Occupancy-Probability Scaling in Diffusion-Limited Aggregation

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A continuous-time random-walk theory of diffusion-limited aggregation yields perimeter occupancy probabilities. Scaling relates the fractal dimension D to the cluster-tip occupancy probabilities. These agree with the analytic probabilities near cusps of a lattice-symmetric array of traps. On a two-dimensional square lattice $D=\frac{5}{3}$, whereas $D=2$ for the Eden model, and $D=\frac{4}{3}$ for the $\eta=2$ dielectric breakdown model. D is not universal: $D=\frac{7}{4}$ for the two-dimensional triangular lattice. The square and triangular lattices bracket ($\pm 2.5\%$) Meakin's large off-lattice simulations ($D=1.71$).

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The irreversible aggregation of colloids[1] and aerosols[2] is often rate limited by diffusion of the particles to the surface of the aggregate. Similar dendritic patterns[3] arise in chemical precipitation from a supersaturated solution or upon crystallization from a supersaturated melt.[4] The multibranched aggregates so formed have been attributed[4] to short-wavelength growth instabilities inherent in these diffusion-limited processes.

Witten and Sander[5,6] have devised a lattice model which simulates such diffusion-limited aggregation (DLA): A cluster is grown by successive accretion of random walkers to perimeter sites. This DLA model is sufficiently simple as to be amenable to numerical simulation.[5-7] These computer experiments have established scale invariance for the clusters and have characterized them by a Hausdorff (fractal) dimension D.

Witten and Sander[6] argue against an upper critical dimensionality for DLA. Several conjectures[7,8] attempt to relate D to the Euclidean dimension d in which the cluster is embedded. While inconclusive as to establishing D, renormalization-group arguments[9] suggest that DLA is in a universality class distinct from both equilibrium lattice animals and the unrestricted random walk. This Letter addresses the theoretical questions of universality and the Hausdorff dimension of DLA clusters.

We first reformulate DLA as a continuous-time random walk (CTRW).[10] Our formal solution for a given cluster yields the perimeter occupancy probabilities. We utilize this probability distribution to generate DLA clusters, without simulating the entire lattice random walk.

Scaling identifies D from the occupancy probability for the maximally extending portion of the cluster. The DLA cluster is thus characterized not by its complicated random interior but rather by the simple growth of its perimeter. We argue that the occupancy probabilities for the unscreened DLA cluster tips may be obtained from occupancy probabilities for similar tips of regular (lattice-symmetric) clusters. The fundamental feature of the growing boundary is its cusp structure; this feature is entirely reproduced by the simpler geometry. Application of the analytic occupancy probabilities for a square to DLA on a two-dimensional square lattice yields $D=\frac{5}{3}$; we similarly obtain $D=\frac{7}{4}$ for a 2D triangular lattice.

For specificity, consider a 2D square lattice. Let $R(\mathbf{s},t)$ be the probability (per unit time) for a random walker just to have arrived at a site $\mathbf{s}$ at time t. Let $\psi(\mathbf{s},\mathbf{s}';t)$ be the joint probability (per unit time) both that the time interval between successive arrivals is t and that the displacement that occurs is from $\mathbf{s}'$ to $\mathbf{s}$. We consider an intermediate stage where the lattice contains N contiguously occupied sites. We distinguish perimeter sites $\mathbf{p}\in\Pi$, which are perfectly absorbing traps, from all other lattice sites, which possess nearest-neighbor transition rates W:

$$\psi(\mathbf{s},\mathbf{s}',t)=\begin{cases}0, & \mathbf{s}'\in\Pi,\\ W(\delta_{\mathbf{s},\mathbf{s}'\pm a\hat{\mathbf{x}}}+\delta_{\mathbf{s},\mathbf{s}'\pm a\hat{\mathbf{y}}})e^{-4WT}, & \mathbf{s}'\notin\Pi,\end{cases} \tag{1}$$

where a is the lattice constant. The Laplace transform of $R(\mathbf{s},t)$ satisfies

