Convection effect on the diffusion-limited-aggregation fractal: Renormalization-group approach

Takashi Nagatani

College of Engineering, Shizuoka University, Hamamatsu 432, Japan

(Received 19 October 1987)

A nonlinear effect of convection on the diffusion-limited-aggregation (DLA) fractal is investigated by use of the real-space renormalization-group method. The renormalization-group equations are derived for two parameters: the diffusion constant on the interface and the Péclet number $\hat{P}$ representing a ratio of the convection effect to the diffusion effect. It is found that the correlation length ξ (limiting the validity of the self-similarity) diverges as $\xi \sim |\hat{P}|^{-\nu}$, where $\nu^{-1} = \ln(\partial \hat{P}' / \partial \hat{P})_{\hat{P}=0} / \ln b$ (b is a scale factor). The correlation-length exponent ν is approximately given by $\nu = d(0)^{-1}$, where $d(0)$ is the surface fractal dimension of the DLA. The aggregate is the DLA fractal on smaller length scales than the correlation length, but becomes a nonfractal structure on large length scales. The renormalization-group method is applied to the formation of solidification patterns in two cases: one is the case of an undercooled melt and the other the case in which the aggregate acts as a heat sink. In the undercooled-melt case the convection acts as a destabilizing force on the interface, but, for the aggregate with a heat sink, the convection acts as a stabilizing force. It is shown that the DLA fractal crosses over to one of the two distinct nonfractal structures depending on the direction of the heat flux.

I. INTRODUCTION

Pattern formation in systems far away from equilibrium is a subject of considerable current interest.^{1,2} Several simplified models have been introduced recently in order to make the originally very complex mathematical problem tractable. One class of these models is based on the assumption that the motion of the surface is merely determined by local properties. Ben-Jacob *et al.*³ proposed the boundary-layer model by assuming the characteristic length of the diffusion field to be much smaller than the local radius of curvature of the interface. Brower *et al.*⁴ presented the geometrical model in which the velocity on the interface was locally determined by the curvature and its derivatives. Although the local models succeeded in explaining a number of important features they do not take into account the nonlocal character of the diffusion field surrounding the growing crystal. On the other hand, Witten and Sander⁵ proposed the diffusion-limited-aggregation (DLA) model to treat the effects of the nonlocal diffusion field. In Monte Carlo simulations⁶⁻⁸ of this model, random fractal clusters were generated as diffusing particles stick to the interface of the growing aggregate. In the DLA model the surface-tension effect was not taken into account in a way consistent with the differential-equation approach. Vicsek⁹ and Kadanoff¹⁰ independently proposed simple generalizations of the DLA model to take into account the stabilizing force of the surface tension. Vicsek succeeded partly in simulation of directional solidification by assuming a local-curvature-dependent sticking probability. Sarkar and Jensen¹¹ studied the evolution of the interface for a one-sided model of directional solidification on a lattice with biased random walkers. Experimentally, when the solidification in an undercooled melt is carried

out at different rates, one finds a reproducible sequence of morphological changes¹² in the asymptotic shape of the interface as a function of the velocity. A generalization of the DLA model has not been made to take into account the effect of the velocity of the moving interface.¹³⁻²⁸ Meakin²⁹ investigated the effect of the drift on the DLA fractal by using biased random walkers. He found a crossover from the DLA fractal on short length scales to a compact structure on longer length scales, with the use of Monte Carlo simulation. Even though there is not a drift of particles, convection is induced in the diffusion field by the moving interface. The convection effect is more important as the moving velocity of the interface is larger.

In this paper, we present a real-space renormalization-group (RG) method taking convection into account. We investigate the effect of the moving velocity at the interface on the DLA structure. The renormalization-group method is applied to the pattern formation of solidification in two cases: one is the case of an undercooled melt and the other is the case in which the aggregate acts as a heat sink.

