

## Aggregation growth in a gas of finite density: Velocity selection via fractal dimension of diffusion-limited aggregation

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The structure and dynamics of an aggregation have been studied when the aggregate grows from a lattice gas with a nonzero gas density  $n_g$ . At low  $n_g$  and for a short length scale up to  $\xi$ , the structure of the aggregation is fractal and similar to the diffusion-limited aggregation (DLA). For a large length scale it is compact and has a nonzero asymptotic density. The steady growth rate  $V$  in  $d$ -dimensional space is inversely proportional to the characteristic length  $\xi$ , and depends on the density as  $V \sim \xi^{-1} \sim n_g^{1/(d-D_f)}$ , with  $D_f$  being the fractal dimension of DLA. Extensive Monte Carlo simulations in two dimensions confirm the above theoretical hypothesis of the velocity selection mechanism with  $D_f = 1.71$ . The interfacial width  $w$  is also found to be compatible with the expectation  $w \propto n_g^{-1/2(d-D_f)}$ .

### I. INTRODUCTION

In nonequilibrium conditions various elegant patterns are formed spontaneously, such as snowflakes, convection patterns, and so on. One of the simplest physical systems that show various regular or irregular patterns is a crystal growing in a diffusion field, which is now under intensive study.<sup>1,2</sup> The main subject of latest research in this area is a characterization of growth patterns as well as determination of the growth rate.

Among various growth patterns, the dendrite is an example of a complicated but regular pattern.<sup>1,3-6</sup> Many works are devoted to find the mechanisms of the velocity and pattern selection. The structure of the dendrite is characterized by the radius  $\rho$  of the parabolic tip, and the dynamics of the growth is characterized by the diffusion length  $l = 2D/V$ , where  $V$  is the velocity of the crystal growing in the diffusion field with a diffusion constant  $D$ . A nonvanishing surface tension introduces in the system an intrinsic characteristic length, i.e., a capillary length  $d_0$ . The marginal stability hypothesis<sup>1,3</sup> gives the proportionality coefficients between  $d_0$  and  $\rho$  or  $l$ . Recently, anisotropy in the surface tension was found to be a prerequisite for stability of the dendritic growth.<sup>4-6</sup>

Without surface tension, the system loses the characteristic length scale  $d_0$  and, therefore, the profile of the crystal becomes irregular. Furthermore, by replacing the diffusion field with its static limit, i.e., the Laplacian field, the system loses dynamics. These two conditions along with random fluctuations lead to an irregular fractal pattern called diffusion-limited aggregation (DLA).<sup>2</sup> The fractal feature of the DLA cluster reveals itself in the relation between the cluster size  $R$  and the mass or the number of solid atoms  $N_s$  that it contains:  $N_s \propto R^{D_f}$ . Here,  $D_f$  is called the fractal dimension and is smaller than the Euclidean dimension of the system  $d$  for fractal objects. The average density of the cluster

$$n_{\text{DLA}}(R) \propto R^{D_f-d} \quad (1)$$

then vanishes in the limit of an infinite cluster size  $R \rightarrow \infty$ , and thus DLA represents growth in the low-density limit of the diffusion field. In other words, the density  $n_g$  of the diffusing particles, which we call gas atoms in this paper, is zero.

When the density  $n_g$  does not vanish and the steady growth is realized, the resultant aggregate has a nonvanishing density  $n_s$ . Thus it cannot ultimately be fractal when its size goes to infinity. There should be a crossover from a fractal to a compact structure at a characteristic length  $\xi$ .<sup>7,8</sup> On the other hand, in the gas of a finite density, gas particles move according to the time-dependent diffusion equation. When the crystal grows with velocity  $V$ , a mass conservation condition at the interface introduces a diffusion length  $l = 2D/V$ .<sup>1</sup> We expect that these two length scales,  $l$  and  $\xi$  are related with each other. Matching of  $l$  and  $\xi$  then determines the growth rate  $V$  of the crystal. This new mechanism of the velocity selection is studied here by means of computer simulation. A brief summary of the present work has been recently published in a letter.<sup>9</sup>

### II. MODEL AND PREDICTIONS

Consider an aggregation growth from a two-dimensional square-lattice gas. Each gas atom makes a random walk on the lattice, until it moves next to solid atoms and stops to form an aggregate.<sup>2</sup> If there are  $N_g$  gas atoms in the system, our time unit is so chosen that one Monte Carlo diffusion trial of a gas atom corresponds to the time increment  $(4N_g)^{-1}$ . By taking the lattice parameter of the square lattice to be unity, the diffusion constant  $D$  also reduces to unity.

