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Progress in DLA research

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Abstract

The diffusion-limited aggregation (DLA) model has become an important paradigm for non-equilibrium pattern formation and provides a basis for understanding a broad range of natural processes. Progress in the simulation and quantitative characterization of DLA clusters is described. Despite advances in this direction, and extensive theoretical studies, the DLA process and the scaling structure of DLA clusters are still not well understood. Clusters generated by the DLA model have a fractal geometry but it is not certain whether the asymptotic structure is self-similar or must be described by a more complex scaling model. Lattice anisotropy has a strong influence on the asymptotic shape of DLA clusters and the asymptotic stucture of DLA clusters with anisotropy (and noise reduction) is relatively well understood.

1. Introduction

It is now well over a decade since the diffusion-limited aggregation (DLA) model was introduced by Witten and Sander [1]. The "Gran Finale" presents an appropriate occasion to review the progress that has been made towards understanding DLA and the problems that still face us.

In the DLA model particles are added, one at a time, to a growing cluster or aggregate of identical particles via random walk paths starting at "infinity". When a particle following a random walk or Brownian path contacts the growing cluster it is incorporated at the position in which it first contacts the cluster. Fig. 1 illustrates a simple square lattice version of the DLA model much like that of Witten and Sander [1]. Subsequent work showed that although the effects of lattice anisotropy are quite subtle for small square lattice DLA clusters these effects become more important as the cluster grows and eventually come to dominate the structure. In recent years attention has been focussed on off-lattice DLA models in which the added particles follow Brownian paths. This model is more appropriate for most natural processes in which DLA-like patterns emerge.

Fig. 2 shows 6 stages in the growth of a 10^7 particle off-lattice DLA cluster. The same cluster is displayed at the stages in which its size reached $10^2, 10^3, 10^4, 10^5, 10^6$ and 10^7 particles. This figure shows that the cluster has a similar shape at all stages of growth but subtle differences between the statistical properties of clusters of different sizes are indicated even by this simple display. This slow approach to statistical self-similarity is at the center of current DLA research. Fig. 3 shows a three-dimensional 5×10^6 particle off-lattice DLA cluster (to my knowledge this is the largest three-dimensional DLA cluster that has, so far, been generated). Clusters have been grown in embedding spaces with dimensionalities d up to $d = 8$ [2] but most work has been carried out for

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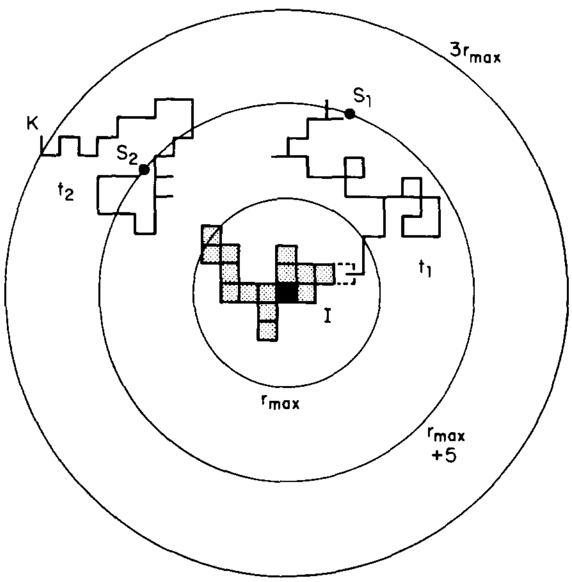

Fig. 1. A simple square lattice DLA model. The initial seed or growth site is shown in black. Sites that have already been added to the growing cluster are shaded. Two typical random walk trajectories originating at random points on the launching circle with a radius of $r_{max} + 5$ lattice units are shown (r_{max} is the maximum cluster radius measured from the initial growth site). Trajectory t_1 starts from point S_1 and eventually reaches an unoccupied perimeter site (dashed borders) that is filled to represent the growth process. Trajectory t_2 started from point S_2 reaches the killing circle with a radius of $3r_{max}$, before reaching the unoccupied perimeter, and is terminated.

$d = 2$ with relative little work for $d = 3$ and almost none for $d \geq 4$.

The DLA model continues to be of considerable interest because it has become one of the most important paradigms for disorderly growth far from equilibrium and because it provides a basis for understanding an extraordinarily large range of natural pattern formation phenomena. Because of these attributes this model has been the subject of intense interest since its discovery. Despite considerable effort and a wide range of different approaches we still seem to be quite far from a satisfactory understanding of DLA (i.e. an understanding on the level that we now have for simple equilibrium fractals such as percolation clusters or the self-avoiding random walk model for polymers).