Here we explain the central idea of the RG method for a DLA. We have presented a hierarchical model for a DLA (Ref. 30) which is a deterministic fractal model on the hierarchical lattice. It shows similar scaling structure to the DLA fractal on the regular lattice. The renormalization procedure can be applied to the model. In the hierarchical model, the growth probability distribution is exactly determined by the recursion relations for the total conductance of the system and the electric field within the growth-bond generator. The total conductance approaches the fixed point after many renormalizations. The electric field on the surface of the aggregate is given by a multiplicative process of that within the growth-

bond generator. The electric field within the generator is also a function of the total conductance. The fractal geometric picture for the position-space renormalization group is demonstrated by the regular random fractal.³¹ It is consistent with the DLA on a hierarchical lattice. We can thus derive the RG method for the DLA.^{32,33}

The organization of the paper is as follows. In Sec. II we present the basic equation governing the convective diffusion. In Sec. III, we start with directly defining the dissipation energy without resorting to the conventional Hamiltonian of phase transition. We present the renormalization group for the coupling constants. In Sec. IV the renormalization procedure is explained. In Sec. V application of the renormalization-group method is given to the pattern formation in solidification. Section VI presents the summary.

II. BASIC EQUATION

We consider the diffusion field of a component α outward from the growing aggregate without surface tension. The density ρ_α of the diffusing component α satisfies the convective diffusion equation under the quasistationary approximation

$$u_i \partial \rho_\alpha / \partial x_i = D_0 \partial^2 \rho_\alpha / \partial x_i^2, \quad (1)$$

where u_i is the i -directional velocity and D_0 the diffusion constant. We assume inviscid flow for the velocity field u_i . From the continuity of total mass the velocity satisfies

$$\partial^2 \Phi / \partial x_i^2 = 0, \quad u_i = -\partial \Phi / \partial x_i \quad (2)$$

where Φ represents the velocity potential. The boundary conditions are the following.

(a) At an infinitely far field from the interface,

$$u_i \rightarrow 0$$

and (3)

$$\rho_\alpha \rightarrow \rho_{\alpha, \infty}.$$

(b) On the interface,

$$u_n \equiv u_s = D_s / (\rho_{\alpha, b} + \rho_{\alpha, s}) (\partial \rho_\alpha / \partial x_n)$$

and (4)

$$\rho_\alpha = \rho_{\alpha, s},$$

where the subscripts n and s indicate the normal component to the surface and the surface, and $\rho_{\alpha, b}$ represents the density of the aggregate constructed from the component α . To cast these equations in dimensionless form we use the following units: $\rho_{\alpha, \infty}$ for density, a for distance, the velocity u_s on the surface for velocity, and D_0 for the diffusion constant. Then Eqs. (1)–(4) become

$$\hat{P} \hat{u}_i \partial \hat{\rho}_\alpha / \partial \hat{x}_i = \hat{D} \partial^2 \hat{\rho}_\alpha / \partial \hat{x}_i^2, \quad (1')$$

$$\partial^2 \hat{\Phi} / \partial \hat{x}_i^2 = 0 \text{ and } \hat{u}_i = -\partial \hat{\Phi} / \partial \hat{x}_i, \quad (2')$$

$$\hat{u}_{i, \infty} \rightarrow 0 \text{ and } \hat{\rho}_\alpha \rightarrow 1, \quad (3')$$

$$\hat{u}_s = 1 \text{ and } \hat{\rho}_\alpha = \hat{\rho}_{\alpha, s}. \quad (4')$$

Here $\hat{P}$ indicates the Péclet number describing the ratio of the convection to the diffusion,

$$\hat{P} \equiv u_s a / D_0 = \hat{D} / (\hat{\rho}_{\alpha, b} + \hat{\rho}_{\alpha, s}) (\partial \hat{\rho}_\alpha / \partial \hat{x}_n). \quad (5)$$

Careted variables are all dimensionless. So the problem is to solve Eqs. (1') and (2') subject to the boundary conditions (3') and (4'), then assign a growth probability proportional to the moving velocity u_s of the interface to the perimeter bond (or site), and to move the interface according to the growth probability. The mass flux is reduced by stronger convection flow, depending on the rate of increase of the growth of the interface. 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.

If $\hat{P}=0$, Eq. (1') merely becomes the diffusion process. This process then gives the DLA model. The process of 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.³⁴ We have derived the renormalization-group scheme for the DLA model.^{32,33} In the RG scheme, the dissipation energy and the diffusion constant on the surface correspond to the Hamiltonian and the coupling constant of conventional critical phenomena.

