Phase Transition in the Multifractal Spectrum of Diffusion-Limited Aggregation

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Based on a novel “exact enumeration” approach, we find evidence suggesting the existence of a phase transition in the multifractal spectrum of diffusion-limited aggregation. Above a critical point $\beta_c$, the moment expansion shows an infinite hierarchy of phases, while below $\beta_c$, we find a single phase. At $\beta_c$, we find fluctuations of all energy scales and singular behavior of the energy and specific heat. We also find that the maximum energy scales with system size $L$ as $E_{\text{max}}(L) \propto L^{3/2} \ln L$. Consequently, for $\beta < \beta_c$, the partition function does not scale with $L$, which implies that the conventional moment expansion must break down.

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The diffusion-limited aggregation (DLA) model has been found to describe a remarkably large number of interesting physical phenomena, from fluid flow in porous media to colloidal aggregation. Nonetheless, there is essentially no theoretical understanding of this model.

The growth of a DLA cluster is determined by the set of growth probabilities $\{p_i\}$, where $p_i$ is the probability that perimeter site $i$ is the next to be added to the cluster. Knowledge of the complete set $\{p_i\}$ for a given cluster at time $t$ is sufficient to describe the statistical properties of clusters at time $t+1$. Hence much attention has focused on how the $\{p_i\}$ scale with system size $L$. In particular, the density-of-states function $D(\epsilon,L)\,d\epsilon$ gives the number of growth sites whose value of $\epsilon \equiv -\ln p/\ln L$ is in the range $[\epsilon,\epsilon+d\epsilon]$. Like other distribution functions, $D(\epsilon,L)$ is characterized by its moments. Motivated by the analogy with thermodynamics, we define the partition function as

$$Z(\beta,L) \equiv \sum_a C_a \sum_i p_{i,a}^\beta = \sum \epsilon D(\epsilon,L) L^{-\beta \epsilon},$$

(1)

where $C_a$ is the weight of configuration $a$, and $p_{i,a}$ is the growth probability of site $i$ of configuration $a$. It has been argued that this density-of-states function is not characterized by a single gap exponent as are familiar density-of-states functions from critical-point systems, in the sense that $F(\beta)$ is not a linear function of $\beta$. Here $F(\beta)$ is defined through

$$F(\beta) \equiv \lim_{L \to \infty} F(\beta,L),$$

(2a)

where

$$F(\beta,L) \equiv -\frac{\ln Z(\beta,L)}{\ln L}.$$  

(2b)

Special attention has focused on the Legendre transform of $F(\beta,L)$, $S(E,L) \equiv \beta E - F(\beta,L)$, where $E = \partial F(\beta,L)/\partial \beta$ is the variable conjugate to $\beta$. It is conventional to call $E$ the energy, $S(E,L)$ the entropy, and $F(\beta,L)$ the free energy (see Table I).

An unsolved problem concerns the behavior of the large-$L$ limit $F(\beta)$ for negative $\beta$. It has been essentially impossible to obtain reliable calculations, and the reason was assumed to be numerical accuracy. Moreover, experiments also give values for $S(E) \equiv \lim_{L \to \infty} S(E,L)$ for large $E$ that disagree with calculations, a fact that has plagued investigators in this field.

In this Letter we propose a resolution of this discrepancy. Specifically, we find a phase transition in DLA in the sense that the energy $E$ undergoes a quite sharp jump near a critical value $\beta_c$. For values of $\beta$ below $\beta_c$, the free energy $F(\beta,L)$ is dominated by the maximum energy term $E_{\text{max}}(L)$ which increases with system size $L$. Hence the partition function $Z(\beta,L)$ does not scale as a power law for $\beta < \beta_c$. Another consequence is that the large $E$ part of $S(E)$ is a straight line with slope $\beta_c$. Furthermore, because of large fluctuations of energy near $\beta_c$, the convergence to this straight line exhibits a “critical slowing down.”

