Effects of particle drift on diffusion-limited aggregation

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With the use of Monte Carlo methods, the effects of particle drift on diffusion-limited aggregation have been investigated. If particle-drift effects are dominant, the particles follow essentially linear trajectories (Hausdorff dimensionality $D_t=1.0$) and the resulting clusters have uniform structure on all but the shortest length scales ($D_c=d=2$ for clusters grown on a two-dimensional lattice). If the effects of drift are small, the particles follow Brownian trajectories ($D_t=2.0$), and the clusters have a Hausdorff dimensionality given by $D_c \simeq 5d/6$ (for small d). For intermediate cases, the clusters have a structure similar to clusters grown with the use of the Witten-Sander model of diffusion-limited aggregation on short length scales ($D_c \simeq 5d/6$) but are uniform on longer length scales ($D_c=d$). All of the simulations reported in this paper have been carried out using two-dimensional square lattices. However, similar results have been obtained with closely related nonlattice models, and we expect that similar results will also be obtained in higher dimensions. A crossover from a fractal structure on short length scales to a uniform structure ($D=d$) on longer length scales should also be observed for the deposition of particles on fibers and surfaces.

INTRODUCTION

At the present time there is considerable interest in the formation of large aggregates or clusters from small particles. A number of models for cluster formation have been proposed[1–7] during the past two decades, and a variety of computer simulations have been carried out to investigate the relationship(s) between the cluster geometry and growth mechanism. There is also considerable theoretical interest in this area.[8–12] Much of the work on cluster-formation processes has been motivated by a need to obtain a better understanding of the formation of biological structures[1,2,13] and the flocculation of colloidal systems.

The two models which are the most relevant for colloid flocculation are the Vold-Sutherland model[5,6,14] (VS) and the Witten-Sander[7] (WS) model for diffusion-limited aggregation. In the VS model, particles are assumed to follow random linear trajectories (Hausdorff[15] or fractal[16] dimensionality $D_t=1.0$). If the particle contacts the cluster, it is incorporated into the cluster and the cluster grows. In the WS model, the particles are assumed to follow Brownian trajectories ($D_t=2.0$). Again the particles are incorporated into the growing cluster on contact. Details concerning the growth of VS clusters on the computer can be found in Refs. 5, 6, and 14, and the growth of WS clusters is described in Refs. 7 and 17.

In many real systems the trajectories of small particles are perturbed by the presence of adventitious or intentionally applied external fields. If the external fields are essentially constant in space and time, and the particles are in a dissipative medium, the trajectories may be described as a random walk with a superimposed drift. On short length scales the particle trajectory behaves like an ordinary random walk with a Hausdorff dimensionality of 2.0, whereas on longer length scales, the drift becomes dominant and the Hausdorff dimensionality of the walk is 1.0. At the present time the relationship between the Hausdorff dimensionality of the cluster and the Hausdorff dimensionality of the particle trajectories is not fully understood. However, it is clear from numerical simulations that the Hausdorff dimensionality of the cluster D_c is *approximately* $5d/6$ (where d is the Euclidean dimensionality of the system) if the particle trajectory is a random walk (Hausdorff dimensionality $D_t=2.0$) for sufficiently small d.[7,8,17,18] Similarly, if the particles follow linear paths ($D_t=1.0$), $D_c \sim d$.[5,6,14] Consequently, if a cluster is grown starting from a single "seed" particle in a system in which the particles follow a drifting random walk, we might expect the cluster to have a morphology similar to that of a WS cluster on short length scales ($D_c \sim 5d/6$) and an essentially uniform density ($D_c \sim d$) on longer length scales.

In this paper, the results of numerical simulations which were carried out to investigate the effects of particle drift on diffusion-limited aggregation are described. The simulations reported here were carried out using two-dimensional lattice models. Similar results have been obtained with a nonlattice model, and we expect our general results to carry over to higher dimensionalities also.

