Fractal structure of drift-diffusion-limited aggregation: Renormalization-group approach

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A real-space renormalization-group method is applied to the drift-diffusion-limited aggregation proposed by Meakin [Phys. Rev. B **28**, 5221 (1983)]. The effects of particle drift on the diffusion-limited aggregation (DLA) are investigated. The aggregation process is transformed from DLA to the ballistic process with increasing drift effect. It is found that at a finite drift velocity the aggregate is compact with a surface fractal dimension between the DLA and ballistic aggregation results with increasing drift effect. The crossover from a fractal structure at short length scales to a compact structure at longer length scales is shown. The multifractal scaling properties of the growth probability distribution on the growing perimeter are found to be described by the set of generalized dimensions and the α - f spectra.

INTRODUCTION

Recently, there has been increasing interest in the problem of geometrical structure in the kinetic growth model.¹⁻¹⁵ A number of models for cluster formation have been proposed and a variety of computer simulations have been carried out to investigate the relationships between the cluster geometry and growth mechanism. The structure of the aggregates strongly depends on the dynamics of the growth process. There is also considerable theoretical interest in this area.¹⁶⁻²⁹ Meakin³⁰ investigated the effects of particle drift on diffusion-limited aggregation (DLA) with the use of Monte Carlo methods. It was found that when particle-drift effects were dominant, the particles followed essentially linear trajectories and the resulting clusters had uniform structure at all but the shortest length scales, and when the effects of drift were small, the particles followed Brownian trajectories and the clusters had the DLA structure. For intermediate cases, the clusters had a structure similar to a DLA cluster at short scales but were uniform at longer length scales. Kapral *et al.*³¹ presented a biased diffusion model. With a Monte Carlo simulation, they found that for thin films there was a crossover between a regime where there were several highly ramified pseudo-one-dimensional clusters and another regime where there was a single cluster which spanned the lattice in both directions.

In this paper, we apply a real-space renormalization-group method³² to the drift-diffusion-limited aggregation model proposed by Meakin.³⁰ We derive the surface fractal dimensions as a function of the Péclet number which represents a ratio of the drift effect to the diffusion effect. The multifractal scaling structure at the growth probability distribution on the growing perimeter is studied with varying Péclet number. We discuss the crossover from a fractal structure at short length scales to a compact structure at longer length scales.

MODEL AND BASIC EQUATION

The concentration C of diffusing particles satisfies the convective diffusion equation under the quasistationary approximation

$$u_i \partial C / \partial x_i = D \nabla^2 C, \quad (1)$$

where u_i is the i -directional velocity and D is the diffusion constant. Under a constant velocity U_x the dimensionless form is as follows:

$$P \partial C / \partial x = \nabla^2 C, \quad (2)$$

where P is the Péclet number, $P = U_x a / D$ (a is the lattice constant), and ∇^2 is the Laplacian. The Péclet number represents a ratio of the drift effect to the diffusion effect. If the Péclet number is large, particle-drift effects are dominant. If one considers convective particle diffusion in the moving frame with constant velocity U_x Eq. (2) becomes the ordinary diffusion equation,

$$\nabla^2 C = 0. \quad (3)$$

This means that the aggregate moves with constant velocity U_x into the diffusion field without convection. We consider the drift-diffusion-limited aggregation process in the picture of a moving aggregate with constant velocity. The process of the formation of the aggregate is purely dissipative without convection in a nonequilibrium steady state. The process is characterized by the theorem of minimum entropy production or minimum dissipation energy.³³

COUPLING CONSTANT IN RENORMALIZATION GROUP

The growth of the aggregate occurs on the perimeter. In the moving frame, the growth probability P_i at the growing-perimeter bond i is given by $P_i \sim -\nabla_i C$, where

