Growth Probability Distribution in Kinetic Aggregation Processes

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A growth process is characterized by the growth-site probability distribution $\{p_i\}_{i\in\Gamma}$, where p_i is the probability that site i on the surface of the cluster becomes part of the aggregate. Equations for the p_i's are solved numerically for diffusion-limited aggregation and the dielectric breakdown models by the standard Green's-function technique, and moments of the distribution are calculated indicating that a hierarchy of independent exponents is required to describe the critical behavior. The absence of a linear relation among the exponents is indicative of a nonconventional scaling for the growth probability distribution.

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What are the relevant parameters to describe fully the essential properties of a kinetic aggregation process? The answer to this question is extremely important in order to be able to understand the complexity and the richness of growth phenomena.

It is clear, for example, that an aggregate cannot be fully characterized by its fractal dimensionality. Diffusion-limited aggregation[1] (DLA) and percolating clusters have the same fractal dimensionality in three dimensions, yet they have completely different structures.

To characterize the aggregate further we consider at each time step the growth-site probability distribution (GSPD) $\{p_i\}_{i\in\Gamma}$, where p_i is the probability that site i becomes part of the aggregate and Γ is the set of perimeter sites. From the GSPD one can obtain detailed information on the capability of each perimeter site to grow and therefore more information on the surface structure.

In DLA the sites that can be occupied more easily are those at the tips of the cluster; very few particles are able to get deep inside the "fjords." The growth probability for the perimeter sites can be regarded as a measure associated to each site. In a similar fashion the voltage drop across the bonds has been selected for random resistor or random superconducting networks,[2,3] or the probability for a given region to be visited has been used for strange attractors.[4]

To be more specific, for an aggregate of macroscopic size L (e.g., L could be the radius of gyration) we consider the following moments as functions of L and their relative exponents:

$$Z(q)=\sum_{i\in\Gamma} p_i^q \sim L^{-(q-1)D(q)}. \quad (1)$$

The set of exponents $(q-1)D(q)$ has been first introduced in the context of turbulence[5-7] and later to characterize the backbone of the percolating cluster in a random resistor network.[2,8] More recently they were used to characterize the GSPD, first by Halsey, Meakin, and Procaccia[9] and independently by Meakin *et al.*[10,11]

Let us rewrite (1) as

$$Z(q)=\int n(p)p^q\,d\ln p=\int e^{F(p,q)}\,d\ln p, \quad (2)$$

where $n(p)\,d\ln p$ is the number of sites characterized by a probability p such that $\ln p$ is in the interval $[\ln p, \ln p+d\ln p]$ and $F(p,q)=\ln n(p)+q\ln p$. Following the approach of Ref. 4 we evaluate the integral in (2) by the steepest-descent method.

If p^* is the value for which $F(p,q)$ has a maximum we have

$$\partial \ln n(p)/\partial \ln p|_{p=p^*}=-q. \quad (3)$$

For each value of q there is a corresponding value of $p^*=p^*(q)$. We can write the following scaling *Ansatz*:[4]

$$p^*\sim A(q)L^{-\alpha(q)}, \quad (4)$$

$$n(p^*)\sim B(q)L^{f(q)}. \quad (5)$$

Therefore

$$Z(q)=e^{F(p^*,q)}\sim L^{-[q\alpha(q)-f(q)]} \quad (6)$$

and from (1)

$$(q-1)D(q)=q\alpha(q)-f(q). \quad (7)$$

Since p^* takes all the values as q varies from $-\infty$ to $+\infty$ we can consider p^* as an independent variable, that from now on we will call p. With use of $\partial/\partial p=(\partial q/\partial p)\partial/\partial q$ from (3) it follows that

$$\partial f/\partial q=q\,\partial\alpha/\partial q \quad (8)$$

and from (7)

$$\partial/\partial q (q-1) D(q) = \alpha(q). \tag{9}$$

Therefore knowing $D(q)$ one can calculate $\alpha(q)$ from (9) and $f(q)$ from (7). The results (7)–(9) were first obtained by Halsey *et al.*[4] Our approach, however, is slightly different from theirs. In particular our definition of $D(q)$ which follows from (1) differs from the one in Ref. 4, based on the method of box counting. Since as $q \to \infty$ only the sites with highest probability of growing contribute to $Z(q)$, from (4) $p_{\max} \sim L^{-\alpha(\infty)}$.