MODEL

The simulation of diffusion-controlled cluster formation in the presence of particle drift was carried out with the use of a simple square lattice. We start out with a single occupied lattice site (shown by the black square in Fig. 1) and allow particles to follow biased random trajectories in the vicinity of the original occupied site or seed. The particle trajectories are generated by first generating a random number in the range 0–1. If this random number is smaller than the "drift probability," the particle is moved one lattice unit in the direction of the drift. If the random

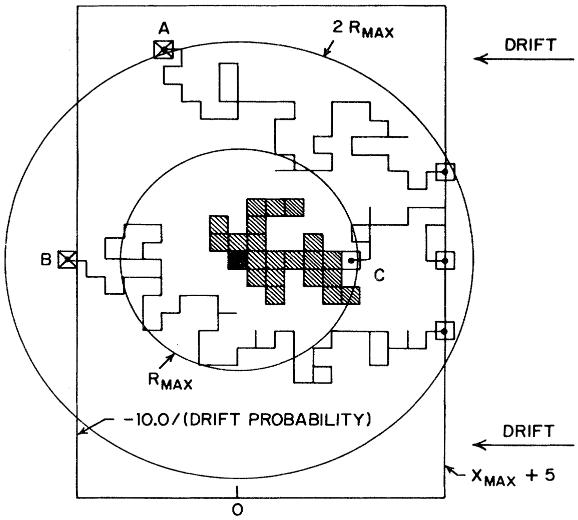

FIG. 1. Schematic representation of the model used to simulate the effects of particle drift on diffusion-limited aggregation.

number is greater than the drift probability, a second random number is generated to select one of the four nearest-neighbor sites, with equal probability, and the particle is moved to this site. If the drift probability has the value 1.0, the particle follows a linear trajectory in the direction of the drift, and if the drift probability is 0.0, the particle follows a random walk on the lattice. If the particle reaches a lattice site which is at a nearest-neighbor position with respect to an already occupied lattice site, it is stopped and incorporated into the growing cluster as an occupied lattice site. Figure 1 shows a schematic representation of the simulation method used in this paper at an early stage of cluster growth.

Since particles which become incorporated into the cluster may be considered to originate at a random point a long way "upstream" from the growing cluster and will cross a line which is upstream from the cluster for the first time at some random position, the particles are started out on a randomly chosen lattice site five lattice sites further upstream than the most upstream lattice site in the cluster (Fig. 1). Since trajectories starting from most such