P_i is the probability that the bond i becomes part of the aggregate and $\nabla_i C$ is the local concentration gradient at the growth bond i . The basic equation governing the aggregate is given by Eq. (3). We merely solve a Laplace equation for an aggregate cluster inside an infinite diffusion field in the moving frame. Since the process is characterized by the theorem of minimum dissipation energy, we consider the dissipation energy in contrast to the free energy (or Hamiltonian). In the discrete version, the dissipation energy is given by

$$\begin{aligned} H_d &= \sum_i D_i (\nabla_i C)^2 \\ &= D_t (\nabla C_t)^2, \end{aligned} \quad (4)$$

where the first equation represents the summation of the local dissipation energy of each bond over total bonds within the diffusion field, and the second indicates the total dissipation energy in the representation of the total diffusion constant D_t and the total concentration drop ∇C_t between the surface of the aggregate and the external boundary. Thus the dissipation energy form (4) describes the interaction between neighboring concentrations by this simple gradient term $\nabla_i C$. The total mass flow, carrying through the surface of the aggregate, is given by

$$J_t = \sum_{i \in s} D_{s,i} (\nabla C)_{s,i},$$

where the sum indicates the summation over all the bonds on the growing perimeter. We can write the dissipation energy in terms of the surface diffusion constant D_s ,

$$H_d = (\nabla C_t) \left[\sum_{i \in s} D_{s,i} (\nabla C)_{s,i} \right], \quad (5)$$

where $D_{s,i}$ indicates the diffusion constant of the bonds on the perimeter of the aggregate. By using a decimation method, one may write the dissipation energy in terms of the coarse-grained variables $D'_{s',i}, (\nabla C)'_{s',i}$ on the new lattice with the lattice spacing b (b is a scale factor),

$$H_d = (\nabla C_t) \left[\sum_{i \in s'} D'_{s',i} (\nabla C)'_{s',i} \right]. \quad (6)$$

Thus the short-range interaction between neighboring concentrations described by the gradient term is renormalized. The surface diffusion constant D_s is transformed to D'_s . This surface diffusion constant plays the role of the so-called coupling constant in the phase-transition Hamiltonian.

RENORMALIZATION-GROUP METHOD

We shall derive the renormalization-group equation for the surface diffusion constant $D'_s = R(D_s)$. We must distinguish between three types of bonds on the lattice before a renormalization procedure: (a) interior bonds which are occupied by diffusing particles and construct the aggregate, (b) growth bonds which are on the surface of the aggregate and can be successively grown, and (c)

exterior bonds which construct the diffusion field and are in the exterior of the aggregate except for the growth bonds. Figure 1(a) shows a part of the aggregate near the surface where the interior, growth, and exterior bonds are respectively indicated by the bold, wavy, and light lines. We partition all the space of the square lattice into cells of edge b (scale factor), each containing $2b^2$ bonds; an example for $b=2$ is shown in Fig. 2. After a renormalization transformation these cells play the role of "renormalized" bonds. The renormalized bonds are then classified into three types of bonds, similarly to bonds before the renormalization. The renormalization procedure is indicated by Fig. 2. If the cell is spanned with the bonds occupied by the interior bond then the renormalized bond is considered to be interior [Fig. 2(a)]. If the cell is not spanned with the interior bond and is nearest neighbor to the aggregate, then the cell is renormalized as the growth bond on the surface [Fig. 2(b)]. When the cell is constructed by the exterior bond only and is not nearest neighbor with the aggregate cluster, the cell is re-

FIG. 1. Example of the renormalization of a part of the surface layer of an aggregate. Bonds constructing the aggregate are indicated by bold lines and bonds on the perimeter of the aggregate by wavy lines. The lattice on the top side is renormalized to that on the bottom side, according to the rule of renormalization.

