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 β_c, the moment expansion shows an infinite hierarchy of phases, while below β_c we find a single phase. At β_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 $E_{max}(L) \propto L^2/\ln L$. 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[1-6] (DLA) model has been found to describe a remarkably large number of interesting physical phenomena, from fluid flow in porous media to colloidal aggregation.[2,3] 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[5,6] $D(\epsilon,L)d\epsilon$ 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

$$Z(\beta,L) \equiv \sum_\alpha C_\alpha \sum_i p_{i,\alpha}^\beta = \sum_\epsilon D(\epsilon,L) L^{-\beta\epsilon}, \quad (1)$$

where C_α is the weight of configuration α, and $p_{i,\alpha}$ is the growth probability of site i of configuration α. 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 β.[4-6] Here $F(\beta)$ is defined through

$$F(\beta) \equiv \lim_{L\to\infty} F(\beta,L), \quad (2a)$$

where

$$F(\beta,L) \equiv -\frac{\ln Z(\beta,L)}{\ln L}. \quad (2b)$$

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 β. It is conventional to call E the energy, $S(E,L)$ the entropy, and $F(\beta,L)$ the free energy[7] (see Table I).

An unsolved problem concerns the behavior of the large-L limit $F(\beta)$ for *negative* β. 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.[8]

In this Letter we propose a resolution of this discrepancy. Specifically, we find a phase transition[9] in DLA in the sense that the energy E undergoes a quite sharp jump near a critical value β_c. For values of β below β_c, the free energy $F(\beta,L)$ is dominated by the maximum energy term $E_{max}(L)$ 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 β_c. Furthermore, because of large fluctuations of energy near β_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 outline by Nagatani[10] 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, 5323, 1.2×10^9, and 3.0×10^{17} configurations (note that the Nagatani calculations are for $L=2,3$). With 3.0×10^{17} 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

TABLE I. Comparison of notation of this paper and that of Refs. 3-6.

$\beta \leftrightarrow q$	$F(\beta) \leftrightarrow \tau(q)$
$E \leftrightarrow \alpha$	$S(E) \leftrightarrow f(\alpha)$

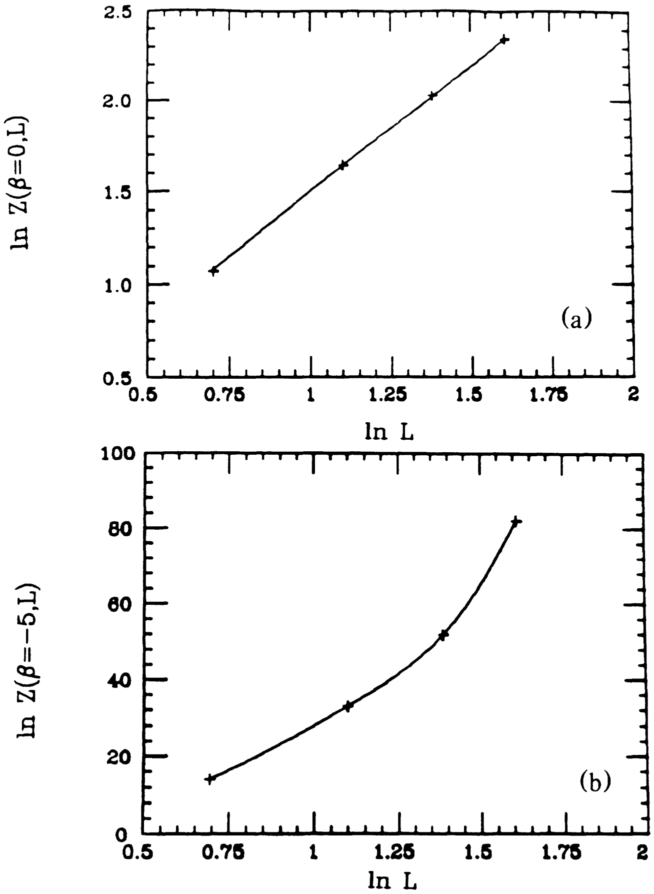

FIG. 1. Dependence of $\ln Z(\beta,L)$ on $\ln L$ for (a) $\beta=0$ ($\beta>\beta_c$) and (b) $\beta=-5$ ($\beta<\beta_c$). The free energy $F(\beta,L)$ is given by the negative of the slope of these plots. One sees that $F(\beta,L)$ is not defined for the $\beta<\beta_c$ case, in contrast to the well-defined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