$$\tilde{R}(\mathbf{s},u)=\sum_{\mathbf{s}'\notin\Pi}\tilde{\psi}(\mathbf{s}-\mathbf{s}';u)\tilde{R}(\mathbf{s}',u)+\delta_{\mathbf{s},\mathbf{s}_0}, \tag{2}$$

with an initial site $\mathbf{s}_0$ for the random walker. Formally,[11]

$$\tilde{R}(\mathbf{s},u)=\tilde{G}(\mathbf{s}-\mathbf{s}_0,u)-\sum_{\mathbf{p}\in\Pi}[\tilde{G}(\mathbf{s}-\mathbf{p},u)-\delta_{\mathbf{s},\mathbf{p}}]\tilde{R}(\mathbf{p},u), \tag{3}$$

where the Green's function for the perfect lattice is given by

$$\tilde{G}(\mathbf{s}-\mathbf{s}_0,u)=\frac{1}{\pi^2}\int_0^\pi d\theta_x\int_0^\pi d\theta_y\frac{\cos m\theta_x\cos n\theta_y}{1+(1/2\sigma)(\cos\theta_x+\cos\theta_y)}, \quad (4)$$

with $\sigma=1+u/4W$ and $\mathbf{s}-\mathbf{s}_0=ma\hat{\mathbf{x}}+na\hat{\mathbf{y}}$. Morita's recursion relations[12] facilitate use of these lattice Green's functions. Evaluating (3) for a perimeter site $\mathbf{p}$, we have

$$\tilde{G}(\mathbf{p}-\mathbf{s}_0,u)=\sum_{\mathbf{p}'\in\Pi}\tilde{G}(\mathbf{p}-\mathbf{p}',u)\tilde{R}(\mathbf{p}',u). \quad (5)$$

The probability that the $(N+1)$st random walker ultimately lands at $\mathbf{p}$ is given by $P(\mathbf{p})=P(\mathbf{p},t\to\infty)=\tilde{R}(\mathbf{p},u\to 0)$, and so we need only invert (5) in the limit $u\to 0$. Thus the CTRW *exactly solves* the DLA growth problem: Given a cluster, we obtain the probability for the next random walker to occupy each perimeter site.

We have incorporated this exact solution into an algorithm for generating DLA clusters, without simulating the entire random walk. Our specific realization of the random walk is accomplished by inverting $R=C(\mathbf{p})$, where R is a random number and $C(\mathbf{p})$ is the cumulant of $P(\mathbf{p})$. In this way, we iteratively build up the cluster; an example ($N=150$) is shown in Fig. 1. The method becomes unwieldly for larger clusters, because of the matrix inversion of (5), the matrix dimensions being proportional to the number of perimeter sites.[13] Also shown in Fig. 1 are the probabilities for subsequent occupancy of the perimeter sites. We emphasize that while our clusters are not large, this method provides the additional information of the occupancy probabilities, which, in the usual technique of brute-force simulation, can only be achieved by many realizations *at each stage of the growth.*

We evaluate D for our clusters from the number of occupied sites within a radius r about the center of mass: $N_{\text{occ}}(r)\sim(r/a)^D$. For the square lattice we find $D\simeq 1.7$, consistent with the largest simulations.[7] Algebraic behavior obtains for $N\gtrsim 20$.

Our solution for the occupancy probabilities motivates a scaling argument for D. As the cluster interior is screened[6] by its exterior branches [screening length ξ^N], interior perimeter sites have negligible probability of subsequent occupancy. Consistent with this screening, the DLA cluster cannot be "fully developed" to its tips: Near the tips, the cluster has fewer occupied sites than it would were it fully developed to its nominal radius $r_0^{(N)}=N^{1/D}a$. As it is fully developed only within a radius $r_-^{(N)}=r_0^{(N)}-\xi^{(N)}$, it must extend further, to a distance[14] $r_+^{(N)}>r_0^{(N)}$ (Fig. 2). In what follows, we use only that $r_0^{(N)}$, $\xi^{(N)}$, and $r_+^{(N)}$ scale similarly with N, which scaling is guaranteed by self-similarity.

