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Multifractal spectrum of off-lattice three-dimensional diffusion-limited aggregation

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We study the multifractal properties of the set of growth probabilities $\{p_i\}$ for three-dimensional (3D) off-lattice diffusion-limited aggregation (DLA) in two distinct ways: (i) from the histogram of the p_i and (ii) by Legendre transform of the moments of the distribution of p_i. We calculate the $\{p_i\}$ for 50 off-lattice clusters with cluster masses up to 15 000. We discover that for 3D DLA, in contrast to 2D DLA, there appears to be no phase transition in the multifractal spectrum. We interpret this difference in terms of the topological differences between two and three dimensions.

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I. INTRODUCTION

The study of growth models has attracted considerable attention recently. In particular, the diffusion-limited-aggregation (DLA) model serves as a paradigm for a vast range of observed growth phenomena, ranging from electrodeposition and viscous fingering to neuron growth. DLA, on the one hand, is appealingly simple to define in terms of random walks sticking to the surface of the growing aggregate but, on the other hand, displays intriguingly complex properties [1, 2].

A detailed description of the growth process in DLA is given in terms of the set of growth probabilities $\{p_i(M)\}$, where $p_i(M)$ is the probability that site i on the perimeter of an aggregate of mass M will be occupied in the next growth step ($M \rightarrow M+1$). The set $\{p_i(M)\}$ displays a wealth of scaling properties that are conveniently described in the framework of the multifractal or thermodynamic formalism.

The multifractal spectrum has been studied extensively for two-dimensional (2D) DLA, both on-lattice and off-lattice. The spectrum displays a systematic mass dependence, corresponding to what is usually termed a "phase transition" (see Sec. II) [3–5]. To our knowledge, Ref. [6] is the only analysis of the $\{p_i\}$ of 3D DLA so far. However, the authors treated *on-lattice* DLA and obtained the p_i using a Monte Carlo technique [7], a method which cannot correctly give small values of p_i.

In this work, we report a calculation and analysis of the $\{p_i\}$ for 3D off-lattice DLA, using a numerical solution to the Laplace equation to determine the p_i. We calculate the multifractal spectrum, as well as specific subsets of $\{p_i\}$, such as the smallest and largest growth probabilities, $p_{\min}$ [3, 4] and $p_{\max}$ [8]. We carefully compare the corresponding results for the 2D and 3D cases, analyzed in the same fashion. We discover that for 3D there appears to be *no* phase transition in the multifractal spectrum, and interpret the difference in terms of the topological differences between two and three dimensions.

II. MULTIFRACTAL FORMALISM

We employ two distinct methods to calculate the multifractal spectrum from the set of growth probabilities $\{p_i\}$ [2].

(i) Histogram method. We calculate the distribution $N(\alpha, M)$ of the quantities $\alpha_i \equiv -\ln p_i/\ln M$; i.e., $N(\alpha, M)d\alpha$ is the number of growth sites in a cluster of mass M with α values in the interval $[\alpha, \alpha + d\alpha]$ [9]. Specifically, we calculate the histogram $\langle N(\alpha, M)\rangle$, where the brackets $\langle\,\rangle$ denote the average over an ensemble of DLA clusters with mass M. We define

$$f_H(\alpha, M) \equiv \ln\langle N(\alpha, M)\rangle / \ln M. \tag{1}$$

Then the "histogram multifractal spectrum" $f_H(\alpha)$ [9] is the limit of $f_H(\alpha, M)$ for $M \rightarrow \infty$, i.e.,

$$f_H(\alpha) \equiv \lim_{M\rightarrow\infty} f_H(\alpha, M). \tag{2}$$

(ii) Legendre transform method. First, we calculate the "partition function" defined as the qth moment of the distribution of p_i

$$Z(q, M) \equiv \left\langle \sum_i p_i^q(M) \right\rangle. \tag{3}$$

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We define the effective scaling exponents $\tau(q,M)$ for fixed q through the local slopes of a log-log plot of $Z(q,M)$ against M,

