

Minimum Growth Probability of Diffusion-Limited Aggregates

S. Schwarzer,⁽¹⁾ J. Lee,⁽²⁾ A. Bunde,⁽¹⁾ S. Havlin,^(1,2) H. E. Roman,⁽¹⁾ and H. E. Stanley⁽²⁾

⁽¹⁾*Institut für Theoretische Physik, Universität Hamburg, D-2000 Hamburg 36, Federal Republic of Germany*

⁽²⁾*Center for Polymer Studies and Department of Physics, Boston University, Boston, Massachusetts 02215*

(Received 28 December 1989)

We calculate the minimum growth probability for diffusion-limited aggregation (DLA) as a function of the cluster mass M , and find a novel singularity of the form $-\ln p_{\min}(M) \sim (\ln M)^y$ with $y \approx 2$. We interpret this result in terms of a simple model for DLA structure, which is characterized by a hierarchy of self-similar voids separated by channels whose diameter increases slower than the cluster diameter.

PACS numbers: 61.50.Cj, 05.40.+j, 64.60.Ak, 81.10.Jt

Since its introduction a decade ago,¹ diffusion-limited aggregation (DLA) has attracted tremendous attention.² On the one hand, it has been found to describe a vast wealth of experimental situations and, on the other hand, a true theoretical understanding has been elusive. Initially, it was imagined that the growing tips of the DLA structure held the key to a complete theoretical understanding. However, it was found that DLA is a multifractal—i.e., the quantities $p_i(M)$ giving the probability that site i of the DLA perimeter is the next to grow do not depend on cluster mass M in the same fashion—so if one knows only the p_i for the tips, one knows essentially nothing about the interior structure of DLA.² A great deal is known about how the maximum $p_i(M)$ (on the tips) scale, while almost nothing is known about the minimum growth probabilities (deep in the fjords). However, if one could learn about the p_i for the bottom of the fjords, one would have important information about the entire structure of DLA (not just the tip region)—since an incoming random walker must “find its way through the entire DLA structure” in order to reach the fjord bottom.

There exist two recently proposed forms^{3,4} for the dependence on M of p_{\min} , the smallest of all the growth probabilities. (i) Blumenfeld and Aharony³ (BA) proposed p_{\min} decreases exponentially with cluster mass M ,

$$p_{\min}(M) \sim \exp(-AM^x). \quad (1a)$$

(ii) Mandelbrot and Vicsek^{4(a)} (MV) and Harris and Cohen^{4(b)} (HC) proposed the power-law form

$$p_{\min}(M) \sim M^{-a_{\max}/d_f}, \quad (1b)$$

where d_f is the fractal dimension of DLA.

In this work we carry out the first simulations⁵ for p_{\min} . We find a surprising result for how p_{\min} depends on cluster mass,

$$-\ln p_{\min}(M) \sim (\ln M)^y \quad (y \approx 2). \quad (1c)$$

We also construct a simple model for DLA structure, which predicts the form (1c) with $y=2$ exactly.

To calculate p_i for each of the perimeter sites, we employ an algorithm⁶ to enumerate *exactly* all random walks which start from an outer circle of radius R_1 and

are trapped *either* at the perimeter sites or at an outer circle of radius $R_2 > R_1$.⁷ The growth probability can be calculated using $p_i = t_i / \sum_i t_i$, where t_i is the probability that the walker is trapped at perimeter site i . For small clusters we confirmed the validity of this approach using a Green's-function technique,⁵ which is accepted to be accurate.

For each cluster, we calculate $p_{\min}(M)$ and the exponent $F(\beta)$ defined from the “partition function”

$$Z(\beta, M) \equiv \sum_i p_i^\beta \equiv M^{-F(\beta)/d_f}. \quad (2)$$

In order to reduce the sample-to-sample fluctuation, we form a “quenched”⁸ average over all the samples studied,

$$p_Q(M) \equiv \exp\{\langle \ln p_{\min}(M) \rangle\}, \quad (3)$$

$$Z_Q(\beta, M) \equiv \exp\{\langle \ln Z(\beta, M) \rangle\}.$$

We first test the possibility of an *exponential* decay, Eq. (1a). Figure 1(a) shows $\ln[\ln p_Q(M)]$ vs $\ln M$ for clusters grown on the triangular lattice, where p_Q is normalized with respect to the p_Q of $M=1$. We note that the slope is decreasing as M increases (see the successive slopes, plotted in the inset), which implies that p_Q decays *slower than exponential*—even slower than a “stretched exponential.”