2. Computer simulation

The first square lattice DLA clusters generated by Witten and Sander [1] contained less than 3000 particles. Since that time quite dramatic progress has been made and clusters of 10^7 particles such as that shown in Fig. 2 are not exceptional. The generation of large DLA clusters is based on the idea that if the randomly walking particle is far from the cluster it can take a long step provided that the step is not large enough for it to actually reach the cluster. If the random walker can be surrounded by an empty region with a simple shape then its random walk path until it leaves that empty region can be replaced by a single step to the surface of the empty region. The most simple procedure is to use a hyperspherical region centered on the current position of the random walker. In this case the trajectory within the hypersphere can be replaced by a single jump to a randomly selected position on its surface. This requires an efficient procedure for determining the smallest distance between the random walker and the cluster [2,4,5]. Once this distance has been found the random walker is allowed to take a step in a randomly selected direction that is (at least) one particle diameter shorter than this minimum distance. The most efficient algorithms use a hierarchy of "maps" of the cluster to determine an upper bound for the smallest distance between the random walker and the cluster [2,5]. The computer time required grows only slightly faster than is the cluster size s (there are logarithmic "corrections"). By the time this article appears it is likely that clusters containing $\geq 10^8$ particles will have been generated using this approach. The algorithm of Ball and Brady [5] was the first to combine long steps for random walkers far from the growing cluster with an efficient way of storing the information needed to obtain a conservative estimate for the lengths of these steps. Essentially all of the progress since that time has come about as a result of the availability of large amounts of time on faster machines with larger "memories".