$$\tau(q,M) \equiv -\partial \ln Z(q,M)/\partial \ln M. \quad (4)$$

The asymptotic behavior of these local slopes is given by

$$\tau(q) \equiv \lim_{M\to\infty} \tau(q,M). \quad (5)$$

When the limit $\tau(q)$ exists, it characterizes the power-law scaling of $Z(q,M)$ as a function of M: $Z(q,M) \sim M^{-\tau(q)}$. In this case $\tau(q)$ corresponds to a free energy in the "thermodynamic formalism." If, in contrast, $Z(q,M)$ as a function of M scales faster than a power law for $q \le q_c$ so that $\tau(q,M)$ diverges, one speaks of a "phase transition" at q_c [3,4]. The multifractal spectrum $f_L(\alpha_L)$ is obtained by Legendre-transformation of $\tau(q)$,

$$f_L(\alpha_L) \equiv q\alpha_L - \tau(q), \quad \alpha_L \equiv \frac{d\tau(q)}{dq}. \quad (6)$$

III. RESULTS

We constructed and analyzed 50 off-lattice clusters with masses up to $M = 15\,000$, which we compare to previous results in 2D from 90 clusters with $M \le 3000$ and 19 clusters with masses up to $M = 21\,000$ [5]. We calculate the p_i by solving the Laplace equation under DLA boundary conditions by discretizing the clusters on a simple cubic lattice for 3D and a square lattice for 2D [5].

(i) Histogram method. The histograms $\langle N(\alpha,M)\rangle$ are shown in Figs. 1(a) and 1(b) for 2D and 3D, respectively. For 3D we found that for large M the distribution seems to converge to a limiting $f_H(\alpha)$. For 2D, however, $f_H(\alpha,M)$ clearly shows a mass dependence and no tendency to converge for large M. The explicit mass dependence in 2D can be seen directly by fitting the data for $\alpha > \alpha_0$ to the form [5]

$$\langle N(\alpha,M)\rangle \sim \exp[-A(\alpha^\gamma - \alpha_0^\gamma)/(\ln M)^\delta], \quad (7)$$

where $\gamma = 2.0 \pm 0.3$, $\delta = 1.3 \pm 0.3$. Here $\alpha_0 = \alpha_0(M)$ locates the maximum of the distribution, while γ and δ are exponents describing the asymptotic (large M and large α) behavior. Note that γ and δ determine the scaling of $p_{\min}$,

$$\ln p_{\min}(M) \sim (\ln M)^y, \quad (8)$$

where $y = (1+\gamma+\delta)/\gamma = 2.15 \pm 0.22$ [5].

(ii) Legendre transform method. We study the partition function $Z(q,M)$ for several M and q values. For 2D [Fig. 2(a)], we observe a distinct upward curvature of the negative moments, indicating that the small growth probabilities vanish faster than a power of the cluster mass M. Hence for 2D, $\tau(q,M)$—the local slope—diverges as a function of M for negative q corresponding to a phase transition at $q = q_c = 0$ in the multifractal spectrum.

In contrast, we find that for 3D an asymptotic power-law relationship $Z(q,M) \sim M^{-\tau(q)}$ holds for *all* measured values of q [Fig. 2(b)], indicating the absence of a phase transition for $d = 3$.

We obtain a sequence of estimates for $\tau(q)$ by linear least-squares fits for large M, which allows us to calculate $f_L(\alpha_L)$ according to Eq. (6). For 2D we find that the right hand part of $f_L(\alpha_L)$ has a tendency to move to larger α_L when $\tau(q)$ is determined from $Z(q,M)$ for larger masses. Such a behavior arises because the divergence of $\tau(q,M)$ for negative q also causes α_L to diverge.

For 3D, $f_L(\alpha_L)$ does not display a systematic mass dependence, so we attribute the deviations between the curves to statistical fluctuations. Moreover, the value of $\alpha_{L,\max}$ from Fig. 2(b) is in agreement with the value of $\alpha_{\max}$ obtained from the scaling of $p_{\min}$ (see below).

