Stochastic hierarchical model for cluster-cluster aggregation

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A stochastic hierarchical model of cluster-cluster aggregation, which is obtained by introducing a sticking probability into the recently proposed hierarchical model by Sornsen and Oh [Phys. Rev. E **58**, 7545 (1998)], is presented. The fractal dimension and aspect ratio calculated using the model give good agreement with those obtained in standard simulations of cluster-cluster aggregates. An interesting result is that the fractal dimension shows a peak as a function of the sticking probability. [S1063-651X(99)17310-0]

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I. INTRODUCTION

An important mechanism of forming fractal aggregates that occur in colloids [1] and aerosols [2,3] is cluster-cluster aggregation. These aggregates are easily generated on a computer and are able to explain the observed structures reasonably well [4–7]. Recently, Sorensen and Oh [8] have proposed a very simple and interesting model (SO model) for diffusion-limited cluster-cluster aggregation (DLCA). The model makes several simplifying assumptions that allow for an analytical calculation of the fractal dimension. The fractal dimension D calculated using the SO model and that obtained from the computer generated aggregates using standard DLCA algorithms show good agreement. However, the *aspect ratio*, which is the divine proportion for analytic calculations of the SO model, is too high compared to the one for computer generated aggregates [8].

For a two-dimensional lattice the assumptions of the SO model can be stated as follows [8]. (The extension to higher dimensions is done along similar lines.)

(i) It is an on-lattice hierarchical model.

(ii) Only side-to-end collisions are allowed. If a cluster is circumscribed by a rectangle commensurate with the lattice, the longest edge of this rectangle is the side, and the shortest edge is the end.

(iii) No part of the circumscribing rectangle of a cluster colliding with the side of the circumscribing rectangle of the second cluster can extend beyond the limits of that side.

(iv) The circumscribing rectangles cannot interpenetrate.

Though the SO model gives the correct fractal dimension for cluster-cluster aggregates, it is based on several assumptions that are not appropriate for the natural and experimental situations and also the standard simulations. This is reflected in the fact that the observed aspect ratio is not in agreement with that obtained by the SO model. However, the SO model is very simple and allows analytic calculation of the fractal dimension and aspect ratio. This, coupled with the good agreement of fractal dimension, prompts one to further investigate the SO model.

In the present paper I study the effect of introducing a stochastic element via the sticking probability, while retaining other simplifying features of the SO model. The stochastic hierarchical model (SH model) presented here leads to several interesting results. I find that the fractal dimension first increases and then decreases as the strength of the stochastic element is increased or the sticking probability decreased. On the other hand, the aspect ratio decreases monotonically as the strength of the stochastic element is increased. Interestingly, as the fractal dimension first increases and then decreases, and when it has the original value, the aspect ratio at that time has the value that is closer to the correct DLCA value. Thus the stochastic model gives a much better agreement with DLCA. (Here, DLCA refers to aggregates generated using the standard algorithm of diffusion-limited cluster-cluster aggregation that involves random aggregation of the polydisperse system of aggregates [6].)

In Sec. II, I introduce the stochastic hierarchical model (SH model) and present the numerical results. Section III concludes the paper.

II. STOCHASTIC HIERARCHICAL MODEL

I now introduce our stochastic hierarchical model (SH model). Instead of a strict hierarchy, as in the SO model, I follow the following procedure.

At any stage of the hierarchy, all the existing clusters are paired. Two clusters in a pair combine to form a new larger cluster with probability q (sticking probability), while they remain as distinct clusters with probability $p=1-q$. Thus p is a parameter that represents the strength of the stochastic element in the hierarchy. We also assume that assumptions (ii), (iii), and (iv) of the SO model are still valid. Thus I retain the simplifying features of the SO model, while making the SH model more realistic in comparison with standard cluster-cluster aggregation. Though analytic calculations are no longer possible, the numerical simulations are very simple. I note that, for $p=0$, the SH model is the same as the SO model. Figure 1 shows a realization of the SH model starting with ten particles or monomers.

With the introduction of the stochastic element in the hierarchy, the volumes of the clusters at a given stage are no longer represented by a Fibonnacci series as in the SO model [8]. Thus I have to rely on numerical calculations for estimating the aspect ratio and fractal dimension [9].

