Effect of growing interface on the diffusion-limited aggregation: Crossover from the diffusion-limited-aggregation fractal

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A generalization of the diffusion-limited-aggregation model is introduced to take into account the effect of the velocity of the moving interface. The growth process is governed by the diffusion field with a convection induced by the growing interface. In the limit of a slow-moving interface, the convection is approximated to be proportional to the local mass flow with changing sign. The convective diffusion field is simulated by the biased-random walker. The development of the distinct morphologies is found with varying strength of the growing interface. The velocity of the moving interface is governed by the dimensionless parameter $\chi = (\rho_g / \rho_a) \Delta C$, where ρ_a is the density of the aggregate, ρ_g the density of the diffusing particles in the diffusion field, and ΔC the concentration difference between the aggregate and the outer boundary. With the increase of χ , the aggregate crosses over to the dense aggregation with the flat interface. The moving interface acts as a stabilizing force on the interface. In the case of a negative χ , the aggregate crosses over to the needle crystal. The growing interface acts as a destabilizing force. The similarities with the pattern formation in the electrochemical deposition and solidification are discussed.

I. INTRODUCTION

Recently, there has been increasing interest in the problem of geometrical structure in the kinetic-growth model.¹⁻¹⁷ 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 mechanisms. The structure of the aggregates strongly depends on the dynamics of the growth process. Examples of pattern formation in a wide variety of diffusive systems include crystal growth,^{18,19} electrochemical deposition,²⁰⁻²² viscous fingering,^{23,24} and dielectric breakdown.²⁵ The diffusion-limited aggregation (DLA) model has been introduced by Witten and Sander¹ for the pattern formation of diffusive systems. The structure of the aggregate has been analyzed by computational,^{1,2,11-14} experimental,¹⁸⁻²⁵ and analytical methods.²⁶⁻³⁶ It has been found that the aggregate was a fractal to have a scale-invariant structure. Vicsek³⁷ and Kadanoff³⁸ independently proposed simple generalizations of the DLA model to take into account the surface tension. A generalization of the DLA model has been little introduced to take into account the effect of the velocity of the moving interface. Experimentally, when the solidification in an undercooled melt is carried out at different rates, one finds a reproducible sequence of morphological changes in the asymptotic shape of the interface as a function of the velocity.³⁹ In experiments of the electrochemical deposition, Grier *et al.*²¹ and Sawada *et al.*²² found that the system was driven from fractal growth, through dense-radial aggregation, to dendritic growth by varying the applied potential and the concentration of the electrolyte.

In this paper, we present a simple generalization of the DLA model to take into account the effect of the velocity

of the growing interface. We study pattern formation in a diffusive system with a convective flow induced by the growing interface. Effects of the velocity of the moving interface on the DLA are investigated by the computational method. The growth process is governed by the diffusion field with a convection induced by the moving interface. The cluster formation in the convective diffusion field is simulated by the biased-random walker. The distinct morphologies develop with varying strength of the growing interface. The dimensionless parameter $\chi = (\rho_g / \rho_a) \Delta C$ governs the velocity of the moving perimeter where ΔC is the concentration difference between the aggregate and the outer boundary. With increasing χ , the aggregate crosses over to the dense aggregation with the flat interface. The moving interface acts as a stabilizing force of the interface. On the other hand, in the case of a negative χ , the aggregate crosses over to the needle crystal. The pattern formation in the electrochemical deposition is corresponding to the case of a positive χ . The pattern formation of the solidification in the undercooled melt is corresponding with the case of a negative χ .

The organization of the paper is as follows. In Sec. II we present the basic equation and model to take into account the effect of the moving interface. In Sec. III we present the computational result. The development of the distinct morphologies is shown in figures. In Sec. IV our model is applied to the problems of the electrochemical deposition and the solidification from the undercooled melt. Section V presents the summary.