Our calculations are based on exact enumeration of all DLA configurations in a $L \times L$ box containing $2L^2 - L$ bonds. This exact enumeration follows the general procedure outlined by Nagatani in connection with a position-space renormalization-group formulation for DLA. The enormous number of possible configurations in a box of $2L^2 - L$ bonds is reduced by many orders of magnitude with use of symmetry considerations. We find that for $L = 2, 3, 4, 5$, we must actually calculate the growth probability exactly for $9$, $523$, $1.2 \times 10^5$, and $3.0 \times 10^{11}$ configurations (note that the Nagatani calculations are for $L = 2, 3$). With $3.0 \times 10^{11}$ configurations, it is impossible to apply the symmetry arguments on a case-by-case basis. Rather, we constructed elaborate computer algorithms to recognize symmetries, reducing

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<th>Table I. Comparison of notation of this paper and that of Refs. 3–6.</th>
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<td>$\beta \leftrightarrow q$</td>
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<td>$E \leftrightarrow a$</td>
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the number of distinct configuration to 3, 14, 259, and 9361, respectively. In particular, we used a burning-type algorithm to remove all the “dead” \( p_{i,a} = 0 \) sites.

Having the \( p_i \) from the exact enumeration procedure for \( L = 2, 3, 4, 5 \), we then form the partition function \( Z(\beta, L) \), which is then used to extract \( F(\beta, L) \) as in Fig. 1. Figure 2(a) shows the free-energy function defined in Eq. (1) as a function of \( \beta \). We see that there is remarkably rapid convergence as a function of \( L \) for \( \beta > \beta_c \) with \( \beta_c \) roughly equal to \( -1 \). On the other hand, for \( \beta < \beta_c \) there appears to be no convergence at all! We also find that the left-hand side of the \( S(E, L) \) plots of Fig. 2(b) converge well but the right-hand sides converge poorly.

What is the origin of this poor convergence? Do the thermodynamic limits (the \( L \to \infty \) limits) of the functions \( F(\beta, L) \) and \( S(E, L) \) exist? To answer these questions, we shall argue that there is a phase transition at a well-defined value of \( \beta_c \). Figure 3 shows the dependence on \( \epsilon \) of the density-of-states function \( D(\epsilon, L) \) weighted by the “Boltzmann factor” \( L^{-\beta \epsilon} \), thus the summand of (1), which shows the contribution of different energy bins. For \( \beta \gg \beta_c \) [Fig. 3(a)] we see that this function is peaked at a characteristic value of the energy which is independent of \( L \). However, as \( \beta \) decreases toward \( \beta_c \), there is no characteristic value of energy and one sees structure on all energy scales [Fig. 3(b)]. Finally, for all \( \beta < \beta_c \), there is a sharp peak. This sharp peak is centered on the value of the energy given by the smallest value of the growth probability, \( E_{\text{max}} = -\ln(p_{\text{min}})/\ln L \).

Moreover, we find from Fig. 4 that \( E_{\text{max}} \) strongly depends on \( L \), with

\[
E_{\text{max}}(L) \propto L^{2/\ln L}.
\]

To see this transition more clearly, we plot the energy \( E(\beta, L) \) as a function of \( \beta \) [Fig. 5(a)]. One can see the sharp variation in \( E(\beta, L) \) near \( \beta_c \approx -1 \). Furthermore, the magnitude of the variation increases with \( L \), which becomes clear when we examine the specific heat \( C(\beta, L) = -\partial E(\beta, L)/\partial \beta \) [Fig. 5(b)]. Moreover, we see that \( \beta_c \) appears to be independent of system size \( L \).

Additional evidence of the phase transition comes from the “data collapse” plots [Fig. 5(c)] in which the

FIG. 1. Dependence of \( \ln Z(\beta, L) \) on \( \ln L \) for (a) \( \beta = 0 \) (\( \beta > \beta_c \)) and (b) \( \beta = -5 \) (\( \beta < \beta_c \)). The free energy \( F(\beta, L) \) is given by the negative of the slope of these plots. One sees that \( F(\beta, L) \) is not defined for the \( \beta < \beta_c \) case, in contrast to the well-defined free energy for the \( \beta = 0 \) and \( \beta > \beta_c \) case.