FIG. 2. Illustration of the renormalization of a $b=2$ cell for the drift-diffusion-limited aggregation. The renormalization procedure in the vertical direction is shown. There are three types of bonds: interior bonds indicated by bold lines, growth bonds by wavy lines, and exterior bonds by light lines. Examples of the distinct configurations are shown in (a), (b), and (c), which are renormalized as interior, growth, and exterior bonds, respectively.

normalized as the exterior bond [Fig. 2(c)]. Figure 1 shows an example of the renormalization of a part of the aggregate near the surface. The lattice on the top side is renormalized to that on the bottom side, according to the rules of renormalization. We consider the diffusion constants of the two types of bonds, exterior and growth bonds. The diffusion constant of the cell to be renormalized as the exterior bond does not change before and after the renormalization because all the bonds within the cell are occupied by exterior bonds. We consider the diffusion constant of the cell that it is possible to renormalize as the growth bond. We define the diffusion constant of the growth bond as a surface diffusion constant. After many repeated renormalizations, the surface diffusion constant must approach a finite value, but not zero, because the total diffusion constant D_t of the system has a finite value except for zero. The nonlocal nature of the concentration field is taken into account as the diffusion constant of the growth bond (surface diffusion constant). If one considers the renormalization in the vertical direction, we shall take a periodic boundary condition in the lateral direction. The constant voltage is vertically applied. Figure 3 shows all the configurations of the cell that it is possible to renormalize as the growth bond. Let us consider the configurational probability $\tilde{C}_\alpha$ with which a particular configuration α appears. The distinct configurations are labeled by α ($\alpha=0,1,2,3$) in Fig. 3. The configuration (1) is constructed by adding an

FIG. 3. All distinct configurations of the 2×2 cell that it is possible to renormalize as the growth bond. The configuration (1) is constructed by adding an interior bond to the configuration (0). In addition, by adding an interior bond to the configuration (1), the configurations (2) or (3) occur. Also the configuration (3) is directly constructed from the configuration (0).

interior bond to the configuration (0). In addition, by adding an interior bond to the configuration (1), the configurations (2) or (3) occur. These stepwise growth processes are similar to the DLA without the drift. Furthermore, we must introduce another route in which the configuration (3) is directly constructed from the configuration (0). The interface grows uniformly with a finite probability A because the surface of the aggregate moves uniformly with constant velocity. We assume that the ratio of the uniformly growing probability A to the stepwise growing probability $(1-A)$ is proportional to the Péclet number P ,

$$A/(1-A)=P, \quad (7)$$

where $P=U_x a/D$. At the limit of the zero velocity, the uniformly growing probability A becomes zero. The configurational probabilities $\tilde{C}_\alpha$ are given by

$$\begin{aligned} \tilde{C}_1 &= (1-A)\tilde{C}_0(p_{0,1}+p_{0,2}), \\ \tilde{C}_2 &= (1-A)\tilde{C}_1(p_{1,2}+p_{1,3}), \\ \tilde{C}_3 &= (1-A)\tilde{C}_1 p_{1,4} + A\tilde{C}_0, \end{aligned} \quad (8)$$

where $A=P/(1+P)$, and $p_{\alpha,i}$ represents the growth probability of the growth bond i within the cell α . The configurational probability $\tilde{C}_0$ is determined from the normalization condition,

$$\sum_{\alpha} \tilde{C}_{\alpha} = \tilde{C}_0 + \tilde{C}_1 + \tilde{C}_2 + \tilde{C}_3 = 1. \quad (9)$$

The growth probability $p_{\alpha,i}$ on the growth bond i within the cell α is proportional to the concentration gradient $\nabla_i C$ on the growth bond. Consider the Laplace equation for cells which can be renormalized as the growth bond. The concentration field within a cell is determined by the diffusion constants of the growth and exterior bonds and the configuration of the cell. In the configuration labeled by α (see Fig. 3), the growth probabilities $p_{\alpha,i}$ of growth bond i within cell α are given by

$$\begin{aligned} p_{0,1} &= p_{0,2} = \frac{1}{2}, \\ p_{1,1} &= (1 + 3D_s)/(4 + 3D_s), \\ p_{1,2} &= p_{1,3} = p_{1,4} = 1/(4 + 3D_s), \\ p_{2,1} &= p_{2,2} = \frac{1}{2}, \\ p_{3,1} &= p_{3,2} = \frac{1}{2}, \end{aligned} \quad (10)$$