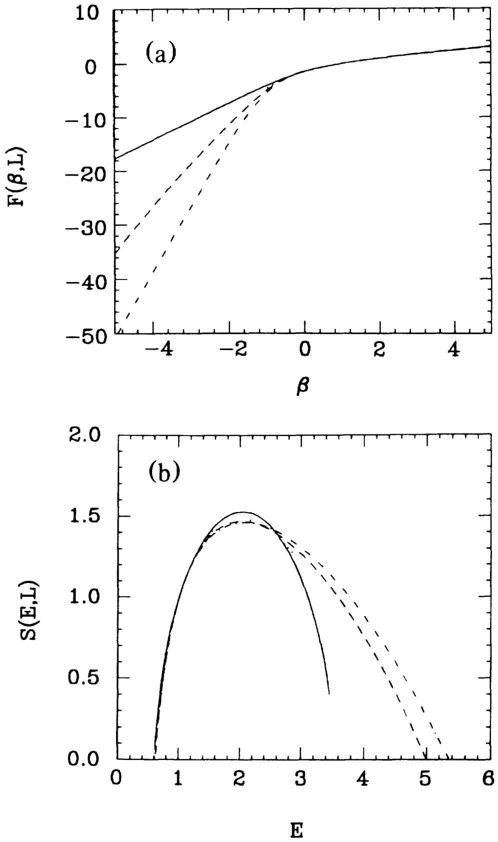

FIG. 2. Effect of cell size L on (a) free energy and (b) entropy. As L increases, the free energy converges for $\beta>\beta_c$, but not for $\beta<\beta_c$. The solid line is for 1×1, the dotted line is for 3×3, the dashed line is for 4×4, and the dashed-dotted line is for 5×5.

the number of distinct configuration to 3, 14, 259, and 9361, respectively. In particular, we used a burning-type algorithm[11] to remove all the "dead" ($p_{i,a}=0$) sites.[12]

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 β. We see that there is remarkably rapid convergence as a function of L for $\beta>\beta_c$ with β_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 β_c. Figure 3 shows the dependence on ϵ 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 β decreases toward β_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 a value of the energy given by the smallest value of the growth probability, $E_{\max}\equiv -\ln(p_{\min})/\ln L$. Moreover, we find from Fig. 4 that $E_{\max}$ strongly depends on L, with

$$E_{\max}(L)\propto L^2/\ln L\,. \qquad (3)$$

To see this transition more clearly, we plot the energy $E(\beta,L)$ as a function of β [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 β_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

