Multiparticle simulation for morphological transitions in diffusion-limited aggregation

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The two models based on multiparticle diffusion-limited aggregation (DLA) are presented to study the morphological changes in diffusive systems. The first model, which takes into account the propagation of cations from the dissolving anode, is proposed for the Hecker effect, which appears in recent experiments of electrochemical deposition. The second model with two components, in which the one component is condensable and the other component is not, is presented to investigate the effect of an incondensable gas on the morphology of DLA. Computer simulation is performed for the deposition process on the plate in two dimensions. In the first model, it is shown that the morphological change of deposits occurs from a less dense structure at the inner region to a more dense structure at the outer region. In the second model, it is found that the morphological transitions appear from the dense structure, through the bay pattern, to the needle structure, with increasing concentration of incondensable gas.

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I. INTRODUCTION

Fractal growth phenomena in diffusive systems have recently attracted considerable attention [1–10]. Pattern-formation examples in diffusive systems include electrochemical deposition, crystal growth, viscous fingering, dielectric breakdown, chemical dissolution, and bacterial colonies [11–18]. An approximation to these phenomena is provided by the Laplacian growth model. The Laplacian growth model can be simulated by the diffusion-limited aggregation (DLA) model. The structure of the aggregate has been analyzed by computational, experimental, and theoretical methods. Several simple generalizations of the DLA model have been proposed to take into account concentration, drift, surface tension, and sticking probability. 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. The morphological changes between the DLA fractal and the nonfractals have been found by computational and experimental methods. The morphological changes have also been analyzed by the real-space renormalization-group method [19–22].

Very recently, morphological changes with a distinct structure transition boundary were found in electrochemical experiments by Sander [23], Garik *et al.* [24], Lam and co-workers [25,26], Melrose, Hibbert, and Ball [27], and Fleury, Rosso, and Chazalviel [28]. For copper experiments, where this transition has been observed, the structure of the aggregate changes from a less dense structure at the inner region of the structure transition boundary to a more dense structure at the outer region. This transition was sometimes referred to as the Hecker effect in the existing literature. Melrose, Hibbert, and Ball [27] have explained that the transition occurs when an acid front migrating in from the anode meets the deposit. Fleury, Rosso, and Chazalviel [28] gave experi-

mental evidence and numerical result showing that the transitions were due to the propagation of charged impurities leaving the dissolving anode at the beginning of the growth and hitting the deposit at later times. By using the mean-field-type simulation, they showed the contour of the transition zone. However, until now, the computer simulation of such a structure transition has never been performed from a point of view of DLA. The morphological transition such as the Hecker effect has not been found in the DLA-type simulation.

It has also been known that one cannot obtain high-quality vapor-deposited thin films under experimental conditions of poor vacuum. In order to make thin films highly qualitative, it has been necessary to obtain the experimental condition of high vacuum. Inert gases in the system avoid making highly qualitative thin films. The inert gases have an important effect on aggregation and deposition processes. However, until now, there are no simulations for analyzing the effect of the inert gases on the morphology of DLA. Open questions include the morphological changes of DLA by an incondensable gas.

In this paper, we present two simple models showing morphological changes in diffusive systems: the first model for the Hecker effect in electrochemical depositions and the second model for the morphological transitions induced by the incondensable gas. The two models are based on the multiparticle DLA simulation. In the multiparticle DLA models, the growth rate of the deposit and the propagation of cations from the dissolving anode can be simulated. However, in the single-particle DLA simulation, the growth rate and the propagating cation cannot be taken into account. Also the single-particle DLA model cannot simulate the deposition process from gas mixture of two components in which the one component is condensable and the other component is not. In the first model, we study the morphological evolution induced by the propagating of cations from the dissolving anode. We show that the morphological change occurs from a less dense structure at the inner region to a more

dense structure at the outer region if the growing deposit meets cations propagating from the anode. We find the criterion of the morphological change. In the second model, we study the effect of an incondensable gas on DLA. With increasing concentration of the incondensable gas, it will prevent the condensable gas from depositing. We try to simulate the effect of the incondensable gas on the morphology of deposits by using a multiparticle simulation model [29,30]. We show that the morphology of deposits changes from the dense structure, through the bay structure, to the needle structure with increasing concentration of the incondensable particles. Above the critical value (near the percolation threshold) of the concentration of the incondensable particles, the growth of the deposit stops. We find the diagram of the morphology for the deposit growing into the incondensable gas.