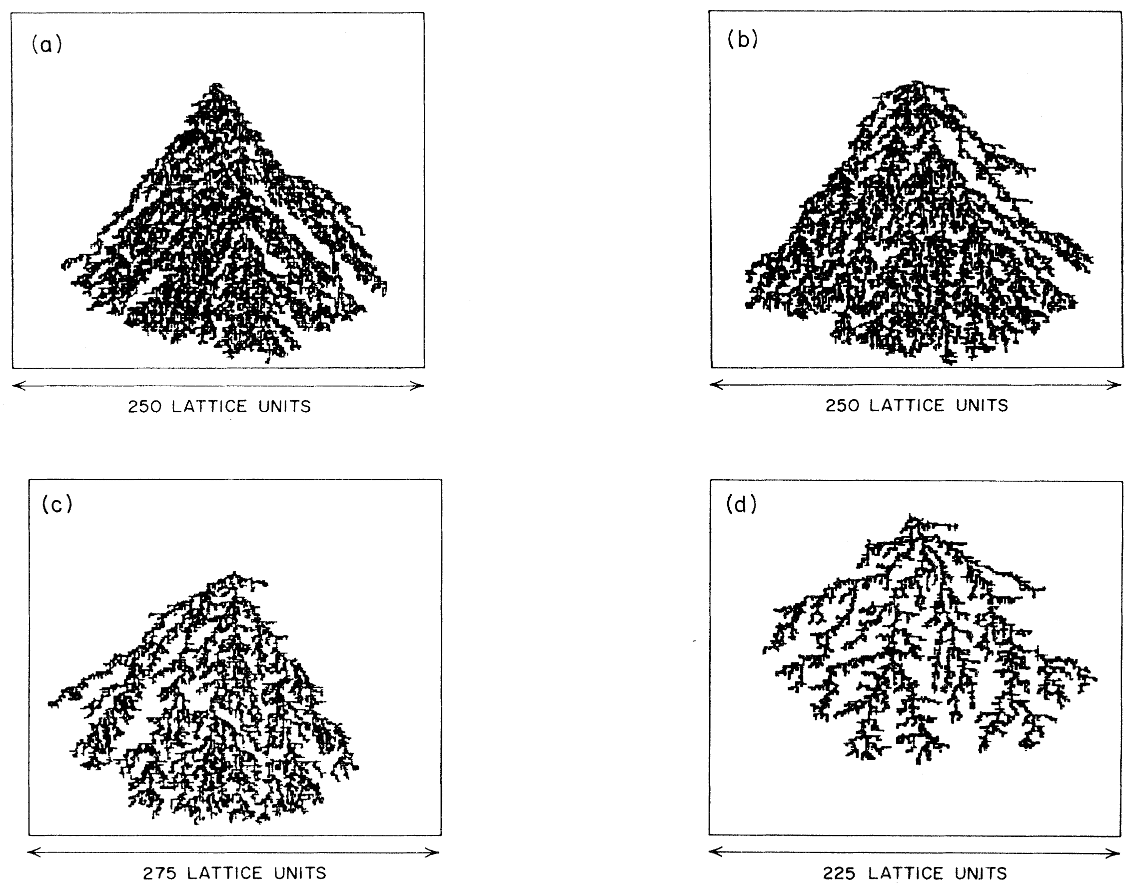

FIG. 2. Typical clusters grown with the use of the model shown in Fig. 1, and described in text, with relatively large drift probabilities. (a) Cluster of 9582 particles grown with the use of a drift probability of 1.0 (linear particle trajectories in the drift direction). (b) Cluster of 9742 particles grown with the use of a drift probability of 0.5. Particles are drifting from the bottom of the figure. (c) Typical cluster of 7902 particles obtained from a simulation with the use of a drift probability of 0.25. (d) Cluster of 4629 particles grown with the use of a drift probability of 0.1. Local structure resembles that of a cluster grown in two dimensions using the WS model.

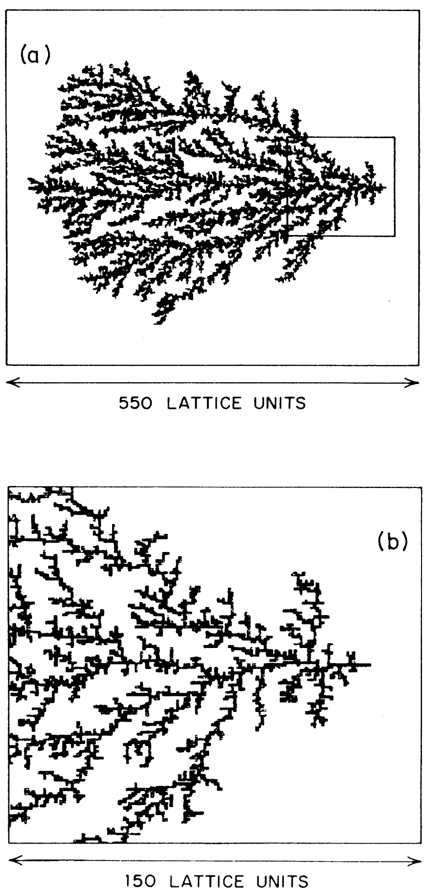

FIG. 3. Shows a cluster of 24 711 particles, or occupied lattice sites, grown with the use of a drift parameter of 0.05. (a) Entire cluster is shown which indicates that the cluster is quite uniform on large length scales. The region enclosed by the small rectangle is shown in (b). (b) Part of the large cluster of 24 711 particles shown in (a). This figure shows the local structure which closely resembles the local structure in a statistically self-similar WS cluster ($D \approx \frac{5}{3}$).