We now show that the occupancy-probability density $p_N(r)$ determines D. Given an N cluster, the probability that the $(N+1)$st random walker will land at the outermost portion of the cluster [the annulus at $r_+^{(N)}$] is

$$P_{\max}=\int_{r_+^{(N)}-a}^{r_+^{(N)}}p_N(r)\,dr, \quad (6)$$

in which case the cluster grows: $r_+^{(N+1)}=r_+^{(N)}+a$. If the $(N+1)$st random walker lands deeper within the cluster [but still in the edge region $r_-^{(N)}<r<r_+^{(N)}$], the cluster does not grow: $r_+^{(N+1)}=r_+^{(N)}$. Combining

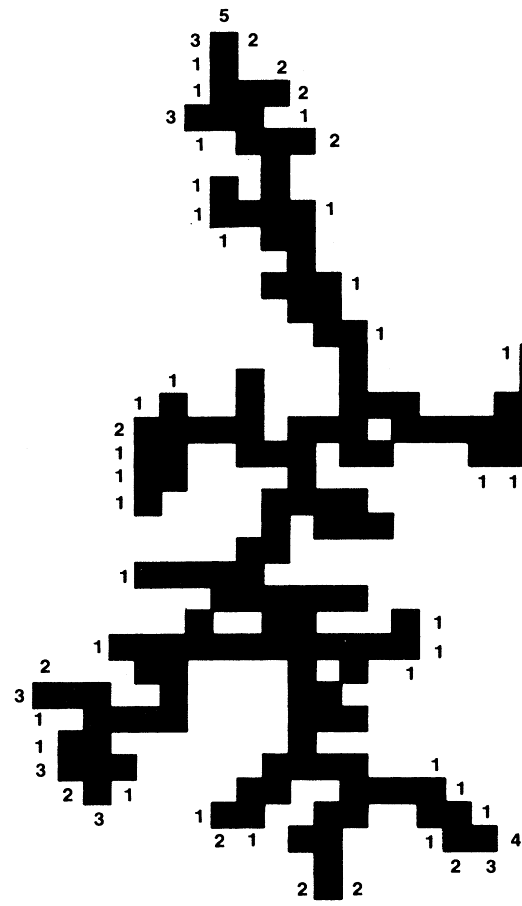

FIG. 1. A cluster ($N=150$) grown according to the CTRW algorithm. Shown also are occupancy probabilities (in percent) for all perimeter sites with $P(\mathbf{p})>0.005$. Appreciable occupancy probability occurs only at the tips of the cluster; the interior is screened.

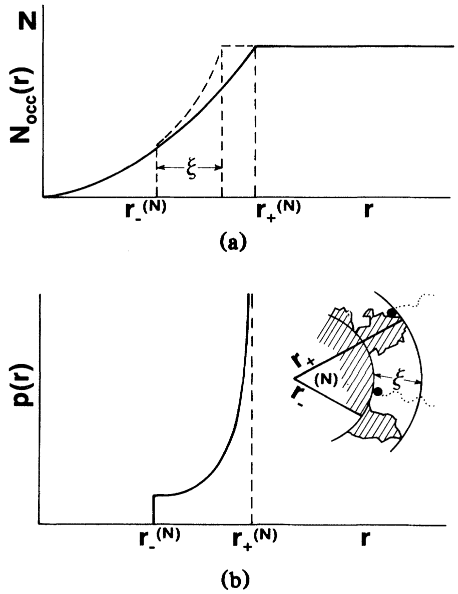

FIG. 2. (a) Schematic of the number of occupied sites $N_{occ}(r)$ within distance r from the center of mass. The cluster is "fully developed" $[N_{occ}(r) \sim (r/a)^D]$ for $r < r_-^{(N)}$ with a lower-density "edge" region for $r_-^{(N)} < r < r_+^{(N)}$. A cluster with no edge region would extend to a nominal radius $r_0^{(N)}$. All growth takes place in this edge region. (b) Schematic of the occupancy probability density $p_N(r)$ in the edge region. No growth occurs for $r < r_-^{(N)}$: $p_N(r) = 0$. $p_N(r)$ is singular at $r_+^{(N)}$, reflecting maximal growth probability at the tips. Inset: Two possible random walkers, one adhering near a cluster tip ($r_+^{(N)}$), and the other adhering deep within the edge region ($r_-^{(N)}$).