Initially we fill a rectangular box with gas atoms of a density  $n_g$ . The seed of the aggregate takes the form of a

line at the bottom of the box. When a randomly chosen gas atom sits or diffuses next to the aggregate, it solidifies. If an aggregate grows to a predetermined level from the top of the box, a thin gas layer of a constant density is added at the top, so as to keep the density far from the top of the aggregate constant. This one-dimensional growth geometry is chosen to realize a steady growth.<sup>10</sup> Mass conservation is ensured in the steady state if the average density of the solid deep inside the aggregate  $n_s$  is equal to that of the gas far from the solid  $n_g$ . The growth situation thus corresponds to that under a unit supersaturation,  $\Delta=1$ . At low densities our simulation is similar to that of Voss,<sup>7</sup> who found a crossover length  $\xi$  but did not reach a steady state. The existence of this steady state is essential to our theoretical argument.

A macroscopic analysis of the crystal growth at  $\Delta=1$  leads to the result that a crystal can grow steadily with a flat interface, but that the growth rate cannot be determined. In the simulation, on the other hand, we find that the aggregate grows steadily at a constant velocity. Figure 1(a) shows an aggregate grown from a diffusive gas with the density  $n_g=0.15$ . The width of the system  $L$  is 1024, and a periodic boundary condition in the  $x$  direction is applied. The height of the box is increased from an initial value of 200 to a final value of 1500. The top of the box is separated from the top of the aggregate by at least a height 100 at this density. The density profile along  $y$  axis is shown in Fig. 1(b). The gas density far from the aggregation clearly has the prescribed value  $n_g=0.15$ . The solid density decays from the initial value  $n_s(y=0)=1$  at the bottom, and it relaxes to the steady-state value  $n_s=0.15=n_g$  at a distance about 60. When one looks into the detailed structure of the aggregate, however, it is not homogeneous but has an intrinsic structure with a characteristic length  $\xi_s$ .

Since the aggregate absorbs all the gas atoms around it, the gas density at the interface turns out to be zero while it grows, as long as  $n_g$  is not too large. In the boundary

layer around the aggregate diffusing particles are rare just as in the DLA model, and the aggregate forms a fractal structure. The solid density decreases as in Eq. (1) when the scale  $R$  increases. But when  $R$  exceeds a characteristic length  $\xi_s$ , the aggregate becomes "compact" with an average density  $n_s=n_g$ . Therefore, the crossover length  $\xi_s$  is determined by  $n_g \sim \xi_s^{D_f-d}$ , or

$$\xi_s \sim n_g^{-1/(d-D_f)}. \quad (2)$$

If one considers the problem from a dynamical point of view, one gets another aspect of the density depletion layer. The number of atoms solidified to the aggregate per unit time  $n_s V$  should be supplied from the gas phase by the diffusion flow as  $D \nabla n_g \sim D(n_g - 0)/\xi_g$ , where  $\xi_g$  is the thickness of the density suppression layer. Therefore, the characteristic length  $\xi_g$  is about half of the diffusion length  $l=2D/V$ . (Here we have used the steady growth condition  $n_s=n_g$ .) Assuming that  $\xi_g$  be the same order with the previously defined characteristic length in the solid  $\xi_s$ , the growth velocity of the aggregate in the steady state is determined as

$$V \sim \xi_g^{-1} \sim \xi_s^{-1} \sim n_g^{1/(d-D_f)}. \quad (3)$$

The result is remarkable since the fractal dimension of the DLA cluster  $D_f$  controls the steady-state growth velocity  $V$ . We hereafter write the characteristic length simply as  $\xi$  unless discrimination between  $\xi_g$  and  $\xi_s$  is necessary. Note that the argument is valid only for  $\xi \gg 1$ , that is, for low gas densities. With increasing density the characteristic length decreases, and when  $\xi \sim 1$ , i.e.,  $V \sim 1$ , the above growth law will break down. The interface kinetics take over the control of the growth since concentration of the gas atoms on the solid surface becomes finite. In the high-density limit  $n_g=1$ , our model is equivalent to the Eden model: Diffusion does not take place but solidification proceeds randomly at the very surface of the solid. Near  $n_g=1$  the system behaves like an Eden model with migrating vacancies. These vacancies hinder the solidification, and the growth velocity will decrease as their population increases.

### III. SIMULATION AND RESULTS

#### A. Structure

According to Eq. (2) the characteristic length in an aggregate  $\xi_s$  should decrease with increasing gas density. This is obvious in Figs. 1 and 2, where the aggregates grown at various gas densities  $n_g=0.08, 0.10, 0.15$  (see Fig. 1), 0.20, 0.40, 0.80, and 1.0 are shown. For low-density samples the branching structure at small scales looks quite similar to that of a DLA cluster. As the density increases, the separation between main branches decreases. The width of the system  $L$  at low densities ( $n_g \leq 0.20$ ) is 1024, and that at high densities ( $n_g \geq 0.30$ ) is 256. The height increases in the simulation up to a maximum of 1500. We have also performed a series of simulations with the maximum box size  $400 \times 4100$  and  $100 \times 10100$  for both high and low densities. At low den-