(iii) $p_{\min}$ and $p_{\max}$. A further test of the possibility of a phase transition can be obtained as follows. Suppose there exist mass independent values of $\alpha_{\min}$ and $\alpha_{\max}$ so that $f_H(\alpha)$ calculated from the histogram $\langle N(\alpha,M)\rangle$ is

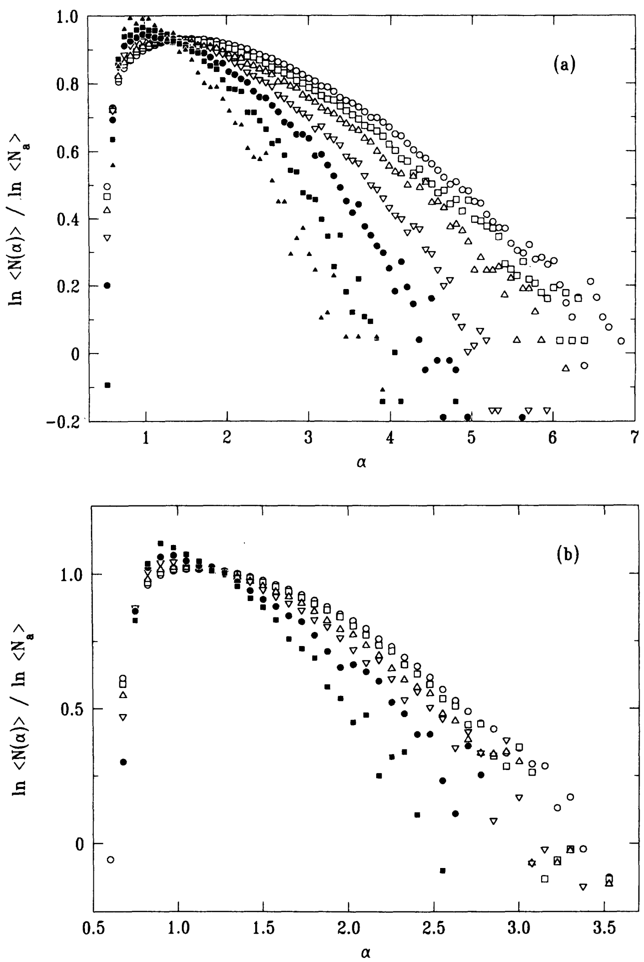

FIG. 1. $\ln\langle N(\alpha,M)\rangle/\ln\langle N_a\rangle$ for (a) 2D DLA. N_a is the number of growth sites with $p_i > 0$ and is asymptotically proportional to the cluster mass M, so that the plots reflect also the scaling properties of $f_H(\alpha)$. The 2D data points are obtained by averaging over ensembles of 90 clusters for $M = 247$ (filled triangles), 505 (■), 1030 (•), 2100 (▽), and from ensembles with 19 clusters for $M = 5250$ (△), 10 500 (□), 21 000 (○). (b) The 3D data are obtained from a set of 50 off-lattice clusters with $M = 165$ (■), 435 (•), 1117 (▽), 2892 (△), 7502 (□), and 15 015 (○).

zero for $\alpha < \alpha_{\min}$ and $\alpha > \alpha_{\max}$. Then $p_{\min} \sim M^{-\alpha_{\max}}$, $p_{\max} \sim M^{-\alpha_{\min}}$, and all other p_i display power-law scaling. Thus also the scaling behavior of $Z(q,M)$ with M must be of the power-law type and a phase transition due to the divergence of $\tau(q)$ cannot occur. In contrast, it is a sufficient condition for a phase transition if $p_{\min}$ scales faster than a power law with increasing M. Then, for $M \to \infty$ and $q < 0$, $p_{\min}^q$ dominates $Z(q,M)$ [Eq. (3)] and $\tau(q)$ [Eq. (4)] is divergent for $q < 0$. Thus a phase transition at $q_c = 0$ occurs.