III. DISSIPATION ENERGY AND RENORMALIZATION GROUP

We start by defining directly the dissipation energy without resorting to the conventional Hamiltonian of phase transition. The net dissipation energy is given by the multiplication of the mass flux of the component α by the thermodynamic force,

$$\hat{H}_d = \hat{J}_i \hat{X}_i \\ = \int_V d\hat{V} [\hat{D} (\partial \hat{\rho}_\alpha / \partial \hat{x}_i)^2 - \hat{P} \hat{u}_i \hat{\rho}_\alpha (\partial \hat{\rho}_\alpha / \partial \hat{x}_i)], \quad (6)$$

where the first relation represents the multiplication of the total mass flux by the force $\hat{X}_i = 1 - \hat{\rho}_{\alpha, s}$ and the second relation indicates the integration of the local energy dissipation over the volume of the diffusion field. The stationary value of the functional $(\partial \hat{H}_d / \partial \hat{\rho}_\alpha = 0)$ reproduces the convective diffusion equation (1'). If $\hat{P}=0$, the second derivative $(\partial^2 \hat{H}_d / \partial \hat{\rho}_\alpha^2)$ becomes a positive value.

Then this is consistent with the principle of minimum entropy production (or dissipation energy) in a nonequilibrium steady state without convection. Generally, when there is convection ($\hat{P} \neq 0$), the sign of the second derivative is not definite. We transform the volume integral of Eq. (6) to the surface integral. The total mass flow, carrying through the surface of the aggregate, is given by

$$\hat{J}_s = \hat{J}_t = \int_{\hat{S}} d\hat{S} [\hat{D}(\partial\hat{\rho}_\alpha/\partial\hat{x}_i)_n - \hat{P}\hat{\rho}_{\alpha,s}] . \quad (7)$$

We can write the net dissipation energy in terms of the surface integral,

$$\hat{H}_d = \hat{X}_t \left[\int_{\hat{S}} d\hat{S} [\hat{D}(\partial\hat{\rho}_\alpha/\partial\hat{x}_i)_n - \hat{P}\hat{\rho}_{\alpha,s}] \right] , \quad (8)$$

where $D(\hat{D} \equiv D/D_0)$ indicates the diffusion constant on the perimeter of the aggregate. The diffusion constant on the surface is given by the bare diffusion constant D_0 before renormalization, but it is renormalized to the dressed value. By using a decimation method explained below, one can express the dissipation energy in terms of the coarse-grained variables on the new lattice with the lattice spacing b . These variables are indicated by primes:

$$\hat{H}_d = \hat{X}_t \left[\int_{\hat{S}'} d\hat{S}' [\hat{D}'(\partial\hat{\rho}'_\alpha/\partial\hat{x}'_i)_n - \hat{P}'\hat{\rho}'_{\alpha,s}] \right] . \quad (9)$$

Thus the short-range interaction between neighboring densities caused by the gradient term is renormalized. The diffusion constant $\hat{D}$ and the Péclet number $\hat{P}$ on the interface are transformed:

$$\hat{D}' = R_D(\hat{D}, \hat{P}) \quad (10)$$

and

$$\hat{P}' = R_P(\hat{D}, \hat{P}) .$$

The relationships (10) present the renormalization-group equations. The two parameters $\hat{D}$ and $\hat{P}$ play the role of the so-called coupling constants in the Hamiltonian of phase transition. When $\hat{P}=0$, Eq. (10) reduces to the RG equation of the DLA where self-similarity is valid. We can identify $\hat{P}=0$ as a critical point. Only right at the critical point, where self-similarity is valid, do we have $\hat{P}'=\hat{P}=0$. In general we know that the correlation length ξ limits the validity of similarity. The correlation length is described as follows:

$$\xi \sim \hat{P}^{-\nu} , \quad (11)$$

where ν indicates the exponent of the correlation length. When the Péclet number approaches zero the correlation length diverges. The correlation length is the same in both the original and the renormalized lattices, namely, $\xi = \xi'$. When in the original lattice ξ is given by $\xi = (\text{const}) \times \hat{P}^{-\nu}$, then in the renormalized lattice with lattice constant b we have $\xi' = (\text{const}) \times b \times \hat{P}'^{-\nu}$. Taking the logarithm of both sides in $b\hat{P}'^{-\nu} = \hat{P}^{-\nu}$, we arrive at

$$\nu^{-1} = \ln(\partial\hat{P}'/\partial\hat{P})^* / \ln b ,$$

with $(\partial\hat{P}'/\partial\hat{P})^* \equiv (\partial\hat{P}'/\partial\hat{P})_{\hat{P}=0}$, (12)

$$(\partial\hat{P}'/\partial\hat{P})^* \equiv (\partial\hat{P}'/\partial\hat{P})_{\hat{P}=0} ,$$

