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Statistical Theory of Diffusion-Limited Growth in Two Dimensions

Yoshinori Hayakawa

Research Institute of Electrical Communication, Tohoku University, Sendai 980-77, Japan

Shinichi Sato

Department of Physics, Faculty of Science, Shizuoka University, Shizuoka 422, Japan

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A formalism of irreversible aggregation processes is presented in terms of statistical mechanics. Thermodynamical variables that include the degeneracy of the histories, which we call "history entropy," are introduced by taking into account all possible growth histories. By considering the thermodynamic properties of the growth history and harmonic measure, we find the condition of the most probable history for diffusion-limited aggregation (DLA) as a function of mass fractal dimension of the resulting clusters. Families of fractal growth generators are utilized for the estimation of the history entropy, and the fractal dimension of observable DLA clusters in two dimensions is evaluated in accordance with numerical and experimental results. [S0031-9007(97)03509-6]

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In the past couple of decades, diffusion-limited aggregation (DLA) [1] has been recognized as one of the most plausible models for many varieties of nonequilibrium growth phenomena including crystal growth, electrochemical deposition, colloidal aggregation, fingering of viscous fluid, and so on [2]. By intensive experimental studies relating to the DLA model, it has been known that there is a certain class of growth phenomena in nature which is often referred to as Laplacian fractals. In order to characterize such phenomena, several statistical properties, such as fractal geometry of obtained clusters, scaling properties of growth probability distribution on cluster in terms of multifractal measure, have been investigated.

In studies of irreversible growth like DLA, one of the most essential and remaining problems is the morphology selection mechanism during stochastic growth processes. For instance, an aggregation process of Brownian particles may happen to produce a sparse stringy or very dense cluster by chance, while it is known that DLA clusters have some definite fractal dimension which depends only on the dimension of space. How probable is the form we observe in computer simulations or experiments? What about the fluctuation of morphology during growth? To answer these questions, one would have to consider the ensemble of total growth histories, rather than observed clusters as a snapshot, which is the subject of statistical mechanics. To our knowledge, in this point of view a few works have been reported so far, e.g., renormalization group approaches [3,4], fixed-scale transformation theory [5], and general statistical mechanics treatment of far-equilibrium growth systems [6,7]. In this Letter, we introduce a thermodynamical formalism for stochastic growth problems, which is similar to that for chaotic dynamical systems, as recently studied by Elezgaray *et al.* [6]. Based on the formalism, we estimate the most probable fractal dimension of DLA clusters in two and

three dimensions by means of asymptotic analysis of the families of fractal generators.

At first we define the history probability for the irreversible growth processes of particles [6,7]. Let σ_n be a conformation of the cluster with n particles after $n - 1$ successive iterations of a growth rule starting from a seed σ_1 . A resulting cluster after $(m - 1)$ -step growth has a set of conformations $\{\sigma_1, \sigma_2 \cdots \sigma_m\}$, which we call growth history. We simply denote this history as $\{\sigma\}^m$. In general, the number of the possible histories resulting in a certain conformation σ_m would be tremendously large as m increases. However, one can still define the probability $P(\{\sigma\}^m)$ for the realization of a history $\{\sigma\}^m$.

For the growth process of DLA, it is known that the growth probability p_g^i at the i th growth site is scaled as $p_g^i \sim L^{-\alpha_i}$, where L is the characteristic length of the cluster and α the singularity exponent of growth probability [8]. Thus, if a DLA cluster maintains a fractal structure with mass fractal dimension D during the growth, i.e., $m \sim L^D$, relation $p_g^i \sim m^{-\alpha_i/D}$ should be held for large mass m . Therefore, the history probability of such growth can be represented with p_g as $P(\{\sigma\}^m) = \prod_{i=1}^{m-1} p_g^i \sim \prod_{i=1}^{m-1} m^{-\alpha_i/D}$ from its definition. More generally, if α_i/D is invariant during the growth, it would be plausible to assume that the history probability has an asymptotic scaling form on the mass m of cluster as [7]

$$P(\{\sigma\}^m) \sim \mu^{-m} (m!)^{-\gamma}, \quad (1)$$

where μ and γ are the characteristic variables of the history. Only the factorial dependency is significant in large m limits in the right-hand side (rhs) of Eq. (1). For the set of growth histories we can write a partition function with an inverse temperature q :

$$Z_q(m) = \sum_{\{\sigma_i\}} P(\{\sigma_i\}^m)^q. \quad (2)$$

Accordingly the generalized entropy of the q th order can be defined as

$$K_q = -\lim_{m \rightarrow \infty} \frac{1}{q-1} \frac{\ln Z_q(m)}{\ln m!}. \quad (3)$$

