Diffusion-Controlled Deposition: Cluster Statistics and Scaling

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Diffusion-controlled deposition in dimension $d = 2$ is studied by Monte Carlo simulation, and the number of clusters of size s is found to scale as $n_s \sim s^{-\tau}$ with $\tau \approx 1.35$. The inequality $\tau < 2$ is shown to imply for a deposit of N particles per nucleation site that the exponents in the scaling *Ansatz* $n_s(N) \sim s^{-\tau} f(s^{\sigma}/N)$ satisfy the scaling law $\sigma = 2 - \tau$. If the scaling properties of deposits on a surface are related to those of an aggregate grown on a seed particle, $\tau = 1 + (d-1)/D$ is obtained, where D is the fractal dimension of the aggregate.

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Studies of the geometrical properties of highly ramified clusters formed by nonequilibrium growth processes are of importance in understanding phenomena like the sol-gel transition,[1] the early stages of nucleation,[2] dendritic crystal growth, the coagulation of smoke particles,[3] red blood cell aggregation,[4] etc. One of the simplest nonequilibrium growth processes which generates branching structures characterized by a fractal dimension (D) different from the Euclidean dimensionality (d) is diffusion-controlled aggregation introduced by Witten and Sander.[5] In this model, a cluster is grown from a seed particle as randomly walking particles launched from distant points arrive at and stick to the surface of the aggregate. Theoretical studies of the resulting clusters have so far concentrated on determining D, and the values obtained from Monte Carlo[5,6] (MC), mean-field,[7] and renormalization-group[8] calculations roughly agree with each other.

An interesting consequence of the fractal nature of the aggregates grown by the above rule is that the density correlations fall off with distance as a power law[5] suggesting an analogy with clusters in equilibrium systems at a critical point. In order to explore further this analogy and to point out some differences arising from the nonequilibrium nature of the aggregates, we studied the cluster statistics of objects grown by diffusion-controlled deposition. The choice of cluster statistics is motivated by the large body of work existing in this field for equilibrium systems[9,10] and by the fact that it seems to be useful in characterizing nonequilibrium systems[11,12] as well. The reason for preferring diffusion-controlled deposition[13] to aggregation[5] is that in the former case an ensemble of clusters appears naturally and, as we show below, the scaling properties of aggregates can be easily related to those of the deposits.

Our MC experiments and phenomenological scaling arguments lead to the conclusion that the analogy with equilibrium systems holds at the level of cluster statistics, too. The cluster-size distribution $n_s(N)$ as a function of the cluster size s and of the number of deposited particles per nucleation site N scales just like that in an equilibrium system near a critical point. The nonequilibrium nature of the deposits can, however, also be seen in this scaling since the critical exponents acquire values which cannot occur in an equilibrium system. Furthermore, the normalization of the cluster distribution yields an extra scaling law, thus reducing the number of independent exponents to one. We show that this one exponent can be expressed through the fractal dimension of a corresponding aggregate.

Diffusion-controlled deposition[13] differs from aggregation[5] only by the boundary condition: A surface of nucleation sites is used instead of a seed particle. As MC simulations show, the presence of the nucleation surface and the competition for the incoming particles result in the growth of a forest of treelike structures (see Fig. 3 in Ref. 13). For our purposes a cluster can be defined as a tree (collection of particles connected to the same nucleation site through nearest neighbors) and then the study of cluster statistics becomes the calculation of the size distribution of the trees in the forest.

In our MC experiments, 6000 to 9000 particles were deposited on a line of $L = 1000$ nucleation sites of a square lattice by use of a standard procedure described in Ref. 13. If we count the number of clusters $\mathfrak{N}_s(N)$ containing s particles, the cluster-size distribution function (the probability of finding an s-site tree on a nucleation site) is defined as $n_s(N) = \mathfrak{N}_s(N)/L$. The results are displayed in Fig. 1.