FIG. 2. Effect of cell size \( L \) on (a) free energy and (b) entropy. As \( L \) increases, the free energy converges for \( \beta > \beta_c \), but not for \( \beta < \beta_c \). The solid line is for \( 1 \times 1 \), the dotted line is for \( 3 \times 3 \), the dashed line is for \( 4 \times 4 \), and the dashed-dotted line is for \( 5 \times 5 \).
FIG. 3. Energy fluctuations above, near, and below the phase transition. Shown is the density of states multiplied by \textquotedblright Boltzmann factor\textquotedblright $L^{-\beta E}$ for the case $L=5$ and (a) $\beta=1.0$, (b) $\beta=-1.0$, and (c) $\beta=-2.0$.

Density of states $D(\epsilon,L)$ is scaled by $\ln L$, giving the entropy function $S(E,L)$ apart from a normalization factor. Because good data collapse is found, we argue that there is a well-defined entropy function $S(E)$ at every scale of $E$. Furthermore, the part of the entropy function with $E>5$ is a fairly straight line, which also suggests the existence of a phase transition.

Thus we argue that there is a phase transition at a well-defined critical value $\beta=\beta_c$. Our argument is based on the following four distinct pieces of evidence: A maximum energy that displays a strong dependence on $L$: $E_{\text{max}}(L) \propto L^2/\ln L$, large energy fluctuations for $\beta=\beta_c$, sharp variation in $E(\beta,L)$ near $\beta_c$, and the straight line portion of the curve $\ln D(\epsilon)/\ln L$ for $\epsilon > \epsilon_c$.

One consequence of this transition is the breakdown of power-law scaling for $\beta < \beta_c$. Since for $\beta < \beta_c$, $F(\beta,L)$ is dominated by $E_{\text{max}} = -\ln(\rho_{\text{min}})/\ln L$, and $E_{\text{max}}(L)$ diverges in the $L \to \infty$ limit, it follows that $F(\beta,L)$ cannot converge for $\beta$ below $\beta_c$. This is consistent with our finding that the moment $Z(\beta,L)$ does not scale as a power law for $\beta < \beta_c$ [e.g., Fig. 1(b)].

Because we find contributions from all energy scales for $\beta = \beta_c$, the conventional derivation of $S(E,L)$ from $F(\beta,L)$ must be called into question. This derivation uses the method of steepest descents, which assumes most of the contribution to an energy integral comes from energies close to the saddle-point energy $E^\star$. Our results show this assumption fails for $\beta \approx \beta_c$. Specifically, we find exceptionally large energy fluctuations for $\beta \approx \beta_c$ [Fig. 3(b)], which cause a significant slowing down of the convergence.

Corresponding to this difficulty, we find poor convergence for the part of $S(E,L)$ corresponding to $\beta \approx \beta_c$; this is the right-hand side of Fig. 2(b), which is expected to converge to a straight line of slope $\beta_c$.

In summary, by using an exact enumeration approach to DLA, we find that there is a critical point $\beta_c$ above which we find the usual infinite hierarchy of phases (the conventional multifractal spectrum) but below which we find a single phase. This phase is characterized by a...
maximum energy that increases with system size $E_{\text{max}}(L) \propto L^{2/\ln L}$. We believe that this is the reason that multifractal analysis must fail below $\beta_c$.

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12Note that the dead sites are removed, so that $E_{\text{max}}$ arises from sites with small but nonzero values of $p_{\text{on}}$. 

FIG. 5. Dependence of (a) energy $E(\beta, L)$ and (b) specific heat $C(\beta, L)$ on $\beta$, displaying features near $\beta=\beta_c$ characteristic of a phase transition. (c) Data collapse plot showing dependence of $\ln D(\epsilon, L)$ scaled by $\ln L$ on $-\ln p$ scaled by $\ln L$. The solid line is for $2 \times 2$, the dotted line is for $3 \times 3$, the dashed line is for $4 \times 4$, and the dashed-dotted line is for $5 \times 5$. 

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