I now present the numerical results. I start with k clusters of single monomers and follow the SH model procedure with

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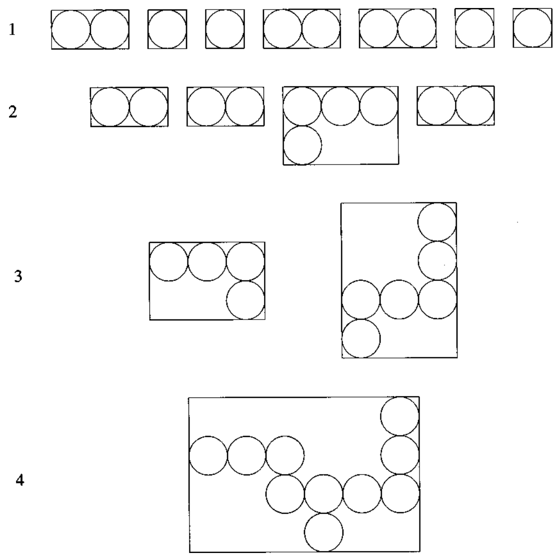

FIG. 1. Four stages of stochastic hierarchical growth in two dimensions leading to a ten-monomer cluster are shown. The starting point is ten single-monomer clusters.

a given p to obtain larger clusters [10]. The results are averaged over several realizations of the stochastic hierarchy for every set of values of k and p. I first consider aggregates generated on a two-dimensional lattice. To calculate the fractal dimension I obtain the average length scale s for clusters having the same mass m (number of monomers). Here the length scale is defined as the larger edge of the circumscribing rectangle. Figure 2 shows the plot of m against s for $p = 0.05$. The log-log plot is linear over more than three orders of magnitude and the slope of the graph gives the fractal dimension $D = 1.442 \pm 0.004$. The uncertainty in D is found to depend on p. It is negligible near $p = 0$, is ± 0.0015 near $p = 0.015$ and increases to ± 0.004 near $p = 0.05$, and is ± 0.006 at $p = 0.2$.

Figure 3 shows the fractal dimension D as a function of p.

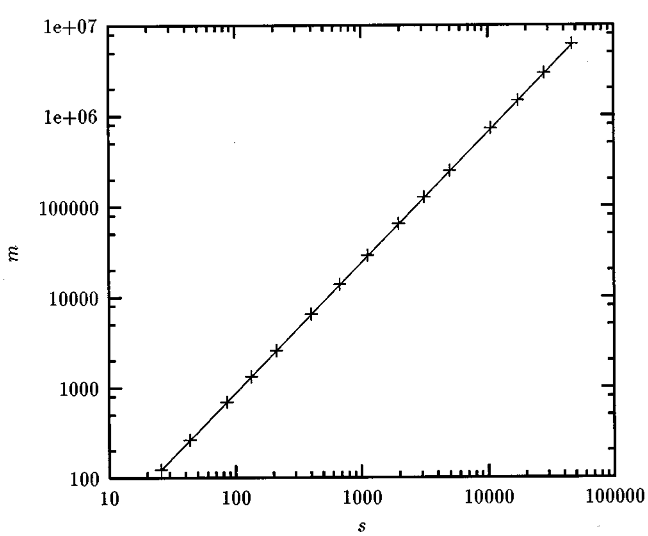

FIG. 2. Log-log plot of the mass m vs the average length scale s, of clusters grown using the SH model in two dimensions for $p = 0.05$. The average is taken over 1000 realizations. The solid line is a straight-line fit and the slope gives the fractal dimension $D = 1.442 \pm 0.004$.