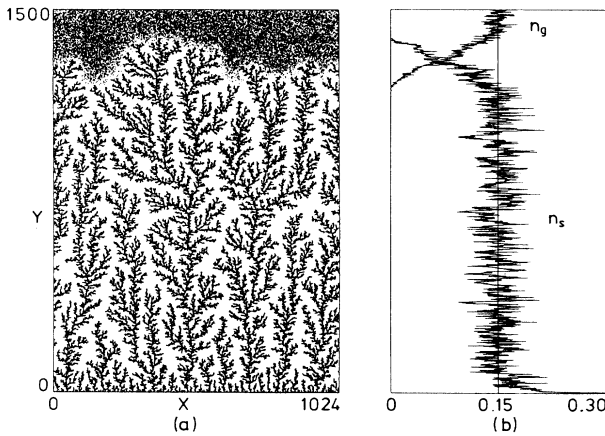


FIG. 1. (a) Configuration of an aggregate grown from diffusing gas particles at a gas density  $n_g=0.15$ . Box size is  $1024 \times 1500$ . (b) The density profile along  $y$  axis.

sities the number of main trees in the aggregate is few, indicating the long correlation length  $\xi_s$ . For gas densities  $n_g$  smaller than 0.08,  $\xi_s$  seems to become comparable to the maximum width  $L=1024$ , and the growth is influenced by the periodic boundary condition: The main "tree" begins to interact with the self-image due to the periodic boundary condition. In order to study only the intrinsic feature of the problem, we have to restrict our simulations at relatively high densities above  $n_g=0.08$ . On increasing the gas density the DLA-like structure be-

comes less obvious, since the characteristic length  $\xi_s$  decreases to the lattice constant. At very high densities the solid is dense and uniform, only with many vacancies.

### B. Velocity

The number of solid atoms  $N_s$  in the aggregate increases almost linearly in time after the solid density reaches the steady-state value as is shown in Fig. 3. Our main concern is the velocity of the aggregate height,