We next try the second possibility, a *power-law* decay of (1b). Figure 1(b) shows a log-log plot of $p_Q(M)$ vs M . The slight curvature implies a weak deviation from power-law behavior, which becomes more apparent from the graph of successive slopes (inset). Since the successive slopes show an increasing trend as M increases, we conclude $p_Q(M)$ decays *faster than power law*. Indeed, the successive slopes increase almost linearly, which is consistent with the possibility (1c).

Since $\ln p_Q(M)$ decays faster than M but slower than M^x [cf. Eqs. (1a) and (1b)], we considered the possibility $(\ln M)^y$. Figure 1(c) shows $\ln[\ln p_Q(M)]$ vs $\ln(\ln M)$, and displays a straight-line part for a wide range of M . The least-squares fit for this part has slope $y=1.97$. Since $y \approx 2$, we show in Fig. 1(d), a plot of $\ln p_Q$ vs $(\ln M)^2$, and find a remarkably large region of straight line (roughly two decades). One can also see from the

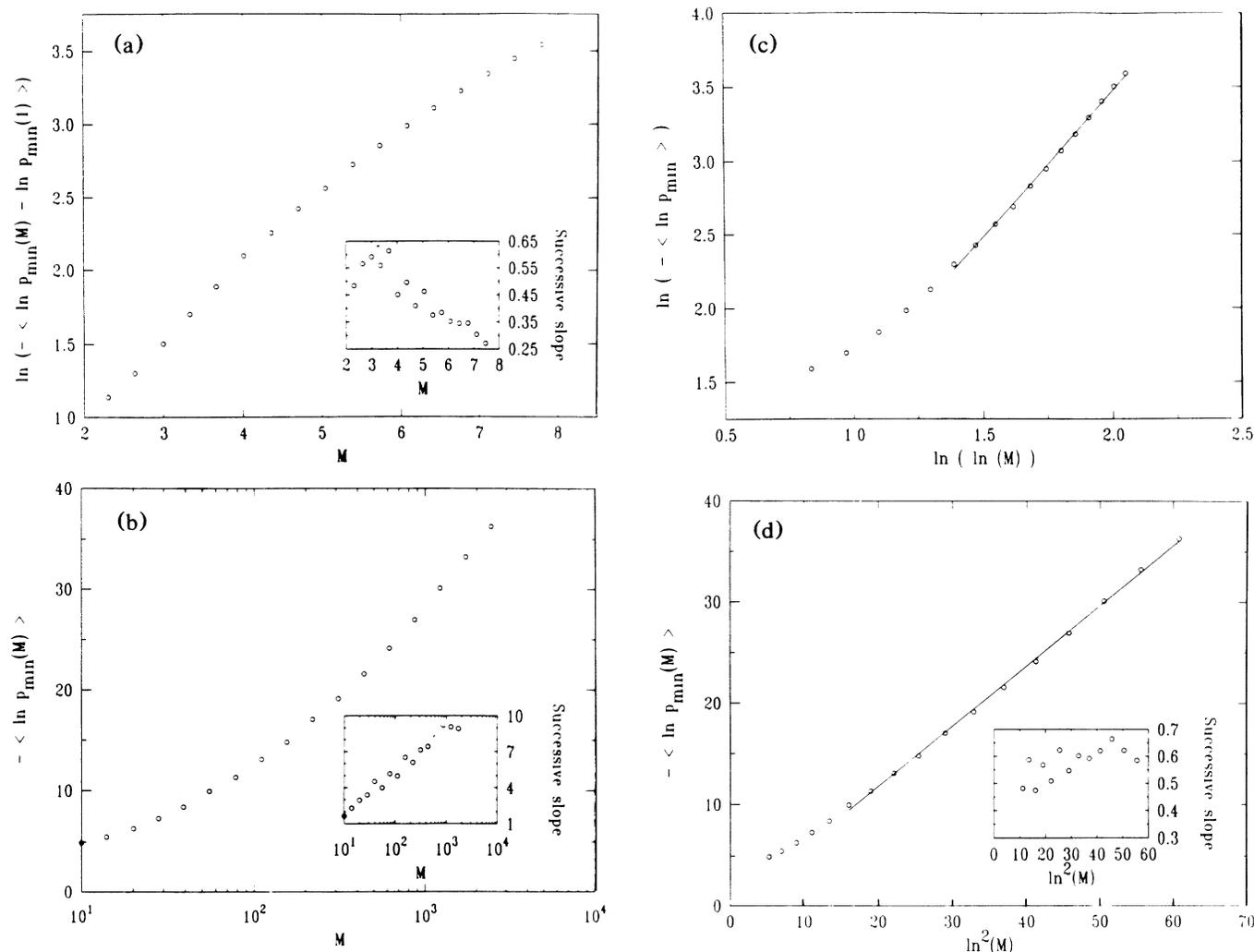


FIG. 1. (a) A $\ln[-\ln p_{\min}(M)]$ vs $\ln M$ plot (triangular lattice) made to test for the exponential behavior of Eq. (1a). One can see the deviation from a straight-line behavior. The successive slopes, plotted in the inset, are monotonically decreasing. Here successive slopes are obtained by drawing line segments between successive points of the graph. (b) A log-log plot of $p_Q(M)$ vs M made to test for the power-law behavior of Eq. (1b). The successive slopes, plotted in the inset, are almost linearly increasing. (c) A $\ln[-\ln p_Q(M)]$ vs $\ln(\ln M)$ plot made to test the logarithmic singularity of Eq. (1c). One can identify a region of straight-line behavior. The least-squares slope is $\gamma = 1.97$ (~ 2). (d) The $\ln[p_Q(M)]$ vs $(\ln M)^2$ plot. One can see quite a large linear region in the graph. Also the successive slopes, plotted in the inset, show no systematic trend.