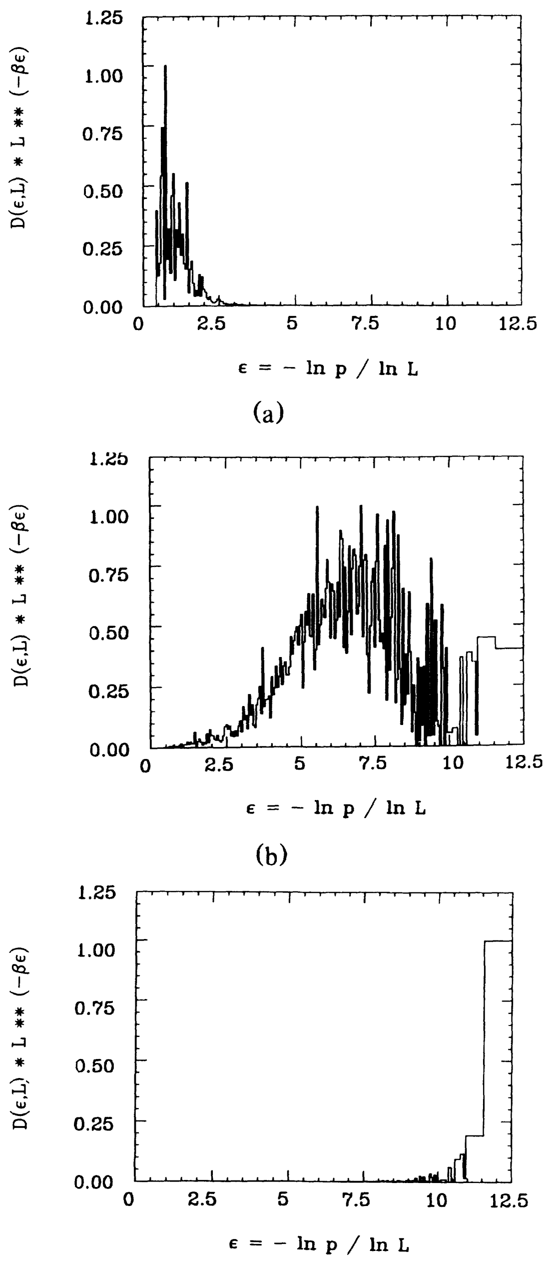

FIG. 3. Energy fluctuations above, near, and below the phase transition. Shown is the density of states multiplied by "Boltzmann factor" $L^{-\beta\epsilon}$ for the case $L=5$ and (a) $\beta=1.0$, (b) $\beta=-1.0$, and (c) $\beta=-2.0$.

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

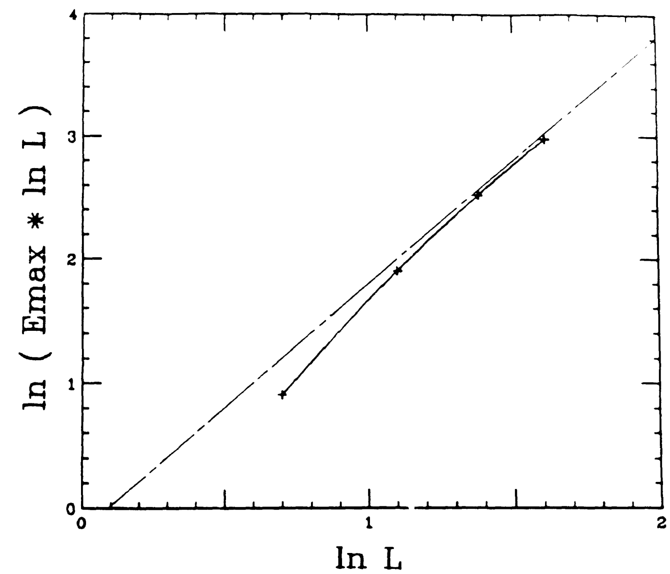

FIG. 4. Dependence of $\ln(E_{\max}\times\ln L)$ on $\ln L$, where $E_{\max}=-\ln(p_{\min})/\ln L$. The asymptotic slope of two supports the form (3).

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: $E_{\max}(L)\propto L^2/\ln L$, large energy fluctuations for $\beta\approx\beta_c$, sharp variation in $E(\beta,L)$ near β_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 $E_{\max}\equiv-\ln(p_{\min})/\ln L$, and $E_{\max}(L)$ diverges in the $L\to\infty$ limit, it follows that $F(\beta,L)$ cannot converge for β below β_c.[12] 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 $\beta\approx\beta_c$, 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^*. 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 β_c.

In summary, by using an exact enumeration approach to DLA, we find that there is a critical point β_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