Following the thermodynamical formulation for multifractal measures by use of a steepest decent approximation [9–11] and combining the scaling ansatz in Eq. (1), one can introduce a pair of new variables γ and h :

$$\gamma(q) = \frac{d}{dq} \tau_q, \quad (4)$$

$$h(q) = q\gamma(q) - \tau_q, \quad (5)$$

where $\tau_q = (q-1)K_q$ and $dh/d\gamma = q$. The variable $h(\gamma)$ represents the degeneracy of the history indexed with γ ; that is, the number of history scales as $N_\gamma \sim (m!)^{h(\gamma)}$, whereas for the history probability $P_\gamma \sim (m!)^{-\gamma}$. Therefore $\nu = h(\gamma) - \gamma$ gives the escape rate of the growth path having γ , and $\nu \leq 0$ has to be always held. The growth path to be realized, i.e., the most probable history, satisfies $\nu = 0$.

Direct calculation of $\gamma(q)$ and $h(q)$ from the history probability distribution would be almost impossible due to the complexity and diverging number of histories, though the definitions are clear. Here we return to the meaning of the history probability in the case of DLA. The growth probability distribution on the surface of DLA clusters can be described with the harmonic measure [8]. It has been mathematically proven that the information dimension D_1 for the harmonic measure in two-dimensional space is exactly 1, independent of the shape of connected objects [12]. The information dimension D_1 can be interpreted as the average value of growth probability singularity over the perimeter sites, and “active” growth sites sit on an invariant set whose fractal dimension is unity. Starting from a cluster $\sigma_m(D)$ of mass fractal dimension D , therefore, the growth probability p_g at the active sites decays as $p_g \sim L^{-1}$, where L is the size of the cluster. Conditional probability for one-step growth then follows $P(\sigma_{m+1}(D) | \sigma_m(D)) \sim m^{-1/D}$. From the definition of γ , for Laplacian fractal growth in two dimensions, we can conclude that the value of observable γ^* is related to the fractal dimension D^* for the corresponding realization of cluster as $\gamma^* = 1/D^*$.

The observable history of fractal growth in $m \rightarrow \infty$, therefore, has to satisfy

$$\nu(D^*) = h(D^*) - 1/D^* = 0 \quad (6)$$

for diffusion-limited growth in two-dimensional space [7]. From this relation, one can determine the fractal dimension for the most probable history, if the dependency of h on D can be calculated.

In order to evaluate the history entropy as a function of D , it might be helpful to consider a fractal growth which is represented with simple generators. First we show one example of the evaluation of $h(D)$. As the

fractal generator, let us choose a set of five cells in which one is at the center and the others surrounding it as shown in Fig. 1(a). We select one of the four outer cells as the seed (gray circle in the figure) from which growth starts. After the seed cell is occupied, the central cell connected to the seed can grow next, and then the remaining three cells are the subject for the successive growing process. Now we regard these cells as the coarse grained parts of a fractal structure in a sense of position-space renormalization group (PSRG) methods; that is, each cell recursively contains the same structure as the generator itself [Fig. 1(b)]. It is clear that the resulting fractal object under this generating rule has a simply connected structure of fractal dimension $D = \ln 5 / \ln 3$. How can the entropy for such growth process? One should note that at any resolution of space, the seed cell numbered 1 in Fig. 1(a) has to be always the first cell to be occupied, and the central cell [2 in Fig. 1(a)] is the second. After that, there are three choices of successive growth. Let us consider the number of possible paths $n(m)$ under this construction rule with the cell which contains m “elementary” particles. Because one can choose a growth site out of these three cells which have totally $3m$ particles inside, it is easily found that $n(m)$ satisfies the following relation between the successive levels of the pattern generation for large m :

$$n(5m) = n(m)^5 \frac{(3m)!}{(m!)^3}. \quad (7)$$

The fractional part in the rhs represents the degree of freedom at branching, which causes a nontrivial contribution to growth entropy.

If one uses a family of similar generators with branches consisting of k cells each as shown in Fig. 1(c), the number $n_k(m)$ of growth paths will satisfy the following equation as well:

$$n_k[(4k+1)m] = [n_k(m)]^{(4k+1)} \frac{(3km)!}{\{(km)!\}^3}. \quad (8)$$

In Fig. 1, solid bars represent the adjustable “linear parts” of generators in which the number of cells varies

FIG. 1. Simple generators of fractal growth process. (a) A simple generator consisting of a five unit cell of fractal generation (circles): Gray circle represents the starting cell of growth. A simply connected fractal object ($D = \ln 5 / \ln 3$) is constructed by the recursive procedure explained in text. (b) is the next recursive step of (a). (c) An extended generator with which fractal dimension of generated structure is adjustable by changing the number k of cells in branches (bars). Growth always begins at cells in the gray colored bar. (d) Another example of adjustable generator with six branches.