where the asterisk indicates the fixed point. The Péclet number plays the role of the reduced relative temperature $(T - T_c)/T_c$ in the conventional phase transition. The Péclet number is also proportional to the applied field $(\rho_{\alpha,\infty} - \rho_{\alpha,s})$ between the aggregate and the external boundary. As the applied field approaches zero (the critical point), the structure of the aggregate becomes self-similar and the DLA structure appears. Near $\hat{P}=0$, the aggregate is the DLA fractal on smaller length scales than the correlation length, but becomes a nonfractal structure on large length scales. Since the convection acts as a nonlocal stabilizing force on the interface, the nonfractal structure will be composed similarly to the drift-diffusion-limited aggregation.²⁹ In Sec. IV we show the fixed point to be given by $\hat{D}^* = \hat{D}_{\text{DLA}}^*$ ($\hat{D}_{\text{DLA}}^*$ is the value of the DLA) and $\hat{P}^* = 0$.

IV. RENORMALIZATION PROCEDURE

We derive the renormalization-group equations (10) for the diffusion constant on the surface and the Péclet number with the use of a decimation method. For simplicity, we perform the decimation procedure on the square lattice. We use the discrete versions of Eqs. (1')–(4') for the renormalization-group transformation. The dissipation energy (8) is transformed to

$$\hat{H}_d = \hat{X}_t \left[\sum_{i \in S} [\hat{D}_i(\nabla\hat{\rho}_\alpha)_i - \hat{P}_i\hat{\rho}_{\alpha,i}] \right] , \quad (13)$$

where the sum indicates the summation over all the bonds on the perimeter, and $(\nabla\hat{\rho}_\alpha)_i$ is the difference of the densities on the bond i . We must distinguish between three types of bonds on the lattice before a renormalization procedure: (a) interior bonds which are occupied by diffusion particles and form 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 by interior bonds 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 interface [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 renormalized 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 in (a) is renormalized to that in (b), according to the rules of re-

normalization. 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 and the Péclet number of the cell that are 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 to a finite value but not zero. The nonlocal nature of the concentration field is taken into account in determining the diffusion constant of the growth bond and the Péclet number. If we consider the renormalization in the vertical direction we shall take a periodic boundary condition in the lateral direction. We perform renormalizations for the vertical and horizontal bonds independently.

Figure 3 shows all the configurations of the cell that are possible to renormalize as the growth bond. Let us consider the configurational probability C_β with which a

FIG. 1. An example of the renormalization of part of the surface layer of an aggregate. Bonds constituting the aggregate are indicated by bold lines and bonds on the perimeter of the aggregate by wavy lines. The lattice in (a) is renormalized to that in (b), according to the rule of renormalization.

FIG. 2. Illustration of the renormalization of a $b=2$ cell for the convective-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.

particular configuration (β) appears. The distinct configurations are labeled by $\beta=0,1,2$ in Fig. 3. Configuration (1) is constructed by adding an interior bond to configuration (0). In addition, by adding an interior bond to configuration (1), configuration (2) occurs. These stepwise growth processes are similar to the DLA without convection. The configurational probabilities C_β are given by

$$\begin{aligned} C_1 &= C_0(p_{0,1} + p_{0,2}) = 2C_0p_{0,1}, \\ C_2 &= C_1(p_{1,2} + p_{1,3} + p_{1,4}) = 3C_1p_{1,2}, \end{aligned} \quad (14)$$

where $p_{\beta,i}$ represents the growth probability of the growth bond i within the cell β . The configurational probability c_0 is determined from the normalization condition,

FIG. 3. All distinct configurations of the 2×2 cell that are possible to renormalize as the growth bond. Configuration (1) is constructed by adding an interior bond to configuration (0). In addition, by adding an interior bond to configuration (1), configuration (2) occurs.

$$\sum_{\beta} C_{\beta} = C_0 + C_1 + C_2 = 1. \quad (15)$$

The growth probability $p_{\beta,i}$ on the growth bond i within the cell β is proportional to the mass flux j_i on the growth bond.