The organization of the paper is as follows. In Sec. II we present the first model for the Hecker effect. We show the simulation results and the condition for the Hecker effect. In Sec. III we present the second model for the morphological transitions induced by incondensable gas. We show the simulation result and the diagram of the morphologies. Finally, Sec. IV contains a brief summary.

II. SIMULATION FOR HECKER EFFECT

We consider the electrochemical deposition process with the dissolving anode. We give the basic equation, boundary conditions, and initial condition for the electrodeposition process on the plate in parallel geometry. The constant voltage is applied between the cathode and the anode. Initially, the concentration of cations holds constant. Cations diffuse toward the deposition plate. With growing aggregate, the new cations dissolve into solution from the anode and propagate toward the deposition plate. Then, the concentration C of a depositing component satisfies the unsteady convective diffusion equation

$$\frac{\partial C}{\partial t} + v \frac{\partial C}{\partial y} = D \frac{\partial^2 C}{\partial x^2} + D \frac{\partial^2 C}{\partial y^2}, \quad (1)$$

where v is the y -directional velocity resulting from the migration, D is the diffusion constant, x axis is parallel to the plate, and y axis is vertical to the plate. The initial concentration of cations holds a constant value C_0 over all the solution. The initial condition is given by

$$C(x, y, 0) = C_0. \quad (2)$$

The diffusing cation sticks to the aggregate as soon as it reaches the deposit. The boundary condition on the deposit is given by

$$C = 0 \quad (3)$$

(on the deposit). The concentration on the dissolving anode is assumed to be a constant value C_a . The concentration C_a holds to be higher than the initial concentration C_0 ($C_a > C_0$). The boundary condition on the anode is given by

$$C = C_a \quad (4)$$

(on the anode). The unsteady convective diffusion equation (1) with initial condition (2) and the boundary conditions (3) and (4) can describe the deposition process taking into account the propagation of cations from the dissolving anode. Especially, the unsteady term $\partial C / \partial t$ in Eq. (1) represents the propagation of cations from anode. It is important. The deposition process described in terms of Eq. (1) with (2)–(4) cannot be simulated by the single-particle DLA model.

We will try to simulate the deposition process by means of the multiparticle DLA model [29,30]. We here consider the simulation of the deposition process on the plate satisfying Eq. (1) with (2)–(4). The convectively diffusing particle governed by the mass transport equation can be simulated by using multiparticle biased-random walkers. The diffusive cations of high concentration, propagating from the dissolving anode, cannot be simulated by means of the single-particle DLA since there is no unsteady term in the Laplace equation. In what follows we summarize the algorithm of the simulation process. The simulation is carried out with the use of a simple square lattice. We consider a subset of the square lattice enclosed by a box with 300×200 (units). We choose a line seed at the bottom of the box. The lateral boundary is periodic. First we distribute particles (cations) randomly with density C_0 : particles occupy each site with probability C_0 . The particles are diffusing in a box by exchanging their sites with their neighboring sites. We choose the time scale such that one Monte Carlo attempt corresponds to a time increment of $(4N_g)^{-1}$ for a system with N_g particles. By choosing the lattice parameter as a length unit, the diffusion constant D reduces to unity. When a particle reaches a point adjacent to a site already occupied by a particle, it solidifies and the aggregate grows. As the aggregate grows, we add a thin particle layer with a constant density C_a at the top of the box. This addition of particles is chosen so as to realize the boundary condition (4). The height of the aggregate increases in the simulation run up to a maximum of 150. The maximum number of solid particles is about 15 000. Figures 1(a)–1(c) show, respectively, typical patterns of the deposits on the plate for the strengths of drift $v/D = 0.05, 0.1$, and 0.2 where the initial concentration $C_0 = 0.02$ and the concentration $C_a = 0.4$ on the anode. Each pattern of the deposits is grown by varying the drift v/D . Each pattern on the deposits has a distinct structure transition boundary. The morphologies change from a less dense structure at the inner region of the structure transition boundary to a more dense structure at the outer region of the structure transition boundary. The structure transition is due to the propagation of cations of high concentration from the anode. Figure 2 indicates the plot of the particle number of the deposit against the height for the patterns in Fig. 1. The particle number increases with steeper slope near the structure transition boundary. The height where the slope changes is indicated by the dotted line. The structure transition boundary corresponds to the position where the density of the deposit changes steeper slope. The position de-