First we consider $p_{\min}$ for 2D and 3D DLA. In Figs. 3(a) and 3(b) we show the mass dependence of $\langle \ln p_{\min} \rangle$ for the 2D and 3D cases. From the log-log plot for 3D, we find that power-law scaling holds, with $\alpha_{\max} = 4.3 \pm 0.2$. For 2D, we see deviations from straight line behavior in a log-log plot, favoring faster than power-law scaling. The inset of Fig. 3(a) shows $\langle \ln p_{\min} \rangle$ vs $(\ln M)^{2.15}$ [see Eq. (8)], which is linear over more than two decades of M ($100 < M < 21\,000$). The 2D and 3D results may be summarized by writing

$$\ln p_{\min} \sim -A(\ln M)^y, \begin{cases} y = 2.15 \pm 0.22 & \text{for 2D,} \\ y = 1, \; A = \alpha_{\max} & \text{for 3D.} \end{cases} \tag{9}$$

Next we consider $p_{\max}$ [inset of Fig. 3(b)]. For 3D DLA, we find $\alpha_{\min} = 0.59 \pm 0.01$. The exponent $\alpha_{\min}$ is related to the fractal dimension d_f by $d_f = 1/(1-\alpha_{\min})$ [8]. From our value of $\alpha_{\min}$ we obtain $d_f = 2.44 \pm 0.06$, which is slightly smaller than the value 2.52, which was obtained from scaling of the radius of gyration of off-lattice 3D DLA with cluster mass [2].

IV. DISCUSSION

The importance of the multifractal spectrum is based on its connection to characteristic quantities describing the growth process.

Specifically, the study of $\alpha_{\max}$ provides information

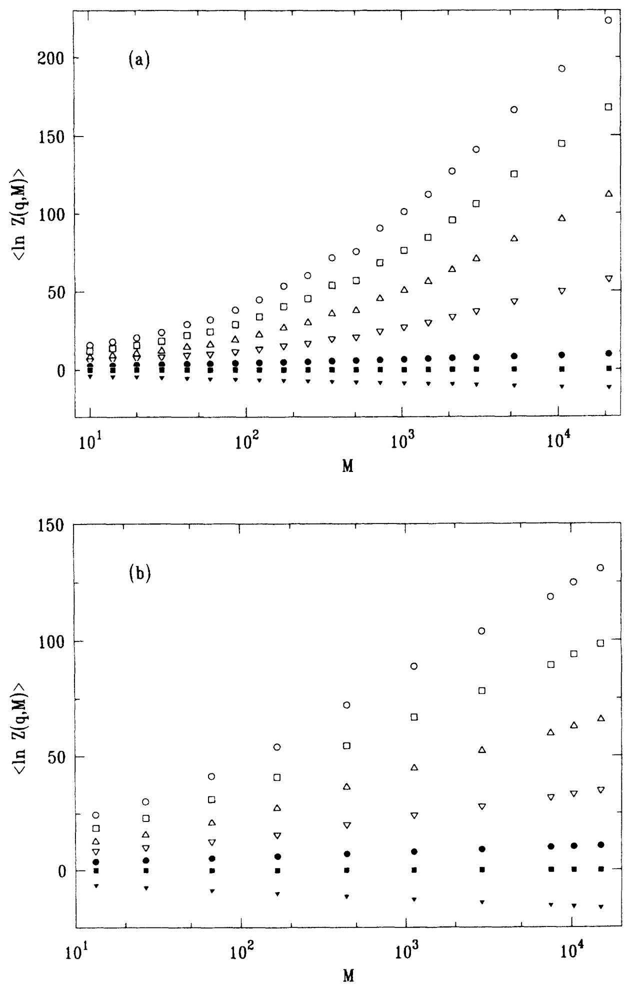

FIG. 2. Plot of $\langle \ln Z(q,M) \rangle$ as a function of N_a for (a) 2D DLA and (b) 3D DLA. Shown are ten moments $q = -4$ (○), -3 (□), -2 (△), -1 (▽), 0 (•), 1 (■), and 2 (filled triangles) for a range of masses M up to 21 000 (2D) and 15 015 (3D). The number of clusters analyzed for each mass is given in the caption to Fig. 1.