according to k , circle is a unit cell. Branching from a circle takes place only when the growth inside the circle has been completed so as to represent an irreversible sticking process of particles. For any integer $k > 0$, patterns constructed with this generating rule hold an open and simply connected fractal structure of fractal dimension

$$D_k = \frac{\ln(4k + 1)}{\ln(2k + 1)}. \quad (9)$$

Using Stirling's formula, one can easily obtain the asymptotic form of $n_k(m)$ as

$$n_k(m) \sim C^m (m!)^{h_k}, \quad (10)$$

$$h_k = \frac{3k \ln 3}{(4k + 1) \ln(4k + 1)} + O\left(\frac{\ln m}{m}\right), \quad (11)$$

where C is a constant. In the large m limit, the number of available paths has a power of factorial dependency on m , which is consistent with the scaling assumption in Eq. (1).

Here we have an implicit relation between the history entropy h_k and the fractal dimension of cluster D_k for one representation of the fractal growth process. Next we estimate the entropy h whose value is equal to the inverse of the dimension of generating fractal. To do that we shall extend the domain of k into any positive real number, although the yielding structure might be incomplete for fractional k . By putting $h_k = 1/D_k$, one can solve this nonlinear equation numerically, e.g., by the Newton method, and obtain one unique solution $k^* = 0.262$ and $D_{k^*} = 1.701$. This estimation of the most probable fractal dimension gives a nice agreement with that of realized clusters obtained by large scale off-lattice computer simulations [13,14]. In Fig. 2 dependency of h_k on D_k is shown for this four-branch generator.

FIG. 2. Estimated growth entropy h for the generator families shown in Fig. 1(c) (black circles) and Fig. 1(d) (white circles) as a function of fractal dimension D . Solid line shows the dependency of γ on D in the case of two-dimensional DLA growth. Dotted and dashed lines are the extrapolation curves by adjusting parameter k , and the crossing point of those curves gives an estimated value of fractal dimension.

By using a similar generator with five arms shown in Fig. 1(d), we obtain the estimate as $D_{k^*} = 1.726$ at $k^* = 0.677$, which is also in nice agreement with known results (see Fig. 2). One should note that the known fractal dimension of 2D-DLA clusters, which approaches to around 1.715 [14], is larger than D_{k^*} for simple four-branch generators [Fig. 1(c)], and smaller than that for five-branch ones [Fig. 1(d)]. This would correspond to the fact that a number of main branches of DLA clusters are between 4 and 5, where “main branches” could be regarded as the branches observed at the most coarse grained resolution of view.

The families of generators chosen here are only special cases among all possible Laplacian growth having the same fractal dimension. The number of total history $N_\gamma(m)$ for given γ and m is evaluated as

$$N_\gamma(m) \sim \sum_{i \in G} n_\gamma^i(m),$$

where G denotes the set of every pattern generator and $n_\gamma^i(m)$ the number of histories for generator i . Fixing the number m of particle, the number of possible generators should be less than that of lattice animal $N_a(m)$ with m occupied sites. Therefore the inequality

$$N_\gamma(m) < N_a(m) \max_{i \in G} n_\gamma^i(m)$$

should be held. It is known that the number of animals has an exponential dependency on m , i.e., $N_a(m) \sim \lambda^m$; thus the dominant contribution to N_γ will be made by the maximum n_γ^i having $(m!)^h$ dependency on m .

In order to evaluate the value of entropy properly, therefore, one has to find out the generator which gives the maximum entropy at a given fractal dimension. In general, in the processes of particle aggregation, multiply connected clusters may happen to be generated. However, as far as DLA-like growth processes, due to the screening effects of Brownian particles that prevent the clusters from making loops at every length scale greater than the particle size, it is enough to consider simply connected structures as the ensemble of statistics, at least, in the discussion of observable histories. Since the growth entropy is brought by the redundancy at branching processes, the more symmetric branching structure is, the better the choice of generator will be. In this point of view, examples of generator family in Fig. 1 might be very simple, while the derivation of fractal dimension seems to be successful. Under the scheme presented here, evaluation of entropy would not be very sensitive to the choice of generator families.