creases with increasing drift v/D . Figure 3 shows the time evolution of the pattern in Fig. 1(b). Figures 3(a)–3(c) indicate, respectively, the patterns obtained at the times $t = 259.25$, 289.5 , and 304.5 . Both gas and solid particles are shown in Fig. 3. Also, the concentration profile $C(y)$ along the y axis are shown on the right-hand side in each figure. Since the cation particles adjacent to the deposit solidify, the cation particles vanish around the deposit. At $t \approx 290$, the cations of high concentration propagating from the anode meet the deposit. Then, the deposit changes from a less dense structure to a more dense structure.

We consider the roughness of the interface of deposits. The roughness of growing interface has recently attracted considerable attention [31]. Evertsz [32] has investigated the roughness of the generalized DLA model (η model). In Fig. 4, we show the log-log plot of the roughness ξ against the average height $\langle h \rangle$ for the patterns in Fig. 1(b). Here we define the roughness (or thickness) of the interface as the root mean square of the height fluctuation. Near the height h_c where the structure transition

FIG. 1. Typical patterns of the deposits on the plate by varying the drift v/D where the initial concentration $C_0 = 0.02$ and the concentration $C_a = 0.4$ on the anode. (a) $v/D = 0.05$ and the particle number of the deposit $N_a = 9174$. (b) $v/D = 0.1$ and $N_a = 10\,001$. (c) $v/D = 0.2$ and $N_a = 13\,000$.

FIG. 2. The plot of the particle number against the height for the patterns in Fig. 1.

occurs, the roughness remains constant irrespective of the increase of the height (see Fig. 2). Then, the roughness also increases with the height. The position of the flat in Fig. 4 agrees with that where the slope of the density of the deposit changes in Fig. 2. The thickness ξ scales as

FIG. 3. The time evolution of the pattern in Fig. 1(b). The patterns are on the left-hand side and the corresponding concentration profiles on the right-hand side. (a) $t = 259.25$ and $N_a = 2000$ particles. (b) $t = 289.5$ and $N_a = 4000$ particles. (c) $t = 304.5$ and $N_a = 6000$ particles.

FIG. 4. The log-log plot of the interface thickness ξ against the average height $\langle h \rangle$ for the pattern in Fig. 1(b).

$$\xi \approx \langle h \rangle^\beta \text{ for } \langle h \rangle < h_c, \quad (5)$$

where $\beta=0.49$. The value 0.49 of the scaling exponent is larger than $1/3$ of the ballistic deposition and smaller than $\frac{3}{4}$ of the DLA [32]. The value 0.49 may be characteristic of the DLA with a finite concentration and a drift. It is found that the structure transition of the deposit appears not only in the density but also in the roughness.