Consider the convective diffusion equation for cells which can be renormalized as the growth bond. The concentration field within a cell is determined by the surface diffusion constant, the Péclet number, and the cell configuration. In the configuration labeled by β (see Fig. 3), the growth probabilities $p_{\beta,i}$ (see the Appendix for details) of growth bond i within cell β are given by

$$\begin{aligned} p_{0,1} &= p_{0,1} = \frac{1}{2}, \\ p_{1,1} &\approx \frac{(\hat{D} - \hat{P})(1 + 3\hat{D} - 3\hat{P})}{[\hat{D}(4 + 3\hat{D}) - \hat{P}(4 + 15\hat{D} - 3\hat{P})]}, \\ p_{1,2} &= p_{1,3} = p_{1,4} \approx \frac{[\hat{D} - \hat{P}(1 + 3\hat{D} - 3\hat{P})]}{[\hat{D}(4 + 3\hat{D}) - \hat{P}(4 + 15\hat{D} - 3\hat{P})]}, \\ p_{2,1} &= p_{2,2} = \frac{1}{2}, \end{aligned} \quad (16)$$

where $\hat{D}$ and $\hat{P}$ indicate the surface diffusion constant and the Péclet number on the growth bond, and the diffusion constant of exterior bonds is set as the unit value.

We consider the diffusion constant and the Péclet number of the cells renormalized as the growth bonds. The surface diffusion constant $\hat{D}'_{\beta}$ and the Péclet number $\hat{P}'_{\beta}$ of the cell with configuration β are renormalized as follows (see Appendix for details):

$$\begin{aligned} \hat{D}'_0 &= 2\hat{D}/(1 + \hat{D} - \hat{P}), \quad \hat{P}'_0 = 2\hat{P}, \\ \hat{D}'_1 &= \hat{D}(4 + 3\hat{D} - 3\hat{P})/(1 + 3\hat{D} - 3\hat{P}), \quad \hat{P}'_1 = 4\hat{P}, \\ \hat{D}'_2 &= 2\hat{D}, \quad \hat{P}'_2 = 2\hat{P}. \end{aligned} \quad (17)$$

Here we must take an appropriate configurational average of the renormalized parameters. The renormalized parameters will be assumed to be given by the most probable values:

$$\begin{aligned} \hat{D}' &= \exp[C_0 \ln(\hat{D}'_0) + C_1 \ln(\hat{D}'_1) + C_2 \ln(\hat{D}'_2)], \\ \hat{P}' &= \exp[C_0 \ln(\hat{P}'_0) + C_1 \ln(\hat{P}'_1) + C_2 \ln(\hat{P}'_2)]. \end{aligned} \quad (18)$$

The relationships (17) and (18) present the renormalization-group equations $\hat{D}' = R_D(\hat{D}, \hat{P})$ and $\hat{P}' = R_P(\hat{D}, \hat{P})$. Equations (14)–(18) are simultaneously solved. We find a nontrivial fixed point ($\hat{D}^* = \hat{D}^*_{\text{DLA}} = 2.326$, $\hat{P}^* = 0$) from $\hat{D}^* = R_D(\hat{D}^*, \hat{P}^*)$ and $\hat{P}^* = R_P(\hat{D}^*, \hat{P}^*)$. The fixed point of the diffusion constant becomes that of the DLA. Figure 4 schematically shows the behavior of the renormalization functions. It displays the streamlines which indicate how the renormalization functions move various points in the parameter space. The fixed point becomes a saddle point on the parameter space. It is unstable. At the saddle point the DLA fractal appears to have the so-called characteristics of self-similarity. As the Péclet number increases, the DLA fractal crosses over into the nonfractal structure. The correlation length ξ diverges as $\xi \sim \hat{P}^{-\nu}$. With the use of Eq. (12), the exponent ν of the correlation length is given by

FIG. 4. Schematic behavior of the renormalization functions, the surface diffusion constant $\hat{D}$ and the Péclet number $\hat{P}$. The streamlines indicate how the renormalization functions move various points in the parameter space. The fixed point is indicated by $\bullet$. At the fixed point the DLA fractal appears and crosses over into the compact structure.

$$\nu = d(0)^{-1}, \quad (19)$$

where $d(0)$ indicates the surface fractal dimension. It is found that the exponent of the correlation length equals the inverse of the surface fractal dimension. An infinite hierarchy of generalized dimensions $d(q)$ is given by^{32,33}

$$d(q) = -(q-1)^{-1} \ln \left\langle \sum_i p_{\beta,i}^{*q} \right\rangle_c / \ln b, \quad (20)$$

where $\langle \rangle_c$ indicates the average over the configurations β and $p_{\beta,i}^*$ represents the cell's growth probability evaluated at the fixed point. The fractal dimension d_f is given by $d(\infty)$. The partition of $d(q)$ into a density of singularities $f(q)$ with singularity strength $\alpha(q)$ is introduced by Halsey *et al.*²⁷ and Amitrano *et al.*²⁸ The multifractal scaling structure can be shown similarly to the DLA model. See Refs. 32 and 33 for the details.