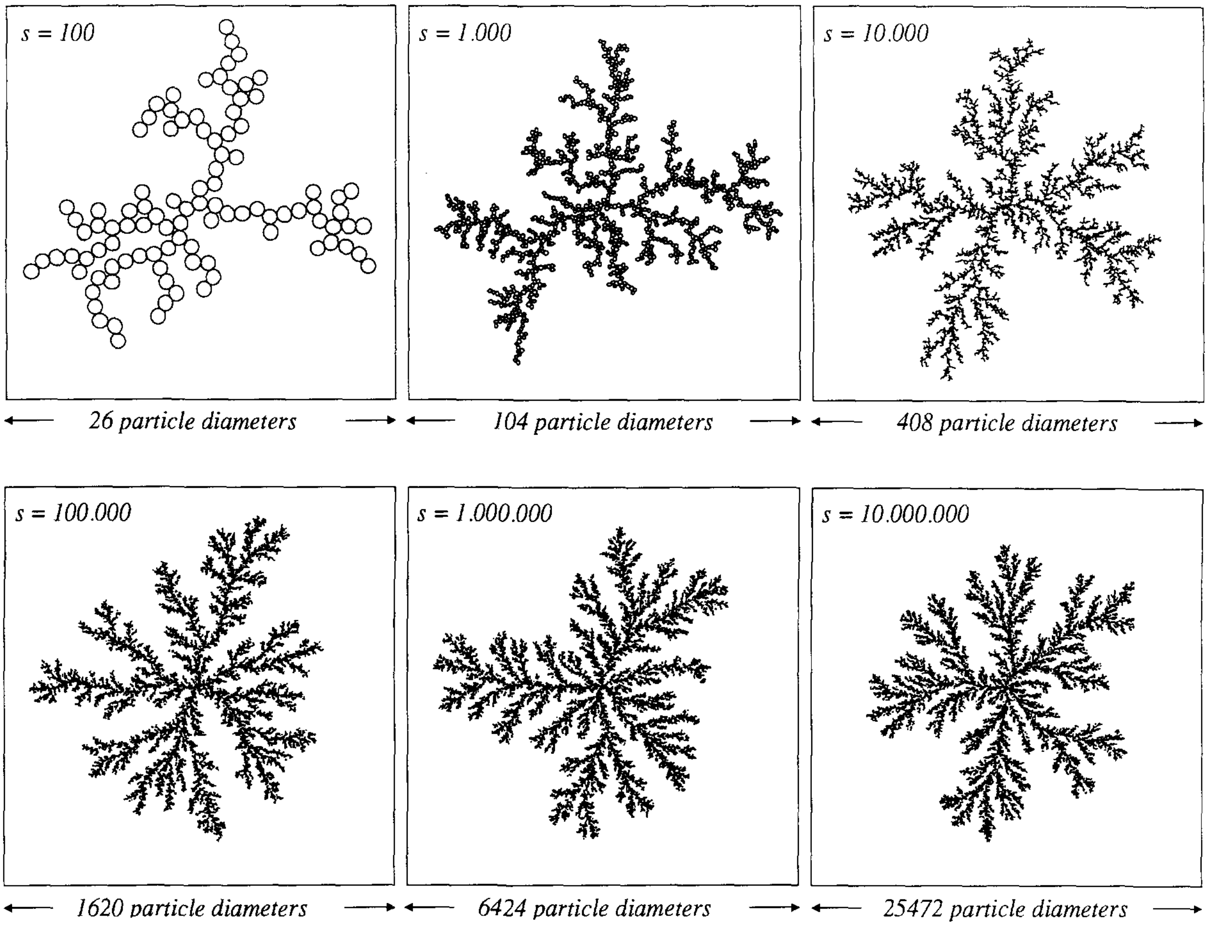

Fig. 2. Six stages in the growth of a 10^7 particle off-lattice DLA cluster. This figure was provided by Thomas Rage.

3. Quantitative characterization of DLA clusters

The first characterization of DLA clusters grown on two-dimensional spaces and lattices was based on the growth of the radius of gyration $R_g(s)$ with increasing cluster size (number of particles) s and the two point density-density correlation function or radial distribution function $C(r)$. Both of these quantities were found to have simple algebraic forms on length scales ℓ larger than the particle size (diameter) d_o and smaller than the cluster diameter L ($d_o \ll \ell \ll L$).

$$\langle R_g(s)\rangle \sim s^{\beta} \tag{1}$$

and

$$C(r) \sim r^{-\alpha}. \tag{2}$$

The correlation function $C(r)$ is defined as

$$C(r) = \langle\langle \rho(\boldsymbol{r}_o)\, \rho(\boldsymbol{r}_o + \boldsymbol{r})\rangle\rangle_{|\boldsymbol{r}|=r}\,, \tag{3}$$

where $\rho(\boldsymbol{r})$ is the density at position $\boldsymbol{r}$ in the d-dimensional embedding space. In Eq. (3) $\langle\langle\cdots\rangle\rangle$ implies averaging over all origins ($\boldsymbol{r}_o$) and all orientations.

Fig. 4 shows the dependence of the exponent β on the cluster size s (obtained by fitting the dependence of $\log R_g$ on $\log s$ by Eq. (1) for clusters with sizes in the range $s_1 - s_2$ with $s_2/s_1 = 1.05$ where $s = (s_1 + s_2)/2$).

The observation that the fractal dimensionalities corresponding to the power law relationships in Eqs. (1) and (2) ($D_\beta = 1/\beta$ and $D_\alpha = d - \alpha$, where d is the Euclidean dimensionality of the embedding space

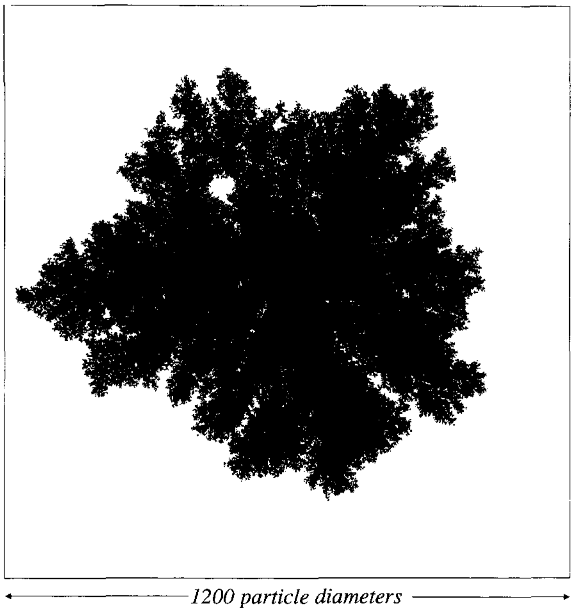

Fig. 3. A three-dimensional off-lattice DLA cluster containing 10^7 particles. A projection onto a plane of 1.25×10^6 of the 10^7 particles is shown. This figure was provided by Thomas Rage.

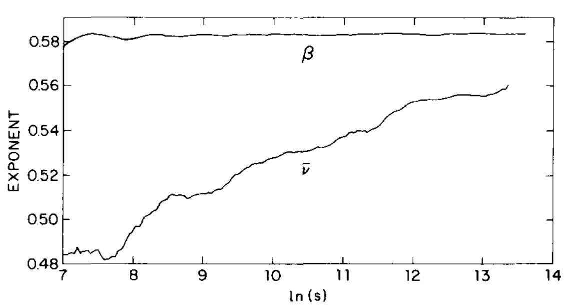

Fig. 4. Dependence of the exponents β and $\overline{\nu}$ that describe the growth of the radius or gyration and width of the active zone, respectively, with increasing cluster size s for dimension off-lattice DLA. The data displayed here were obtained from 1000 10^6 particle clusters.

in which the cluster is grown) were equal (within the uncertainties of the measurements at that time) and that small portions taken from different parts of the cluster looked "similar" to each other led to the idea that DLA clusters were homogeneous self-similar fractals. Since that time our picture of DLA has become more complex.