successive slope plot (inset) that there is no systematic deviation from this straight-line behavior.

Strong evidence supporting this novel possibility is found from the behavior of the “partition function.” Figure 2(a) is a log-log plot of $Z_Q(\beta, M)$ vs M for four different values of β ($-3, -2, -1, 0$). One sees a clear deviation from power-law behavior as β decreases from 0. Furthermore, a plot of $Z_Q(\beta = -3, M)$ vs $(\ln M)^2$ [Fig. 2(b)] shows a large portion of straight-line behavior.⁹ This is consistent with p_Q , since the smallest growth probability dominates the behavior of $Z_Q(\beta, M)$ for large negative β values.

We also made similar plots for 55 clusters grown on the *square* lattice. Comparing these with Figs. 1 and 2 suggests that our results are universal (inherent to

DLA), and not artifacts of the special anisotropy of the triangular lattice.

A physical interpretation of our result (1c) follows from the observation that (1c) implies the inequality

$$Z(\beta, M) \equiv \sum_i p_i^\beta \geq p^\beta \sim \exp[-B\beta(\ln M)^2].$$

Combining this with the definition (2), we find¹⁰

$$F(\beta)/d_f \equiv -\ln Z(\beta, M)/\ln M \leq B\beta \ln M. \quad (4)$$

Thus for any value of $\beta < 0$, $F(\beta)$ diverges logarithmically as $M \rightarrow \infty$, resulting in a phase transition in the multifractal spectrum of typical DLA aggregates at $\beta_c = 0$ between two “phases” characterized by finite and infinite $F(\beta)$.¹¹ Furthermore, a singularity in typical