One of the notable features of Fig. 1 is the scaling behavior $n_s \sim s^{-\tau}$ which is similar to that occurring in equilibrium systems at a critical point. Another important outcome is that $\tau < 2$. This result is to be contrasted with the situation in equilibrium systems where the existence of the thermodynamic limit requires $\tau > 2$. Surprisingly, the inequality $\tau < 2$ follows from the existence of the "thermodynamic limit" $N \to \infty$ in our case as well. Indeed, since $n_s(N)$ is obviously bounded by $0 < n_s(N) < 1$ and since it is unlikely that $n_s(N)$ would have oscillatory or irregular behavior in the limit $N \to \infty$, one expects that $n_s(N)$ converges to some function n_s. Now taking the limit $N \to \infty$ of the normalization condition

$$N = \sum_s s n_s(N), \tag{1}$$

we find that if $n_s \sim s^{-\tau}$ for $s \to \infty$ then $\tau < 2$.

Now, to see a direct consequence of the inequality $\tau < 2$, let us remember that in the limit $N \to \infty$, scale-independent correlations appear, i.e., we may consider the system to be at a critical point. Then considering N^{-1} as the parameter measuring the deviation from the critical point, we may assume the following two-exponent scaling form for $n_s(N)$:

$$n_s(N) \sim s^{-\tau} f(s^\sigma/N). \tag{2}$$

Here $f(x)$ is a cutoff function $[f(x) \approx 1$ for $x \ll 1$ while $f(x) \approx 0$ for $x \gg 1]$ and Eq. (2) is supposed to be valid for large s and N. Equation (2) is the usual scaling *Ansatz* borrowed from the theory of thermal[14] and geometrical[15] critical points and extended to a far-from-equilibrium system. This extension is suggested by our MC results, and an additional justification of it comes from a regular fractal model of diffusion-limited deposition[16] where Eq. (2) can be shown[17] to be satisfied exactly. Now the effect of the inequality $\tau < 2$ appears through the sum rule of Eq. (1). In contrast to equilibrium systems where $\tau > 2$, the sum on the right-hand side is dominated by the large ($s \to \infty$) clusters; thus Eq. (2) can be used for its evaluation:

$$N \sim \int^\infty s^{1-\tau} f(s^\sigma/N)\, ds \sim N^{(2-\tau)/\sigma},$$

leading to the scaling law $\sigma = 2 - \tau$.

Obviously, if it is possible to express a quantity through n_s then its critical exponent can be related to τ. As an example, we computed the mean cluster size $S \sim N^\gamma$:

$$S = \frac{\sum s^2 n_s(N)}{\sum s n_s(N)} \sim N^{-1} \int^\infty s^{2-\tau} f(s^\sigma/N)\, ds \sim N^{1/(2-\tau)}$$

resulting in the scaling relationship $\gamma = 1/(2-\tau)$. This result differs from the corresponding scaling law $\gamma = (3-\tau)/\sigma$ in the theory of percolation. The difference occurs because in percolating systems $\sum s n_s(N)$ is not divergent at the percolation threshold and the $\sigma = 2 - \tau$ scaling law does not hold there. Our MC results for S (Fig. 2) show that the relationship $\gamma = 1/(2-\tau)$ is satisfied within the accuracy of the measurement. Note, however, that there is a curvature on the $\ln S$ vs $\ln N$ plot and the above scaling law is satisfied with some effective exponents only. The curvature in Fig. 2 suggests that $\gamma > 1.9$ and, consequently, for the true value of τ we should have $\tau > 1.5$. This indicates that much larger N should be used in order to reach the asymptotic regime.

Now we show how the one independent exponent τ characterizing a d-dimensional deposit on a $(d-1)$-dimensional plane can be related to the fractal dimension D of a d-dimensional aggregate grown on a seed particle. For this purpose, first the root mean square (rms) thickness $X(N)$