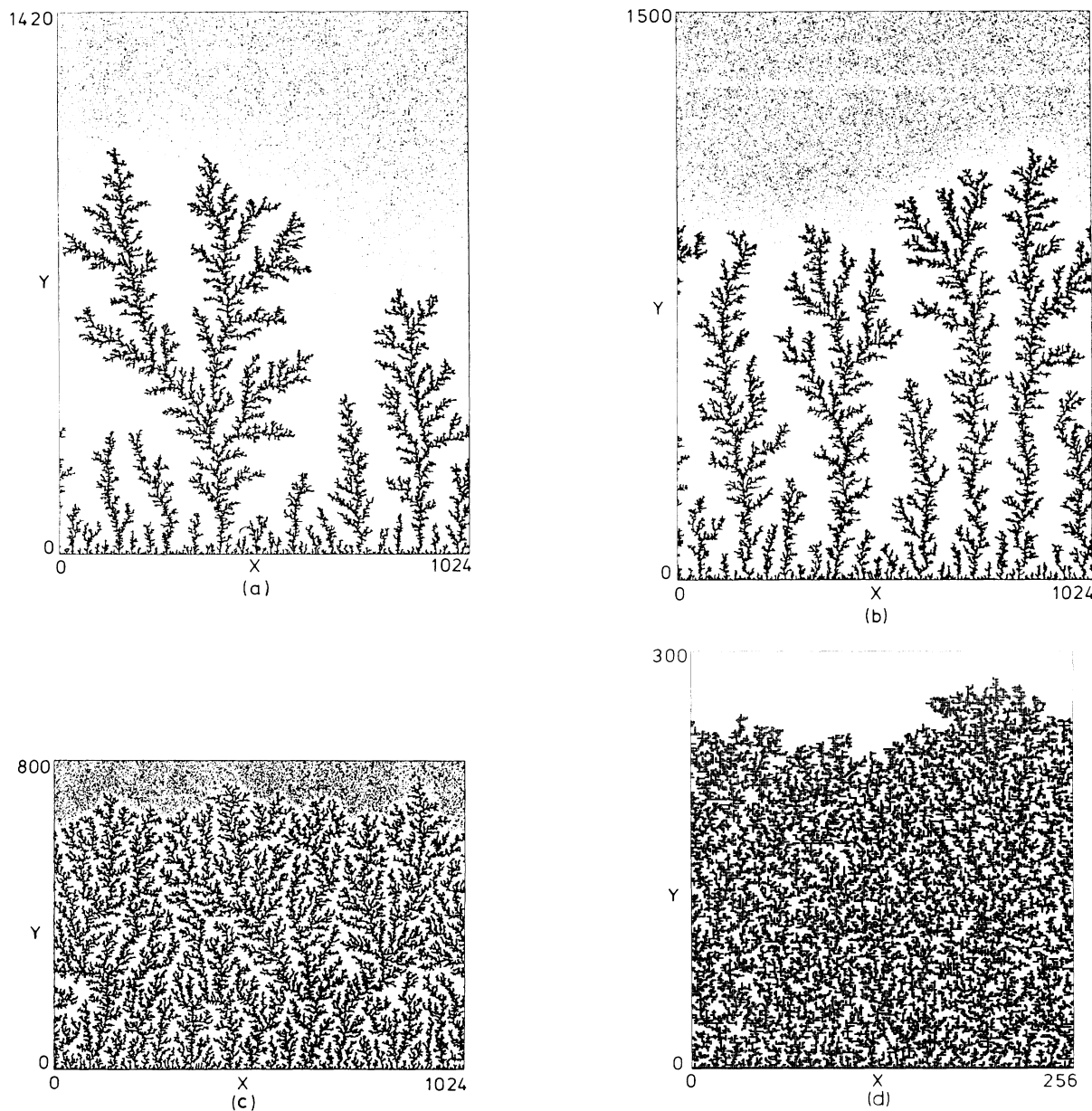


FIG. 2. Aggregations from various gas densities,  $n_g =$  (a) 0.08, (b) 0.10, (c) 0.20, (d) 0.40, (e) 0.80, and (f) 1.0. (f) Only solid atoms are shown.

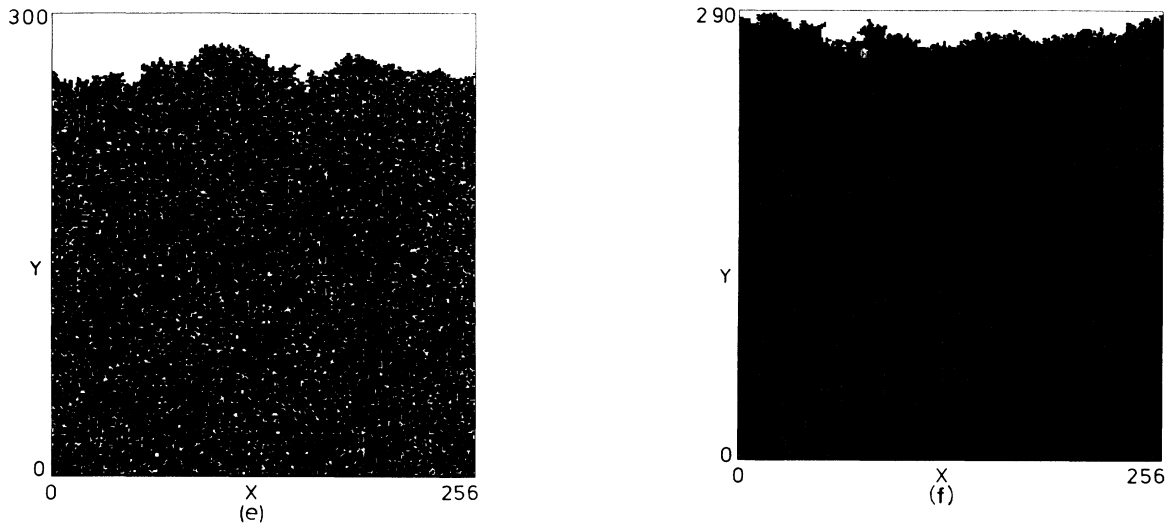


FIG. 2. (Continued).

which is defined by

$$V = \frac{\Delta N_s}{n_g L \Delta t} \quad (4)$$

Here  $\Delta N_s$  is the increase of  $N_s$  in a time interval  $\Delta t$ , and  $L$  is the width of the box. Velocity thus defined increases as a function of gas density  $n_g$ , as is shown in Fig. 4. At low densities  $0.08 \leq n_g \lesssim 0.35$ , the velocity satisfies the relation

$$V = 58 n_g^{3.50} \quad (5)$$

(From the least-squares fitting a straight line to the data in a logarithmic plot we obtain  $49 n_g^{3.40} \sim 69 n_g^{3.60}$ .) Com-

paring with Eq. (3) we get the fractal dimension

$$D_f = 1.71 \pm 0.01, \quad (6)$$

which agrees with the one obtained previously:  $D_f = 1.695 \pm 0.002$  for square-lattice DLA<sup>11</sup> and  $D_f = 1.715 \pm 0.002$  for off-lattice DLA.<sup>12</sup> Therefore, our matching hypothesis between the static length scale  $\xi_s$  in

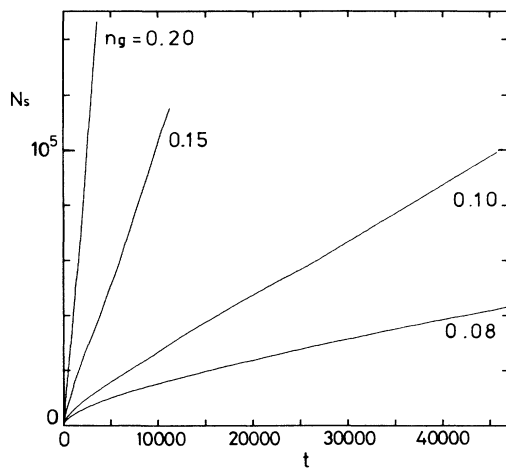


FIG. 3. Number of solid atoms vs time at low densities with  $L = 1024$ .