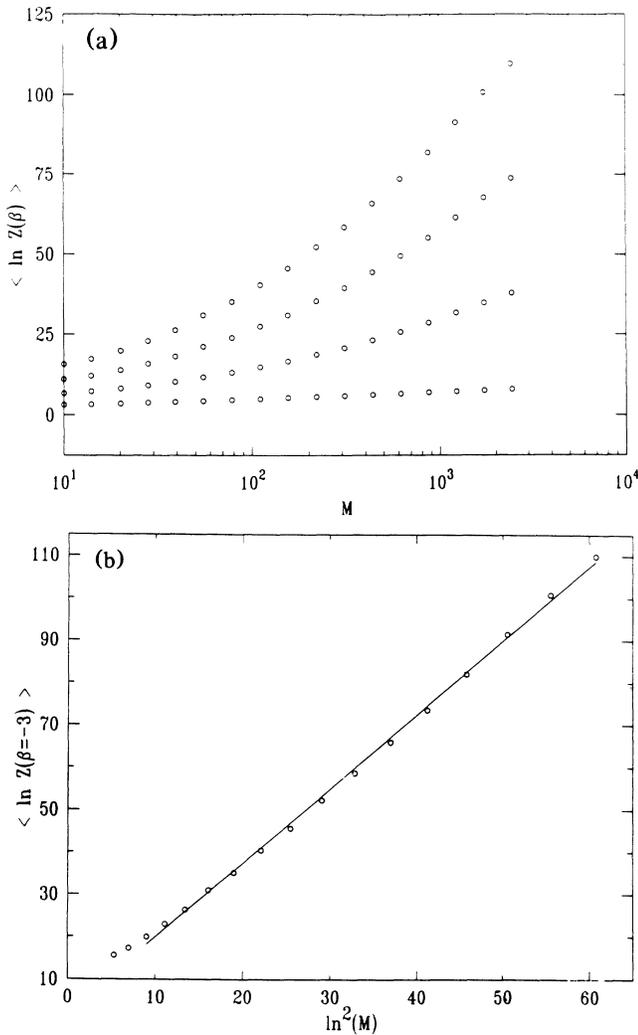


FIG. 2. (a) The β th moment $Z(\beta, M)$ ($\beta = -3, -2, -1, 0$) for the triangular lattice. As β is decreased from 0, they show clear deviation from power-law behavior. (b) $\ln Z(\beta, M)$ vs $(\ln M)^2$ plot (triangular lattice). One can clearly see the wide region of straight line, which supports $(\ln M)^2$ behavior.

DLA implies a transition in the multifractal spectrum obtained by averaging all the possible clusters.¹²

Next we argue that the form of p_{\min} enables one to construct models for the structure of DLA, which in turn serves to justify the significance of focusing attention of the p_{\min} function. It is our thesis that the problem of DLA structure is strongly connected to the problem of the structure of a fjord.

Perhaps the simplest model of a DLA fjord is that of a "tunnel" [Fig. 3(a)], which leads immediately to the predicted form of Eq. (1a). A second model is that of a *wedge-shaped fjord* (not to be confused with a *wedge-shaped tip*¹³). This model predicts p_{\min} should be of the form of Eq. (1b). We have demonstrated [e.g., in Figs. 1(a) and 1(b)] that *neither* Eq. (1a) *nor* Eq. (1b) is sup-