We consider the condition for appearance of the Hecker effect. The structure transition occurs when the deposit meets the cations of high concentration propagating from the anode. We consider whether or not the structure transition occurs within the deposit with the height h_a . The time to be necessary for the deposit to grow until the height h_a is given by h_a/v_g where v_g is the growth rate of deposit. The time to be necessary for cations to propagate until the distance h_0-h_a is given by $(h_0-h_a)/v_d$ where v_d is the propagation speed of the cations from the anode and h_0 is the height between the cathode and the anode. When the time $(h_0-h_a)/v_d$ is shorter than the time h_a/v_g , the structure transition occurs. The condition for occurrence of the transition is given by

$$h_a/v_g > (h_0-h_a)/v_d. \quad (6)$$

Until now, in the case of no drift (only diffusion), it is known that the growth rate v_g [29] and the propagation speed v_d are given, respectively,

$$v_g \approx n_g^{1/(d-d_f)} \text{ and } v_d \approx D/(h_0-h_a), \quad (7)$$

where n_g is the gas density, d the dimension of the space, and d_f the fractal dimension of the aggregate. The growth rate of the deposit increases with the gas density. Therefore, at a low initial concentration, the structure transition will occur since the condition (6) is satisfied by a low initial concentration. We expect that whether or not the structure transition occurs can be controlled by the initial concentration C_0 . We perform the simulation for the case of no drift by varying the initial concentration C_0 . Figure 5 shows the typical patterns obtained at the high initial concentration $C_0=0.2$ ($C_a=0.6$). Figures 5(a) and 5(b) indicate, respectively, the patterns at $t=92.75$ and 183. Since the growth rate of the deposit is higher than the propagation of cations from the anode,

(a)

(b)

FIG. 5. The typical patterns obtained at the high initial concentration $C_0=0.2$ ($C_a=0.6$). Since the deposit does not meet the cations of high concentration from the anode, the structure transition cannot occur. (a) $t=92.75$ and $N_a=4000$ particles. (b) $t=183$ and $N_a=8000$ particles.

(a)

(b)

FIG. 6. The typical patterns obtained at the low initial concentration $C_0=0.05$ ($C_a=0.6$). Since the deposit meets the cations of high concentration from the anode, the structure transition can occur. (a) $t=354$ and $N_a=1000$ particles. (b) $t=697.75$ and $N_a=5500$ particles.

the deposit does not meet the cations of high concentration from the anode. Therefore, the structure transition cannot occur. On the other hand, Fig. 6 shows the typical patterns obtained at the low initial concentration $C_0=0.05$ ($C_a=0.6$). Figures 6(a) and 6(b) indicate, respectively, the patterns at $t=354$ and 697.75 . Since the growth rate of the deposit is lower than the propagation of cations from the anode, the deposit can meet the cations of high concentration from the anode. Therefore, the structure transition can occur.

III. EFFECT OF INCONDENSABLE GAS

We consider the effect of the incondensable gas on the morphology of deposits in diffusive systems. We give the basic equations and the boundary conditions for the deposition process into an incondensable gas. We consider the deposition process on the plate in parallel geometry in two dimensions. Initially, each concentration of condensable and incondensable gases holds constant. As soon as the condensable gas reaches the plate or the deposit, it solidifies. With growing deposit, the condensable gas diffuses toward the deposition plate. Then, the concentrations C_1 and C_2 of the depositing component and the incondensable gas satisfy, respectively, the unsteady diffusion equations with concentration-dependent diffusion coefficients

$$\frac{\partial C_1}{\partial t} = \nabla \cdot [D_1(C_1, C_2) \nabla C_1], \quad (8)$$

$$\frac{\partial C_2}{\partial t} = \nabla \cdot [D_2(C_1, C_2) \nabla C_2], \quad (9)$$

where D_1 and D_2 are, respectively, the diffusion coefficients of each component depending on concentrations. For simplicity we consider the case of $D_1=D_2=D$. At a low concentration, the diffusion coefficients will be assumed to be given by