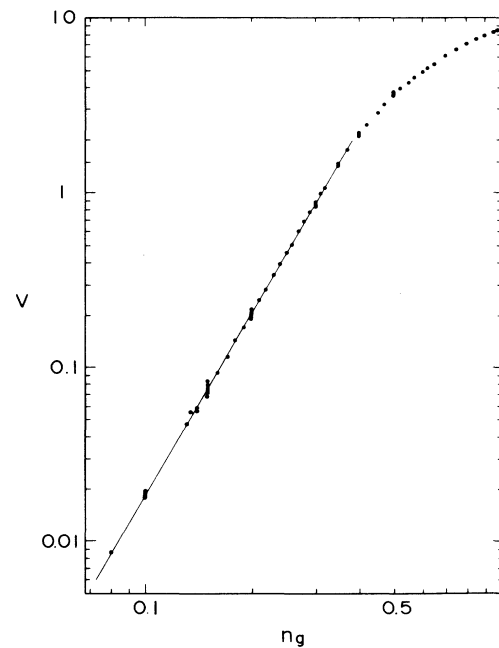


FIG. 4. Growth rate  $V$  vs gas density  $n_g$  in a logarithmic scale. The best fit  $V = 58 n_g^{3.50}$  for  $n_g \leq 0.35$  is shown by the solid line.

solid and the dynamical one  $\xi_g$  in gas seems to be powerful for the determination of the growth rate of the aggregate.

Recently we were informed of work by Meakin,<sup>13</sup> who performed a simulation study on two similar models. In the first model I, solidification takes place only when an attempt is made to move a gas particle onto a site occupied by a solid atom. Therefore, the growth rate is always smaller in model I than in our model, where the solidification takes place whenever a gas atom comes in contact with solid atoms. In model I Meakin obtained the asymptotic linear growth of the total mass of solid as

$$N_s \propto n_g^{3.8} t$$

at low densities. ( $n_g$  and  $N_s$  corresponds to  $\rho$  and  $M$  in Meakin's notation.) In our terminology, the velocity of average height is then

$$V = \frac{\Delta N_s}{n_g L \Delta t} \propto n_g^{2.8}, \quad (7)$$

whose exponent 2.8 is smaller than our result 3.50.

In the second model II which is the same with Voss's model,<sup>7</sup> any gas particles connected to the freshly solidified atom via nearest-neighbor links also become part of the aggregate, and therefore at high densities a percolation transition occurs. The growth rate in model II is always larger than our model, and at low-density limit both models become equivalent to the usual DLA model. From the scaling plot of  $n_s^{3.0} N_s$  versus  $n_g^{8.0} t$  in Ref. 13, the asymptotic growth rate may be written as

$$V \propto n_g^{4.0}. \quad (8)$$

The exponent 4.0 is larger than our value 3.50. In Meakin's paper, however, the simulation does not seem to be long enough at low densities ( $n_g \lesssim 0.2$ ) to warrant the linear growth law ( $N_s \propto t$ ). Furthermore, at late stages the scaling plot does not fall on a single curve, indicating the inaccuracy in the choice of the exponents.

We now consider the growth at high densities. When density is higher than 0.6 the asymptotic growth velocity is higher than the initial velocity. At high densities the gas atoms do not have much space to move, and the growth is mainly controlled by the interface kinetics. At the start the interface is flat and  $n_g L$  sites next to the solid seed are occupied by gas atoms. Among the remaining  $(1-n_g)L$  empty sites,  $n_g/4$  are filled by gas atoms via the diffusion from the second next layer. Thus the number of solid atoms increases as  $\Delta N_s = 4[n_g + (1-n_g)n_g/4]L\Delta t$  and the initial velocity should be  $V = 5 - n_g$ . The later increase in the growth velocity is the result of the increased number of the gas atoms at the growth front due to the spatial fluctuation of the interface. The steady growth velocity approaches the limiting value  $8.64 \pm 0.08$  for  $n_g = 1$ . In the high-density limit of this model ( $n_g = 1$ ), the gas atoms are packed densely and cannot diffuse at all. Crystallization progresses via random selection of surface gas atoms. This is the growth process of the Eden model. The growth rate of the Eden model in our time unit is four times the average

number of gas atoms next to the solid per column (ratio of the perimeter length to the width  $L$ ). Thus the steady-state velocity  $V(n_g = 1) \approx 8.64$  means that the interface is about 2.16 times longer than the simple flat interface. This enhancement factor agrees with that obtained previously in the Eden model simulations.<sup>14,15</sup> Hirsch and Wolf<sup>15</sup> found that the true steady-state value  $2.180 \pm 0.007$  is reached when the interface is as high as 2000 for  $L \gtrsim 90$ . Our steady-state value for  $n_g = 1$  is taken when the height is comparable to the width  $L = 400$ . It is 1% lower than their asymptotic value but agrees with that at the same height given in Ref. 15.