points will almost certainly miss the cluster, the starting position is also restricted to a position which is within a distance of $xR_{\max}$ lattice units ($x > 2.0$) from the origin ($R_{\max}$ is the maximum distance from any occupied lattice site in the cluster to the origin). If $xR_{\max}$ is less than 20 lattice units, points on a line perpendicular to the drift direction, five lattice units upstream from the cluster, which are within 20 lattice units of the origin, are chosen at random to start the trajectories. Figure 1 shows the three possible results of a particle trajectory. Trajectory A eventually moves the particle a long distance from the cluster (greater than $xR_{\max}$ from the origin, or 20 lattice units from the origin if $R_{\max} < 20$). In Fig. 1, x is 2.0. In this case, the trajectory is stopped to conserve computer time and a new trajectory is started 5 lattice units upstream from the cluster. This procedure does not introduce any significant error since most trajectories which travel a long distance from the cluster will never return to the cluster because the drift component will carry them "downstream" past the cluster. Trajectory B misses the cluster and eventually arrives at a position which is so far down stream that a return to the vicinity of the cluster is very improbable. This trajectory is also terminated to conserve computer time and a new trajectory is started. In most of the simulations, a trajectory is terminated if it reaches a position 10.0/(drift probability) lattice units down stream from the cluster origin. Trajectory C soon reaches a position which is a nearest-neighbor site to an occupied lattice site in the cluster. The particle is now incorporated into the cluster. In most of our simulations, the parameter x was set to a value of 2.0, but larger values were used in some cases to check on our results.

RESULTS

Figure 2 shows results obtained for a few relatively small clusters using the procedures outlined above. The cluster shown in Fig. 2(a) was obtained using a drift probability of 1.0 (i.e., linear particle trajectories in the direction of the drift). Overall the cluster looks like a sector with uniform density on all but very short length scales. The opening angle of the sector (which cannot be measured directly from Fig. 2 because of distorters in the computer graphics) is about 75°. A considerably smaller opening angle is found with a corresponding nonlattice model.

Figure 2(b) shows a cluster of 9742 particles grown with the use of a drift parameter of 0.5, and Fig. 2(c) shows a cluster of 7902 particles grown with the use of a drift parameter of 0.25. The structure is now becoming more open, and density fluctuations are beginning to extend to a longer range. As the drift probability is reduced still further to a value of 0.1 [Fig. 2(d)], the local structure begins to resemble that seen in two-dimensional WS clusters.

As the drift probability is lowered further and further, the WS structure extends to longer and longer length scales. To investigate the effects of drift on diffusion-limited aggregates, the largest clusters which were practical on a DEC VAX 11/780 computer were generated. Figure 3(a) shows a cluster of 24 711 occupied lattice sites generated with the use of a drift parameter of 0.05 and a sticking probability of 1.0 at nearest-neighbor positions only. The portion of the cluster contained in the rectangle in Fig. 3(a) is shown in Fig. 3(b) so that the local structure can be seen.

Figure 4 shows density-density correlation functions obtained from four large two-dimensional clusters grown using drift probabilities of 1.0, 0.2, 0.1, and 0.05. For comparison, the density-density correlation function obtained from a WS cluster (drift probability $=0$) is also shown in Fig. 4.

DISCUSSION

The crossover in the fractal dimensionality of the cluster from $D_c \sim \frac{5}{3}$ on short length scales to $D_c \sim 2.0$ on long length scales is associated with a similar crossover in the dimensionality of the particle trajectory from $D_t = 2.0$ on short length scales to $D_t = 1.0$ on long length scales. For a random walk on a Euclidean lattice we have