II. BASIC EQUATION AND MODEL

We consider the aggregation process growing on a surface in two dimensions, namely, the one-sided model. The diffusion field of a component is considered outward

from the growing aggregate without surface tension. The growing interface induces the convection flow in the diffusion field outward from the aggregate. The concentration C of the diffusing component satisfies the convective diffusion equation under the quasistationary approximation (in the moving frame):

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

where u_i is the i -directional velocity, $\langle u_i \rangle$ the average velocity of the growing aggregate, and D the diffusion constant. We consider the diffusion field within the square with $L \times L$. The aggregate grows from the lower boundary toward the upper boundary. The boundary conditions in the horizontal direction are periodic in order to simulate an infinite half-plane. In the vertical direction the boundary conditions are given by the constant concentrations C_u and C_a , where C_u and C_a are the concentrations on the upper and lower boundaries. In the limit of a slow convective flow, we shall assume that the induced flow by the moving interface is proportional to the local mass flow with changing sign. The velocity field is given by

$$u_i = (\rho_g / \rho_a) D \partial C_0 / \partial x_i \quad (2)$$

with $\partial^2 C_0 / \partial x_i^2 = 0$, where ρ_g is the density of the diffusing particles in the diffusion field and ρ_a the density of the aggregate. To cast these equations in dimensionless form we use the following units: The concentration difference $\Delta C (= C_u - C_a)$ for the concentration and L for distance. Then Eq. (1) becomes

$$\begin{aligned} \chi (\partial \hat{C}_0 / \partial \hat{x}) \partial \hat{C} / \partial \hat{x} + \chi (\partial \hat{C}_0 / \partial \hat{y} - \langle \partial \hat{C}_0 / \partial \hat{y} \rangle) \partial \hat{C} / \partial \hat{y} \\ = \partial^2 \hat{C} / \partial \hat{x}^2 + \partial^2 \hat{C} / \partial \hat{y}^2 \end{aligned} \quad (3)$$

with

$$\partial^2 \hat{C}_0 / \partial \hat{x}^2 + \partial^2 \hat{C}_0 / \partial \hat{y}^2 = 0,$$

where $\chi = (\rho_g / \rho_a) \Delta C$ and $\hat{C} = (C - C_a) / \Delta C$. The boundary conditions are given by $\hat{C}_0 = 0$ and $\hat{C} = 0$ on the aggregate, and $\hat{C}_0 = 1$ and $\hat{C} = 1$ on the upper boundary. Cared variables are all dimensionless.

We find the important dimensionless parameter $\chi = (\rho_g / \rho_a) \Delta C$ representing the strength of the velocity of the moving interface. With a positive χ , the mass flux is reduced by the stronger convection flow as the growth rate of the interface increases. At places where the growth rate is larger, the mass flux is more reduced than at places with smaller growth rate. Convection acts as a nonlocal stabilizing force which flattens the interface. On the other hand, for a negative χ , the mass flux is increased by the stronger convection flow as the growth rate of the interface increases. At places where the growth rate is larger, the mass flux is more increased than at places with smaller growth rate. Convection acts as a nonlocal destabilizing force which enhances the roughness of the interface.

Now we consider the simulation of the aggregation process following Eq. (3). The diffusing particles undergo random walks except for a bias resulting from the convective flow induced by the moving interface. We simu-

late the aggregation process by using the biased-random walkers. The bias varies from position to position. The local strength of the bias is step-by-step calculated by solving the Laplace equation under the boundary conditions of the instantaneous aggregate. The biased-random walker continues to move until he arrives at the surface of the aggregate. The probability to visit the site (i, j) satisfies the equation:

$$\begin{aligned} p(i, j) = & (\frac{1}{4} + \alpha_x) p(i+1, j) \\ & + (\frac{1}{4} - \alpha_x) p(i-1, j) + (\frac{1}{4} + \alpha_y) p(i, j+1) \\ & + (\frac{1}{4} - \alpha_y) p(i, j-1) \end{aligned} \quad (4)$$

with

$$\begin{aligned} \alpha_x = & \chi [\hat{C}_0(i+1, j) - \hat{C}_0(i-1, j)] / 8, \\ \alpha_y = & \chi [\hat{C}_0(i, j+1) - \hat{C}_0(i, j-1) - \langle \hat{C}_0(i, j+1) \\ & - \hat{C}_0(i, j-1) \rangle_s] / 8, \end{aligned} \quad (5)$$

where $\hat{C}_0$ is given by solving the Laplace equation under the boundary conditions of the instantaneous aggregate, and $\langle \rangle_s$ indicates the average value of the concentration gradient over the interface.