As the gas density decreases, the growth rate decreases linearly from the limiting value  $V = 8.64$  and is approximately given by

$$V \approx 8.64(1 - 0.64n_g). \quad (9)$$

This decrease is attributed to the increasing number of vacancies at the interface, which hinder the solidification. In order to see simply the effect of the vacancy, let us insert vacancies with a concentration  $(1-n_g)$  in a configuration of an Eden model. The number of gas atoms which is contingent to solid atoms decreases approximately by a factor  $n_g^2$  relative to that of an Eden model. From the definition Eq. (4) of the velocity of the aggregate height, then, the reduction in the velocity  $V(n_g)$  from that of the Eden model  $V(n_g = 1)$  is expected to be proportional to the density  $n_g$ . Since the interface is not planar, the proportionality coefficient is not unity.

### C. Characteristic length $\xi_g$ in gas phase

In order to confirm our hypothesis about the length scales for the low density growth, we analyze the characteristic length in the gas and in the solid. The local density distribution of gas atoms around the growth points is calculated and it is found<sup>9</sup> that the gas density relaxes exponentially with a characteristic length  $\xi_g$  for  $n_g < 0.2$ . Macroscopic theory of crystal growth tells us that the gas density relaxes from 0 to  $n_g$  exponentially with a screening length  $D/V = \xi_g = l/2$  for a flat surface.  $D/\xi_g$  obtained from the simulation is about half of the growth rate  $V$ . (see Fig. 5.) The enhancement factor  $\sim 2$  of the growth rate  $V$  may be attributed to the rough and wandering interface, which increases the length of the effective growth front about twice to that of the flat interface.

### D. Characteristic length $\xi_s$ in solid aggregate

The solid density correlation function  $g(r) = \langle n_s(0)n_s(r) \rangle$  is expected to decrease from 1 at  $r=0$  in a power law  $g(r) \sim r^{D_f-d}$  for distances  $r$  up to the crossover length  $\xi_s$ , and then saturates at the value  $n_s$  for larger  $r$ . Instead of calculating  $g(r)$  directly, we divide the system into square cells of the linear dimension  $b$  and count the number of solid atoms in a cell  $N_s = \sum_{i \in \text{cell}} n_s(i)$ . Its second moment is transformed as

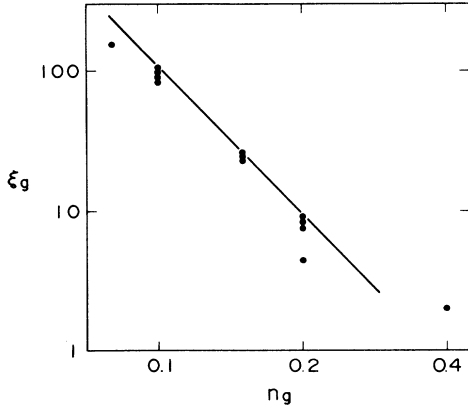


FIG. 5. The characteristic length  $\xi_g$  vs gas density  $n_g$  in a logarithmic scale. The solid line indicates  $2/V$ .

$$\begin{aligned} \langle N_s^2 \rangle_b &= \left\langle \sum_i \sum_{j \neq i} n_s(i)n_s(j) + \sum_i n_s(i) \right\rangle_b \\ &\approx \langle N_s \rangle_b \left[ \int_1^b g(r) d^d \mathbf{r} + 1 \right]. \end{aligned} \quad (10)$$

Therefore, the quantity defined by

$$n(b) = (\langle N_s^2 \rangle_b / \langle N_s \rangle_b - 1) b^{-d} \quad (11)$$

is essentially an average of  $g(r)$  in a cell with the dimension  $b$ . The subtraction of unity in Eq. (11) removes the strong autocorrelation. Reflecting the crossover of  $g(r)$  at  $r = \xi_s$ ,  $n(b)$  is expected to be proportional to  $b^{D_f - d}$  for  $b \lesssim \xi_s$ , and  $n(b) = n_s$  for  $b \gtrsim \xi_s$ . Figure 6 depicts  $n(b)$  as a function of the cell size  $b$ , taken from the central part of aggregates where the local solid density  $n_s$  stays homogeneous and equal to  $n_g$  on the average. The crossover of  $n(b)$  in  $b$  dependence is apparent. From the data at

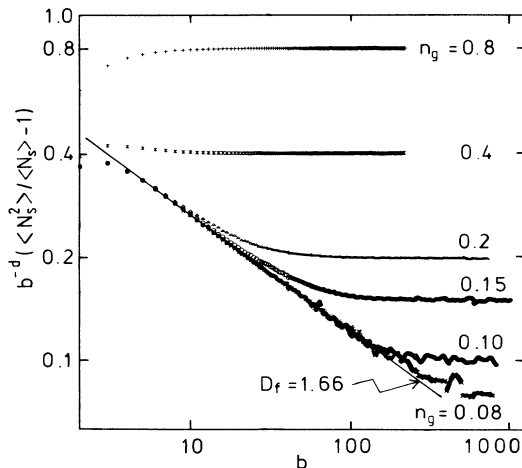


FIG. 6. Log-log plot of correlation of solid atom numbers in a cell  $n(b)$  vs the cell dimension  $b$ . The power-law decrease of  $n(b)$ , which characterizes the DLA, changes into a constant at a crossover length  $\sim \xi_s$ .

