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ATOMS, MOLECULES, OPTICS

Diffusion-Limited Aggregation: A Continuum Mean Field Model

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Abstract—Mean field theory is used as a basis for a new approach to analyzing fractal pattern formation by diffusion-limited aggregation. A coarse time scale is introduced to take into account the discrete nature of DLA clusters. A system of equations is derived and solved numerically to determine the fractal dimension and density of a cluster as a function of distance from its center. The results obtained are in good agreement with direct numerical simulations. © *2005 Pleiades Publishing, Inc.*

1. INTRODUCTION

Diffusion-limited aggregation (DLA) is the generic name for a class of models describing the formation of a cluster by addition of randomly walking particles sticking to it. The original model introduced in [1] has drawn continuing attention (e.g., see reviews in [2, 3]), because it applies to a variety of physical processes, including dielectric breakdown, solute aggregation, and growth of bacterial colonies. Moreover, the DLA cluster is of mathematical interest as an object with fractal dimension.

To date, detailed numerical simulations of the growth and scaling properties of DLA clusters have been performed. In particular, the aggregate density field was found to exhibit fractal behavior: the aggregate mass is a power of its characteristic radius, with exponent $d = 1.71$ and 2.50 in the two- and three-dimensional continuum models, respectively [4].

Furthermore, the statistical properties of an individual branch of the DLA cluster have been analyzed by examining quasi-one-dimensional random walk as a model of fractal aggregate growth in a domain of length much greater than its width. A summary of early studies in this area was presented in [5]. It was found that $d = 0.66$ for the direction of power-law density profile. This exponent was corrected in extensive numerical experiments: $d = 0.72$ [6]. Currently, diffusion-limited aggregation in the absence of external forces or interaction between particles is a well-studied process that may serve as a benchmark test for new numerical methods [7].

In theory, the most substantial progress has been achieved by applying renormalization-group methods, but only in discrete DLA of two types. In one approach, a parameter is sought that leaves invariant the mean occupancy distribution in a lattice DLA model [8]. In the other [9], the cluster is generated by using a conformal map of the unit circle such that the random walker is mapped to a randomly chosen point on the growing cluster perimeter at each step of an iterative process. A modification of the latter approach was used in [6] to simulate one-dimensional diffusion-limited aggregation in channel geometry.

However, it still remains unclear if DLA is tractable as a continuum model formulated in terms of differential equations. In the original model proposed in [10], a mean field theory that relates the continuous aggregate density distribution to the probability distribution of a random walker coming from infinity was developed by using a power series expansion in cluster density. A refined mean field theory was proposed in [11]. These models provided a qualitative explanation of the process, but the predicted dimension was substantially lower than the measured one. For this reason, various modified mean field approaches were proposed in subsequent studies.

In [12], the growth rate was assumed to be proportional to the gradient of the distribution of the randomly walking particle, rather than to the distribution itself. In [13], the growth rate was represented as a power of the field variable with a phenomenological exponent greater than unity to cut off growth at small density. As a consequence, density buildup was observed in the cluster front zone, and a higher fractal dimension is obtained. In [14], the latter model was substantiated by showing that the growth rate is proportional to a quadratic combination of density and its derivatives if the probabilities of attachment and interaction between random walkers are equal. This result was obtained by replacing the Boltzmann collision integral with a system of differential equations.

Thus, even though the understanding of aggregation kinetics has improved owing to progress in the framework of mean field theory, the "final solution" has never been found. In this paper, we show how the mean field