One of the first indications that DLA might not be so simple came with the studies of the "active zone"

Table 1
Progress in the simulation of two-dimensional DLA clusters: 1991–1993

Year	Reference	Size[1]	Number	Type[2]
1981	T.A Witten and L.M. Sander [1]	3000	$\simeq 10$	L
1982				
1983	P. Meakin [3]	10^4	$\simeq 100$	L,O
1984				
1985	P. Meakin [4]	10^5	$\simeq 100$	L,O
	R.C. Ball and R. M. Brady [5]	10^5	$\simeq 10$	L
1986				
1987				
1988				
1989	S. Tolman and P. Meakin [2]	10^6	$\simeq 1000$	O
		10^7	$\simeq 10$	L
1990				
1991	P. Ossadnik [6]	6×10^6	?[3]	O
1992	B. B. Mandelbrot [7]	3×10^7	?[3]	O
1993	R.F. Voss [8]	2×10^7	?[3]	O
	P. Ossadnik [9]	5×10^7	1	O

[1] Number of sites or discs.
[2] L $\Rightarrow$ lattice model, O $\Rightarrow$ off-lattice model.
[3] Unknown, probably small.

(the region where growth is occurring) by Plischke and Rácz [10]. The growth of a DLA cluster can be thought of in terms of the propagation of an active zone or growth front leaving behind a "dead" or "frozen" structure that does not grow after the front has passed. Plischke and Rácz measured the radius r_a and the width w_a of the active zone (where w_a^2 is the variance in the deposition radius measured from the center of mass) as a function of the cluster size. Using both square lattice and cubic lattice models they found that

$$r_a(s) \sim s^{\nu} \tag{4}$$

and

$$w_a(s) \sim s^{\overline{\nu}}, \tag{5}$$

where the exponent ν in Eq. (4) is equal to the exponent β in Eq. (1) but $\overline{\nu} < \nu$ (for 2d DLA they found that $\nu = 0.584 \pm 0.02$ and $\overline{\nu} = 0.48 \pm 0.01$). This implies the existence of two characteristic length scales ξ_1 and ξ_2 with a ratio ξ_1/ξ_2 that diverges as

$s \to \infty$. Although more recent work [11,2] with both lattice and off-lattice models indicates that the exponents converge as the cluster size increases, this convergence is very slow and it is not certain that the exponents ν and $\overline{\nu}$ are asymptotically equal. Fig. 4 shows the cluster size dependence of the exponent $\overline{\nu}$. More recent numerical work [9,12] suggests that the width of the active zone and the radius of gyration may be related by

$$w_a(s) \sim R_g(\log s)^{-1/2}. \tag{6}$$

Another indication of a more complex structure for DLA comes from the work of Meakin and Havlin [13] on the fluctuation and distribution of the masses contained within a distance r measured from particle centers in a DLA cluster. These quantities can be describes by $P_r(s)$, the probability that a disc of radius r will contain s particles. For simple self-similar fractals $P_r(s)$ is expected to conform the scaling form

$$P_r(s) = s^{-1} f(s/r^D) \tag{7}$$

and the "moment ratios"

$$Z^n(r) = \langle s^n \rangle_r / [\langle s^1 \rangle_r]^n, \tag{8}$$

where $\langle s^n \rangle_r = \sum s^n P_r(s)$ are expected to have constant (r-independent) values for $d_\circ \ll r \ll L$. The distributions $P_r(s)$ obtained for statistically self-similar fractals (such as aggregates generated by cluster–cluster aggregation models) can be described quite well in terms of Eq. (7) and the moment ratios $Z^n(r)$ defined in Eq. (8) are essentially independent of r. However, if the probability distribution $P_r(s)$ is measured in the central, frozen region of off-lattice DLA clusters then these distributions cannot be scaled using the scaling form shown in Eq. (7) and the moment ratios $Z^n(r)$ decrease with increasing r indicating that the DLA structure is more uniform on larger length scales than on smaller length scales [13]. Fig. 5 shows the dependence of the moment ratios $Z^n(r)$ on r for $n = 2$–10 obtained from clusters grown using both the diffusion-limited cluster–cluster aggregation model and the DLA model. In the former case $Z^n(r)$ is essentially constant (except for lengths r that approach the overall cluster size) while for DLA $Z^n(r)$ depends strongly on r.