We summarize the algorithm of the simulation process. We start by setting the concentrations $\hat{C}_0 = 0$ and $\hat{C}_0 = 1$ on the aggregate and the upper boundary. We solve the Laplace equation under the boundary conditions. We evaluate the local bias over the diffusion field by Eq. (5). Particles are introduced one at a time at a randomly chosen point on the outer boundary. The particle performs a biased-random walk following Eq. (4). The particle continues to move until it either reaches a point adjacent to a site already occupied by a particle or until it reaches the outer boundary. If the particle sticks on the aggregate, we solve the Laplace equation under the boundary conditions with the configuration of the aggregate. We repeat the procedure.

III. SIMULATION RESULTS

We have carried out calculations for a range of χ [$= (\rho_g / \rho_a) \Delta C$] values, on square arrays of size 70×70 . The clusters are grown until the height reaches 35 units. The patterns formed in the diffusive system are shown in Fig. 1 for a number of positive χ . The pattern of the cluster shown in Fig. 1(a) was obtained for $\chi = 0.0$. Overall the cluster looks like the DLA fractal. Figures 1(b)–1(e) show, respectively, the aggregates for $\chi = 16.0, 64.0, 96.0$, and 240.0 . The aggregates consist, respectively, of 1(a) 264, 1(b) 586, 1(c) 845, 1(d) 974, and 1(e) 1139 particles. With increasing χ the interface of the aggregates becomes flat overall. The crossover from the DLA fractal to the flat-dense aggregation is found with increasing parameter χ . For a positive χ the growing interface acts as a stabilizing force which flattens the interface. The mass flux is reduced by the stronger convection flow as the growth rate of the interface increases. At places where the growth rate is larger, the mass flux is more reduced than at places with smaller growth rate. Convection induced

by the moving interface acts as a stabilizing force which flattens the interface.

We consider the drift-diffusion-limited aggregation⁴⁰ for comparing with our modified DLA model. In the drift-diffusion-limited aggregation model, particles move on a lattice towards the surface with probability P_0 , and perform a random walk with probability $(1-P_0)$. The

aggregation process is transformed from DLA to the ballistic process with increasing drift effect. We allow particles to follow biased-random trajectories and perform a biased-random walk with probability P_0 towards the surface. The patterns formed in the drift-diffusion-limited aggregation are shown in Fig. 2. Figures 2(a)–2(e) show, respectively, for $P_0=0.0, 0.1, 0.2, 0.5,$

FIG. 1. For a positive χ typical aggregates grown on a surface. (a) $\chi=0.0$; (b) $\chi=16.0$; (c) $\chi=64.0$; (d) $\chi=96.0$; (e) $\chi=240.0$. The aggregate crosses over from DLA fractal to the flat-dense aggregation with increasing χ , where $\chi[(\rho_g/\rho_a)\Delta C]$ represents the dimensionless parameter relating to the strength of the velocity of the moving interface.

FIG. 2. Typical aggregates grown in the diffusion process with the drift P_0 . (a) $P_0=0.0$; (b) $P_0=0.1$; (c) $P_0=0.2$; (d) $P_0=0.5$; (e) $P_0=1.0$. The aggregate crosses over from DLA fractal to the dense aggregation. The interface of the aggregates remain rough.

and 1.0. The aggregates consist, respectively, of 2(a) 264, 2(b) 473, 2(c) 670, 2(d) 828, and 2(e) 988 particles. The clusters crossover from DLA fractal to the dense aggregation with increasing P_0 . For $P_0=0.0$ we obtain the DLA model and for $P_0=1.0$ the ballistic model. In this model the interface of the aggregates remain rough. In contrast to the drift-diffusion-limited aggregation, the interface obtained from our modified DLA model is very flat.

For a negative χ we carry out calculations. The patterns obtained are shown in Fig. 3. Figure 3(a)–3(d) show, respectively, the aggregates for $\chi=0.0$, -1.6 ,

(a)

(b)

(c)

(d)

FIG. 3. For a negative χ , typical aggregates grown on a surface. (a) $\chi=0.0$; (b) $\chi=-1.6$; (c) $\chi=-4.0$; (d) $\chi=-8.0$. The aggregate crosses over from DLA fractal to the needle crystal with decreasing χ .