where the D_s indicates the diffusion constant of the growth bond (the surface diffusion constant), and the diffusion constant of exterior bonds is set as the unit value. We consider the diffusion constant of the cells renormalized as the growth bonds. The surface diffusion constant $D'_{s,\alpha}$ of the cell with configuration α is renormalized as follows:

$$\begin{aligned} D'_{s,0} &= 2D_s/(1 + D_s), \\ D'_{s,1} &= D_s + 3D_s/(1 + 3D_s), \\ D'_{s,2} &= 2D_s, \\ D'_{s,3} &= 2D_s. \end{aligned} \quad (11)$$

The renormalized diffusion constant D'_s of the growth bond will be assumed to be given by the most probable value,

$$D'_s = \exp[C_0 \ln(D'_{s,0}) + C_1 \ln(D'_{s,1}) + C_2 \ln(D'_{s,2}) + C_3 \ln(D'_{s,3})]. \quad (12)$$

The relationships (11) and (12) present the renormalization-group equation $D'_s = R(D_s)$. Equations (8)–(12) are simultaneously solved. We find a nontrivial fixed point D_s^* from $D_s^* = R(D_s^*)$ as a function of the Péclet number P . Figure 4 shows a schematic behavior of the renormalization function $D'_s = R(D_s)$ as a parameter of the Péclet number. When the Péclet number approaches an infinite value, the fixed point approaches 3. In the limit of a zero Péclet number, the fixed point becomes that of DLA: $D_s^* = 2.326$. The renormalization-group (RG) equation has a fixed point between the DLA result and 3 with increasing Péclet number. At the fixed points the derivatives dR/dD_s have positive values less than 1. They are stable fixed points. These results represent stable steady states and are consistent with the entropy production minimum theorem or the dissipation energy minimum theorem.³⁴

FIG. 4. A schematic behavior of the renormalization function $D'_s = R(D_s)$ as a parameter of the Péclet number P . The RG equation has a stable fixed point between the DLA and ballistic model results with increasing Péclet number.

RANDOM MULTIPLICATIVE PROCESS

We consider the growth probability on any growth bond. We define the two growth probabilities as (a) the growth probability $P_i(L)$ over the total perimeter bonds of the aggregate with the size L , and (b) the growth probability $p_{\alpha,i}$ on the growth bond i , within the cell α . Before and after renormalization, the growth probability $P_i(L)$ on any growth bond i is given by

$$P_i(L) = p_{\alpha,i} P_{\alpha}(L/b), \quad (13)$$

where L represents the size of the system, and b is the scale factor. The cell's growth probability $p_{\alpha,i}$ is represented by a function of the surface diffusion constant as a parameter of the Péclet number. After many repeated renormalizations, the cell's growth probability $p_{\alpha,i}$ approaches that evaluated at the fixed point $p_{\alpha,i}^*$. The total growth probability $P_i(L)$ assigned to each growth bond is then given by a random multiplicative process of the cell's growth probabilities evaluated at the fixed point. In the limit of L sufficiently large, an infinite hierarchy of generalized dimensions $D(q)$ is given by

FIG. 5. Surface fractal dimensions as a function of the Péclet number. In both limits of $P = 0$ and ∞ , they present the values of the DLA and ballistic aggregation.