$b \lesssim \xi_s$  the fractal dimension is calculated to be  $D_f = 1.66 \pm 0.02$ , a little smaller than the expectation<sup>16</sup> 1.71. The discrepancy may be attributed to the insufficiency of the true solid region to get enough statistics: At low densities the diffusing gas particles remain deep in the solid, and the completely solidified region is rather small. Considering the unsettled controversy<sup>17</sup> on the fractal dimension of DLA, however, we do not exclude the possibility that the discrepancy is real.

Voss<sup>7</sup> analyzed density correlation of similar aggregates grown from a point seed and found  $\xi_s \propto n_g^{-2}$ , although other possibilities were not excluded. For this geometry the true steady state does not exist because of a finite radius effect, and more thought is required to make any definite conclusion (see Ref. 8). The present geometry ensures that  $g(r) = n_s = n_g$  for large enough  $r$ . Therefore the fractal behavior in short distances  $g(r) \sim r^{D_f - d}$  automatically leads to the density dependence of the crossover length Eq. (2).

### E. Width of a growing region

Since the aggregation growth takes place at the interface between the gas and the aggregate, positions of newly grown aggregate atoms reflect the structure of the interface. These newly grown atoms have heights fluctuating from the average as is shown in Fig. 7(a). The average height fluctuation over every  $M$  growing points

$$W^2(\bar{t}) = \frac{1}{M} \sum_{i=1}^M \left[ y_{j+i} - \frac{1}{M} \sum_{i=1}^M y_{j+i} \right]^2 \equiv \overline{(y - \bar{y})^2} \quad (12)$$

at the average time  $\bar{t} = \sum_{i=1}^M t_{j+i} / M$  has large fluctuation, but it tends to reach a certain steady-state value. Since  $W^2(t)$  contains the systematic drift of the average height during the time interval  $\Delta t$  for the growth of  $M$  points, the true width of the growing region  $w$  needs correction. In the steady state, the probability distribution of the height of the growth points is a function of the difference  $y - Vt$ . Therefore,

$$\begin{aligned} W^2(t) &= \frac{1}{\Delta t} \int_{\bar{t} - \Delta t/2}^{\bar{t} + \Delta t/2} \overline{(y - Vt)^2} dt \\ &= \frac{1}{\Delta t} \int \overline{[(y - Vt) + V(t - \bar{t})]^2} dt \\ &= w^2 + (V\Delta t)^2 / 12 = w^2 + (M / Ln_g)^2 / 12. \end{aligned} \quad (13)$$

The corrected width of the growing region  $w^2$  is shown in Fig. 7(b) for systems with various widths  $L = 100, 400$ , and 1024.

If we assume that the system is strongly correlated within the correlation length  $\xi$  but uncorrelated for large separations, the interface may be interpreted to consist of blobs with a size  $\xi$  and these blobs perform a random walk in the "time" of  $L/\xi$  steps. The value of the averaged interface width  $w$  is estimated from the analogy to the Brownian motion, with  $\xi$  the step unit length and  $L/\xi$  the time,

$$w^2 \propto \xi^2 (L/\xi)$$

or

$$w \propto (\xi L)^{1/2} \propto n_g^{-1/2(d-D_f)} L^{1/2}. \quad (14)$$

We now compare the simulation results with the above theoretical hypothesis. At high densities ( $n_g \gtrsim 0.3$ ) one can clearly observe the size dependence  $\sim L^{1/2}$  for  $L=100$  and  $400$ . In the limit of Eden model ( $n_g=1$ ) our results agree with a previous extensive simulation study<sup>18</sup> of the interface width which predicts  $w \approx 0.42\sqrt{L}$ . In order to reach this asymptotic behavior, however, the height should reach as high as  $\bar{y} \sim 0.4L^{3/2}$ . For narrow systems with  $L=100$  and  $400$  we have taken the maximum box height at  $h_{\max}(L=100)=10\,100$  and

$h_{\max}(L=400)=4100$ , which satisfies the above criterion. For a wider system with  $L=1024$ , the above criterion requires the height more than  $13\,000$  which is out of scope of our CPU time. For example, to grow one sample at  $n_g=0.1$  up to the height  $h_{\max}=1500$ ,  $8.5 \times 10^9$  Monte Carlo steps were necessary. The interface width at this height does not yet reach the steady-state value, and the data shown in Fig. 7(b) for  $L=1024$  is rather tentative.

As the density decreases, the width  $w$  increases. But our data does not seem to be sufficient for quantitative discussions. At low densities the crossover length  $\xi$  increases and thus the number of "time steps"  $L/\xi$  decreases quickly. Thus the data for narrow system as  $L=100$  is strongly influenced by the periodic boundary condition in the horizontal direction. The widest system with  $L=1024$  again suffers from the shortage of height and the data values have very large error bars. With all these limitations, however, one can observe that the slope in  $w$  versus  $n_g$  becomes steeper as the system width  $L$  increases. For  $L=400$  and  $L=1024$ , simulation results of the interface width  $w$  at low densities are compatible with the hypothesis Eq. (14) with  $D_f=1.71$ .

