Breakdown of Multifractal Behavior in Diffusion-Limited Aggregates

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Analytic arguments are presented, concerning the "phase transition" to nonmultifractal behavior of the qth moment, M_q, of growth probabilities in diffusion-limited aggregation, found numerically by Lee and Stanley. Assuming the existence of exponentially small growth probabilities, for a single growing aggregate, we find a transition at $q=0$. For aggregates of size L, this transition splits into two at $q_0(L) < q_c(L) < 0$. Quantitative analysis of $q_0(L)$ yields information on the tail of the growth probability distribution. Averaging M_q over all aggregates may yield a finite q_0.

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The diffusion-limited aggregation (DLA) model[1] has been studied intensively in recent years due to its wide range of physical applications.[2] The development of a DLA structure is governed by the growth probability of each site that belongs to its accessible perimeter. This growth probability density (GPD) is believed to be well described by the multifractal formalism,[3-9] which basically assumes scaling of each and every part of the GPD. The moments of the GPD are therefore assumed to have the form,

$$M_q = \sum_i p_i^q = \sum_p n(p) p^q \simeq A(q) L^{-\tau(q)}, \qquad (1)$$

where L is the linear size of the growing aggregate, and $n(p)$ is the number of sites having a growth probability p. Multifractal analysis often uses the Legendre transform of $\tau(q)$, $f(\alpha)$, where

$$\alpha = \frac{d\tau}{dq}, \quad f = q\alpha - \tau. \qquad (2)$$

$f(\alpha)$ is usually upward convex, with a maximum at $f = D_g$ (the fractal dimension of the accessible sites on the growth perimeter).

The mathematical form of (1) led several authors[10-12] to formulate an analogy between M_q and a statistical-mechanical partition function. In this analogy q corresponds to the inverse "temperature" and $\tau(q) = -\ln M_q/\ln L$ represents a "free energy." This analogy is meaningful as long as $\tau(q)$ and $f(\alpha)$ are independent of L. Indeed this size independence is assumed in the usual picture of multifractals for sufficiently large L. However, problems arose[11-13] for negative values of q [and $\alpha > \alpha(q=0)$]. Large fluctuations prevented accurate calculations and results for $\tau(q)$ became size dependent. This led to the anticipation[11,12] of a "phase transition" at some negative value of q, q_c. Specifically, Lee and Stanley[12] (LS) used exact enumeration to find a transition at a well defined critical value, $q_c \simeq -1$. LS also found an exponential decay of the smallest growth probability with size.

In the present Letter we relate analytically, the nonmultifractal phenomena for negative q to the possible existence of small growth probabilities that decay faster than any power of L.[13] First, we consider a single aggregate at different stages of its development. Assuming the existence of exponentially small growing probabilities, we identify two *size-dependent* critical thresholds, $q_0(L) < q_c(L) < 0$, both approaching zero as $L \to \infty$. The usual multifractal formalism, with size-independent exponents, applies only for $q > q_c(L)$. For $q_0(L) < q < q_c(L)$, the exponents depend on L, as shown in Fig. 1. For $q < q_0(L)$, the exponents f and α are independent of q. For $L \to \infty$, we predict a phase transition in $f(q)$ at $q=0$, which is either first or third order. Both $-\tau(q,L)$ and $\alpha(q,L)$ diverge to ∞ for $L \to \infty$ and $q<0$. Averaging over aggregates may yield a nonvanishing value of q_c if sites with exponentially small growth probability appear *only* in clusters whose occurrence probability is also exponentially small. The value of q_0, on the other hand, may be finite even if such growth probabilities appear in typical clusters. This may explain the size-independent finite threshold found by LS, who also averaged M_q over *all* aggregates with $L \le 5$.