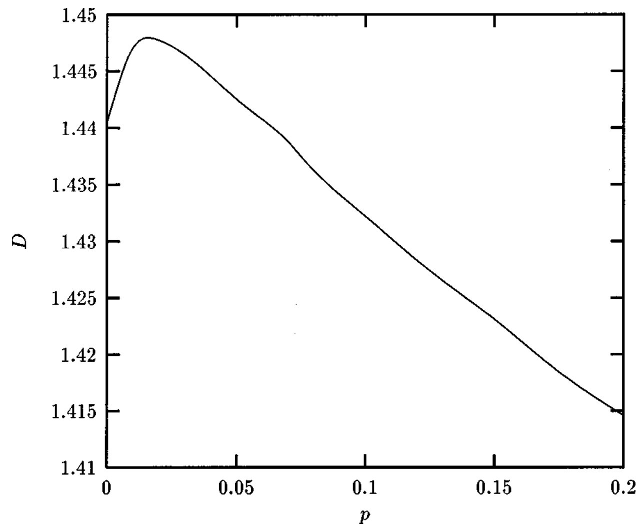

FIG. 3. Graph of the fractal dimension D vs the probability p for clusters in two dimensions. The fractal dimension shows a maximum near $p = 0.016$.

The figure givies a very surprising and interesting result. As p increases, the fractal dimension D first increases, goes through a maximum at $p \simeq 0.016$, and then decreases monotonically. Near $p = 0.061$, the fractal dimension again has the same value as for $p = 0$, i.e., 1.4404.

I now consider another important parameter of the system, the aspect ratio R, which is the ratio of the larger and smaller edges of the circumscribing rectangle. The aspect ratio is almost independent of N, the number of monomers in an aggregate, for $N > 50$. Figure 4 shows the plot of R as a function of p. There is a monotonic decrease of R as p increases. For $p = 0.061$, I have $R = 1.53 \pm 0.02$. The values of fractal dimension and aspect ratio for DLCA are $D = 1.44 \pm 0.03$ [11,12] and $R = 1.51 \pm 0.06$ [8], respectively. The corresponding values for the SO model are $D = 1.44\ldots$ and $R = 1.618\ldots$. Note the remarkable agreement of the aspect ratio for the SH model with $p \simeq 0.061$ and DLCA and also the good agreement of the fractal dimension for the SH

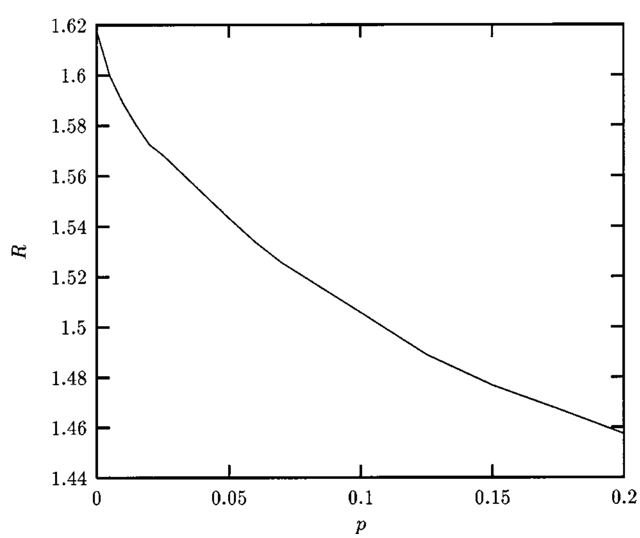

FIG. 4. Plot of the aspect ratio R vs the probability p for clusters in two dimensions. The aspect ratio decreases monotonically as p increases.

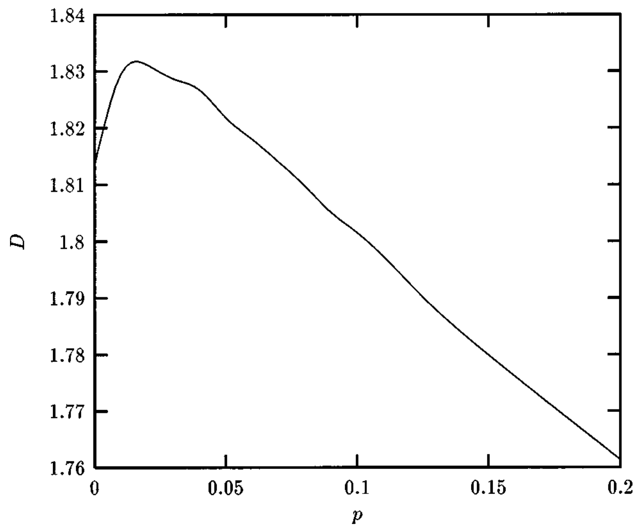

FIG. 5. Graph of the fractal dimension D vs the probability p for clusters in three dimensions. The fractal dimension shows a maximum near $p=0.016$ as in the two-dimensional case (Fig. 2).

model with both the SO and DLCA models.