these alternatives,

$$r_+^{(N+1)} = P_{\max}(r_+^{(N)} + a) + (1 - P_{\max}) r_+^{(N)}.$$

For large N,

$$dr_+^{(N)}/dN = P_{\max}(N)a. \qquad (7)$$

The growth of the cluster is thus driven by the occupancy probability of the tips. If particles are added at the tips, the cluster grows; if particles are added to the interior, the cluster does not grow. With cluster growth, the occupancy probability for the next random walker is altered; otherwise the next random walker experiences essentially the same perimeter of traps. Hence (7) also self-consistently expresses the change in occupancy-probability density as particles are added to the cluster. Since $r_+^{(N)} \sim N^{1/D}a$, (7) and the divergence of $p_N(r)$ at r_+^N determine D.

As we require $p_N(r)$ only at the tips, the intricacies of the random, ramified cluster interior are irrelevant—*all the growth takes place at the tips.* We thus may utilize *any* object possessing the same cusp structure as a DLA cluster. This is the fundamental feature of our approach to the probability scaling: D does not depend on the cluster's random, irregular structure but rather on *how it grows.* We can thus utilize a regular object with the symmetry of the lattice—for the square lattice, a square with sides $2L$—i.e., we consider a random walker in the presence of a square array of absorbing traps. We compute the occupancy probabilities as a function of distance δs away from the corners of the square.

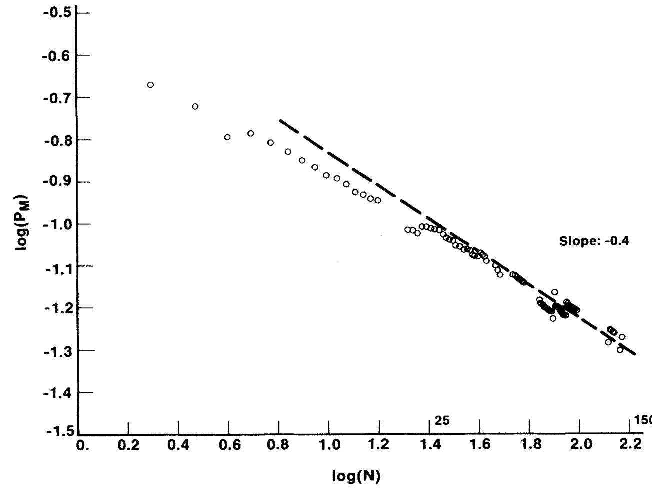

FIG. 3. $P_{\max}(N)$ for the growing cluster of Fig. 1, scaling as predicted.

For $t \to \infty$, the random-walk diffusion problem reduces to Laplace's equation with perfectly absorbing walls. This is equivalent[15] to the electrostatic problem $\mathbf{E} = -\nabla\phi$ of a conducting square held at $\phi = 0$ with a conducting circle at infinity held at $\phi = 1$. The probability density,

$$p(\mathbf{r}) = |\mathbf{E}(\mathbf{r})| / \int_\Pi |\mathbf{E}(\mathbf{r})| ds, \qquad (8)$$

is normalized over the perimeter Π of the square.