Starting from a single typical aggregate we first establish the existence of a threshold, q_c. For all $q \ge 0$, there exists a large amount of evidence[7-9] that $\tau(q)$ is finite and independent of L. Particularly, $\tau(0) = -D_g$. Hence, $q_c \le 0$. For large negative q, M_q is dominated by the smallest growth probability, $p_{\min}$, with degeneracy $n(p_{\min})$. This probability is associated with sites that are deep inside "fjords." The electrostatic field (proportional to the growth probability) at the bottom of a narrow straight slit decays exponentially with its depth.[14] Hence it is conceivable that sites within a tortuous fjord of the same depth are even more screened, and we assume $p_{\min}$ to decay, at least, exponentially with L, i.e.,

$$\lim_{L \to \infty} \ln p_{\min}(L)/\ln L = -\infty. \qquad (3)$$

Intuitively, we expect (3) to hold for *any typical aggregate*. In any case, all the following arguments apply only to aggregates obeying (3). On a single aggregate we al-

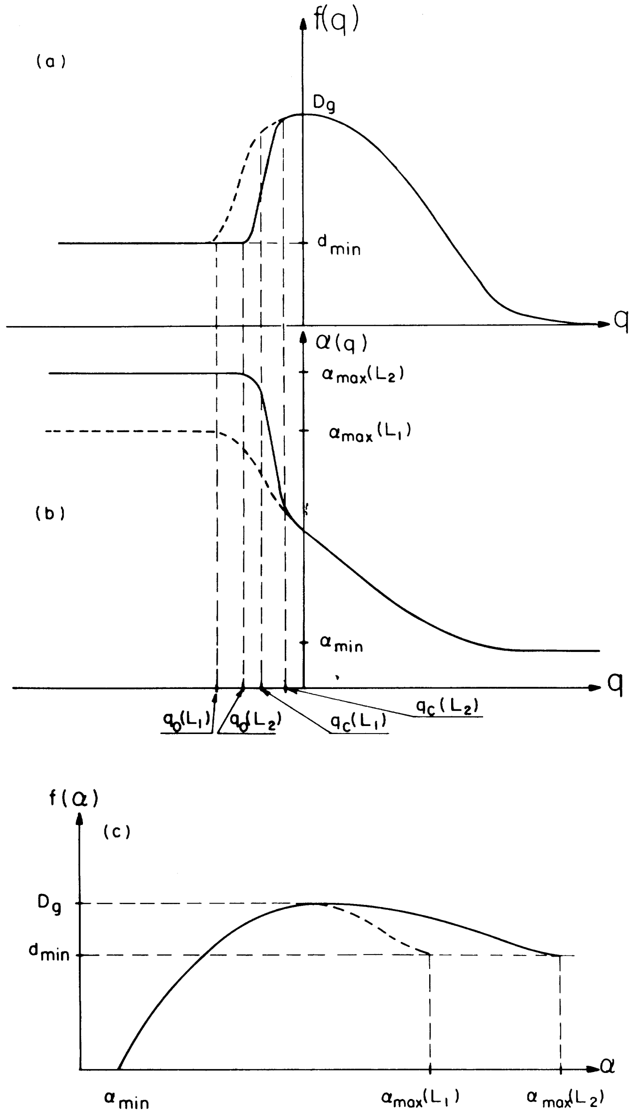

FIG. 1. Schematic dependence of (a) $f(q,L)$, (b) $\alpha(q,L)$, and (c) $f(\alpha,L)$, for a single growing aggregate at sizes L_1 (dashed line) and $L_2>L_1$ (solid line). $d_{\min}$ is the fractal dimension of the growth sites with the smallest growth probability. $\alpha_{\max}(L)$ and $q_0(L)$ are given in Eqs. (6) and (9).

ways have $1\le n(p_{\min})\le M_0$, hence we expect that (for large L) $n(p_{\min})\sim L^{d_{\min}}$. Here we should comment that although we assume a fractal behavior of DLA clusters, such a behavior has not been established for $L\to\infty$.[2] Everything that follows applies only to the range of length scales where DLA exhibits this behavior. Thus for $q<0$ we have $M_q\ge n(p_{\min})p_{\min}^q$, and

$$-\tau(q,L)\simeq \ln M_q/\ln L\ge d_{\min}+q\ln p_{\min}(L)/\ln L\,. \quad (4)$$

Given (3), Eq. (4) implies that $\lim_{L\to\infty}\tau(q,L)\to-\infty$ for all $q<0$, hence $q_c(\infty)=0^-$.