FIG. 6. The generalized dimension $D(q)$ of the drift-diffusion-limited aggregation at a Péclet number. The curves indicate, respectively, the results for $A = P/(1+P) = 0, 0.3, 0.6, 0.9, 0.99$.

$$D(q) = -(q-1)^{-1} \ln \left\langle \sum_i p_{\alpha,i}^{*q} \right\rangle_c / \ln b, \quad (14)$$

where $\langle \rangle_c$ indicates the configurational average and $p_{\alpha,i}^*$ represents the cell's growth probability evaluated at the fixed point. The surface fractal dimension d_s is given by $D(0)$. A dependence of the surface fractal dimension on the drift effect is shown in Fig. 5. It shows a continuous function of the Péclet number between 1 (for the infinite-Péclet-number case corresponding to ballistic aggregation) and about 1.526 (for the zero-Péclet-number case corresponding to DLA). The partition of $D(q)$ into a density of singularities $f(q)$ with singularity strength $\alpha(q)$ is introduced,

$$\begin{aligned} \alpha(q) &= (d/dq)[(q-1)D(q)], \\ f(q) &= q\alpha(q) - (q-1)D(q). \end{aligned} \quad (15)$$

The multifractal scaling structure is shown in Figs. 6 and 7; $D(q)$ in Fig. 6 and the α - f spectra in Fig. 7. As the

FIG. 7. The α - f spectra of the drift-diffusion-limited aggregation at Péclet numbers: $A = P/(1+P) = 0, 0.3, 0.6, 0.9, 0.99$.

Péclet number increases, the α - f spectrum becomes a more steep curve. This is analogous to the generalized DLA model (η model).^{26,29,35}

CLUSTER-TOP OCCUPANCY PROBABILITY

We now show that the cluster-top occupancy probability p_a determines the fractal dimension d_f , following Turkevich's method.^{36,37} Given a (L, M) aggregate which consists of M bonds and has size L , we obtain

$$\Delta M = (1/p_a)\Delta L = [(1-A)/p_{\max} + A/p_U]\Delta L, \quad (16)$$

where $p_{\max}$ and p_U indicate, respectively, the maximum growth probability and the uniformly growing probability. The following scalings hold:

$$\begin{aligned} (1/p_{\max}) &\sim L^{(d_{\text{DLA}}-1)}, \\ (1/p_U) &\sim L, \end{aligned} \quad (17)$$

where d_{DLA} is the fractal dimension of DLA. We obtain the fractal dimension $d_f = 2$ from $M \sim L^2$ for sufficiently large L . Hence the interior of the aggregate is compact at large length scale. It is found that at a finite drift velocity the aggregate is compact with a surface fractal dimension between the DLA and ballistic aggregation results with increasing drift effect. We note that $[D(\infty) + 1]$ is not identical with the fractal dimension d_f for the drift-diffusion-limited aggregation.

CROSSOVER

We consider the crossover phenomenon found by Meakin³⁰ with the use of Monte Carlo simulation. We obtain the following scaling form from Eq. (16):

$$M \sim L^{d_{\text{DLA}}} f(L/\xi), \quad (18)$$

where the scaling function $f(x)$ must become 1 for $x \ll 1$ and vary for $x \gg 1$ as $x^{d-d_{\text{DLA}}}$. ξ is regarded as the crossover radius from the DLA to the uniform cluster. We find the crossover from the DLA at short length scales to a compact structure at longer length scales than a crossover radius ξ from Eq. (16). The crossover radius ξ is given by

$$\xi \sim A^{-1/(d-d_{\text{DLA}})}, \quad (19)$$

where $A = P/(1+P)$. Equation (19) tells us that the length scale over which the aggregate exhibits a DLA structure is proportional to $A^{-1/(d-d_{\text{DLA}})}$. This result does not agree with that by Meakin: $\xi \sim A^{-1}$. We find the crossover from the DLA fractal at short length scales to a compact structure at longer length scales with the use of the relationship (16).

SUMMARY

In conclusion, we have applied the real-space renormalization-group method to the drift-diffusion-limited aggregation proposed by Meakin. We have investigated the effects of particle drift on the diffusion-limited

aggregation. We have analytically found the surface fractal dimension and the α - f spectra depending on the Péclet number. We have shown that at a finite drift velocity the aggregate is compact with a surface fractal di-

mension between the DLA and ballistic aggregation results with increasing drift effect. We have found the crossover from the DLA structure at short length scales to a compact structure at longer length scales.

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