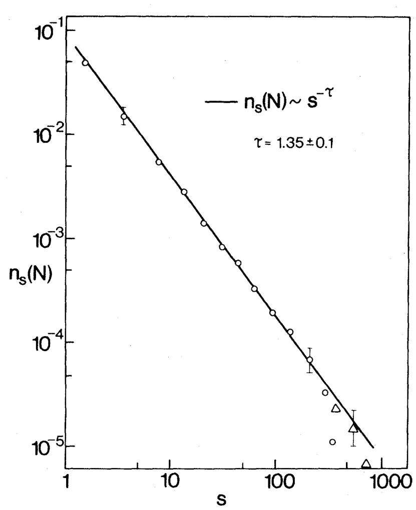

FIG. 1. Number of s-site clusters $n_s(N)$ vs s for deposits grown on $L = 1000$ nucleation sites. The straight line has a slope of $\tau = 1.35$. Circles are the results of statistics made on eight deposits of $N \times L = 6000$ particles while triangles refer to data from three deposits of 9000 particles. For small s values there is no remarkable difference between the results obtained for systems of different size.

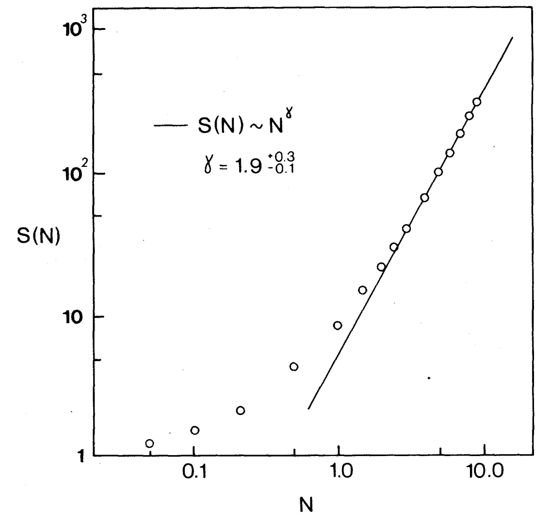

FIG. 2. Dependence of the mean cluster size S on N, the number of deposited particles normalized by the length of the substrate ($L = 1000$). The straight line corresponds to an asymptotic slope of $\gamma = 1.9$.

$\sim N^\epsilon$ of the deposit is calculated. Denoting by x_i the distance of the ith particle from the plane, we have

$$X^2(N) = N^{-1}\sum_i x_i^2 = N^{-1}\sum_s s X_s^2 n_s(N) \sim N^{2\epsilon},$$

where X_s is the average rms thickness of a cluster of s particles. Assuming $X_s \sim s^\theta$ and using Eq. (2), one finds $X(N) \sim N^{\theta/(2-\tau)}$ and so

$$\tau = 2 - \theta/\epsilon. \tag{3}$$

In order to express θ and ϵ through D and to explain why the scaling $X_s \sim s^\theta$ occurs, let us imagine a growing aggregate when its radius of gyration $R(N)$ is large. The tips of its branches grow in the same way as if they were the tips of the trees growing out of a plane. Thus the correlations within the large trees are the same as those within the branches of the aggregate far from the seed particle. As a consequence, for large s, $X_s \sim R(s)$, and since $R(s) \sim s^{1/D}$, we have $\theta = D^{-1}$. This relationship can also be verified[17] in the regular growth model.[16]

The above argument connecting the aggregate and the deposit may be used to calculate the D dependence of ϵ as well. It should be recognized, however, that D is defined by $R(\tilde{N}) \sim \tilde{N}^{1/D}$ where $\tilde{N}$ is the number of deposited particles in the aggregate while ϵ is obtained from $X(N) \sim N^\epsilon$ where N is the number of particles deposited on a unit of area. Since the characteristic length in the aggregate is $R(\tilde{N})$, its effective surface area should be given by $bR(\tilde{N})^{d-1}$ where b is a constant independent of $\tilde{N}$. Thus N and $\tilde{N}$ are related by $N = \tilde{N}/bR(\tilde{N})^{d-1}$, and the condition that the changes in $X(N)$ and $R(\tilde{N})$ are equal provided the number of incoming particles per unit area is the same gives

$$\epsilon = \frac{\delta \ln X(N)}{\delta \ln N} = \frac{\delta \ln R(\tilde{N})}{\delta \ln[\tilde{N} b^{-1} R(\tilde{N})^{1-d}]} = \frac{1}{D-d+1}, \tag{4}$$