If we define

$$x = \ln p / \ln p_{\max}, \tag{10}$$

from (4) for large L, $x = \alpha(q)/\alpha(\infty)$. Therefore for each value of the ratio x there is a corresponding value $q(x)$. From (5) we have

$$n(p) = C(x) L^{\phi(x)}, \tag{11}$$

where $\phi(x) = f(q(x))$ is the exponent and $C(x) = B(q(x))$ is the amplitude, both depending on x. Taking into account that $p_{\max} \sim L^{-\alpha(\infty)}$, (11) can also be written

$$n(p) = C(x) p^{-\phi_1(x)}, \tag{12}$$

where $\phi_1(x) = \phi(x)/x\alpha(\infty)$.

Note that the total set of sites in the aggregate has been partitioned into many fractal subsets, each characterized by a value $x = \ln p/\ln p_{\max}$. Each subset has its own fractal dimensionality given by $\phi(x)$ and is characterized by a singularity $\alpha = x\alpha(\infty)$ which describes the way the value p associated with each subset goes to zero. The partition of $(q-1)D(q)$ into a density of singularities $f(q)$ with singularity strength $\alpha(q)$ was first introduced in the context of DLA by Halsey, Meakin, and Procaccia.[9] The scaling (12) has also been found for the random resistor network[3] and for DLA.[12] For an earlier account of this scaling see Coniglio.[13]

The scaling form (11) and (12) for the GSPD is characterized by a power law where the exponent is function of the ratio $\ln p/\ln p_{\max}$. This is completely different from the scaling laws in ordinary critical phenomena where the exponent is a constant. This peculiarity is a direct consequency of the fact that $f(q)$ and $\alpha(q)$ were assumed to be functions of q. Vice versa it is easy to show that a pure power law with constant exponents, $n(p) \sim L^y p^{-\tau}$, leads to α and f equal to two constants α_1 and f_1 above some value $q_c = \tau$, and two different constants α_2 and f_2 below q_c; in fact in such a case, for $q > q_c$ the moments of the distribution would be dominated by $p_{\max}$ and for $q < q_c$ by $p_{\min}$. On the contrary, in the scaling law (12) there is no characteristic value p that dominates a range of moments of the distribution, and for each moment q there is a corresponding value of p which dominates. In conclusion, then, the fact that $(q-1)D(q)$ is not a linear function of q and α and f are not constant implies the new scaling law (12).

The GSPD for DLA has been measured recently[9,10] in a computer simulation where the experimental probabilities of growth have been determined. The set of exponents computed for the moments $q = 2, \ldots, 8$ seemed to satisfy a linear dependence in q, although a slight deviation from it seemed also to be plausible, and a different dependence was not excluded.[9,10]

A crucial test to give a net answer would consist in computing low and negative moments. This is not very easy to accomplish with the computer simulations of Refs. 9 and 10 because of the fact that sites with small probability of being hit, which contribute to small moments and are determinant in negative moments, are very difficult to probe.

To calculate the GSPD we have used an analytical approach. Using the electrostatic formulation of Niemeier, Pietronero, and Wiesman[14] in the continuum version, the probability density $p(x)$ at site x on the aggregate is given by

$$p(x) = K[\mathbf{n}_x \cdot \nabla_x \Phi(x)]^\eta, \tag{13}$$

where $\mathbf{n}_x$ is the normal at that point and $\Phi(x)$ is the electrostatic potential satisfying the Laplace equation $\nabla^2\Phi = 0$ with the appropriate boundary conditions. K is the normalization constant; η is a parameter which describes different models: $\eta = 1$ corresponds to DLA, $\eta = 0$ gives the Eden model where the growth probabilities are all identical if different from zero, and $\eta = \infty$ produces one-dimensional aggregates.

To evaluate (13) we calculate the charge density on the aggregate $\sigma(x) = \mathbf{n}_x \cdot \nabla_x \Phi(x)$. Namely,

$$\Phi(x) = \int_S \sigma(x') G(x,x')\, da', \tag{14a}$$

where $G(x,x')$ is the Green's function

$$G(x,x') = \begin{cases} 1/|x-x'|^{d-2}, & d>2, \\ \ln|x-x'|, & d=2. \end{cases} \tag{14b}$$

From (14a)

$$\sigma(x) = \int_S \sigma(x') \mathbf{n}_x \cdot \nabla_z G(x,x')\, da'. \tag{15}$$

Equation (15) has the great advantage that it allows one to calculate $\sigma(x)$ without explicitly solving the Laplace equation in all the space outside the aggregate, avoiding the complication of introducing a circle at a finite distance where the potential is taken equal to zero.[14] As a result of this electrostatic analogy the GSPD is related via (13) to the voltage drop on the surface of the aggregate. For this reason we expect the GSPD to have the same properties as the voltage distribution in a random resistor or random supercon-