Now I consider aggregates on a three-dimensional lattice. Figure 5 shows a plot of the fractal dimension D as a function of p. Again, as for the two-dimensional case, as p increases, D first increases, goes through a maximum at $p \simeq 0.016$, and then decreases monotonically. Near $p=0.061$, the fractal dimension has the same value, i.e., 1.813 ± 0.004, as for $p=0$. Note the similarity of the curve with the two-dimensional case. The value of fractal dimension for DLCA is $D=1.80 \pm 0.05$ [13].

Figure 6 shows the plot of R, defined as the ratio of the largest to the next largest edge of the circumscribing cube [8], as a function of p for aggregates on a three-dimensional lattice. There is a monotonic decrease of R as p increases. For $p=0.061$, I have $R=1.40 \pm 0.03$. To compare this value of the aspect ratio with that for DLCA I have generated aggregates using the algorithm proposed by Meakin [6]. The

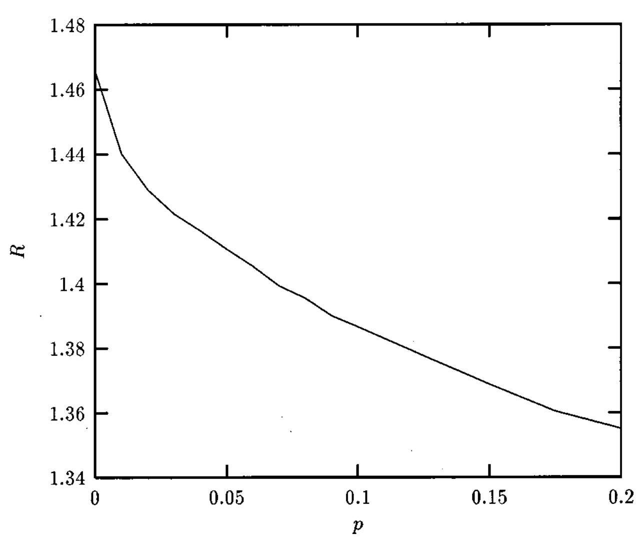

FIG. 6. Plot of the aspect ratio R vs the probability p for clusters in three dimensions. The aspect ratio decreases monotonically as p increases.

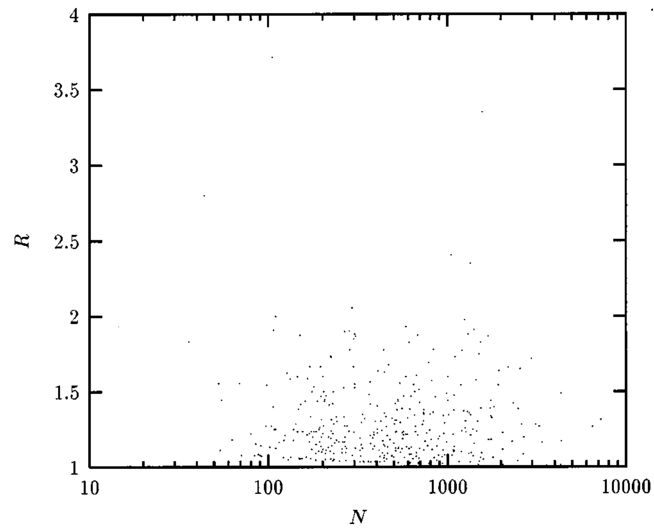

FIG. 7. The aspect ratio R as a function of the number of monomers in a cluster N for DLCA clusters in three dimensions.

maximum lattice size used was $200 \times 200 \times 200$. The log-log plot of the radius of gyration R_g as a function of the number of monomers per aggregate is linear with inverse of the slope giving the fractal dimension 1.8. Figure 7 shows a plot of the aspect ratio R verses N. The aspect ratio shows a large fluctuation with no dependence on N for large aggregates. The average aspect ratio is $R=1.30 \pm 0.09$, which is lower than the value 1.40 for the SH model with $p=0.06$. Note that the SO model gives $R=1.465$. Thus the value $R=1.40$ for the SH model is an improvement over the SO model.