We find that the effect of convection on the DLA fractal is very important. As the velocity of the growing interface increases, the stronger convective flow is induced into the diffusion field. Then at places where the growth rate is larger, the mass flux is more reduced than with the smaller growth rate. Convection acting as a stabilizing force flattens the interface. The DLA fractal crosses over into a compact structure. The correlation length limiting the self-similarity becomes shorter with increasing Péclet number.

V. APPLICATION TO PATTERN FORMATION IN 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. Figure 5 shows a schematic illustration for the two types of solidification. For solidification from a quenched liquid phase the heat 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 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. On the other hand, 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.

We can write the net dissipation energy in place of Eq. (8):

$$\hat{H}_d = |\hat{X}_t| \left[\int_{\hat{S}} dS [\hat{\lambda} |(\partial \hat{T} / \partial \hat{x}_t)_{s,n}| - \hat{P} \hat{T}_s] \right], \quad (21)$$

where $\hat{X}_t = 1 - T_s / T_\infty$ ($T_s = T_m$, T_m is the melting tem-

perature), $\hat{\lambda} = \lambda / \lambda_0$, and

$$\hat{P} \equiv \pm u_s a / (\lambda_0 / \rho c_p) = \hat{\lambda} / (\hat{L} \pm \hat{T}_s) (\partial \hat{T} / \partial \hat{x}_n). \quad (22)$$

Here L ($\hat{L} = L / c_p$) is the latent heat, c_p the specific heat, and the $\pm$ sign depends on whether or not the gradient of the temperature is positive. For solidification from the undercooled melt the Péclet number is negative. For the solidification from the liquid phase above the melting temperature the Péclet number is positive. The renormalization-group equations are obtained by replacing the surface diffusion constant with the surface thermal conductivity in Eqs. (17). The correlation length is given by

$$\xi \sim |\hat{P}|^{-\nu}, \quad (23)$$

where the Péclet number is given by Eq. (22) and the exponent of the correlation length is given by Eq. (12). The correlation length exponent has the same value above and below the critical point ($\hat{P} = 0$).

We consider the effect of the convection on the growth probability. From Eq. (16) the following relation holds for the growth probabilities of the growth bonds within the configuration (1):

$$(p_{1,1} / p_{1,2} \{ \hat{P} > 0 \}) < (p_{1,1} / p_{1,2} \{ \hat{P} = 0 \}) < (p_{1,1} / p_{1,2} \{ \hat{P} < 0 \}), \quad (24)$$

where the first, second, and third terms indicate, respectively, the ratio of the maximum growth probability to the minimum growth probability for the cases where the Péclet number is positive, zero, and negative. In the renormalization-group transformation it is shown that the convection acts as a stabilizing or destabilizing force, depending upon whether or not the Péclet number is positive. In Fig. 6 we display a schematic behavior of the

FIG. 5. Schematic illustration for the two solidifications: (a) solidification from an undercooled melt where the heat flows into the liquid phase and convection acts as a destabilizing force to the interface and (b) solidification from the liquid phase above the melting temperature where heat flows into the aggregate and convection acts as a stabilizing force to the interface.

FIG. 6. Schematic behavior of the renormalization functions is shown by the streamlines which indicate how the renormalization functions move various points in the parameter space. The fixed point is indicated by $\bullet$, which is a saddle point. The DLA fractal at the fixed point crosses over into the two distinct nonfractal structures, depending on whether or not the Péclet number is positive. The positive Péclet number indicates solidification from the liquid phase above the melting temperature. The negative Péclet number indicates solidification from the undercooled liquid.

renormalization-group functions by the streamlines which indicate how the renormalization functions move various points in the parameter space. The DLA fractal at the fixed point crosses over into two distinct nonfractal structures, depending on whether or not the Péclet number is positive. For solidification from the undercooled melt the DLA fractal crosses over into the needle structure since convection enhances the roughness of the interface. For solidification from the liquid phase above the melting temperature the DLA fractal crosses over into the compact structure since the convection flats the interface. We note that convection acts only as a stabilizing force of the interface in the convective diffusion system (see Fig. 4). A negative Péclet number never appears in the convective diffusion system. This is the distinct difference between Figs. 4 and 6.