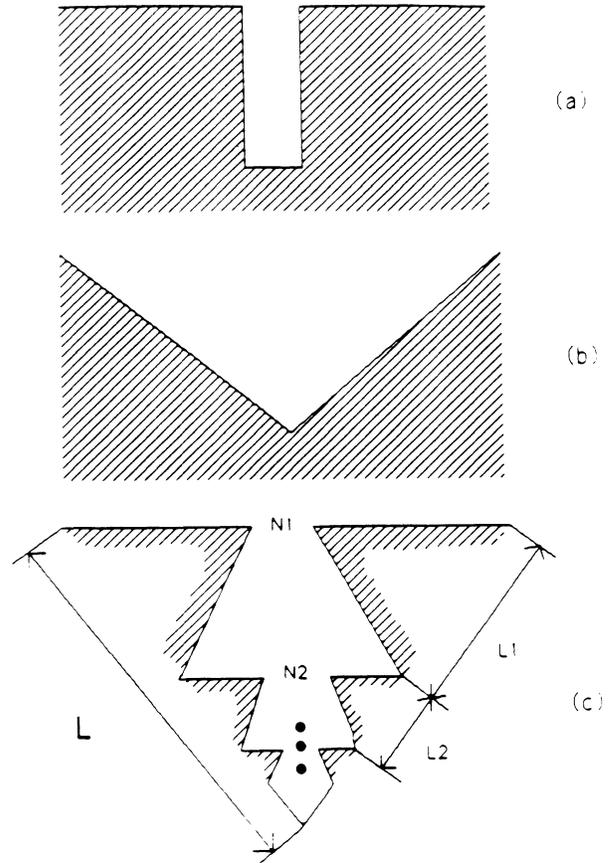


FIG. 3. (a) "Tunnel" model of a DLA fjord, which is consistent with the prediction of Eq. (1a). (b) "Wedge" model of a DLA fjord, which is consistent with the prediction of Eq. (1b). (c) "Hierarchical wedge" model of a DLA fjord, which is shown here to lead to Eq. (1c).

ported by our calculations, suggesting that both the tunnel model and the wedge model of Figs. 3(a) and 3(b) are oversimplifications.

We now introduce a new model, characterized by a hierarchical distribution of open spaces of linear dimension $L_i = a^i L$ in which the incoming random walker can diffuse [Fig. 3(c)]. The key fact is that in order for a random walker to reach the end of the fjord, it must successively move from one open space to another open space through a succession of relatively narrow regions or "channels." The plausibility of such a model is evident upon examination of photographs of *off-lattice* DLA structures. Pictorial resemblance cannot justify a model, but a straightforward heuristic calculation of the predictions of a model *can* be compared directly with the simulation results for DLA itself. Specifically, we can estimate p_{\min} as follows. The probability $\Pi(i, i+1)$ for a random walker at channel N_i to reach channel N_{i+1} scales^{4(b)} as a power $-a$ of the distance L_i separating the two channels, $\Pi(i, i+1) = L_i^{-a}$. We can now calcu-

late p_{\min} recursively,

$$p_{\min} = \Pi(0,1)\Pi(1,2) \cdots \Pi(n-1,n) = \left(\prod_{i=1}^n L_i \right)^{-a}. \quad (5a)$$

For a finite value of L , the hierarchy of open spaces continues only down to a length scale $L_n = 1$. Substituting $\Pi(i, i+1) = L_i^{-a}$ into (5a), we find

$$p_{\min} = a^{-n(n+1)a/2} L^{-an} \sim \exp(-A'n^2 - A''n \ln L). \quad (5b)$$

Since $n = -\ln L / \ln a$, we recover Eq. (1c). Thus the concept of a fractal distribution of open spaces in which diffusion takes place seems to capture the essential features of DLA structure. The only assumptions in our derivation are that the hierarchy of voids connecting the growth site with minimum growth probability to the outside of the cluster are self-similar and are connected to each other by narrow channels whose diameter is *not* self-similar. Hence the result $\gamma = 2$ is possibly more general than the model used to derive it. In particular, it should hold even when the channel width increases with M so long as the exponent is smaller than $1/d_f$.

In summary, we calculated the minimum growth probability for DLA as a function of the cluster mass M , and found a singularity of the form (1c). We interpreted this result in terms of a simple model for DLA structure. The form of (1c) shows that $F(\beta)$ diverges very slowly, hence explaining why previous work had difficulty seeing the singularity in the multifractal spectrum of a "typical" DLA aggregate. The form of (1c) is consistent with the idea that the natural scaling variables are $-\ln p$ and $\ln M$ exactly as in random multiplicative processes; in terms of these natural variables, the logarithmic singularity of (1c) becomes a power law with "critical exponent" γ .

We are grateful for discussions with A. Aharony, P. Alström, R. Blumenfeld, A. B. Harris, D. Stauffer, and T. Vicsek, and for support from NATO, Minerva Gesellschaft für die Forschung m.b.H., ONR, and the NSF-Germany program. We especially thank C. Amitrano and T. C. Halsey for extremely useful discussions; C. Amitrano for permission to use her Green's-function program for smaller values of mass.