The moment ratios Z^n (Eq. (8)) are a measure of the lacunarity [14] of a structure. A much more vivid illustration of the scale-dependent lacunarity of DLA clusters has been obtained by Mandelbrot [7]. In this study a region of radius r (centered on the cluster origin) is taken from the frozen, inactive center of a very large DLA cluster and scaled to a circular region of unit area. The ϵ neighborhood of this scaled region is then obtained (it is the union of the circular regions with a radius of ϵ centered on each particle in the region). For a simple self-similar fractal the filling factor or density $\rho(\epsilon, r)$ (fraction of the scaled circular region of unit area that is occupied) should be independent of the size r of the region that is selected. Instead (Fig. 6) Mandelbrot finds that $\rho(\epsilon, r)$ increases with increasing r using a range of values for ϵ. The number of "arms" in the cluster appears to increase with increasing size in accord with the decrease in w_a/r_a found by Plischke and Rácz [10]. Based on this work Mandelbrot proposed two geometric scenarios; a "massive transient" and a "limitless drift". The massive transient scenario would be consistent with Eq. (6). The dependence of the mass (number of particles) $s(r)$ contained within a distance r of the cluster origin can be represented by [7]

$$s(r) = \Lambda_s r^D , \tag{9}$$

where Λ_s is a prefactor that measures the lacunarity. In order to retain the linear dependence of $\log s(r)$ on $\log r$ (one of the few characteristics of DLA clusters that appears to be firmly established by computer simulations), it is necessary for Λ_s to have the form

$$\Lambda_s \sim r^{-\Delta D} \tag{10}$$

so that the exponent D_{sr} obtained from $s(r)$ assuming that $s(r) \sim r^{D_{sr}}$ is related to the fractal dimensionality D in Eq. (9) by [7]

$$D_{sr} = D - \Delta D. \tag{11}$$

A variety of other studies indicate that all is not well with the simple self-similar model for DLA. These include discrepancies between different ways of estimating the "fractal dimensionality" [15,16], numerical evidence for inhomogeneity [17] and different

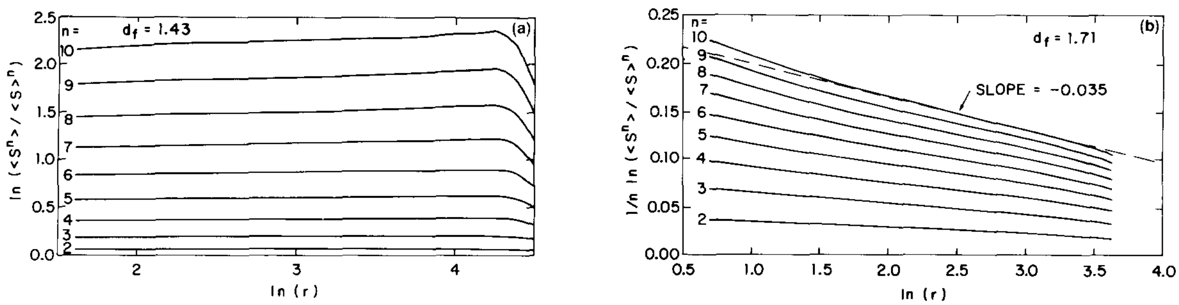

Fig. 5. Dependence of the moment ratio $Z^n(r) = \langle S^n(r)\rangle / \langle S^1(r)\rangle^n$ on r for off-lattice clusters generated using (a) the diffusion-limited cluster–cluster aggregation model and (b) the off-lattice DLA model. For the DLA clusters the distance r is measured with respect to particles in the "dead" interior region of the cluster.

