Active Zone of Growing Clusters: Diffusion-Limited Aggregation and the Eden Model

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Growth is described in terms of an active zone defined as the region where new particles join the existing cluster. This zone is characterized by the probability $P(r,N)\,dr$ that the Nth particle is deposited a distance r from the center of mass. Our Monte Carlo simulations for two-dimensional diffusion-limited aggregation and the Eden process show that, for large N, $P(r,N)=[(2\pi)^{1/2}\xi_N]^{-1}\exp[-(r-\overline{r}_N)^2/2\xi_N^2]$, where $\overline{r}_N \sim N^{\nu}$ and $\xi_N \sim N^{\overline{\nu}}$. We find $\overline{\nu}<\nu$ indicating the presence of two distinctly diverging lengths in these processes.

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In recent years considerable effort has gone into the study of clusters grown by various processes. The reason for this interest is twofold. First, a wide variety of nonequilibrium phenomena such as the aggregation of smoke particles, electric breakdown, the early stages of nucleation and tumor growth, polymerization, etc., can be described[1] in terms of simple models in which a single cluster grows through the addition of individual particles. Second, some of these growth processes, such as diffusion-limited aggregation[2] (DLA), lead to structures which are fascinating in their own right since they are scale invariant and have a fractal dimension (D) different from the Euclidean dimension (d) of the space in which they are grown. To date most theoretical work has focused on the calculation of the fractal dimension.[3-8] Although numerical methods[9-12] have yielded well converged values of D for several models, no systematic analytic method of calculating this quantity has emerged.

One of the obstacles to theoretical progress is the lack of understanding of the surface structure of the growing clusters. Observing growth in various circumstances, one can easily recognize a characteristic common to all growth processes, namely, that there exists an "active" region, usually the outer part of the surface, which collects practically all the new particles. This active zone moves outward and leaves behind a frozen structure. Since the aim is to derive the properties of the frozen bulk from the dynamical equations describing the process and since the bulk is built in the active zone, clearly, effort should be concentrated on characterizing and understanding this region of the cluster. Here, we report on an attempt to characterize the growing interface in DLA and in the Eden process.[13] In particular, we have carried out extensive Monte Carlo simulations to calculate the probability $P(r,N)\,dr$ that the Nth particle is deposited within a shell of width dr at a distance r from the center of mass of the $(N-1)$-particle cluster. Figure 1 shows $P(r,N)$ for DLA in $d=2$ for selected values of N. The solid lines are Gaussian fits of the form

$$P(r,N)=\frac{1}{(2\pi)^{1/2}\xi_N}\exp\left[-\frac{(r-\overline{r}_N)^2}{2\xi_N^2}\right], \quad (1)$$

and we see that the fit to the simulations is quite remarkable. From this we conclude that the active

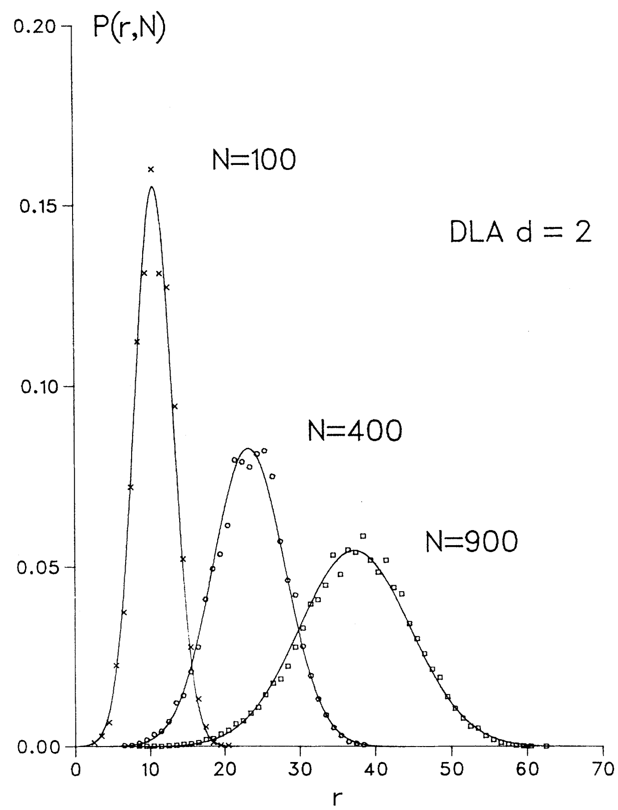

FIG. 1. Probability of the Nth particle being attached at a distance r from the center of mass of the clusters in DLA. The solid lines are Gaussian fits to the averages over 200 clusters.