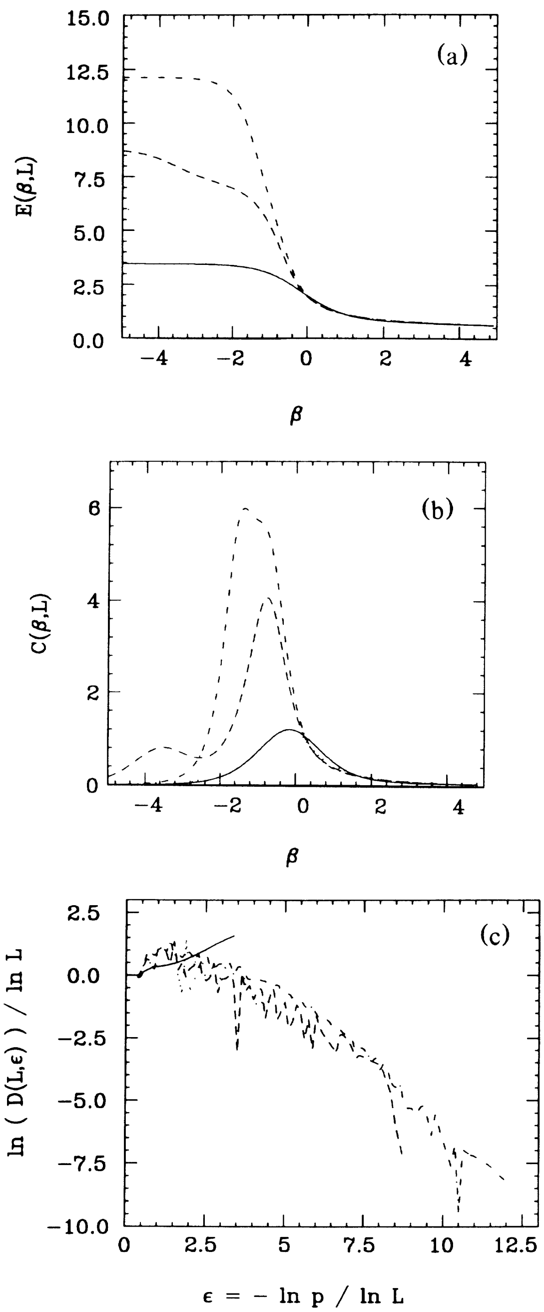

FIG. 5. Dependence of (a) energy $E(\beta,L)$ and (b) specific heat $C(\beta,L)$ on β, displaying features near $\beta=\beta_c$ characteristic of a phase transition. (c) Data collapse plot showing dependence of $\ln D(\epsilon,L)$ scaled by $\ln L$ on $-\ln P$ scaled by $\ln L$. The solid line is for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

maximum energy that increases with system size $E_{\max}(L)\propto L^2/\ln L$. We believe that this is the reason that multifractal analysis must fail below β_c.

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[1]T. A. Witten and L. Sander, Phys. Rev. Lett. **47**, 1400 (1981).

[2]For background, see J. Feder, *Fractals* (Pergamon, New York, 1988); *Random Fluctuations and Pattern Growth: Experiments and Models,* edited by H. E. Stanley and N. Ostrowsky (Kluwer Academic, Dordrecht, 1988); T. Vicsek, *Fractal Growth Phenomena* (World Scientific, Singapore, 1989).

[3]Many applications are described by P. Meakin, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. L. Lebowitz (Academic, Orlando, 1988), Vol. 12.

[4]T. C. Halsey, P. Meakin, and I. Procaccia, Phys. Rev. Lett. **54**, 854 (1986).

[5]C. Amitrano, A. Coniglio, and F. di Liberto, Phys. Rev. Lett. **57**, 1016 (1986).

[6]P. Meakin, A. Coniglio, H. E. Stanley, and T. A. Witten, Phys. Rev. A **34**, 3325 (1986).

[7]M. J. Feigenbaum, J. Stat. Phys. **46**, 919, 925 (1987); T. Bohr and D. Rand, Physica (Amsterdam) **25D**, 387 (1987).

[8]See, e.g., the discussion on this point by K. J. Måløy, F. Boger, J. Feder, and T. Jøssang, in *Time Dependent Effects in Disordered Materials,* edited by R. Pynn and T. Riste (Plenum, New York, 1987) and by P. Alstrøm, Phys. Rev. A **37**, 1378 (1988); see also, J. Nittmann, H. E. Stanley, E. Touboul, and G. Daccord, Phys. Rev. Lett. **58**, 619 (1987); H. E. Stanley and P. Meakin, Nature (London) **335**, 405 (1988).

[9]The concept of a phase transition in multifractal spectra was found in the study of simple systems, such as the logistic map {T. Bohr and M. Jensen, Phys. Rev. A **36**, 4904 (1987); M. Duong-van, in Proceedings of the International Conference on the Physics of Chaos and Systems far from Equilibrium, Monterey, California, 1987 [Nucl. Phys. B Proc. Suppl. **2**, 521 (1987)]}, charged needle [T. C. Halsey, M. H. Jensen, L. P. Kadanoff, I. Procaccia, and B. I. Shraimann, Phys. Rev. A **33**, 1141 (1986)], and Julia sets (T. Bohr, P. Cvitanovic and M. Jensen, to be published). However, the present work presents the first evidence for a phase transition in a realistic model system.

[10]T. Nagatani, Phys. Rev. A **36**, 5812 (1987), and J. Phys. A **20**, L381 (1987).

[11]H. J. Herrmann, D. C. Hong, and H. E. Stanley, J. Phys. A **17**, L261 (1984).

[12]Note that the *dead sites* are removed, so that $E_{\max}$ arises from sites with small but nonzero values of $p_{i,a}$.