$$D(C_1, C_2) = D_0(1 - C_1 - C_2), \quad (10)$$

where D_0 is the diffusion constant in the limit of $C_1 \rightarrow 0$ and $C_2 \rightarrow 0$. With increasing concentration C_2 of the incondensable gas, the diffusion coefficient of the depositing component decreases. The expression (10) of the diffusion coefficient can take into account the effect that the incondensable gas prevents the condensable gas from diffusing toward the plate. The diffusion equations (8) and (9) with the concentration-dependent diffusivity can be simulated by the multiparticle DLA model with two components. The initial conditions are given by

$$C_1(x, y, 0) = C_{1,0}, \quad C_2(x, y, 0) = C_{2,0} \quad (11)$$

where $C_{1,0}$ and $C_{2,0}$ are the initial concentrations of each component. The condensable particle sticks the deposit or the lower plate as soon as it reaches the deposit or the lower plate. The boundary condition of the condensable gas on the deposit or the lower plate is given by

$$C_1(x, y, t) = 0 \quad (12)$$

(on the deposit or the lower plate). The incondensable

component does not stick the deposit or the lower plate. The boundary condition of the incondensable gas on the deposit or the lower plate is given by

$$\frac{\partial C_2}{\partial y} = 0 \quad (13)$$

(on the deposit or the lower plate), where the x axis is parallel to the plate and the y axis is vertical to the plate. Both gases do not deposit on the upper plate. The boundary conditions on the upper plate are given by

$$\frac{\partial C_1}{\partial y} = 0 \text{ and } \frac{\partial C_2}{\partial y} = 0 \quad (14)$$

(on the upper plate). The deposition process governed by Eqs. (8) and (9) with the concentration-dependent diffusivity (10) cannot be simulated by a single-particle DLA model with two components since the single-particle DLA model cannot take into account the avoiding effect of the incondensable gas from diffusing of the condensable gas (the concentration-dependent diffusivity).

We try to simulate the deposition process governed by Eqs. (8) and (9) by using the multiparticle DLA model with two components. We simulate the deposition process with an incondensable gas by the multiparticle DLA model. The multiparticle DLA model with one component [29,30] is extended to that with two components where the one component is condensable and the other component is incondensable. The multiparticle DLA model with two components corresponds to the deposition model on the plate satisfying Eqs. (8) and (9). In what follows we summarize the algorithm of the simulation process. The simulation is carried out with the use of a simple square lattice. We consider a subset of the square lattice enclosed by a box with 300×300 (units). We choose a line seed at the bottom of the box. The lateral boundary is periodic. First we distribute condensable and incondensable particles randomly with the densities $C_{1,0}$ and $C_{2,0}$: condensable and incondensable particles occupy, respectively, each site with probabilities $C_{1,0}$ and $C_{2,0}$. Particles are diffusing in a box by exchanging their sites with their neighboring sites. We choose the time scale such that one Monte Carlo attempt corresponds to a time increment of $(4N_g)^{-1}$ for a system with N_g particles. By choosing the lattice parameter as a length unit, the diffusion constant D_0 reduces to unity. When a condensable particle reaches a point adjacent to a site already occupied by the deposit, it solidifies and the deposit grows. When an incondensable particle reaches the deposit, it is reflected. When each particle reaches the upper plate, it is reflected. The height of the deposit increases in the simulation run up to a maximum of 150. The maximum number of solid particles is about 10 000. Figure 7 shows typical patterns of the deposits on the plate obtained by varying concentration $C_{2,0}$ of incondensable particles under constant concentration $C_{1,0}=0.1$ of condensable gas. The patterns (a)–(d) in Fig. 7 indicate, respectively, the deposits for $C_{2,0}=0, 0.2, 0.3$, and 0.31 . With increasing concentration of incondensable gas, the morphology of the deposit changes

from the dense structure [Figs. 7(a) and 7(b)], through the bay structure [Fig. 7(c)], to the needle structure [Fig. 7(d)]. Above the critical concentration $C_{2,0,c} \approx 0.32$, the deposit does not grow. The critical concentration $C_{2,0,c}$ increases with the concentration $C_{1,0}$ of condensable gas. Figure 8 shows typical patterns of the deposits obtained by varying concentration $C_{2,0}$ under higher concentration $C_{1,0} = 0.2$ of condensable gas. The patterns (a)–(d) in Fig. 8 indicate, respectively, the deposits for $C_{2,0} = 0, 0.3, 0.4$, and 0.45 . Similarly to the patterns in Fig. 7, the morphology of deposits changes from the dense structure