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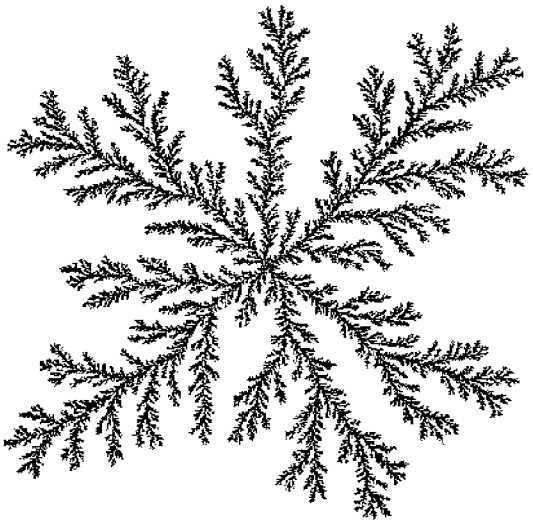

Fig. 1. Typical DLA cluster.

theory can be modified to provide adequate description of the scaling behavior of the system.

2. MODEL

In this section, we formulate a mean field DLA model based on the equations proposed in [11]. We present the complete derivation of a system of equations and demonstrate how a coarse time scale can be introduced.

2.1. Preliminary Analysis

Generally, two-dimensional aggregation kinetics are modeled as follows. While an immobile seed of diameter a is located at the origin, a particle born at a distance R from the seed executes a random walk inside the circle of radius R until it either escapes from the circle or comes in contact with the seed. The particle crossing the circle reappears at a random point on it. The particle that contacts the seed sticks to it, and then a new particle is launched from a random point on the circle. As the process repeats, each new particle can stick to any particle on the cluster boundary. To minimize the effects due to the finite size of the system, the birth radius R is assumed to be much larger than the characteristic radius of the cluster.

A typical DLA cluster has a highly ramified fractal structure (see Fig. 1). New particles are captured in its front zone. In mean field theory, the structure is smeared, and the average perimeter of a cluster is difficult to define.

To describe the growth of a cluster, we introduce the characteristic function $\hat{\rho}(r, \phi)$, equal to unity at the locations occupied by the aggregate and zero elsewhere. The characteristic function $\hat{u}(r, \phi)$ is defined analogously for the random walker. In mean field theory, these functions are ensemble averaged by assuming that the particle distribution is isotropic. As a result, we obtain probability density profiles $\rho(r)$ and $u(r)$ for the cluster and the random walker, respectively.

The entire two-dimensional domain is partitioned into square cells of area a^2, where a is the particle diameter. For each cell containing a particle with probability one, we set $\rho = 1/a^2$, so that its integral over the cell is unity. To facilitate analysis, we perform the change $\rho \longrightarrow a^2\rho$, $u \longrightarrow a^2u$ so that ρ or u is unity in each cell occupied by a particle. Then, the number of particles in the cluster is expressed as

$$N = \frac{1}{a^2}\int_D \rho\, ds, \tag{1}$$

where ds is an area element and D is the region occupied by the cluster.

In the model proposed in [11], the cluster growth is modeled by the system of equations

$$\begin{aligned} \frac{\partial \rho}{\partial t} &= u(\rho + a^2\Delta\rho), \\ \frac{\partial u}{\partial t} &= a^2\Delta u - u(\rho + a^2\Delta\rho), \end{aligned} \tag{2}$$

supplemented with the boundary condition $u(R, t) = c$, which represents a source of random walkers located at the birth radius. The kinetic equation for the random-walker distribution describes diffusion and adsorption onto the aggregate. In the kinetic equation for the cluster density distribution, the terms $u\rho$ and $a^2\Delta\rho$ represent the contributions of random-walker–cluster contact and the lateral growth due to nonlocal interaction, respectively.

Since random walk is much faster than cluster growth, we set $u_t = 0$:

$$\begin{aligned} \frac{\partial \rho}{\partial t} &= u(\rho + a^2\Delta\rho), \\ 0 &= a^2\Delta u - u(\rho + a^2\Delta\rho). \end{aligned}$$

As mentioned in the Introduction, this model contains the basic features of the model, but fails to predict the correct fractal dimension, because it ignores the discrete nature of the DLA cluster.