To obtain a quantitative estimate of the size dependence of q_c, divide the M_0 points in the GPD into K groups, each containing $M_0(L)/K$ growth probability values. Choosing a representative probability value p_k for each group (e.g., the average), we have $p_{\min}=p_1<p_2<\cdots<p_K$, with corresponding degeneracies $n(p_k)$. For large L, $n(p_k)\sim L^{d_k}$, with $0\le d_k\le D_g$. Since $p_{\min}$ decays exponentially with L, and $p_K\sim p_{\max}\sim L^{-\alpha_{\min}}$, where $\alpha_{\min}$ is L independent,[5,7,9] we expect to find a value k_s (which may depend on L) such that $\ln p_k/\ln L$ is independent of L for $k>k_s$ and approaches $-\infty$ with $L\to\infty$ for $k\le k_s$. We can now write $M_q=M_q^>+M_q^<$, with

$$M_q^<=\sum_{k=1}^{k_s}n(p_k)p_k^q\,.$$

For large negative q, $M_q\simeq M_q^<$ is dominated by $n(p_{\min})p_{\min}^q$, and (4) becomes an equality:

$$-\tau(q,L)\simeq d_{\min}+q\ln p_{\min}(L)/\ln L\,. \quad (5)$$

Combining (2) and (5) implies

$$\alpha(q,L)=\alpha_{\max}(L)=-\ln p_{\min}(L)/\ln L,\quad f(q,L)\equiv d_{\min}\,. \quad (6)$$

In this range of q we thus predict an L-independent value of f, associated with a q-independent value of α which increases strongly with L. The right-hand side point in the $f(\alpha)$ curve, $\alpha_{\max}$, moves to the right at constant f, as L increases. Assuming $q_0(L)$ to be the value below which (5) and (6) hold, we identify $q_0(L)$ by requiring that the term with $k=2$ in $M_q^<$ becomes comparable to that with $k=1$. Thus

$$q_0(L)\simeq\frac{(d_2-d_{\min})\ln L}{\ln p_{\min}(L)-\ln p_2(L)}\,. \quad (7)$$

If we assume[13]

$$p_{\min}\sim\exp(-A_1L^x),\quad p_2\sim\exp(-A_2L^y)\,, \quad (8)$$

with $x\ge y$, then we predict that for large L

$$q_0(L)\simeq-C(d_2-d_{\min})L^{-x}\ln L\,, \quad (9)$$

with $C=1/A_1$ (if $x>y$) and $C=1/(A_1-A_2)$ (if $x=y$). Equation (8) also yields

$$\alpha_{\max}(L)\simeq A_1L^x/\ln L\propto 1/q_0(L)\,. \quad (10)$$

As q increases above q_0, M_q will still be dominated by $M_q^<$ up to $q=q_c(L)$. Since $q_0\le q_c\le 0$, Eqs. (7) or (9) can be used as lower bounds on the approach of $q_c(L)$ to zero: $|q_c(L)|\le|q_0(L)|\sim L^{-x}\ln L$. For $q_0<q<q_c$, we can use steepest descent to estimate $M_q^<\sim n(p_x)p_x^q$, with $n(p_x)\sim L^{d_x}$, hence

$$-\tau(q,L)=d_x+q\ln p_x(L)/\ln L\,,$$
$$\alpha(q,L)=-\ln p_x(L)/\ln L\,, \tag{11}$$
$$f(q,L)=d_x,\ \text{for}\ q_0<q<q_c\,.$$