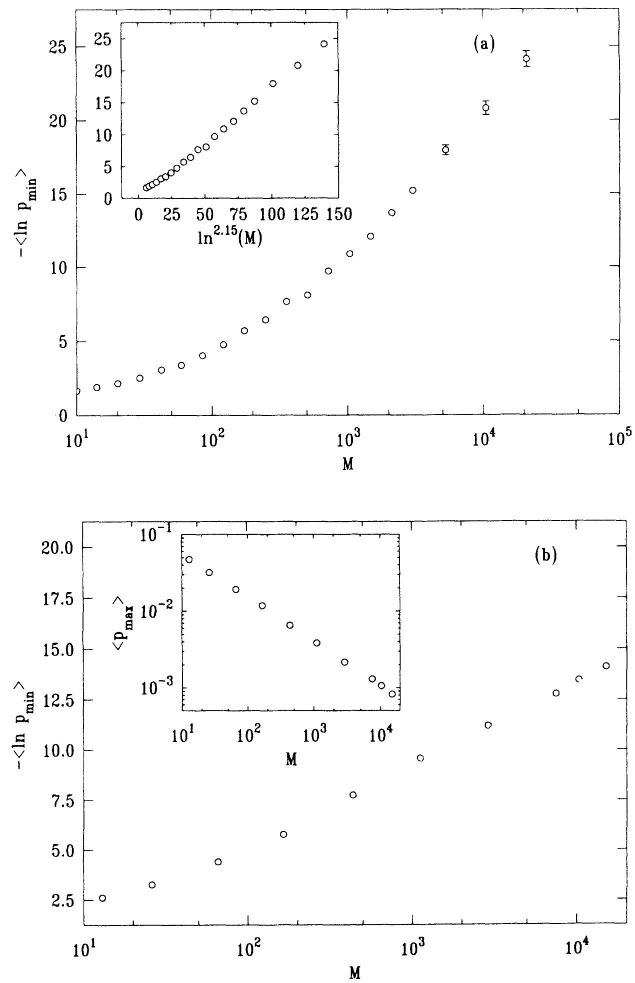

FIG. 3. Scaling of the minimum growth probability in (a) 2D and (b) 3D. $\langle \ln p_{\min} \rangle$ is plotted as a function of the cluster mass M. The inset of (a) shows the same quantity, but plotted vs $(\ln M)^{2.15}$. Inset of (b): log-log plot of $\langle p_{\max} \rangle$ as a function of the cluster mass M, averaged over 50 off-lattice 3D clusters.

about the internal structure of DLA [10–12]. A recent picture for 2D DLA structure regards DLA as a succession of self-similar "voids," separated by narrow "necks" that scale slower than the linear size of the associated "void" [4, 13]. In both 2D and 3D, "necks" are created by side branches in DLA that grow closer and closer until their growth probabilities become so small that no further narrowing occurs. This observed phenomenon can perhaps be better understood if one notes that the $\{p_i\}$ of a given DLA cluster are identical to normalized values of the electric field $\{E_i\}$ on the surface of a charged conductor whose shape is identical to the given DLA cluster. Thus as side branches of the DLA "conductor" grow closer to each other, the electric field at their surface must become smaller and smaller (since $E_i \propto \nabla\phi_i$, where $\phi \equiv$ const on the surface of the conductor). Consequently we observe a peculiar scaling behavior of the smallest growth probability, which leads to the phenomenon of a phase transition in 2D DLA.

In 3D, even if there are points where tips from different branches of the aggregate come close or meet, there is no significant screening of growth due to this configuration, because no volume is cut off from the exterior and particles can enter the cluster from a direction perpendicular to the loop. Simply stated, one cannot cut off a volume with branches in the same way that one can cut off an area. Thus we interpret the apparent absence of a phase transition for 3D as the effect of the topological differences between two and three dimensions. We further note that as d increases, d_f becomes closer to $d-1$ [14]; the higher d is, the less dense the clusters are, since $\rho(R) \sim R^{d_f - d}$. Thus it is tempting to conjecture that $d = 2$ is a "lower critical dimension" in the sense that there is a phase transition for $d = 2$ but power-law scaling for all $d > 2$.