This explanation is corroborated by numerical analyses of the following model [15, 16]. The region occupied by the cluster is divided into annuli of width a. The annulus of radius r_n is characterized by the largest number M_n of particles that can be placed inside it (estimated as $M_n \sim 2\pi r_n a/a^2$) and the actual number N_n of particles contained in it. The probability of adsorption of a random walker onto the annulus is N_n/M_n. A ran-

dom walker contained in the nth annulus can be adsorbed on the $(n-1)$th, $(n+1)$th, or nth annulus. If $N_n/M_n \ll 1$, then the total adsorption probability can be approximated as

$$\sum_{k=-1}^{1} \frac{N_{n+k}}{M_{n+k}}.$$

When a random walker is adsorbed onto the nth annulus, the value of N_n increases by unity. Otherwise, it drifts into an adjoining annulus with a probability of $1/2 \pm a/r_n$, where the plus and minus signs in $\pm a/r_n$ correspond to the outer and inner annuli, respectively.

This model is simpler than direct numerical simulation, because it makes use of the axial symmetry of the cluster and retains information about the number of particles in each annulus. Nevertheless, the predicted fractal dimensions agree with measured ones, 1.65 [15] and 1.72 [16].

Returning to the mean field theory and taking into account the discrete nature of the cluster, we define $u(r, \tau)$ as the probability distribution for a particle to be located at a distance r from the origin at an instant τ. Furthermore, we introduce an initial distribution $u(r, 0)$ and impose the impermeability condition $u_r|_{r=0,R} = 0$. We use a random-walk time variable τ and a discrete cluster-growth time n to allow for the disparity between the corresponding time scales, because it is obvious that the adsorption of a single random walker does not result in any significant change in the overall cluster geometry.

To derive a kinetic equation for $u(r, \tau)$, we use simple partition into cells in the (x, y) plane. We introduce the probability $\rho(x, y)$ of finding a cluster particle in the cell with coordinates x and y and the probability $u(x \pm a, y \pm a, \tau)$ that the random walker occupies a neighboring cell (see Fig. 2). Treating the random walk followed by adsorption onto the cluster and the ensuing evolution of cluster density as independent processes, i.e., assuming that the change in cluster density over the random-walker lifetime is negligible, we write the following difference equation for $u(x, y, \tau)$:

$$u(x, y, \tau+1) = (1-\rho(x, y))[u(x-a, y, \tau) + u(x+a, y, \tau) + u(x, y-a, \tau) + u(x, y+a, \tau)]/4. \quad (3)$$

Then, we introduce a time increment $\delta t \sim a^2$ and use a Taylor series expansion to rewrite (3) as

$$\frac{\partial u}{\partial \tau} = \frac{a^2}{4}\Delta u - \rho\left(u + \frac{a^2}{4}\Delta u\right), \quad (4)$$

where the Laplacian to be calculated in Cartesian coordinates reduces to

$$\Delta = \frac{\partial^2}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r}$$

under the assumption of axial symmetry.

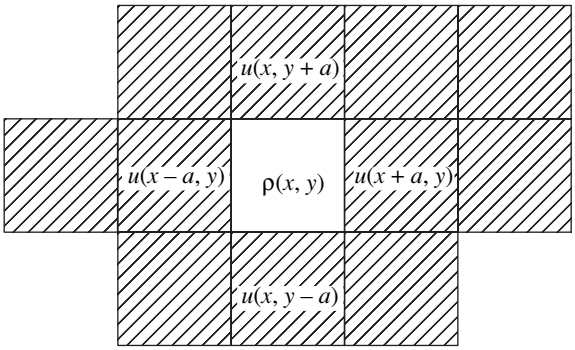

Fig. 2. Lattice model.

The resulting equation is similar to the second one in (2) up to second-order terms, but it allows for nonlocal adsorption, vanishing diffusion through fully occupied regions (where $\rho_n(r) = 1$), and free diffusion into unoccupied regions (where $\rho_n = 0$). We believe that this equation provides a more accurate description.