where the limit $N \to \infty$ is taken and $D^{-1} = \delta \ln R(\tilde{N})/\delta \ln \tilde{N}$ has been used. Equation (4) can be shown[17] to be valid in the regular growth model[16] and it can also be compared with MC results. D is measured accurately[5,6] and the values $D(d=2) = 1.68 \pm 0.05$ and $D(3) = 2.50 \pm 0.05$ imply $\epsilon(d=2) = 1.47 \pm 0.10$ and $\epsilon(3) = 2.00 \pm 0.20$. The corresponding MC values[13] $\epsilon(2) = 1.30 \pm 0.075$ and $\epsilon(3) = 1.70 \pm 0.20$ are systematically smaller than the predictions following from Eq. (4). It should be noted, however, that the MC results for ϵ are quite unreliable since N is not large enough to be in the scaling region. The curvatures on the $\ln X(N)$ vs $\ln N$ plots of Ref. 13 indicate that the actual values of ϵ might be considerably higher.

A quantity which is measured with good statistics and provides an independent estimate of $\epsilon(3)$ is the radius of gyration exponent of deposits on a fiber $[R(N) \sim N^\delta,\ \delta = 0.665 \pm 0.030]$.[13] This exponent can be related to $\epsilon(3)$ by the same arguments used for the derivation of Eq. (4). The result $\epsilon(3) = \delta/(1-\delta) = 1.99 \pm 0.03$ is in excellent agreement with our previous estimate, thus giving confidence in the validity of Eq. (4).

Once ϵ and θ are known, τ is obtained from Eq. (3):

$$\tau = 1 + (d-1)/D. \tag{5}$$

With the MC estimates of D, this equation predicts $\tau(d=2) = 1.59 \pm 0.02$ and $\tau(3) = 1.80 \pm 0.02$. The value $\tau(2) \approx 1.6$ differs from our MC estimate $\tau(2) \approx 1.35$ but it is in agreement with the inequality $\tau(2) > 1.5$ derived from the scaling law $\gamma = (2-\tau)^{-1}$ together with the inequality $\gamma > 1.9$ suggested by the curvature on Fig. 2.

We close with two notes. First, D and, as a consequence of Eq. (4), τ are expected to be functions of d only. If, e.g., Muthukumar's mean-field expression[7] $D = (d^2+1)/(d+1)$ is used, then $\tau = 2d^2/(d^2+1)$. Second, Eq. (4), together with the inequality $\tau < 2$, yields $d-1 < D$. This result is consistent with Muthukumar's result[7] quoted above.

After we had submitted the manuscript, P. Meakin informed us that his large-scale MC simulations[18] of the $d = 2$ deposition model yielded $\epsilon(2) = 1.55 \pm 0.10$ and $\tau(2) = 1.55 \pm 0.05$. Both of these results are in excellent agreement with our predictions.

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[1] H. J. Herrmann, D. P. Landau, and D. Stauffer, Phys. Rev. Lett. 49, 419 (1982).

[2] W. Klein, Phys. Rev. Lett. 47, 1569 (1981).

[3] S. R. Forrest and T. A. Witten, Jr., J. Phys. A 12, L109 (1979).

[4] F. W. Wiegel and A. S. Perelson, J. Stat. Phys. 29, 813 (1982).

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

[6] P. Meakin, Phys. Rev. A 27, 1945 (1983).

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

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

[9] D. Stauffer, Phys. Rep. 54, 1 (1979).

[10] J. W. Essam, Rep. Prog. Phys. 43, 833 (1980).

[11] K. Binder and D. Stauffer, Adv. Phys. 25, 343 (1976).

[12] N. Jan, T. Lookman, and D. Stauffer, J. Phys. A 16, L117 (1983).

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

[14] M. E. Fisher, Rep. Prog. Phys. 30, 615 (1967).

[15] D. Stauffer, Phys. Rev. Lett. 35, 394 (1975).

[16] T. Vicsek, to be published.

[17] Z. Rácz and T. Vicsek, to be published.

[18] P. Meakin, to be published.