[Figs. 8(a) and 8(b)], through the bay structure [Fig. 8(c)], to the needle structure [Fig. 8(d)] with increasing $C_{2,0}$. Above the critical concentration $C_{2,0,c} \approx 0.46$, the deposit does not grow. With higher concentrations $C_{1,0}$ of condensable gas, the critical concentration $C_{2,0,c}$ approaches the percolation threshold $p_c = 0.5$ on the square lattice. Figure 9 shows the morphological diagram in $C_{1,0}$ – $C_{2,0}$ plane. The dense structure obtained by simulations is indicated by the circle, the bay structure by the square, and the needle structure by the triangle. The region of no growth is also indicated by the cross. The needle struc-

FIG. 7. Typical patterns of the deposits obtained by varying concentration $C_{2,0}$ of the incondensable gas under constant concentration $C_{1,0} = 0.1$ of the condensable gas. The morphology changes from the dense structure [patterns (a) and (b)], through the bay structure [pattern (c)], to the needle structure [pattern (d)]. (a) $C_{2,0} = 0$. (b) $C_{2,0} = 0.2$. (c) $C_{2,0} = 0.3$. (d) $C_{2,0} = 0.31$. Above the critical concentration $C_{2,0,c} \approx 0.32$, the deposit does not grow.

FIG. 8. Typical patterns of the deposits obtained by varying concentration $C_{2,0}$ under constant concentration $C_{1,0} = 0.2$. The morphological changes occur from the dense structure [patterns (a) and (b)], through the bay structure [pattern (c)], to the needle structure [pattern (d)]. (a) $C_{2,0} = 0$. (b) $C_{2,0} = 0.3$. (c) $C_{2,0} = 0.4$. (d) $C_{2,0} = 0.45$. Above the critical concentration $C_{2,0,c} \approx 0.46$, the deposit does not grow.

FIG. 9. The morphological diagram of the deposits grown into the incondensable gas. The horizontal and vertical axes indicate, respectively, the concentrations of the incondensable and condensable gases. The dense structure obtained by simulations is indicated by the circle, the bay structure by the square, and the needle structure by the triangle. The region of no growth of deposits is also marked by the cross.

ture appears near the critical concentration $C_{2,0,c}$. The DLA fractal appears in the limit of $C_{1,0} \rightarrow 0$ and $C_{2,0} \rightarrow 0$. We find that an incondensable gas has an important effect on the morphology of DLA.

IV. SUMMARY

We present the two models based on the multiparticle diffusion-limited aggregation in order to study the morphological changes in diffusive systems. In the first model, we present the simulation model showing a structure transition boundary to mimic the morphological changes (the Hecker effect) appearing in particular realizations of electrochemical deposition experiments. We find that the morphology changes from a less dense structure at the inner region to a more dense structure at the outer region if the deposit meets cations of high concentration propagating from the anode. We give the condition for occurring the structure transition. In the second model, we study the effect of an incondensable gas on the morphology of the diffusion-limited deposition by using the multiparticle DLA model with two components where the one component is condensable and the other component is not. We find that with increasing concentration of incondensable gas the morphological changes occur from the dense structure, through the bay structure, to the needle structure. Above the critical concentration $C_{2,0,c}$ (near the percolation threshold), the deposit does not grow. We show the morphological diagram of the deposit.