zone of the cluster can be characterized by two parameters, the mean deposition radius $\bar{r}_N$ and the width of the active zone ξ_N. It should be noted that $\bar{r}_N$ and ξ_N can also be defined without any reference to the actual form of $P(r,N)$:

$$\bar{r}_N = \frac{1}{M}\sum_{i=1}^{M} r_N(i), \quad \xi_N^2 = \overline{(r_N - \bar{r}_N)^2}, \tag{2}$$

where $r_N(i)$ is the deposition radius of the Nth particle in the ith cluster. We have obtained the same values of $\bar{r}_N$ and ξ_N for $N \gtrsim 100$ whether fitting a Gaussian form or using Eqs. (2) and hence the following analysis of the scaling properties of $\bar{r}_N$ and ξ_N is not biased by the Gaussian form (1) of $P(r,N)$.

The double-logarithmic plot of $\bar{r}_N$ and ξ_N, calculated by averaging over 4000 clusters, is displayed in Fig. 2. The simulation points fall on straight lines at least over the range $200 < N < 2500$ indicating that the functional form is

$$\bar{r}_N \approx r_0 N^{\nu}, \quad \xi_N \approx \xi_0 N^{\bar{\nu}}. \tag{3}$$

A least-squares fit of the data by these functions produces the estimates $\nu = 0.584 \pm 0.02$ and $\bar{\nu} = 0.48 \pm 0.01$.

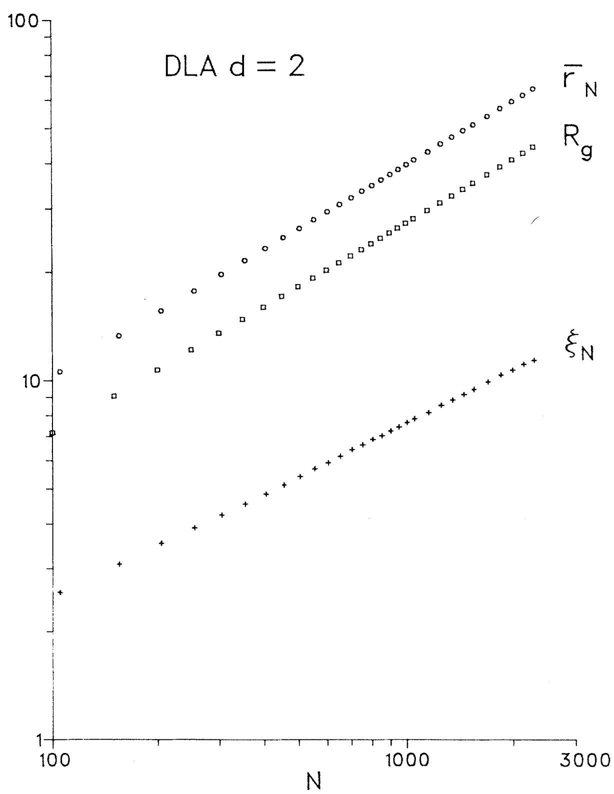

FIG. 2. The mean deposition radius ($\bar{r}_N$) of the Nth particle, the width of the active zone ξ_N, and the radius of gyration $R_g(N)$ for DLA.

Since $\bar{r}_N$ describes the cluster expansion as the particles are added, one expects that ν is related to the fractal dimension D of the frozen structure. This expectation is confirmed by also plotting the radius of gyration $R_g(N) \sim N^{1/D}$ in Fig. 2. The curves $\bar{r}_N$ and $R_g(N)$ are parallel and we have $\nu = 1/D$. This result can easily be obtained by calculating the density of particles at a distance r from the center of mass

$$\rho(r,N) = \frac{1}{S_d r^{d-1}} \int_0^N dN' \, P(r,N'), \tag{4}$$

where S_d is the surface area of a d-dimensional unit sphere. In the limit $N \to \infty$ and r large, one can evaluate the integral (4) by the method of steepest descent if the functional forms (3) are used. The result is $\rho(r, N=\infty) \sim r^{-d+1/\nu}$ yielding the expected relationship $\nu = 1/D$. Note also that requiring the density to be finite in the large-N yields $\bar{\nu} \leq \nu$. Our simulation results support the *inequality,* i.e., the growing clusters are characterized by two distinct lengths $\bar{r}_N$ and ξ_N which scale differently.