In Fig. 3 we illustrate a few more complex examples of generator families. Every generator has no cell at one of the corners of its hexagonal outline so that the condition for simply connected structure is preserved when $k \geq 1$. For large size generators, it becomes difficult to check if the resulting patterns have no loop and they

FIG. 3. A few examples of generators that still result in simply connected fractal patterns. A circle represents the generator itself and a bar a set of k cells. Examples of (a) 17 cells and 16 adjustable bars (b) 45 cells, 44 bars, and (c) 78 cells, 78 bars are illustrated, respectively. The cell at the lower right-hand side of its hexagonal outline should not be present in the generators so as to prevent reconnection of cells during growth.

are connected, and it is also a hard task to find out the growth history that maximizes h because of the increasing number of possible growth paths inside the generators. Furthermore, when the degeneracy of the growth history is not large enough for a chosen generator, we may not obtain the solution of the balancing equation $h_k = 1/D_k$ within $k > 0$. We summarize estimated values of fractal dimension in Table I. It seems clear that the larger number of cells gives a better estimation of fractal dimension. More systematic calculations and analysis of asymptotic behavior by means of some other extrapolation methods will be needed in our future study.

In the three- or higher-dimensional case, no exact relation between D_1 and the form of objects has been known mathematically. By dimensional analysis, Matsushita *et al.* claim that the generalized dimension of growth probability distribution on a d -dimensional DLA surface would obey $D_1(d) = d - 1$ [15]. Assuming that $D_1(3) = 2$ here, i.e., $h(D^*) = 2/D^*$, one can estimate D^* for three-dimensional DLA as well. When a three-dimensional generator family with one “ball” and six adjustable arms attached to the ball is used, $D_{k^*} = 2.266$ is obtained at $k^* = 0.245$. For one ball and twelve arms at nearest neighbor locations of a closest packed structure in three dimensions, $D_{k^*} = 2.423$ at $k^* = 0.862$, which is close to numerical results $D \approx 2.5$ [13]. The evaluation of D^* seems to also work even in three dimensions, if one assumes $D_1(3) = 2$. Practically it would be, however, difficult to construct larger size three-dimensional generator families of simply connected fractals.

In conclusion, by taking into consideration the history of irreversible growth, we present a thermodynamical formalism and define some thermodynamical quantities which characterize the growth process. For DLA, two-

TABLE I. Estimation of fractal dimension by means of families of fractal growth generators. Generators are categorized with the pair of number of nodes (cells) and links (group adjustable cells). Equation $h_k = 1/D_k$ is satisfied at k^* .

Generator (nodes, links)	Fractal dimension	
	k^*	D_{k^*}
(1,4) [Fig. 1(c)]	0.262	1.701
(1,5) [Fig. 1(d)]	0.677	1.726
(17,16) [Fig. 3(a)]	0.104	1.732
(45,44) [Fig. 3(b)]	0.0998	1.722
(78,78) [Fig. 3(c)]	0.513	1.715

dimensional space has a special property because the active growth surface has always the same fractal dimension ($= 1$) with no regard to the shape of objects. Based on this knowledge, we show the relation between the degree of degeneracy in branching processes h and the geometrical property, i.e., the fractal dimension D^* , for the observable DLA histories. We present a PSRG-like approach to evaluate the history entropy for fractal growth by considering families of fractal generators, and estimate the fractal dimension of the observable DLA clusters, which is in good agreement with the known results of large scale computer simulations.

- [1] T. A. Witten and L. M. Sander, Phys. Rev. Lett. **47**, 1400 (1981).
- [2] T. Vicsek, *Fractal Growth Phenomena* (World Scientific, Singapore, 1992), 2nd ed.
- [3] T. Nagatani, J. Phys. A **20**, L381 (1987).
- [4] P. W. Barker and R. C. Ball, Phys. Rev. A **42**, 6289 (1990).
- [5] L. Pietronero, A. Erzan, and C. Evertz, Phys. Rev. Lett. **61**, 861 (1988).
- [6] J. Elezgaray, J. E. Muzy, F. Argoul, and A. Arneodo, Phys. Rev. Lett. **71**, 2425 (1993).
- [7] S. Sato and Y. Hayakawa, Phys. Rev. E (to be published).
- [8] T. C. Halsey, P. Meakin, and I. Procaccia, Phys. Rev. Lett. **56**, 854 (1986).
- [9] T. C. Halsey, M. H. Jansen, L. P. Kadanoff, I. Procaccia, and B. I. Shraiman, Phys. Rev. A **33**, 1141 (1986).
- [10] M. Sano, S. Sato, and Y. Sawada, Prog. Theor. Phys. **76**, 945 (1986).
- [11] J. P. Eckmann and I. Procaccia, Phys. Rev. A **34**, 659 (1986).
- [12] N. G. Makarov, Proc. London Math. Soc. **51**, 369 (1985).
- [13] S. Tolman and P. Meakin, Phys. Rev. A **40**, 428 (1989).
- [14] P. Ossadnik, Physica (Amsterdam) **176A**, 454 (1991).
- [15] M. Matsushita, Y. Hayakawa, S. Sato, and K. Honda, Phys. Rev. Lett. **59**, 86 (1987).