To obtain a more quantitative estimate for q_c, we use steepest descent to write

$$M_q^{>}=\sum_{k=k_s+1}^{K}n(p_k)p_k^q\sim L^{-a(q)}\,.$$

We expect $a(q)=\tau(q)$ if the sum is dominated by $p^*\sim L^{-a(q)}>p_{k_s}$. At q_c, $M_{q_c}^{<}\sim M_{q_c}^{>}$, hence $-[a(q_c)+d_c]\ln L=q_c\ln p_c(L)$, where $p_x=p_c$ dominates $M_{q_c}^{<}$. Assuming that $|q_c|\ll 1$, and using $a(q_c)=\tau(q_c)=q_c\alpha(q_c)-f(q_c)$, we now expand α and f around $q=0$, noting that $f(0)=D_g$ and $(df/dq)_{q=0}=0$. Thus,

$$q_c(L)=(D_g-d_c)/[\alpha(0)+\ln p_c(L)/\ln L]\,. \tag{12}$$

Since both p_c and d_c depend on q_c, (12) is still implicit.

The detailed q dependence of f and α is determined by the value of $d_{\min}$. If $d_{\min}<D_g$, then f drops quickly from $f(q_c)\simeq D_g$ to $d_{\min}$ as q decreases from q_c to q_0 and remains constant for $q<q_0$ [Fig. 1(a)]. In the limit $L\to\infty$, $f(q,L)$ undergoes a *first-order* transition at $q=0$. The function $\alpha(q,L)$ increases quickly from $\alpha(q_c)\simeq\alpha(0)$ to $\alpha_{\max}(L)$ in the same narrow range $q_0<q<q_c$, and $\alpha_{\max}(L)\to\infty$ as $L\to\infty$ [Fig. 1(b)]. In the corresponding $f(\alpha)$ curve [Fig. 1(c)], the finite drop in f (from D_g to $d_{\min}$) occurs over a very wide range of α [from $\alpha(q_c)$ to $\alpha_{\max}(L)$]. The variation of f with α therefore seems very slow, approaching an almost flat line. A gradual increase with L of $f(\alpha)$, towards a flat line, was indeed observed by Meakin (Ref. 2) in Ref. 12 and in references cited there. If $d_{\min}=D_g$ (a finite fraction of the growth sites have the minimal growth probability $p_{\min}$), then $f(q,L)\equiv D_g$ for all $q<0$. The transition at $q=0$ (for $L\to\infty$) then becomes *third order.*

We can now discuss (real or computer) experiments. For $q<q_0$, and fixed L, (5) implies that $\tau(q,L)$ varies linearly with q, with a slope of $\alpha_{\max}(L)$. Measuring this slope for several cluster sizes yields $p_{\min}(L)$, via (6), and allows a check of our theory. Near $p_{\min}$, this implies $n(p)\sim(-\ln p)^{d_{\min}/x}$.

So far we have discussed a *single* growing aggregate. To analyze an average over aggregates we define $\rho(\gamma)$ as the occurrence probability (OP) of the configuration γ. Since for $q\geq 0$ the averaged M_q scales as a power of L, then clearly there exists a set of configurations, Γ_r, whose OP is *not* exponentially small with L. We also define the complementary set, Γ_e, provided it exists. As for a single aggregate, we consider the smallest possible value of $p_{\min}$ over *all* cluster configurations, $P_{\min}$. In fact, LS found that $P_{\min}\sim\exp(-AL^2)$, confirming our arguments, at least for the *average* case. We can now write

$$\langle M_q\rangle=\sum_{\gamma\in\Gamma}\rho(\gamma)\sum_p n_\gamma(p)p^q\sim L^{-a(q)}+\langle n(P_{\min})\rangle P_{\min}^q\,. \tag{13}$$

