from the Yamada Foundation for a visit to Kyoto University for collaboration on this work.

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Kinetics of Formation of Randomly Branched Aggregates: A Renormalization-Group Approach

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(Received 22 November 1982)

The first renormalization-group approach for irreversible growth models of randomly branched aggregates is presented. The main result is that the Witten-Sander diffusion-limited aggregation model, a discrete version of a dendritic growth model, is in a different universality class than "equilibrium" lattice animals. Also calculated is the fractal dimension for the Witten-Sander model and the Eden model (a model developed for the study of biological structures).

PACS numbers: 68.70.+w, 05.70.Jk, 64.60.Cn, 82.70.Rr

There has been considerable recent interest in the physical mechanisms governing the structure of randomly branched aggregates or clusters formed by an irreversible kinetic process. Much of this interest is due to the applicability of these mechanisms to a variety of problems such as branched polymers and the sol-gel transition,[1] coagulation of smoke particles,[2,3] turbulence,[4,5] the early stages of nucleation,[6] and the growth of tumors.[7,8] Much of our understanding of the structure of clusters formed by irreversible growth processes has been obtained by computer simulation. Although such a procedure can yield accurate results and many insights into the structure of such clusters, it nevertheless is not usually sufficient to establish the universality classes for kinetic aggregation nor to determine how the structure of such clusters might differ from clusters formed by a random "equilibrium" process[9]; e.g., percolation clusters which model gelation and random lattice animals which model dilute branched polymers. Here we develop a renormalization-group method applicable to kinetic aggregation problems.

One measure of the structure of an aggregate is the manner in which N, the total number of particles in the aggregate, scales with the linear dimension R of the aggregate, $N \approx R^D$ where D is the Hausdorff or fractal dimension of the aggregate.[10] If D is less than the spatial dimension d, the aggregates are ramified; for $D = d$ they are compact.

A kinetic growth model for which D has been determined by computer simulation was proposed recently by Witten and Sander[2] and extensively studied by Meakin.[3] In this "diffusion-limited" aggregation model, the initial state at $t = 1$ is a seed particle located at the center of a large hypersphere. At $t = 2$ a particle is released on the surface of this sphere and undergoes a random walk until it visits a site adjacent to the seed (a perimeter site) and joins the cluster. If a walking particle reaches the surface of the sphere, it is removed and another released. This process continues until a large cluster of $t = N$ sites has been formed. Since sites deep inside the cluster are "screened," this growth process favors treelike or dendritic structures (see Fig. 1 of Ref. 2). Computer simulations suggest that D may be a "superuniversal" quantity for $d = 2$–6 given approximately by[3] $D_{\mathrm{WS}} \cong \frac{5}{6} d$.

In contrast to the Witten-Sander model, perimeter sites in the Eden cluster-growth model[7] are not screened. In this model a seed site is occupied at $t = 1$, and the cluster grows by occupying at $t = 2$ a randomly selected perimeter site. This process is repeated until a large cluster is formed. Because of the absence of screening it has been hypothesized that Eden clusters are compact, i.e., $D = d$. Monte Carlo simulations in $d = 2, 3$ are consistent with this hypothesis.[7]

Since there is no Hamiltonian formulation of either the Eden or Witten-Sander models, we adopt a position-space renormalization-group (PSRG) approach[9] in which the change in connectivity of the cluster, upon repeated length rescaling, is determined. The lattice is divided into cells of linear dimension b, a weight or fugacity K is associated with each occupied site in the cluster, and the cells are rescaled to a single site. The renormalization transformation $K' = R(K)$ is introduced by defining a cell to be occupied if a connected path "spans" the cell; $R(K)$ includes all spanning configurations in a cell of length b. D is given by[9] $D = \nu^{-1} = \ln\lambda_K / \ln b$, where $\lambda_K = (\partial K'/\partial K)_{K=K^*}$ and K^* is the critical fixed point.