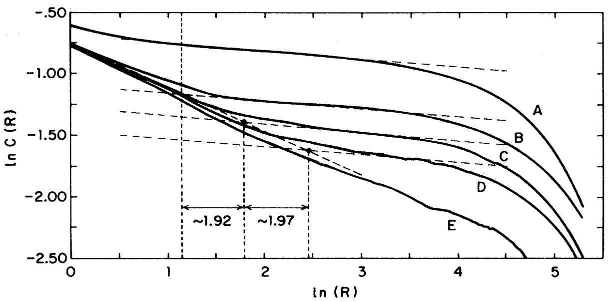

FIG. 4. Density-density correlation functions shown in the form of log-log plots for four clusters grown with the use of different drift probabilities (P). A—40 000 occupied lattice sites, $P=1.0$. B—40 000 occupied lattice sites, $P=0.2$. C—27 210 occupied lattice sites, $P=0.1$. D—24 711 occupied lattice sites, $P=0.05$. Curve E is the density-density correlation function obtained from a WS cluster. At small distances the slope of curves C—E, are close to $-\frac{1}{3}$.

$$\langle R^2 \rangle^{1/2} = AN^{1/2} , \qquad (1)$$

where $\langle R^2 \rangle^{1/2}$ is the rms displacement and N is the number of steps in the walk. For a square lattice $A=1.0$. The particle displacement resulting from the drift is given by

$$\langle x \rangle = PN , \qquad (2)$$

where $\langle x \rangle$ is the average displacement and P is the drift probability. Consequently we expect a crossover from $D_t=2.0$ to $D_t=1.0$ when $PN \sim AN^{1/2}$ or when the walk length (Pythagorean length) is given by

$$l \sim A^2/P . \qquad (3)$$

Equation (3) tells us that the length scale over which the cluster exhibiting a WS structure is proportional to $1/P$. This seems to be supported by our results shown in Fig. 4. A similar crossover from a fractal to a uniform geometry as the length scale increases should also be observed in related phenomena, such as the deposition of particles on fibers and surfaces.[19]

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[1]M. Eden, in *Proceedings of the Fourth Berkeley Symposium on Mathematical Statistics and Probability, Berkeley, 1961,* edited by G. Neyman (University of California Press, Berkeley, 1961), Vol. 4, p. 223.

[2]T. Williams and R. Bjerknes, Nature (London) 236, 19 (1972).

[3]P. A. Rikvold, Phys. Rev. A 26, 647 (1982).

[4]Y. Sawada, S. Ohta, M. Yamazaki, and H. Honjo, Phys. Rev. A 26, 3557 (1982).

[5]M. J. Vold, J. Colloid Sci. 18, 684 (1983).

[6]H. P. Hutchison and D. M. Sutherland, Nature (London) 206, 1036 (1965); D. N. Sutherland, *ibid.* 226, 1241 (1970); J. Colloid Interface Sci. 22, 300 (1966); 25, 373 (1967); D. N. Sutherland and I. Goodarz-Nia, Chem. Eng. Sci. 26, 2071 (1971).

[7]T. A. Witten and L. M. Sander, Phys. Rev. Lett. 47, 1400 (1981).

[8]H. Gould, H. E. Stanley, and F. Family, Phys. Rev. Lett. 50, 686 (1983).

[9]M. Muthukumar, Phys. Rev. Lett. 50, 839 (1983).

[10]M. Nauenberg, R. Richter, and L. M. Sander, Phys. Rev. B 28, 1649 (1983).

[11]R. Ball, M. Nauenberg, and T. A. Witten (unpublished).

[12]M. Sahimi and G. R. Gerauld, J. Phys. A (in press).

[13]P. Tautu, Z. Krebsforsch. 91, 223 (1978).

[14]P. Meakin, J. Colloid Interface Sci. (in press).

[15]F. Hausdorff, Math Annalen 79, 157 (1919).

[16]B. B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, San Francisco, 1982).

[17]P. Meakin, Phys. Rev. A 27, 604 (1983); 27, 1495 (1983).

[18]It is not known at the present time if any unique relationship exists between the dimensionality of the particle trajectories and the dimensionality of the clusters. This question will be the subject of additional simulation studies.

[19]P. Meakin, Phys. Rev. A 27 2616 (1983).