FIG. 4. The crossover lines separated between the distinct morphologies. When the ratio of the densities or the concentration difference increase, the aggregate crosses over from DLA fractal to the dense aggregation. On the other hand, for a negative χ when the ratio of the densities or the absolute value of the concentration difference increase, the aggregate crosses over from DLA fractal to the needle crystal.

-4.0 , and -8.0 . The aggregates consist, respectively, of 3(a) 264, 3(b) 153, 3(c) 94, and 3(d) 67 particles. With decreasing χ , the aggregate crosses over from DLA fractal to a single needle. Convection flow induced by the moving interface enhances the roughness of the interface and makes a needlelike cluster. The mass flux is increased by the stronger convection flow as the growth rate of the interface increases. At places where the growth rate is larger the mass flux is more increased than at places with smaller growth rate. Convection acts as a destabilizing force which enhances the roughness of the interface.

The χ value separated DLA fractal from the flat-dense aggregation is approximately given by

$$\chi_c \simeq 40, \quad (6)$$

where $\chi = (\rho_g/\rho_a)\Delta C$. On the other hand, the χ value separated DLA fractal from the single needle is approximately given by

$$\chi_c \simeq -5.0. \quad (7)$$

The crossover phenomena are governed by the two parameters: the ratio of the densities (ρ_g/ρ_a) and the concentration difference ΔC . Figure 4 shows the crossover lines separated between the distinct morphologies. When the ratio of the densities or the concentration difference increases, the aggregate crosses over from DLA fractal to the flat-dense aggregation. On the other hand, for a negative χ , when the ratio of the densities or the absolute value of the concentration difference increase, the aggregate crosses over from DLA fractal to the single needle.

IV. ELECTROCHEMICAL DEPOSITION AND SOLIDIFICATION

We shall apply our result to the electrochemical deposition and solidification. First we consider the electro-deposition.

A. Electrodeposition

In the electrodeposition system the growth rate is controlled by the electric current flowing between the aggregate and the outer boundary. In the quasistationary approximation, the process is determined by the conservation law of ion current. By replacing the concentration $\hat{C}_0$ with the electrostatic potential $\hat{\Phi}_0$ in Eq. (3), we obtain the governing equation

$$\chi(\partial\hat{\Phi}_0/\partial\hat{x})\partial\hat{C}/\partial\hat{x} + \chi(\partial\hat{\Phi}_0/\partial\hat{y} - \langle\partial\hat{\Phi}_0/\partial\hat{y}\rangle)\partial\hat{C}/\partial\hat{y} = \partial^2\hat{C}/\partial\hat{x}^2 + \partial^2\hat{C}/\partial\hat{y}^2 \quad (8)$$

with

$$\partial^2\hat{\Phi}_0/\partial\hat{x}^2 + \partial^2\hat{\Phi}_0/\partial\hat{y}^2 = 0,$$

where $\chi = (L/Dze\rho_a)\sigma\Delta\Phi$ (D is the diffusion constant, σ is the electric conductivity, ze is the charge per ion), $\hat{\Phi}_0 = (\Phi_0 - \Phi_{0,a})/\Delta\Phi$ and $\hat{C} = (C - C_a)/\Delta C$.

The boundary conditions are given by $\hat{\Phi}_0 = 0$ and $\hat{C} = 0$ on the aggregate, and $\hat{\Phi}_0 = 1$ and $\hat{C} = 1$ on the upper boundary. The applied voltage in the electrodeposition experiments is corresponding with the potential difference $\Delta\Phi$. In general the potential difference $\Delta\Phi$ is a positive value in the electrochemical deposition process. The dimensionless parameter χ is always positive. In the electrodeposition the DLA fractal crosses over only to the flat-dense aggregation. The crossover line is given by

$$\sigma\Delta\Phi \simeq 40ze\rho_a D/L, \quad (9)$$

where $\Delta\Phi$ is the applied voltage. Generally, the electric conductivity σ in solution increases with the concentration of ion. With increasing applied voltage or concentration, the DLA fractal crosses over to the flat-dense aggregation. The velocity of the moving interface effects to flatten the interface. By the effect the pattern will become circular in the deposition from a single seed. The radial-dense aggregation will appear.