To illustrate the method we consider the random-animal problem[11] for which all geometrically distinct clusters of s particles are weighted by a factor of K^s. We discuss the square lattice with $b = 2$, and define a cell to be occupied only if a connected path exists in both directions. We choose the starting point to be the lower-left corner of the cell. Enumerating the spanning configurations [Fig. 1(a)], we find $K' = 3K^3 + K^4$ and $D = 1.66$. For $b = 3$, $D = 1.60$.[11]

A one-parameter PSRG treatment of the Eden model proceeds as follows: $R(K)$ includes all the spanning configurations that can be *grown* from an initial seed site. In Fig. 1(b) we show the four different ways that the fully occupied configuration of four sites can be generated using the same corner rule; there are also four different ways of growing three-site clusters. Thus

$$K' = 4K^3 + 4K^4, \tag{1}$$

and $D = 1.72$. For $b = 3$ we find $D = 1.73$. Monte Carlo calculations by Peters *et al.*[7] for the Eden model have found slow convergence of the exponent ν $(= D^{-1})$ to its asymptotic value $\nu = 1/d$. Large-cell PSRG calculations are necessary to study the asymptotic behavior by the present method.

In contrast to the Eden model, a PSRG treatment of the Witten-Sander model requires at least two parameters, a fugacity K associated with an occupied site and a fugacity W associated with each step of the random walk of the added particle. The renormalization transformation for K' can be written in the form

$$K' = \textstyle\sum_{s,t} c_{st} K^s W^t , \tag{2}$$

where c_{st} is the number of different ways of growing a spanning cluster of s sites generated by random walks totaling t steps. Since the seed site occupies the lower-left corner of the cell, we allow the random walks to enter the cell only

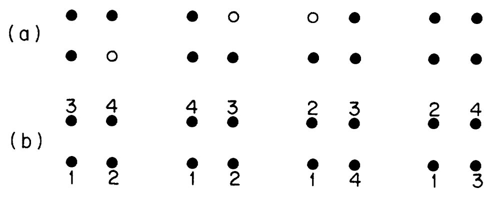

FIG. 1. (a) Spanning random-animal configurations for a $b = 2$ cell on a square lattice. (b) The four different ways in the Eden model that a cluster of four occupied sites can be grown from the seed at site A. Occupied sites are indicated by full circles and given weight K.

from the north and east. Examples of the generation of several spanning clusters for the case of a $b=2$ site-bond cell are shown in Fig. 2(a). We first enumerate all the possible ways of growing the two-site clusters (AB and AD) beginning with a seed particle at site A. In Fig. 2(a) we show the three ways of growing cluster AB, with their associated weights. The weight associated with both two-site clusters is $2K^2W(1+2W)$. For the three distinct three-site clusters, we consider all possible walks into a cell with a two-site cluster already present. If we enumerate all possible random walks in the cell and all possible spanning clusters, we obtain the recursion relation

$$K'=6K^3W^2(1+2W)+8K^4W^3(1+2W). \tag{3}$$

Next consider the recursion relation for W. To avoid the problem of enumerating the infinite number of spanning random walks on a finite cell, we note that at the critical fugacity only walks of length $\xi \approx N^{1/2}$ are important (N is the number of steps in the walk). Hence walks with more steps than the square of the end-to-end displacement can be neglected [see Fig. 2(b) and Ref. 12], and

$$W'=W^2+2W^3+5W^4+14W^5. \tag{4}$$

The fractal dimension of the Witten-Sander diffusion-limited aggregates is found from (3) and (4) with λ_K evaluated at $K=K^*$ and $W=W^*$. The results $D=1.71$ for $b=2$, $D=1.67$ for $b=3$, and $D=1.64$ for a cell-to-cell transformation[9] agree with the Monte Carlo result[2] $D=1.67$.

An important unanswered question is whether the ramified clusters of Witten-Sander are fundamentally different from equilibrium random animals for which $D=1.56$.[11] A comparison of the numerical values of the corresponding fractal dimensions does not allow any definite conclusions, since both numerical values are based on approximation procedures whose accuracy is difficult to gauge. In order to investigate the relation between the kinetic and equilibrium clusters, we generalize the model of Witten and Sander so that now the role of a seed is played by a random *animal* rather than a single *site*. For a given seed we add a particle to a random unoccupied site within a large hypersphere and allow the particle to undergo a random walk until it reaches a perimeter site or joins the cluster. This process continues until the cluster size becomes sufficiently large. Our generalized model is similar to that of Sander and Witten[13] who have shown, using Monte Carlo methods, that the structure of diffusion-limited aggregates is unchanged in the presence of finite-sized seeds.