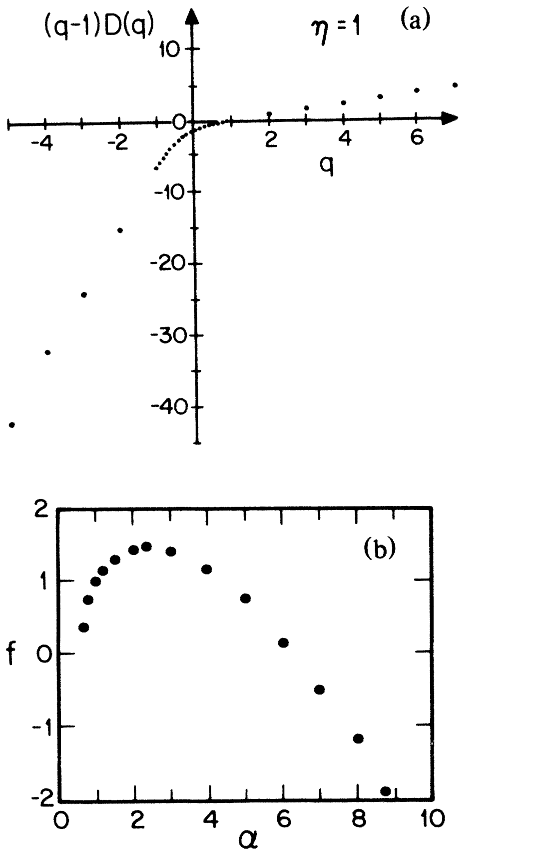

FIG. 1. (a) Exponents of the moments q of the GSPD vs q; (b) fractal dimensionality f vs fractal singularity α.

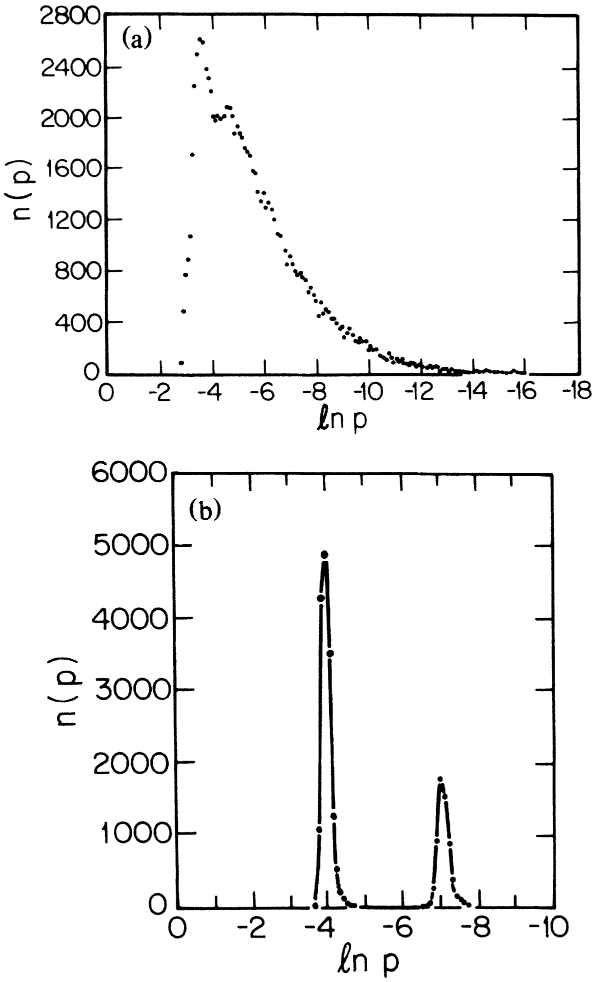

FIG. 2. GSPD for 1000 realizations of $N = 100$-particle clusters plotted vs $\ln p$: (a) $\eta = 1$ (DLA); (b) $\eta = 0.1$. The second peak starts to appear for $\eta = 0.7$ at the end of the tail. It does not develop from the shoulder.

ducting network.[2] In particular we expect a hierarchy of exponents $(q-1)D(q)$ with a departure from the linear dependence in q.