If $P_{\min}$ resides in a cluster that belongs to Γ_r, then as $L\to\infty$ $\langle n(p_{\min})\rangle\sim L^{\langle d_{\min}\rangle}$ and the moments will diverge at $q_0\sim\langle d_{\min}\rangle\ln L/\ln P_{\min}\to 0^-$. If, on the other hand, $P_{\min}$ belongs to Γ_e, then $\langle n(P_{\min})\rangle$ may be as small as L^{-L^z} with $0\leq z\leq D$, where D is the fractal dimension of DLA. Hence,

$$q_0\simeq L^z\ln L/\ln P_{\min}\,. \tag{14}$$

The asymptotic behavior of q_0 is determined by (14). If $P_{\min}$ is given by (8), and if $z\simeq x$, then q_0 depends very weakly on L. This seems to be the case for currents in random resistor networks.[13] Such a situation may cause the apparent L independence of the threshold, found by LS. It is straightforward to extend the analysis for q_c as for a single cluster, and to find that asymptotically $q_c\neq 0$ *only when all the exponentially small growth probabilities appear in exponentially rare aggregates.* The L-independent threshold observed by LS is probably to be identified with our $q_0(L)$. The data of LS do not allow a distinction between q_0 and q_c. It is possible that such a distinction is evident only for $L\gg 5$.

Real experiments and Monte Carlo simulations of DLA may not be able to probe the innermost regions of the aggregate, corresponding to $p_{\min}$. However, if the smallest growth probability which is actually probed decays exponentially with L, then we still expect our results to hold, with $d_{\min}$ replaced by its effective measured value. The decrease in $|q_c|$ thus serves as an indicator to the efficiency in fjords examination. Moreover, if the unprobed sites comprise a very large fraction then the measured value of D_g is lower then its actual value. This may explain why D_g is found to be lower then the fractal dimension of the mass in DLA[7] and may even cause $\tau(q>0)$ to shift downward a little.

Even if (8) were exact, one might expect large fluctuations in the coefficient A_1 (and probably in x as well) between different aggregates. These result in fluctuating values of $q_0(L)$ and $\alpha_{\max}(L)$, and explain the practical difficulties in obtaining reliable data for negative q,[2,12] or in comparing $f(\alpha)$ curves from different experiments. Large fluctuations will then occur when one averages M_q over many clusters. One way to reduce them is to average over $\ln M_q$, i.e., over $\tau(q,L)$, as done for quenched averages in glassy statistical systems (and not to use the annealed average over M_q, as done usually[12]). This tactic indeed leaps to mind in view of the analogy with thermodynamics of random systems.[15]

In summary, our arguments offer a full explanation for the nonmultifractal behavior of DLA. For finite size, L, we find, in general, two transitions at $0\leq q_c\leq q_0$ with $q_0(L)\sim L^{-x}$ for a single cluster. It is true that our analysis is based on the assumption that exponentially small probabilities exist for most clusters. This assumption is supported by the exact solution to the field inside a long and narrow slit,[14] and by results for, at least, some small clusters.[12] One should bear in mind that screened sites are prominent in large clusters, and small scale simulations as in Ref. 12 may not be able to see *large typical configurations.* Thus simulations for larger clusters are needed, though we note that these sites are inherently difficult to measure. We expect similar scenarios to apply to other "phase transitions" discussed in the literature.[11,13] We hope that accurate (computer or real) measurements will be able to test our predictions concerning the two thresholds $q_c(L)$ and $q_0(L)$, and the size variation of the multifractal spectrum $f(\alpha)$.

After submitting the first version of this Letter we received a report by J. Lee, P. Alstrøm, and H. E. Stanley, which extends Ref. 12 and elaborates on $P_{\min}(L)$ and on the "phase transition." As we state above, there is no disagreement between them and ourselves as regards the transition on the "average" aggregate. It remains to be seen if the *typical* aggregate also has a similar transition (as we believe) or not.

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