{d}$ are consistent with the available simulation results, and these can be used to get information about the exponent δ. Meakin and Stanley[4] give estimates of $\tilde{d}$ for diffusion-limited aggregates ranging from 1.1 to 1.45 in two dimensions, and from 1.2 to 1.64 in three. Using these, we infer for diffusion-limited aggregates that $\delta < 1.4$ in two dimensions and $\delta < 1.67$ in three.

It would be valuable to have accurate measurements of the exponent δ relating N to L. This would give precise predictions of $\tilde{d}$ and allow the Aharony-Stauffer argument to be checked. It would also be helpful to examine the statistics of backbone branches and of paths to the boundary in order to check our assumptions of homogeneity. Our arguments should be equally applicable to the elastic response[21] of a network composed of branched structures, such as silica gel.

We thank D. Pearson, P. Pincus, and I. Webman for helpful comments on the manuscript.

[1]S. Alexander and R. Orbach, J. Phys. (Paris) Lett. **43**, L625 (1982).

[2]R. Rammal and G. Toulouse, J. Phys. (Paris) Lett. **44**, L13 (1983).

[3]J. C. Angles d'Auriac, A. Benoit, and R. Ramal, J. Phys. A **16**, 4039 (1983).

[4]P. Meakin and H. E. Stanley, Phys. Rev. Lett. **51**, 1457 (1983).

[5]F. Leyvraz and H. E. Stanley, Phys. Rev. Lett. **51**, 2048 (1983).

[6]T. A. Witten and L. M. Sander, Phys. Rev. Lett. **47**, 1400 (1981); T. A. Witten and L. M. Sander, Phys. Rev. B **27**, 5686 (1983).

[7]P. Meakin, Phys. Rev. Lett. **51**, 1119 (1983).

[8]M. Kolb, R. Botet, and R. Julien, Phys. Rev. Lett. **51**, 1123 (1983).

[9]Y. Gefen, A. Aharony, and S. Alexander, Phys. Rev. Lett. **50**, 77 (1983).

[10]B. B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, San Francisco, 1982).

[11]P. Meakin, Phys. Rev. A **26**, 647 (1982).

[12]P. Meakin, Phys. Rev. A **29**, 997 (1984).

[13]S. Havlin, Z. Djordevic, I. Majid, H. E. Stanley, and G. H. Weiss (unpublished).

[14]F. Family and A. Coniglio, J. Phys. A **17**, L285 (1984).

[15]J. Vannimenus, J. P. Nadal, and H. Martin, J. Phys. A **17**, L351 (1984).

[16]S. Havlin and R. Nossal, J. Phys. A **17**, L427 (1984).

[17]Thus, e.g., for a square lattice of spacing "a" in two dimensions $M(L) \leqslant \pi (L/a)^2 \leqslant \pi N_{\min}^2 \leqslant \pi \bar{N}^2$. But $M(L)$ can be related to the maximum length Δ; $M(L) \geqslant \sum_{j=0}^{k} 2^j \Delta \geqslant 2^k \Delta$ with $k = \bar{N}/\Delta$. Combining, we find $\pi \bar{N}^2 \geqslant \Delta 2^{N/\Delta}$. Since $\Delta \leqslant \bar{N}$, this means $\Delta \geqslant \bar{N} \ln 2/\ln(\pi \bar{N})$.

[18]Our conclusion about the resistance would break down if Δ_j increased without bound as j increased. Thus, in a structure where all jth-order branches have length 2^j, resistance would increase only logarithmically with N.

[19]S. Alexander, G. Grest, H. Nakanishi, and T. A. Witten, J. Phys. A **17**, L185 (1984).

[20]A. Aharony and D. Stauffer, Phys. Rev. Lett. **52**, 2368 (1984).

[21]Y. Kantor and T. A. Witten, J. Phys. (Paris) Lett. (to be published); Y. Kantor and I. Webman, Phys. Rev. Lett. **52**, 1891 (1984).