The actual value of $\bar{\nu}$ is of importance since at present there are several conjectures concerning the interface of DLA clusters. Mean-field–type theories[3,4,6] of calculating D begin with an assumption about the surface: the screening length which can be identified with ξ_N is assumed to scale as ρ^{-1} or $\rho^{-1/2}$ implying $\bar{\nu} = \nu d - 1$ or $\bar{\nu} = (\nu d - 1)/2$. In $d=2$ this gives $\bar{\nu} \approx 0.18$ or $\bar{\nu} \approx 0.09$ in contrast to our result $\bar{\nu} \approx 0.48$. Recently, Sander[14] suggested that there is only one diverging length scale in DLA clusters which would mean $\xi_N \sim \bar{r}_N$. Our results seem to exclude the possibility $\bar{\nu} = \nu$ although $\nu - \bar{\nu} \approx 0.1$ is small and one must be cautious about small differences obtained in Monte Carlo simulations. Note, e.g., that requiring $P(r,N)$ to satisfy Eq. (1) could introduce a bias against finding $\bar{\nu} = \nu$. Our analysis based on Eq. (2), however, does not involve $P(r,N)$ and is free from such bias.

We have also studied the two-dimensional Eden model from the same point of view. Once again the formula (1) is found to fit the probability $P(r,N)$ very accurately. The functions $\bar{r}_N$, ξ_N, and $R_g(N)$ calculated from averages over 4000 clusters are plotted in Fig. 3, and as in the case of DLA we find $\bar{r}_N \sim N^{\nu}$ and $\xi_N \sim N^{\bar{\nu}}$ with $\nu = 0.495 \pm 0.005$ and $\bar{\nu} = 0.18 \pm 0.03$.

The Eden model is thought to be a space-filling process in $d=2$ and one can calculate $\bar{r}_N$ and $R_g(N)$ under this assumption. One obtains in the large-N limit $\bar{r}_N = (N/\pi)^{1/2}$ and $R_g(N) = (N/2\pi)^{1/2}$. These functions are also displayed in Fig. 3

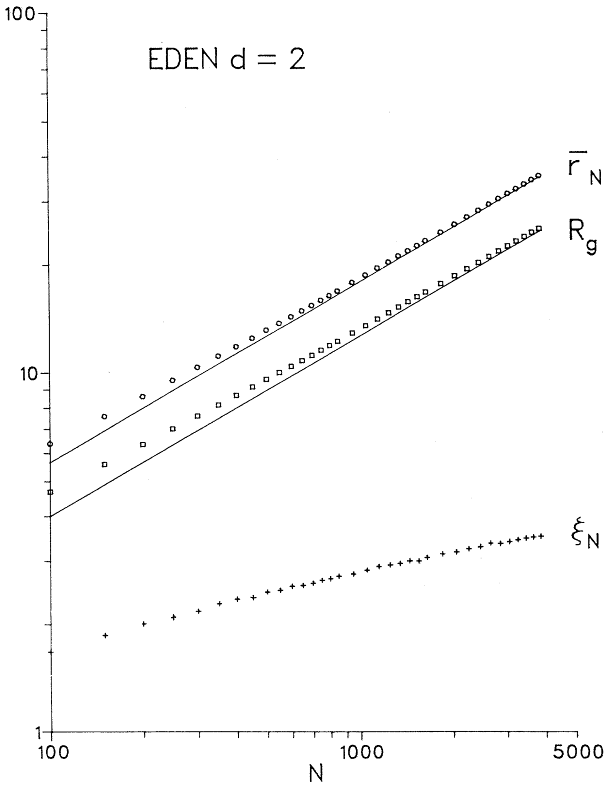

FIG. 3. The same as Fig. 2 but for the Eden clusters. The two solid lines are $(N/\pi)^{1/2}$ and $(N/2\pi)^{1/2}$, respectively.

and we see that the data points do approach these curves for large N. As in the case of DLA our results indicate that the growth process is governed by two exponents ν and $\bar{\nu}$ rather than one as has been generally assumed. In the Eden model the statistical uncertainties are larger and it seems to be more difficult to reach the asymptotic regime. Since there is evidence of downward curving in the $\ln\xi_N$ vs $\ln N$ plot, we cannot rule out the possibility that $\bar{\nu}=0$. Indeed, the functional form $\xi_N = 0.14(\ln N)^{1.5} + 0.19$ fits the data as well as formula (2). Much larger simulations will be needed to resolve this equation.

We now comment on the relation of our work to previous Monte Carlo studies of surfaces of growing clusters.