2.2. Refined Model

Since the asymptotic solution to the diffusion–absorption equation derived above is such that

$$u(\tau) \le \exp(-\rho\tau),$$

there exists a bounded function

$$U(r) = \frac{1}{T^*}\int_0^{T^*} u(r, \tau)d\tau$$

describing, up to normalization, the time-averaged random-walker distribution in space. The integral of Eq. (4) with respect to τ from zero to the random-walker lifetime T^* combined with the condition $u(T^*) = 0$ yields

$$-u(r, 0) = \frac{a^2}{4}\Delta U(r) - \rho\left(U(r) + \frac{a^2}{4}\Delta U(r)\right).$$

This equation is supplemented with the impermeability condition

$$\frac{\partial}{\partial r}U(r)|_{r=0,R} = 0.$$

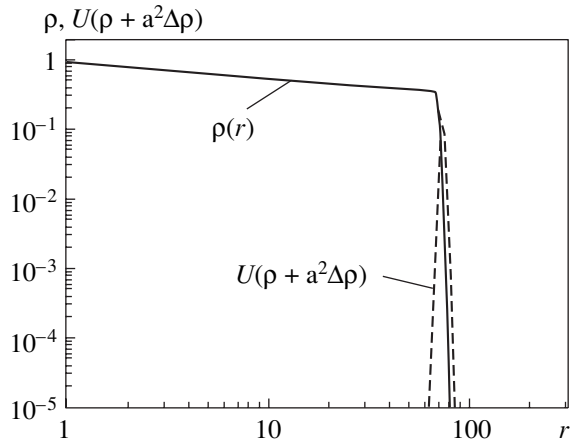

Fig. 3. Cluster density ρ and density of a newly adsorbed single-particle layer.

The equation for the cluster density ρ corresponding to the first equation in (2) can be written as

$$\frac{\partial\rho}{\partial n} = CU(\rho + \Delta\rho),$$

where C is a normalization factor and n is the number of an iteration step. In the conventional mean field theory, the random walker hitting the cluster "spreads" over its perimeter, which results in a slight increase in the cluster size. According to (1), the ensuing change in the cluster density is such that

$$\int[\rho(n+1) - \rho(n)]2\pi r dr/a^2 = 1.$$

Hence,

$$C_0 = \frac{a^2}{2\pi\int U(\rho + \Delta\rho)rdr}.$$

A numerical analysis of the corresponding system of equations leads to results analogous to those reported in [11]; in particular, the fractal dimension of the 2D cluster is unity (see Section 3.1).

However, the adsorption of a particle at a point on the real cluster perimeter has a negligible effect on the probability of particle addition at other locations. Therefore, we can introduce a coarse time scale by assuming that the density of a cluster changes only after its entire perimeter is covered by newly adsorbed random walkers. Then, we obtain

$$C = \frac{2\pi R_b}{a}C_0 = \frac{2\pi R_b}{a}\frac{a^2}{2\pi\int U(\rho + \Delta\rho)rdr}, \qquad (5)$$

where R_b is the characteristic distance from the origin to the cluster boundary.

Figure 3 shows a typical density profile and a curve $U(\rho + \Delta\rho)$ characterizing the location of new adsorbed particles. The latter curve has a sharp peak in the neighborhood of the cluster boundary and vanishes outside it. Accordingly, we can approximately replace R_b with r in expression (5) and write the final system of equations as follows:

$$\begin{aligned}\rho_{n+1} &= \rho_n + CU(\rho + \Delta\rho),\\ -u(r,0) &= \Delta U(r) - \rho_n(r)\left(U(r) + \frac{a^2}{4}\Delta U(r)\right),\end{aligned} \qquad (6)$$

where

$$C = \frac{2\pi r}{a}\frac{a^2}{2\pi\int U(\rho + \Delta\rho)rdr},$$

and n is the number of an iteration step at which the cluster is covered by a new layer of adsorbed random walkers and its boundary shifts by an increment on the order of particle diameter.