#### IV. SUMMARY AND DISCUSSIONS

Our model of aggregation growth from a lattice gas interpolates the DLA and the Eden model in a unified way. At the high density it approaches the Eden model. The characteristic length is the lattice constant, and the solid is always compact. The growth is controlled by the interface kinetics, and the growth velocity  $V \sim 8.64$  is determined by the number of interface atoms per width. ( $V \approx 8.72$  given in Ref. 15 is probably a better value.) At low densities, the growth is controlled by diffusion and the characteristic length  $\xi_g$  of the gas is essentially the diffusion length  $l=2D/V$ . This length  $l$  also characterizes the structure of the aggregate. As the size increases beyond  $l$ , a DLA-like fractal structure of the aggregate changes to a compact structure. Thus  $l$  should match with the crossover length  $\xi_s$  determined by the fractal dimension of the DLA. Despite the lack of physical time and dynamics in the DLA, its static structure determines the growth velocity of an aggregation from the diffusion field. This length matching is a new mechanism of velocity selection in unidirectional diffusion growth in the absence of the surface-tension effect. To summarize, the steady-state growth velocity turns out, in practice, to be a very sensitive and useful measure of the structural quantities, such as the length of the interface and the crossover length in the bulk of an aggregation.

The transition from an open fractal to a compact structure is seen in nature. In crystal growth, by increasing the supercooling and thus increasing  $V$  and decreasing  $\xi$ , a dendritic crystal with an open structure changes to a compact one.<sup>19</sup> This compact dendrite is often seen in crystal growth, and it is essentially the same as our uniformly packed fractal clusters. Shigematsu and Komatsu grew dendrites of ionic crystals from a solution in a two-dimensional geometry and observed the solute concentration by an optical method.<sup>20</sup> The range of the solute depletion near the crystal in their photograph is about the

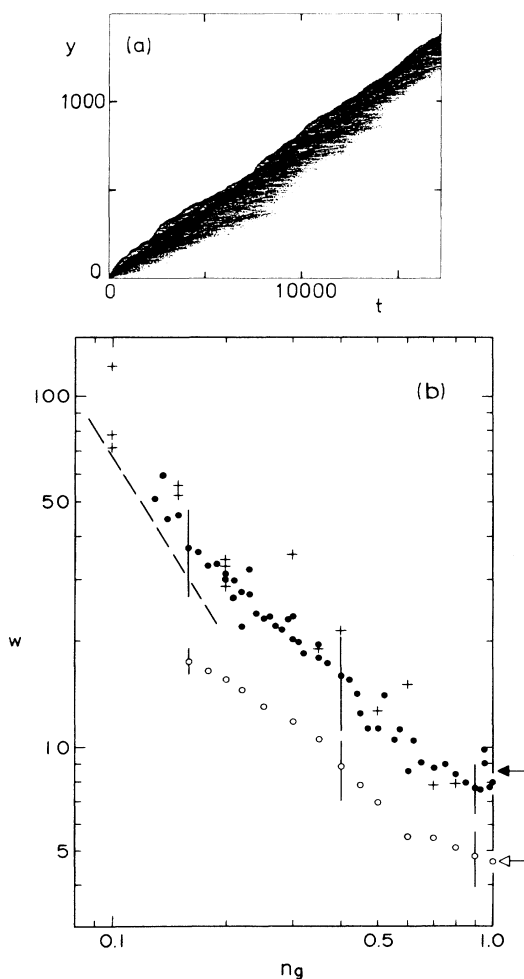


FIG. 7. (a) Height  $y$  of the newly grown aggregate sites vs time  $t$  for  $n_g=0.15$ . (b) Average width  $w$  of the interface vs gas density  $n_g$  for  $L=1024$  (+),  $L=400$  (●), and  $L=100$  (○). The bars are typical fluctuations of the width. The expected slope  $w \propto n_g^{-1/2(d-D_f)}$  is shown by the dashed line. Arrows indicate the values given in Ref. 15 for the Eden model ( $n_g=1.0$ ).

same as the characteristic distance of the main dendrite branches, in agreement with our simulation.

Transition from a DLA to a regular dendritic pattern occurs as anisotropy is introduced. We have neglected the anisotropy of the square lattice since our DLA-like clusters are not so large that the anisotropy has little effect.<sup>12,21</sup> By reducing the randomness in a numerical simulation one can make regular dendritic patterns.<sup>22-24</sup> The same is possible in real experiments: dendritic viscous fingers<sup>25</sup> and a DLA-like crystal<sup>26</sup> have been made by introducing anisotropy and randomness on the wall of the cell, respectively. The study of the anisotropy effect in the present model is now under way.<sup>27</sup> One of the authors (Y.S.) is now simulating a crystal growth from a lattice gas with interactions and also with the pos-

sibility of evaporation.<sup>28</sup> On increasing the chemical potential gain by solidification, a similar transition from fractal to compact structure is observed.

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