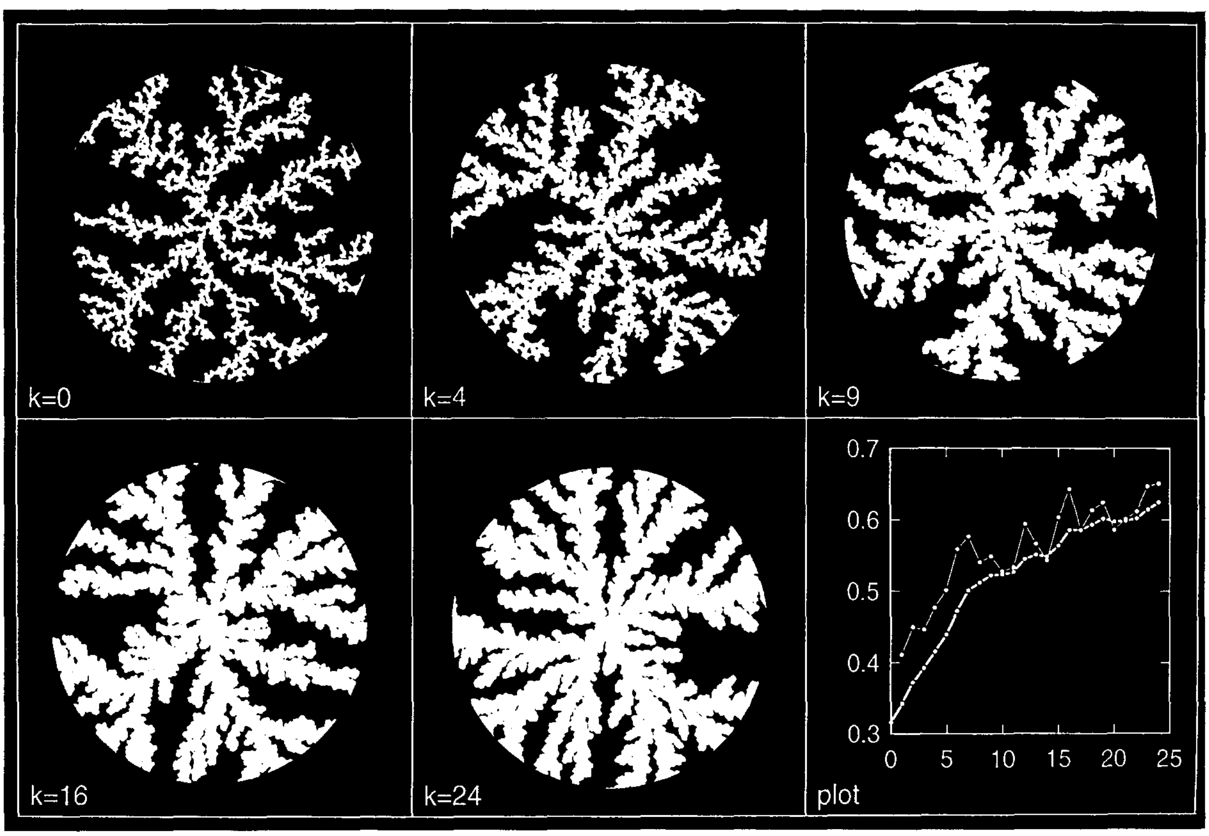

Fig. 6. The ϵ neighborhoods of circular regions of different radii r that have been scaled onto a circle of unit area. In (a)–(e) the radii r are given by $r = r_0 \lambda^k$ with $r_0 = 78$ particle diameters, $\lambda = 1.175$ and $k = 0, 4, 9, 16$ and 24, respectively. In (f) the bold line shows how the density or filling ratio (the fraction of the circle of unit area covered by the ϵ neighborhood) depends on r or k. This figure was provided by Benoit B. Mandelbrot.