3. ANALYSIS IN A PLANAR GEOMETRY

3.1. Numerical Analysis

Using the initial density distribution

$$\rho_0(r) = \exp(-2r/a),$$

we solved Eqs. (6) by successive iteration with steps corresponding to cluster size increments. At each step, the second equation in (6) was computed on a 10^5 point spatial grid by using an explicit finite-difference scheme, the distribution $\rho_n(r)$ obtained at the preceding step, and

$$u_0(r) = \exp(-(r-R)^2/a^2).$$

The resulting solution was substituted into (6) to calculate a normalized increment of the cluster distribution. We set the increment at $r = 0$ to zero, since the region occupied by the seed is impenetrable.

The dashed curve in Fig. 4 is a numerical solution to system (6) for the two-dimensional cluster. Its log–log slope, -0.22 ± 0.02, corresponds to the cluster fractal dimension $d = 1.78 \pm 0.02$. The dotted curve predicted by the mean field theory developed in [11] without introducing any coarse time scale corresponds to a lower dimension ($d = 1$).

3.2. Analytical Solution

Since system (6) is amenable to iterative computation, it can be solved analytically by applying the finite Hankel transform

$$U(p_i, t) = \frac{1}{R^2}\int_0^R U(r, t) J_0\left(p_i \frac{r}{R}\right) r dr,$$

where $J(p_i)$ is the Bessel function. The desired solution can be represented by a series expansion:

$$U(r, t) = 2\sum_i U(p_i, t)\frac{J_0\left(p_i \frac{r}{R}\right)}{J_0^2(p_i)},$$

where p_i denotes the roots of the equation $J_1(p_i) = 0$.

Using the initial condition

$$u(r, 0) = \delta(r - R)/2\pi(r/R)$$

and the boundary conditions

$$\frac{\partial}{\partial r} U(r)|_{r = 0, R} = 0,$$

we obtain an infinite algebraic system of equations for $U(p_i, t)$:

$$U(p_i) + \sum_{p_k} U(p_k) K(p_k, p_i) = \frac{2R^2}{\pi a^2}\frac{J(p_i)}{p_i^2},$$

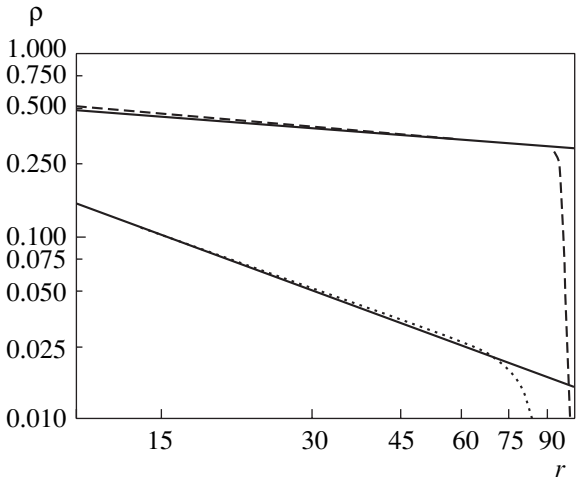

Fig. 4. Cluster density vs. radius predicted in [11] (dotted curve) and by model (6) with coarse time scale (dashed curve). The fractal density is 1.0 and 1.78 in the former and latter cases, respectively.

with

$$K(p_k, p_i) = \frac{8\left(1 - \frac{a^2}{4R^2}p_k^2\right)_R}{a^2 p_i^2 J_0^2(p_k)}\int_0^R \rho(r) J_0\left(p_i \frac{r}{R}\right) J_0\left(p_k \frac{r}{R}\right) r dr.$$