The electrostatics may be solved exactly via conformal transformation, $E_\perp \sim 1/\mathrm{cn}\zeta$, where the variable ζ is given implicitly by

$$z = x + iy = \frac{-iL}{E - K/2}\left[\mathrm{zn}\zeta + \left(\frac{E}{K} - \frac{1}{2}\right)\zeta\right] + L. \qquad (9)$$

Here $\mathrm{cn}\zeta$ and $\mathrm{zn}\zeta$ are Jacobi elliptic functions, and K and E are the usual complete elliptic integrals, all of modulus $k = 1/\sqrt{2}$. Near the tips ($\delta s \to 0$), the probability density (8) diverges as

$$p(\delta s) \sim \frac{(E - K/2)^{2/3}}{\sqrt{2}\pi L}\left(\frac{L}{12\delta s}\right)^{1/3}. \qquad (10)$$

We assert that the scaling of the probability density near a square-lattice DLA tip is reproduced by that of the square near a corner,

$$p_N(r) \sim \tfrac{2}{3}\zeta^{(N)-2/3}(r_+^{(N)} - r)^{-1/3}, \qquad (11)$$

normalized over the screening length $\zeta^{(N)}$. We verify this assertion below (Fig. 3). With use of (11) in (6), $P_{\max} \sim (a/\zeta)^{2/3}$. Substitution into (7) gives $N \sim (r/a)^{5/3}$, i.e., $D=\frac{5}{3}$ for 2D DLA on a square lattice.

We verify the consistency of using (11) to obtain the singular behavior of the DLA probability density near the cluster tips. Figure 3 displays $P_{\max}(N)$ as the cluster of Fig. 1 is grown. Even for such small clusters, $P_{\max} \sim \zeta^{-2/3} \sim N^{-2/5}$. Similar scaling ($P_{\max} \sim N^{-0.39}$) is observed by Meakin[16] in averages over many (10^5) simulations of large clusters ($N \sim 5 \times 10^4$).

We remark that (8) may be trivially extended,

$$p(\mathbf{r}) = |\mathbf{E}(\mathbf{r})|^{\eta} \Big/ \int_{\Pi} |\mathbf{E}(\mathbf{r})|^{\eta}\, ds, \qquad (12)$$

to yield $D=2$ for the Eden cancer model[17] ($\eta=0$) and $D=\frac{4}{3}$ for the $\eta=2$ Brown-Boveri dielectric breakdown model[15] on a 2D square lattice.

We have investigated the lattice dependence of the above results. The random-walk problem on a 2D triangular lattice can also be solved, where we utilize a hexagonal array of traps to obtain the probability density near a DLA cluster tip,

$$p_N(r) \sim \tfrac{3}{4}\xi^{(N)-3/4}(r_+^{(N)} - r)^{-1/4}. \qquad (13)$$

Use of (13) in (6) and substitution into (7) gives $D=\frac{7}{4}$ and $P_{\max} \sim N^{-3/7}$. The results for fourfold- ($D=\frac{5}{3}$) and sixfold-coordinated ($D=\frac{7}{4}$) lattices bracket and are within $\pm 2.5\%$ of the latest[18] numerical results ($D=1.71$) for DLA *without* a lattice, where we expect averaging over coordination.

Finally the random-walk problem on *anisotropic* fourfold-coordinated lattices (2D oblique lattices with arbitrary angle of inclination β) can also be solved by conformal transformation. Near the sharp corner,

$$p_N(r) \sim [\pi/(2\pi-\beta)]\xi^{(N)-x}(r_+^{(N)} - r)^{x-1}, \qquad (14)$$

with $x = \pi/(2\pi-\beta)$. In this case, $D=(3\pi-\beta)/(2\pi-\beta)$; while nonuniversal, D is stringently bracketed: $1.50 < D < 1.67$. Numerical simulations on these anisotropic lattices will provide a strong test of the theory.

In summary, we have presented a CTRW reformulation of DLA. Our solution yields, at each stage of the growth, the perimeter occupancy probabilities of the random walker. Growth occurs predominantly at the cluster tips, and is controlled by the occupancy probability $P_{\max}$ of these maximally extending tips. The scaling of $P_{\max}$ determines the Hausdorff dimension D of the cluster, and may be obtained by use of the singular part of the probability density for a regular object with the same cusps as exhibited by the DLA cluster tips. We find $D=\frac{5}{3}$ for DLA on a 2D square lattice and $D=\frac{7}{4}$ on a 2D triangular lattice, thus evincing nonuniversality. We have verified the predicted scaling, $P_{\max} \sim N^{-2/5}$, for the square lattice.

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