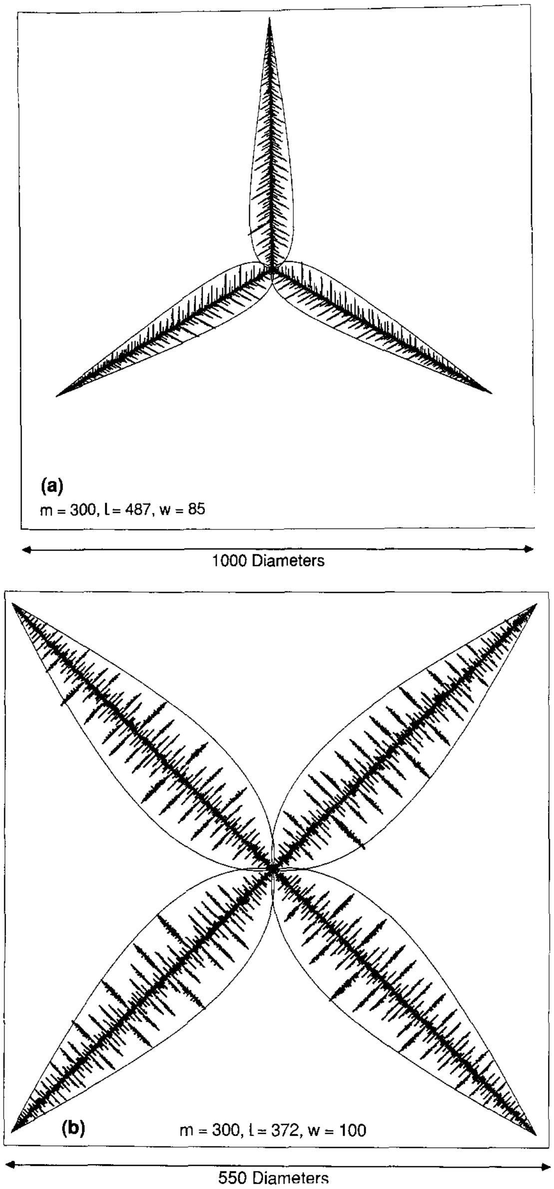

Fig. 7. A comparison of the shapes of DLA clusters grown with a semi-lattice model (the random walkers follow off-lattice trajectories but the direction of attachment to the contacted particle in the cluster is in the closest of n discrete directions). (a)–(c) show clusters grown with $n = 3, 4$ and 5, respectively, with a noise reduction parameter of 300 (a),(b) or 1000 (c).

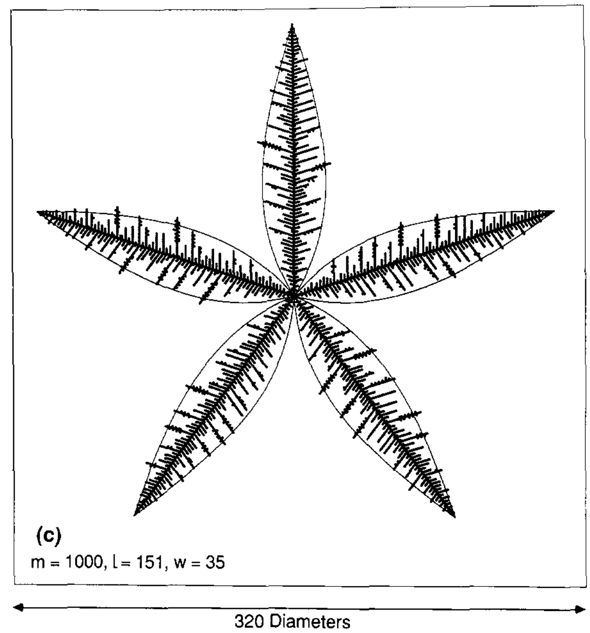

Fig. 7 — continued.

density correlations in the tangential and radial directions [18]. In addition other scaling models have been proposed for DLA based on the ideas of "multiscaling"[19] and "multifractality" [20]. It should be emphasised that computer simulations alone cannot unambiguously define the scaling properties of DLA and the simple self-similar picture may still be asymptotically correct. In fact some numerical evidence indicates that this may be the case [2,9,11,12,16]. In any event a satisfactory theoretical model should explain the "massive transient" or "limitless drift", or establish another scenario. On balance the numerical evidence appears to favor the "massive transient" scenario [2,9,11,12,16]

Compared with two-dimensional DLA there has been little work for $d > 2$. There is some evidence that 3d DLA suffers from the same "problems" as 2d DLA [2,10] and that the 2d behavior is not a consequence of the unique properties of two-dimensional random walks. However there have been no attempts to measure the lacunarity properties of 3d DLA clusters.

Much of the theoretical work that has been carried out to develop a better understanding of DLA is based on the assumption that DLA clusters are self-similar. To the extent that this purported self-similarity has not been established by computer simulations these theoretical approaches must be held in abeyance. The most fruitful theoretical approach to DLA has been that initially proposed by Turkevich and Sher [21] and Ball et al. [22]. This is based on the idea that the growth of a DLA cluster can be thought of in terms of the growth of the cluster "envelope" and the growth velocity of the envelope is given by

$$V(\boldsymbol{r}) \sim \nabla_n \phi(\boldsymbol{r}), \tag{12}$$

where $\phi(\boldsymbol{r})$ is a scalar field that obeys the Laplace equation, $\nabla^2\phi = 0$, subject to the boundary conditions $\phi(\boldsymbol{r}) = 1$ for $|\boldsymbol{r}| \to \infty$ and $\phi(\boldsymbol{r}) = 0$ on the advancing envelope. Here $\nabla_n \phi(\boldsymbol{r})$ is the gradient of $\phi(\boldsymbol{r})$ at position $\boldsymbol{r}$ on the cluster envelope, measured in a direction normal to the envelope surface. Unfortunately the idea of Turkevich and Sher that the shape of the cluster envelope is related in a simple way to the lattice structure, is not supported by computer simulations. Nevertheless this approach has led to important insights into the growth of DLA clusters and related growth processes [23–26]. In particular a quite detailed understanding of "tamed" DLA (DLA with anisotropy and DLA with anisotropy plus noise reduction) has been obtained [25–28] using conformal mapping and renormalization group techniques. In DLA growth on a lattice (Fig. 1) with noise reduction the random walker does not result in growth when it enters an unoccupied perimeter site. Instead the random walk is terminated when it reaches a perimeter site and a record of the events is kept. Growth occurs (the perimeter site is filled) only when it has been reached m times where m is the noise reduction parameter. Fig. 7 shows clusters generated using two-dimensional DLA models with anisotropy and noise reduction. This figure compares the shapes of the clusters arms generated using this model and the shapes expected theoretically using the approach described in Refs. [27] and [28]. However this work has not led to a theory for "wild" (off-lattice) DLA with no long range anisotropy