By analogy with Fredholm integral equation of the second kind, an exact solution to this system can be represented in terms of a resolvent kernel:

$$U(p_i) = \frac{2R^2}{\pi a^2}\left[\frac{J(p_i)}{p_i^2} + \frac{1}{D}\sum_k \frac{J(p_k)}{p_k^2} D(p_i, p_k)\right], \quad (7)$$

where

$$D = 1 + \sum_{m=1}^{\infty}\frac{(-1)^m}{m!}\sum_{\alpha_1=1}^{\infty}\cdots\sum_{\alpha_m=1}^{\infty}\begin{vmatrix} K(p_{\alpha_1}, p_{\alpha_1}) & \cdots & K(p_{\alpha_1}, p_{\alpha_m}) \\ \vdots & \ddots & \vdots \\ K(p_{\alpha_m}, \rho_{\alpha_1}) & \cdots & K(p_{\alpha_m}, p_{\alpha_m}) \end{vmatrix},$$

$$D(p_i, p_k) = K(p_i, p_k) + \sum_{m=1}^{\infty}\frac{(-1)^m}{m!}\sum_{\alpha_1=1}^{\infty}\cdots\sum_{\alpha_m=1}^{\infty}\begin{vmatrix} K(p_i, p_k) & K(p_i, p_{\alpha_1}) & \cdots & K(p_i, p_{\alpha_m}) \\ K(p_{\alpha_1}, p_k) & K(p_{\alpha_1}, p_{\alpha_1}) & & \\ \vdots & & \ddots & \vdots \\ K(p_{\alpha_m}, p_k) & K(p_{\alpha_m}, p_{\alpha_1}) & \cdots & K(p_{\alpha_m}, p_{\alpha_m}) \end{vmatrix}.$$

Substituting the derivative of this solution into the system of kinetic equations, we obtain a recursive procedure for calculating cluster growth. However, since it involves calculations of slowly convergent series and integrals containing Bessel functions, this procedure is more difficult to use for evaluating the fractal dimension, as compared to numerical analysis.

4. CONCLUSIONS

A new mean field approach to analyzing fractal pattern formation by diffusion-limited aggregation is proposed. Following previous studies (e.g., see [11, 14, 15]), we assume that the aggregate is axially symmetric. The key distinction of the present approach is the use of a coarse time scale taking into account the discrete nature of DLA clusters. The coarse time scale is introduced by requiring that the addition of a particle to the cluster does not lead to any significant effect on the continuous radial distribution obtained by changing from the discrete to the continuum model and averaging over the angle. To meet this requirement, we assume that the cluster density changes jumpwise only after the entire accessible perimeter of the cluster is covered by a new layer of adsorbed random walkers. As a result, we derive a difference–differential equation with discrete time step. Furthermore, this form of the governing equation entails the use of a normalization factor C proportional to the accessible cluster perimeter.

The value of the normalization factor depends on the dimension of the spatial domain of aggregate growth. For cluster growth on the surface of a cylinder along its element [5, 18], C = const since the corresponding fractal dimension is evaluated by solving a quasi-one-dimensional problem on a line parallel to the direction of aggregate growth. For the two-dimensional cluster, this factor is proportional to the circle perimeter. For the three-dimensional one, it scales with the volume of a spherical layer with thickness equal to the particle diameter, where random walkers are adsorbed. Since the density profile drops near the cluster boundary (see Fig. 3), the function $\rho + \Delta\rho$ has a sharp maximum, which makes it possible to define absorption region and find the normalization factor.

The approach developed in this study is used to obtain a fractal dimension of 1.78 in the two-dimensional DLA model, in good agreement with direct numerical simulations. We have also considered the quasi-one-dimensional and three-dimensional DLA models. In the former case, our solution of the proposed system of equations yields a fractal dimension of 0.80 ± 0.02, in fair agreement with direct numerical simulations [5, 18]. As the space dimension increases to three and higher, the assumption of isotropy progressively becomes less accurate (even if physically plausible) and entails a systematic overestimation of the fractal dimension of the cluster.

Finally, we note that the proposed approach can be naturally generalized to aggregation in a system of particles of several types. In particular, if we assume that particles of the same type stick together, then the normalization factor in each equation corresponding to a particular type of particles will be proportional to the largest volume fraction that can be occupied by particles of this type in the current spherical layer.

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