We consider the condition for appearance of the DLA structure. The DLA fractal appears in the limit of sufficiently small Péclet number given by Eq. (22). A sufficiently small Péclet number is satisfied by the conditions that the latent heat is sufficiently large and the temperature gradient is sufficiently small. Otherwise, the DLA structure does not appear. In place of the DLA, the needle or compact crystal will appear, depending on whether the latent heat flows into the liquid phase or the solid phase.

VI. SUMMARY

We extend the real-space renormalization-group method to diffusion-limited aggregation with convection. We investigate a nonlinear effect of convection on the DLA fractal. The renormalization-group transformations for the surface diffusion constant and the Péclet number are derived. The DLA fractal appears at a critical point ($\hat{P}=0$) in the nonequilibrium critical phenomenon. The critical point is the saddle point in the $(\hat{P}, \hat{\lambda})$ plane. The DLA fractal always becomes unstable. There is no stable fixed point if one takes account of the moving interface. It is found that the correlation length ξ (limiting the validity of the DLA fractal) diverges as $\xi \sim |\hat{P}|^{-\nu}$. The correlation-length exponent ν is given by the inverse of the surface fractal dimension of the DLA: $\nu = d(0)^{-1}$. The aggregate is the DLA fractal on smaller length scales than the correlation length, but becomes a nonfractal structure on large length scales. We apply the renormalization-group method to the pattern formation of solidification. We find two distinct cross-overs of the DLA fractal to nonfractal structures, depending upon whether or not the liquid phase is undercooled. For solidification from the undercooled melt convection acts as a destabilizing force of the interface and

the DLA fractal crosses over toward a needle crystal. For solidification from the liquid above the melting temperature convection acts as a stabilizing force of the interface and the DLA fractal crosses over toward a compact crystal.

APPENDIX

We derive the renormalization transformation (17) of the surface diffusion constant and the Péclet number for configuration (1) shown by Fig. 3. The growth probabilities of the cell with configuration (1) are derived. The mass flows j_1, j_2, j_3, j_4 , and j_e , carrying through the growth bonds 1, 2, 3, 4, and the exterior bond, are given by

$$\begin{aligned} j_1 &= \hat{D}(\hat{\rho}_s - \hat{\rho}_0) - \hat{P}_1 \hat{\rho}_s, \\ j_2 &= j_3 = j_4 = \hat{D}(\hat{\rho}_s - \hat{\rho}) - \hat{P}_2 \hat{\rho}_s, \\ j_e &= (\hat{\rho} - \hat{\rho}_0) - 3\hat{P}_2 \hat{\rho}, \end{aligned} \quad (\text{A1})$$

where $\hat{\rho}_s$ is the density of the α component on the interface, $\hat{\rho}_0$ the density on the top side of the cell, $\hat{\rho}$ the density at the point crossing the growth bonds 2,3,4. The total mass flow within the cell j' is given by

$$j' = j_1 + 3j_2 = \hat{D}'(\hat{\rho}_s - \hat{\rho}_0) - \hat{P}' \hat{\rho}_s. \quad (\text{A2})$$

From (A1) and (A2) we obtain

$$\begin{aligned} \hat{D}' &= \hat{D}(4 + 3\hat{D} - 3\hat{P}_2)/(1 + 3\hat{D} - 3\hat{P}_2), \\ \hat{P}' &= \hat{P}_1 + 3\hat{P}_2. \end{aligned} \quad (\text{A3})$$

We approximate Eqs. (A3) by

$$\begin{aligned} \hat{D}' &\approx \hat{D}(4 + 3\hat{D} - 3\hat{P})/(1 + 3\hat{D} - 3\hat{P}), \\ \hat{P}' &\approx 4\hat{P}. \end{aligned} \quad (\text{A4})$$

The growth probabilities $p_{1,i}$ of the growth bonds i ($i=1,2,3,4$) within the cell with configuration (1) are given by

$$p_{1,1} = j_1/(j_1 + 3j_2), \quad (\text{A5})$$

$$p_{1,2} = p_{1,3} = p_{1,4} = j_2/(j_1 + 3j_2).$$

By assuming that $\hat{\rho}_0=0$ and $\hat{P}_1=\hat{P}_2$ we obtain

$$\begin{aligned} p_{1,1} &\approx (\hat{D} - \hat{P})(1 + 3\hat{D} - 3\hat{P})/[\hat{D}(4 + 3\hat{D}) \\ &\quad - \hat{P}(4 + 15\hat{D} - 3\hat{P})], \\ p_{1,2} &\approx [\hat{D} - \hat{P}(1 + 3\hat{D} - 3\hat{P})]/[\hat{D}(4 + 3\hat{D}) \\ &\quad - \hat{P}(4 + 15\hat{D} - 3\hat{P})]. \end{aligned} \quad (\text{A6})$$

¹J. S. Langer, Rev. Mod. Phys. **52**, 1 (1980), and references therein.