Using the scaling properties of $\alpha_{\max}$ in 3D, we can construct a qualitative picture for 3D DLA close to the wedge model [8, 11] for 2D DLA. Imagine one fits hollow cones with opening angle ψ into the branched 3D DLA structure (ψ is taken to be the angle between the cone axis and the walls). Using the electrostatic analogy, the decay of the electric field and thus of the p_i in the interior of the cone as the tip is approached is a power law of the depth [15]. Assuming that the depth of the cone L and the mass of the surrounding DLA are related by $L \sim M^{1/d_f}$, and equating powers of M, we find for small ψ

$$d_f \alpha_{\max} = -\frac{1}{2} + \frac{2.405}{\psi}. \tag{10}$$

Our calculated value of $\alpha_{\max}$ for the 3D case yields $\psi \approx 12°$.

For comparison with geometrical studies of 3D DLA, the angle 2ψ can be viewed as the upper limit to the smallest angle characterizing the tendency of DLA branches to diverge with increasing cluster radius R.

In summary, we have calculated the $\{p_i\}$ for 50 off-lattice 3D DLA clusters, and compared our analysis to the 2D case, which is believed to undergo a phase transition. We find that the 3D case is quite different. Specifically, we find that (i) the local slopes $\tau(q, M)$ do not diverge for $q < 0$ (as they do in 2D), (ii) $f_L(\alpha)$ has no systematic mass dependence (as it has in 2D), and (iii) $p_{\min}$ has a power-law singularity in M (in the 2D case, $p_{\min}$ vanishes faster).

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[1] C. Amitrano, A. Coniglio, and F. di Liberto, Phys. Rev. Lett. **57**, 1016 (1986); Y. Hayakawa, S. Sato, and M. Matsushita, Phys. Rev. A **36**, 1963 (1987).
[2] T. Vicsek, *Fractal Growth Phenomena*, 2nd ed. (World Scientific, Singapore, 1992).
[3] J. Lee and H. E. Stanley, Phys. Rev. Lett. **61**, 2945 (1988).
[4] S. Schwarzer, J. Lee, A. Bunde, H. E. Roman, S. Havlin, and H. E. Stanley, Phys. Rev. Lett. **65**, 603 (1990).
[5] S. Schwarzer, J. Lee, S. Havlin, H. E. Stanley, and P. Meakin, Phys. Rev. A **43**, 1134 (1991).
[6] P. Meakin, Phys. Rev. A **35**, 2234 (1987); A. Block, W. von Bloh, and H.J. Schellnhuber, *ibid.* **42**, 1869 (1990).
[7] P. Meakin, H. E. Stanley, A. Coniglio, and T. A. Witten, Phys. Rev. A **32**, 2364 (1985).
[8] L.A. Turkevich and H. Scher, Phys. Rev. Lett. **55**, 1026 (1985).
[9] Note that in the literature α is sometimes defined with respect to linear size L, i.e., $\alpha \equiv -\ln p/\ln L$. Similarly, $\ln N(\alpha)$ is sometimes rescaled with respect to $\ln L$ (and not $\ln M$ as in this work). For comparisons to such work, our values for $\alpha, \alpha_L, f_H, f_L$ must be multiplied by d_f.
[10] C. J. G. Evertsz, P. W. Jones, and B. B. Mandelbrot, J. Phys. A **24**, 1889 (1991).
[11] B. B. Mandelbrot and T. Vicsek, J. Phys. A **22**, L377 (1989).
[12] J. Lee, S. Havlin, and H. E. Stanley, Phys. Rev. A **45**, 1035 (1992).
[13] S. Schwarzer, H.E. Stanley, and S. Havlin (unpublished).
[14] R. Ball and T. A. Witten, Phys. Rev. A **29**, 2966 (1984).
[15] D. Jackson, *Classical Electrodynamics*, 2nd ed. (Wiley, New York, 1975).