(a) Peters *et al.*[12] defined the width (d_N) of the surface layer for the Eden clusters through the moments of the derivative of the density profile $W(r,N) = -\partial\rho(r,N)/\partial r$. They studied clusters of size $N \leq 400$ and obtained $d_N \sim N^{0.5}$ whereas our results would indicate $d_N \sim \xi_N \sim N^{0.18}$. We have attempted to repeat their calculations and found that it is difficult to get convergence for d_N since the derivative $-\partial\rho/\partial r$ is frequently negative as a result of fluctuations in the regions where $\rho \approx 0$ or $\rho \approx 1$. If $W(r,N)$ is set equal to zero whenever this occurs (as it is done in Ref. 12), then one presumably obtains an artificial contribution to d_N which is of the order of $\bar{r}_N$ and $\bar{r}_N$ indeed grows as $N^{0.5}$. Actually, $W(r,N)$ can be expressed through the moments of $\rho(r,N)$ and then the above difficulty does not occur. In this way, stable estimates of d_N can be obtained and one finds that for large N, $d_N \sim \xi_N$ and the effective exponent of d_N is significantly less than 0.5 even in the range $N < 400$.

(b) For DLA, Meakin and Witten[15] have studied a quantity called the "interfacial mass." This quantity is defined as the number of new particles $N_i(N)$ in contact with an existing cluster of N particles when the growth process has been continued long enough that $N_i(N)$ has reached its asymptotic value. We do not see an obvious way of relating $N_i(N)$ and ξ_N. If $N_i(N)$ is assumed to be proportional to the number of particles in the surface layer (as is done in Ref. 15) then $N_i(N) \sim \rho(\bar{r}_N,N) \times \bar{r}_N^{d-1}\xi_N \sim N^{1+\nu+\bar{\nu}} \sim N^{0.9}$ for $d=2$ in contrast to the Monte Carlo result[15] $N_i(N) \sim N^{0.6}$.

(c) Meakin[16] has also carried out studies of deposits which are similar to aggregates except that the seed particle is replaced by a $(d-1)$-dimensional plane of nucleation sites. Deposits can be linked to aggregates by observing that (i) for large $\bar{r}_N$ the surface of an aggregate is indistinguishable from that of a deposit, and (ii) the scaling variable for aggregates is the number of particles in the cluster (N) while for the deposits it is the number of particles (N_1) deposited on a unit of area, and thus for large N, $N_1 \sim N/\bar{r}_N^{d-1}$.

On the basis of (i) and (ii) one can derive scaling laws connecting the exponents of deposits to those of aggregates.[17] These scaling laws can also be obtained on more general grounds[4] and seem to be satisfied within the accuracy limits of the Monte Carlo simulations.[16-18] For our purposes (i) means that the active zone of a deposit can be described by a Gaussian form, while (ii) implies that the average deposition distance from the plane of nucleation sites scales as

$$x(N_1) \sim N_1^{\nu_1} \sim \bar{r}_N \sim N^{\nu} \sim N_1^{1/(D-d+1)}, \quad (5)$$

while the width of the active region behaves as

$$\xi_1(N_1) \sim N_1^{\bar{\nu}_1} \sim \xi_N \sim N^{\bar{\nu}} \sim N_1^{\bar{\nu}D/(D-d+1)}. \quad (6)$$

Meakin[16] measured the average height $h(N_1)$ and the mean-square deviation $\langle[\Delta h(N_1)]^2\rangle$ of the upper surface for diffusion-limited deposits in $d=2$ and 3. These quantities are not exactly equal to

$x(N_1)$ and $\xi_1(N_1)$, but we believe that their scaling properties should be the same, i.e., $h(N_1) \sim x(N_1) \sim N_1^{\nu_1}$ and

$$\langle [\Delta h(N_1)]^2 \rangle^{1/2} \sim \xi_1(N_1) \sim N_1^{\bar{\nu}_1}.$$

Meakin's estimates ($\nu_1 = 1.45 \pm 0.05$ and $\bar{\nu}_1 = 1.32 \pm 0.08$ for $d=2$ and $\nu_1 = 2.1 \pm 0.1$ and $\bar{\nu}_1 \approx 1.8$ for $d=3$) indicate that if Eqs. (5) and (6) are correct then $\bar{\nu} < \nu$ for both $d=2$ and 3 in accord with our observation. Moreover, for $d=2$ Meakin's results yield $\nu = 0.59 \pm 0.03$ and $\bar{\nu} = 0.54 \pm 0.07$, which are in reasonable agreement with our results, especially if one takes into account that for deposits, the large corrections to scaling make it difficult to obtain accurate estimates of the exponents.

In summary, our results suggest that the presence of the interface in growing clusters leads to the existence of two distinct diverging lengths. We believe that this is a quite general feature of growing systems and should be studied in more detail.

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