- [1] *Kinetics of Aggregation and Gelation*, edited by F. Family and D. P. Landau (North-Holland, Amsterdam, 1984).
- [2] *On Growth and Form*, edited by H. E. Stanley and N. Ostrowsky (Nijhoff, The Hague, 1985).
- [3] *Fractals in Physics*, edited by L. Pietronero and E. Tosatti (North-Holland, Amsterdam, 1986).
- [4] H. J. Herrmann, *Phys. Rep.* **136**, 153 (1986).
- [5] P. Meakin, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, New York, 1988), Vol. 12, p. 336.
- [6] R. Julien and R. Botet, *Aggregation and Fractal Aggregates* (World Scientific, Singapore, 1987).
- [7] J. Feder, *Fractals* (Plenum, New York, 1988).
- [8] *Random Fluctuations and Pattern Growth*, edited by H. E. Stanley and N. Ostrowsky (Kluwer Academic, Dordrecht, 1988).
- [9] T. Vicsek, *Fractal Growth Phenomena* (World Scientific, Singapore, 1989).
- [10] H. E. Stanley, A. Bunde, S. Havlin, J. Lee, E. Roman, and S. Schwarzer, *Physica A* **168**, 23 (1990).
- [11] T. A. Witten and L. M. Sander, *Phys. Rev. Lett.* **47**, 1400 (1981); *Phys. Rev. B* **27**, 5686 (1983).
- [12] P. Meakin, *Phys. Rev. A* **26**, 1495 (1983); **27**, 2616 (1983).
- [13] G. Daccord and R. Lenormand, *Nature (London)* **325**, 41 (1987).
- [14] H. Fujikawa and M. Matsushita, *J. Phys. Soc. Jpn.* **58**, 3875 (1989).
- [15] M. J. King and H. Scher, *Phys. Rev. A* **41**, 874 (1990).
- [16] M. Murat and A. Aharony, *Phys. Rev. Lett.* **57**, 1875 (1986).
- [17] C. Evertsz, Ph.D. thesis, University of Groningen, 1989.
- [18] D. Grier, E. Ben-Jacob, Roy Clarke, and L. M. Sander, *Phys. Rev. Lett.* **56**, 1264 (1986).
- [19] T. Nagatani, *Phys. Rev. A* **40**, 7286 (1989).
- [20] J. Lee, A. Coniglio, and H. E. Stanley, *Phys. Rev. A* **41**, 4589 (1990).
- [21] T. Nagatani and H. E. Stanley, *Phys. Rev. A* **41**, 3263 (1990).
- [22] T. Nagatani, J. Lee, and H. E. Stanley, *Phys. Rev. Lett.* **66**, 616 (1991).
- [23] L. M. Sander, in *The Physics of Structure Formation*, edited by W. Guttinger and G. Dangelmayr (Springer, New York, 1987), p. 257.
- [24] P. Garik, D. Barkey, E. Ben-Jacob, E. Bochner, N. Broxholm, B. Miller, B. Orr, and R. Zamir, *Phys. Rev. Lett.* **62**, 2703 (1989).
- [25] L. Lam, R. D. Pocky, and U. M. Castillo, in *Nonlinear Structures in Physical Systems*, edited by L. Lam and H. C. Morris (Springer, New York, 1990), p. 1.
- [26] M. A. Guzman, R. D. Freimuth, P. U. Pendse, M. C. Veinott, and L. Lam, in *Nonlinear Structures in Physical Systems*, edited by L. Lam and H. C. Morris (Springer, New York, 1990), p. 32.
- [27] J. R. Melrose, D. B. Hibbert, and R. C. Ball, *Phys. Rev. Lett.* **65**, 3009 (1990).
- [28] V. Fleury, M. Rosso, and J. N. Chazalviel, *Phys. Rev. A* **43**, 6908 (1991).
- [29] M. Uwaha and Y. Saito, *J. Phys. Soc. Jpn.* **57**, 3285 (1988).
- [30] R. F. Voss, *J. Stat. Phys.* **36**, 861 (1984).
- [31] *Dynamics of Fractal Surfaces*, edited by F. Family and T. Vicsek (World Scientific, Singapore, 1991).
- [32] C. Evertsz, *Phys. Rev.* **41**, 1830 (1990).