²Special issue, Physica D **12** (1984).

³E. Ben-Jacob, N. Goldenfield, J. S. Langer, and G. Schon, Phys. Rev. Lett. **51**, 1930 (1983); Phys. Rev. A **29**, 330 (1984).

⁴R. C. Brower, D. A. Kessler, J. Koplik, and H. Levine, Phys.

Rev. Lett. **51**, 1111 (1983); Phys. Rev. A **29**, 1335 (1984).

⁵T. A. Witten and L. M. Sander, Phys. Rev. Lett. **47**, 1400 (1981); Phys. Rev. B **27**, 5686 (1983).

⁶P. Meakin, Phys. Rev. A **27**, 1495 (1983).

⁷P. Meakin, Phys. Rev. A **27**, 2616 (1983).

⁸H. J. Herrmann, Phys. Rep. **136**, 153 (1986).

- ⁹T. Vicsek, Phys. Rev. A **32**, 3084 (1985).
- ¹⁰L. P. Kadanoff, J. Stat. Phys. **39**, 267 (1985).
- ¹¹S. K. Sarkar and M. H. Jensen, Phys. Rev. A **35**, 1877 (1987).
- ¹²K. Somboonsuk, J. T. Mason, and R. Trivedi, Metall. Trans. A **15**, 967 (1984).
- ¹³*Kinetics of Aggregation and Gelation*, edited by F. Family and D. P. Landau (North-Holland, Amsterdam, 1984).
- ¹⁴*On Growth and Form*, edited by H. E. Stanley and N. Ostrowsky (Nijhoff, The Hague, 1985).
- ¹⁵*Fractal in Physics*, edited by L. Pietronero and E. Tosatti (North-Holland, Amsterdam, 1986).
- ¹⁶*Scaling Phenomena in Disordered Systems*, edited by R. Pynn and A. Skjeltorp (Plenum, New York, 1985).
- ¹⁷*Statistical Physics*, edited by H. E. Stanley (North-Holland, Amsterdam, 1986); Special issue, Physica A **140** (1986).
- ¹⁸P. Meakin, H. E. Stanley, A. Coniglio, and T. A. Witten, Phys. Rev. A **32**, 2364 (1985); **34**, 3325 (1986).
- ¹⁹P. Meakin, Phys. Rev. A **35**, 2234 (1987).
- ²⁰F. Family, T. Vicsek, and B. Taggett, J. Phys. A **19**, L727 (1986).
- ²¹L. Paterson, Phys. Rev. Lett. **52**, 1621 (1984).
- ²²T. Vicsek, Phys. Rev. Lett. **53**, 2281 (1984).
- ²³M. Sahimi and Y. C. Yortsos, Phys. Rev. A **32**, 3762 (1985).
- ²⁴J. D. Sherwood and J. Nittmann, J. Phys. (Paris) **47**, 15 (1986).
- ²⁵P. R. King, J. Phys. A **20**, L529 (1987).
- ²⁶H. Gould, F. Family, and H. E. Stanley, Phys. Rev. Lett. **50**, 696 (1983).
- ²⁷T. C. Halsey, P. Meakin, and I. Procaccia, Phys. Rev. Lett. **56**, 854 (1986).
- ²⁸C. Amitrano, A. Coniglio, and F. di Liberto, Phys. Rev. Lett. **57**, 1016 (1986).
- ²⁹P. Meakin, Phys. Rev. B **28**, 5221 (1983).
- ³⁰T. Nagatani, J. Phys. A **20**, L641 (1987).
- ³¹T. Nagatani, J. Phys. A **19**, L1079 (1986).
- ³²T. Nagatani, J. Phys. A **20**, L381 (1987).
- ³³T. Nagatani, Phys. Rev. A **36**, 5812 (1987).
- ³⁴P. Glansdorff and I. Prigogine, *Structure, Stability, and Fluctuations* (Interscience, London, 1971).