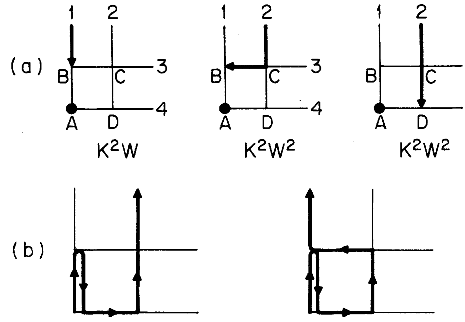

FIG. 2. (a) Possible ways of growing the cluster AB by a diffusion-limited process. (b) Examples of random walks on a $b=2$ bond lattice. The random walk of five steps and a net square displacement of 5 is included in the recursion relation (4); the random walk of six steps and a net square displacement of 4 is excluded from (4).

To construct the recursion relation for K' in our generalized random-animal, Witten-Sander model, we consider all possible random-animal configurations (spanning and nonspanning) as seed clusters in a $b=2$ cell. We then enumerate all possible random walks into the cell and the resultant spanning clusters:

$$K'=3K^3+K^4+6K^3W(1+W+2W^2)$$
$$+4K^4W(1+2W+2W^2+4W^3). \tag{5}$$

Now K' includes all the spanning animal configurations at $W=0$. The recursion relation for W' must also be modified, since certain walks are restricted because of nonspanning clusters already present in the cell. Including all possible spanning random walks in a cell with some sites already occupied, we find

$$W'=W^2+2W^3+5W^4+14W^5$$
$$+2KW^2(1+W+3W^2+5W^3)$$
$$+K^2W^2(1+2W^2). \tag{6}$$

The global flow diagram for the coupled recursion relations (5) and (6) is shown in Fig. 3. The fixed points at $K^*=0.532$, $W^*=0$ and $K^*=0$, $W^*=0.347$ correspond to the equilibrium random animal and unrestricted random walk, respectively. The most unstable fixed point corresponds to that of the "kinetic animal" grown by a diffusion-limited process from the random-animal seed sites. Note that the flow on the critical surface is from the kinetic-animal fixed point to the ran-

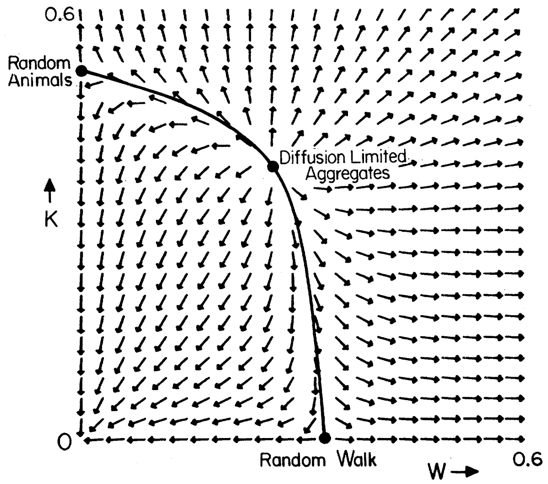

FIG. 3. Global diagram of flow vectors from (5) and (6). Each arrow indicates the local direction of flow, and the important fixed points are labeled.

dom-animal fixed point. Hence we conclude that as a consequence of the specific growth process considered, random animals and diffusion-limited aggregates are in different universality classes.

Our renormalization-group approach for kinetic aggregation is completely general and is not limited to the particular models considered here. Now that we have presented a renormalization-group argument that irreversible kinetic models can be in different universality classes from those of equilibrium random theories, further work should be done to generalize the nature of the kinetic growth models and to use renormalization-group arguments to establish dynamic universality classes. One possible generalization has been proposed recently by Rikvold,[14] who studied a model in which the growth is determined by both a diffusion process and a "surface-tension" term that favors compact clusters. Monte Carlo simulation suggests that crossover between clusters of different fractal dimension occurs as the screening length associated with the diffusion growth process is varied.

We thank C. Unger, H. Nakanishi, W. Klein, L. M. Sander, T. A. Witten, and D. Stauffer for useful discussions. This work was supported in part by National Science Foundation Grant No. DMR-82-08051, and by grants from the U. S. Army Research Office, the National Science Foundation, the U. S. Office of Naval Research, the Research Corporation, and the Emory University Research Fund. One of us (H.G.) was the recipient of a National Science Foundation Faculty Development Fellowship.

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