We have solved numerically a discretized version of Eq. (15), obtained by using standard techniques and the Green's function for the square lattice as developed by Morita.[15] A similar method to obtain a set of discrete equations for the GSPD was independently developed by Turkevich and Sher.[16]

The knowledge of the p_i at a given step allows us to grow the aggregate at the next step by using a random-number generator. We have generated clusters up to $N = 150$. At the same time we are able to calculate the fractal dimensionality d_f, the moments $Z(q)$, and the relative exponents $(q-1)D(q)$. For $\eta = 1$, we find $d_f \simeq 1.71$ and $\alpha(\infty) \simeq 0.7$ in good agreement with the off-lattice result and the theoretical prediction $\alpha(\infty) = d_f - 1$[16,17]; the exponents $(q-1)D(q)$ are plotted in Fig. 1(a). For $q \geqslant 2$ our results are in complete agreement with the existing data[9,10] and with the theoretical prediction $D(1) = 1$[9,18]; a strong deviation from the linear behavior is largely evident for values of q near zero. As a consequence we find that $f(q(\alpha))$ is a convex function whose shape is similar to the ones found for other systems[3,4] [Fig. 1(b)].

Note that $\lim_{q\to 0} D(q) \sim 1.5$ is slightly different from our independent result $d_f \simeq 1.71$. This finding seems to indicate that in DLA the external part of the perimeter where the field is neither zero nor exponentially decreasing with L scales with an exponent $D(0^+) < d_f$.

We can make the same analysis for other values of η. In the limiting case $\eta \to 0$ we expect the aggregate to coincide with the Eden model and to have a fractal dimensionality $d_f = 2$. Since all the probabilities on the surface $\Gamma \sim L^{d-1}$ are equal, $p \sim L^{-(d-1)}$ and $n(p) \sim L^{d-1}$, implying $\alpha(q) = f(q) = D(q) = d-1$.

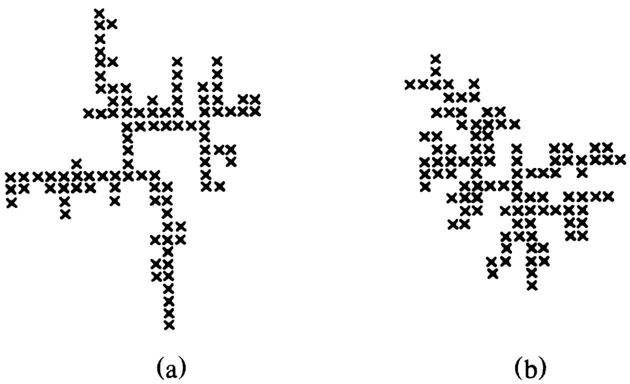

FIG. 3. Two aggregates for (a) $\eta=1$ and (b) $\eta=0.1$. The presence of deep smooth crevices for $\eta=0.1$ gives rise to the distribution of Fig. 2(b).

For $\eta=0.01$ our results are already consistent with this expectation.

In the opposite limit $\eta\rightarrow\infty$ the cluster grows along a straight line $d_f=1$. Moreover, the only growth sites are the ones at the two extremes and each of them grows with probability $p\sim 0.5$. Thus for any q we expect $\alpha(q)=f(q)=D(q)=0$. In fact, for $\eta=10$ and $q>0.2$ this is our result. But, although for small size we always find that the aggregate grows along a straight line, the fact that $D(q)$ deviates from zero for small q shows that eventually there will be a ramification, since the probabilities of occupying a side site is not rigorously zero.

The shape of $\eta(p)$ has very interesting features. For $\eta=1$ we find a pronounced maximum with a small shoulder at a lower value of p and a very long tail, which resembles closely the shape of the voltage distribution in a random resistor network.[2,3] As η becomes smaller a new peak arises in the far end of the tail and develops into a sharp maximum distinctly separated from the main one (Fig. 2). For extremely small values of η the two peaks merge into a single peak. This intriguing behavior has the following interpretation. For $\eta=0$ all the probabilities coincide (Eden model). As η increases from zero smooth crevices start to develop. Inside the crevice the growth probability is very small and very distinct from the one on the tips which have more the shape of fingers (Fig. 3). We believe that this unusual distribution may possibly arise in other physical systems such as viscous fingers[19] with surface tension different from zero (the surface tension playing the role of $1/\eta$).

In summary, we have used the standard Green's-function technique to calculate growth probability distributions in kinetic aggregation processes. We have found strong evidence that in DLA and related models continuously depending on a parameter η the exponents $\alpha(q)$ and $f(q)$ are not constant in q. This implies a new type of scaling for the GSPD different from a pure power law. We also find the interesting result that the GSPD develops two well separated peaks for small values of η. We expect this feature to be relevant also for systems which develop viscous fingers.

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