III. CONCLUSION AND DISCUSSION

I have suggested a simple cluster-cluster aggregation model that uses some simplifying features of the SO model and a stochastic element for the sticking probability. Though the model is very simple, the fractal dimension and aspect ratio obtained using this model give a good agreement with that obtained in the standard DLCA in two dimensions. In three dimensions the aspect ratio is somewhat higher than the DLCA value, though it is an improvement over the SO model. The results establish the importance of stochastic hierarchy in aggregation problems. One might like to conjecture that the simple SH model has picked up the essential aspects of the cluster-cluster aggregation problem; however, much more work needs be done before such a conjecture may be accepted.

An interesting result that I obtain is the peak in the fractal dimension as a function of p. To the best of my knowledge I do not know of any other model that gives a peak in the fractal dimension as a function of the stochastic element. The exact reason for such a peak is not clear. The aspect ratio decreases as p increases. This behavior is natural, since as p increases the probability of collisions of clusters having different sizes increases. In a side-to-end collision, this will lead to a decrease in the aspect ratio. The mass (number of monomers) of the resulting cluster, which is an extensive quantity unlike the aspect ratio, also decreases; but for small p, the mass does not appear to decrease with the same rate as the aspect ratio, resulting in a rise in the fractal dimension. As p

increases further the mass starts decreasing at a faster rate and the fractal dimension starts dropping.

It is now well accepted that noise (or a stochastic element) can play an important role in the dynamics of many processes [14]. One interesting observation is that of stochastic resonance where noise is known to play a constructive role in some nonlinear systems [15,16]. Our SH model can also be treated as a dynamical growth model and we see that the stochastic element introduced by p appears to play an important role for small values of p in enhancing the fractal dimension.

ACKNOWLEDGMENT

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[1] D. A. Weitz and M. Oliveria, Phys. Rev. Lett. **52**, 1433 (1984).

[2] R. J. Samson, G. W. Mulholland, and J. W. Jentry, Langmuir **3**, 273 (1987).

[3] R. A. Dobbins and C. M. Megaridis, Langmuir **3**, 254 (1987).

[4] *Kinetics of Aggregation and Gelation*, edited by F. Family and D. P. Landau (North-Holland, Amsterdam, 1984).

[5] J.-F. Gouyet, *Physics and Fractal Structures* (Springer, New York, 1996).

[6] P. Meakin, Phys. Rev. Lett. **51**, 1119 (1983).

[7] M. Kolb, R. Botet, and R. Jullien, Phys. Rev. Lett. **51**, 1123 (1983).

[8] C. M. Sorensen and C. Oh, Phys. Rev. E **58**, 7545 (1998).

[9] It must be noted that, though I have not been able to obtain analytic results, the numerical calculations are very simple and take very little computer time.

[10] When there are odd numbers of clusters at any stage of hierarchy, one cluster cannot be paired. This cluster may be retained for the next stage of hierarchy or may be discarded. The final results do not seem to depend on either procedure. However, fluctuations are reduced if such isolated clusters are discarded.

[11] R. Jullien, M. Kolb, and R. Botet, J. Phys. Lett. **45**, L211 (1984).

[12] P. Meakin, Phys. Lett. **107A**, 269 (1985).

[13] C. M. Sorensen and G. D. Feke, Aerosol. Sci. Technol. **25**, 328 (1996).

[14] *Noise in Dynamical Systems*, edited by F. Moss and P. V. E. McClintock (Cambridge University Press, Cambridge, 1989), Vols. 1–3.

[15] R. Benzi, A. Sutera, and A. Vulpiani, J. Phys. A **14**, 1453 (1981).

[16] L. Gammaitoni, P. Hänggi, P. Jung, and F. Marchesoni, Rev. Mod. Phys. **70**, 223 (1998).