B. Solidification

We consider pattern formation in solidification from the melt. We consider two cases: (a) the case of an undercooled melt and (b) the case in which the aggregate acts as a heat sink. In solidification from the undercooled melt the latent heat released at the interface flows into the liquid. On the other hand, in the case in which the aggregate acts as a heat sink, the temperature of the liquid is above the melting temperature and the latent heat flows toward the heat sink into the aggregate. We assume an infinite heat conductivity for the aggregate. The process is determined by the conservation law of heat energy. We first consider the solidification process from an undercooled melt. By replacing the concentration field with the temperature field in Eq. (3), we obtain

$$\chi(\partial\hat{T}_0/\partial\hat{x})\partial\hat{T}/\partial\hat{x} + \chi(\partial\hat{T}_0/\partial\hat{y} - \langle\partial\hat{T}_0/\partial\hat{y}\rangle)\partial\hat{T}/\partial\hat{y} = \partial^2\hat{T}/\partial\hat{x}^2 + \partial^2\hat{T}/\partial\hat{y}^2 \quad (10)$$

with

$$\partial^2\hat{T}_0/\partial\hat{x}^2 + \partial^2\hat{T}_0/\partial\hat{y}^2 = 0,$$

where

$$\chi = -(\rho_g/\rho_a)(c/L)\Delta T, \quad \Delta T = T_a - T_u,$$

$\hat{T}_0 = (T_{0,a} - T_0)/\Delta T$, $\hat{T} = (T_a - T)/\Delta T$ (c , specific heat; L , latent heat). The boundary conditions are given by $\hat{T}_0 = 0$ and $\hat{T} = 0$ on the aggregate, and $\hat{T}_0 = 1$ and $\hat{T} = 1$ on the upper boundary. In the solidification from an undercooled melt, the dimensionless parameter χ is always negative. The DLA fractal crosses over to the needle crystal. The crossover line is given by

$$(\rho_g/\rho_a)(c/L)\Delta T \simeq 5.0, \quad (11)$$

where ΔT represents the degree of the undercooling, and ρ_g and ρ_a are the densities of the melt and solid. For solidification from a quenched liquid phase the heat flux is increased by the stronger convective flow as the growth rate of the interface increases. At places where the growth rate is larger the heat flux is more increased than at places with smaller growth rate. Convection acts as a nonlocal destabilizing force which enhances the roughness of the interface. The needle crystal appears.

We here consider the solidification process in the case where the aggregate acts as a heat sink. Heat flows toward the aggregate. We obtain the governing equation (10) with $\hat{T} = (T - T_a)/\Delta T$, $\chi = (\rho_g/\rho_a)(c/L)\Delta T$, and $\Delta T = T_u - T_a$. The dimensionless parameter χ is always positive. The DLA fractal crosses over to the flat-dense aggregation. The crossover line is given by

$$(\rho_g/\rho_a)(c/L)\Delta T \simeq 40, \quad (12)$$

where $\Delta T = T_u - T_a$, T_a is the temperature of the aggregate and T_u the temperature of the melt far away the aggregate ($T_u > T_a$). For solidification from the liquid phase above the melting temperature, the heat flux is reduced by the stronger convection flow as the growth rate of the interface increases. At places where the growth rate is larger, the heat flux is more reduced than at places with smaller growth rate. Convection acts as a nonlocal stabilizing force which flattens the interface. The flat-dense aggregation appears.

V. SUMMARY

We extend the DLA model to take into account the effect of the velocity of the moving interface. We find that in the diffusive systems the convection induced by the moving interface is very important to form the pattern of the aggregates. The growth process is governed by the diffusion field with a convective flow induced by the growing interface. We present the modified DLA model to simulate the growth process. We find the important dimensionless parameter $\chi [= (\rho_g/\rho_a)\Delta C]$ governing the effect of the moving interface. With the increase of a positive χ , the aggregate crosses over to the dense aggregation with flat interface. The moving interface acts as a nonlocal stabilizing force on the interface. With the decrease of a negative χ , the aggregate crosses over to the needle crystal. The growing interface acts as a destabilizing force on the interface. We apply our result to both problems of the electrodeposition and solidification. We find a possibility explaining the morphologies appearing in the diffusive systems.

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