Note added.—After this work was submitted, we learned of several related works bearing on the results of this work: (i) Wolf¹⁴ confirmed the behavior of Fig. 1(a) for $M < 65$, and (ii) Barabasi and Vicsek¹⁵ studied the channel-size distribution function.

¹T. A. Wittmann and L. Sander, Phys. Rev. Lett. **47**, 1400 (1981); Phys. Rev. B **27**, 5686 (1983).

²There have appeared many books, reviews, and conference proceedings dealing with the fractal and multifractal properties

of DLA—the most recent of these is *Fractals in Physics: Essays in Honor of B. B. Mandelbrot*, edited by A. Aharony and J. Feder (North-Holland, Amsterdam, 1990).

³(a) R. Blumenfeld and A. Aharony, Phys. Rev. Lett. **62**, 2977 (1989); (b) P. Trunfio and P. Alström, Phys. Rev. B **41**, 896 (1990).

⁴(a) B. Mandelbrot and T. Vicsek, J. Phys. A **20**, L377 (1989); (b) A. B. Harris and M. Cohen, Phys. Rev. A **41**, 971 (1990).

⁵The system size is very large compared to previous accurate calculations of the growth site probability distribution. For example, the classic work of C. Amitrano, A. Coniglio, and F. di Liberto [Phys. Rev. Lett. **57**, 1016 (1986)] is limited to clusters of $M < 150$. Our systems are more than an order of magnitude larger. The central processing unit (CPU) time needed to calculate all the p_i accurately (even p_{\min}) is about 2 h for a cluster of mass 2429 using the IBM 3090 with a vector facility. In this calculation, the main loop is fully vectorized. We made a sequence of cluster sizes up to mass 2429 for 74 clusters on the triangular lattice and 55 clusters on the square lattice, or about 300 h of IBM 3090 CPU time.

⁶S. Havlin and B. L. Trus, J. Phys. A **21**, L731 (1988).

⁷In the simulation, we chose $R_1 = 4R_0 + 7$ and $R_2 = 5R_0 + 5$, where R_0 is the maximum span of the cluster for a given size M . We found that further increase of R_1 and R_2 has a negligible effect on the p_i .

⁸The idea of quenched average comes from the study of disordered systems. Consider, e.g., a system with some disorder in it. If the disorder is quenched (frozen), one must estimate thermodynamic quantities based on measurements on few samples. One could average the partition function Z_i for sample i and so calculate the "annealed" free energy as $F_A \equiv -kT \times \ln \langle Z_i \rangle$. Since the variation of Z_i between different samples is huge (typically several orders of magnitude), F_A is dominated by the sample which has the largest Z_i . Therefore, F_A cannot be used as a "typical" free energy. The way to get around this problem is to average $\ln Z_i$ instead of Z_i . Since the sample-to-sample fluctuations for $\ln Z_i$ is quite small, the "quenched" free energy $F_Q \equiv -kT \langle \ln Z_i \rangle$ is not dominated by a single configuration. Therefore, we use the quenched average as a representative of a typical configuration. See T. C. Halsey, in *Fractals: Physical Origin and Properties*, Proceedings of the 1988 Erice Workshop on Fractals, edited by L. Pietronero (Plenum, London, 1990); Ref. 3(a).

⁹Here $\beta = -3$ is the smallest value of β we can choose without overflow error.

¹⁰Our results for large negative β [Fig. 2(b)] suggest that (4) becomes an equality for large M .

¹¹In the language of statistical mechanics, some workers view $Z(\beta, M)$ as a *partition function*, $F(\beta)$ as a *free energy*, and β as an *inverse temperature*. If in the thermodynamic limit $M \rightarrow \infty$ the free energy $F(\beta)$ diverges for β below a critical moment β_c , then there is a singularity in the free energy at β_c which is customarily termed a "phase transition."

¹²J. Lee and H. E. Stanley, Phys. Rev. Lett. **61**, 2945 (1988).

¹³L. A. Turkevich and H. Scher, Phys. Rev. Lett. **55**, 1026 (1985).

¹⁴M. Wolf (to be published).

¹⁵A. Barabasi and T. Vicsek, J. Phys. A (to be published).