4. Summary

There was no space in this short survey to discuss progress in the experimental growth of DLA patterns. There are now many examples of natural and laboratory phenomena that generate patterns that can be understood in terms of the DLA model (even though the DLA model is not itself well understood from a theoretical point of view). Several reviews of "DLA experiments" have appeared [29–31] and new examples are still being found.

Unfortunately a satisfactory theoretical understanding of DLA still eludes us and computer simulations alone cannot provide completely unambiguous answers. It is becoming apparent that DLA is a difficult problem that poses a major theoretical challenge.

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References

[1] T.A. Witten and L.M. Sander, Phys. Rev. Lett 47 (1981) 1400.
[2] S. Tolman and P. Meakin, Phys. Rev. A 40 (1989) 428.
[3] P. Meakin, Phys. Rev. A 27 (1983) 604, 1495.
[4] P. Meakin, J. Phys. A 18 (1985) L661.
[5] R.C. Ball and R.M. Brady, J. Phys. A 18 (1985) L809.
[6] P. Ossadnik, Physica A 176 (1991) 454.
[7] B.B. Mandelbrot, Physica A 191 (1993) 95.
[8] R.F. Voss, Fractals 1 (1993) 141.
[9] P. Ossadnik, Physica A 195 (1993) 319.
[10] M. Plischke and Z. Rácz, Phys. Rev. Let. 53 (1984) 415–418.
[11] P. Meakin and L.M. Sander, Phys. Rev. Let. 54 (1985) 2053.
[12] C. Amitrano, A. Coniglio, P. Meakin and M. Zannetti, preprint (1993).
[13] P. Meakin and S. Havlin, Phys. Rev. 36 (1987) 4428–4433.
[14] B.B. Mandelbrot, The fractal geometry of nature (Freeman, New York, 1983).
[15] F. Argoul, A. Arneodo, G. Grasseau and H.L. Swinney, Phys. Rev. Lett. 61 (1988) 2558–2561.
[16] G. Li, L.M. Sander and P. Meakin, Phys. Rev. Lett. 63 (1988) 1322.
[17] T.C. Halsey and P. Meakin, Phys. Rev. A 32 (1985) 2546–2548.
[18] P. Meakin and T. Vicsek, Phys. Rev. A 32 (1985) 685–688.
[19] C. Amitrano, A. Coniglio, P. Meakin and M. Zannetti, Phys. Rev. B 44 (1991) 4974.
[20] T. Vicsek, F. Family and P. Meakin, Europhys. Lett. 12 (1990) 217–222.

[21] L.A. Turkevich and H. Scher, Phys. Rev. Lett. 55 (1985) 1026–1029.
[22] R.C. Ball, R.M. Brady, G. Rossi and B.R. Thompson, Phys. Rev. Lett 55 (1985) 1406–1409.
[23] T.C. Halsey, P. Meakin and I. Procaccia, Phys. Rev. Lett. 56 (1986) 854–857.
[24] R.C. Ball, M.J. Blunt and O. Rath Spivack, Proc. R Soc. (London) 423 (1989) 123–132.
[25] R.C. Ball, P.W. Barker and R. Blumenfeld, Europhys. Lett. 16 (1991) 47–52.
[26] R.C. Ball, Physica A 140 (1986) 62–69.
[27] J.-P. Eckmann, P. Meakin, I. Procaccia and R. Zeitak, Phys. Rev. A 29 (1989) 3185–3195.
[28] J.-P. Eckmann, P. Meakin, I. Procaccia and R. Zeitak, Phys. Rev. Lett. 65 (1990) 52–55.
[29] P. Meakin, in: Phase Transitions and Critical Phenomena, Vol. 12, C. Domb and J.L. Lebowitz, eds. (Academic Press, New York, 1988) p. 335.
[30] J. Feder, Fractals (Plenum, New York, 1988).
[31] T. Vicsek, Fractal Growth Phenomena, 2nd ed. (World Scientific, Singapore, 1992).
[32] M. Matsushita, in: The Fractal Approach to Heterogeneous Chemistry: Surfaces, colloids, polymers, D. Avnir, ed. (Wiley, Chichester, 1989).