PHYSICAL REVIEW A VOLUME 35, NUMBER 12 JUNE 15, 1987

Structure of large two-dimensional square-lattice diffusion-limited aggregates: Approach to asymptotic behavior

Paul Meakin
Central Research and Development Department, E.I. du Pont de Nemours and Company, Wilmington, Delaware 19898

Robin C. Ball
Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, United Kingdom

P. Ramanlal and L. M. Sander
Department of Physics, University of Michigan, Ann Arbor, Michigan 48109

(Received 20 October 1986)

Efficient algorithms have been used to grow large (4×10^6 site) diffusion-limited aggregation (DLA) clusters on two-dimensional (2D) square lattices. As the clusters grow larger, their envelope grows, from a more or less round shape characteristic of small clusters, through a diamond shape characteristic of clusters containing about 10^5 sites, into a cross shape. Results from about 25 clusters indicate that the exponents describing the length l and width w of the four major arms vary continuously with M (the cluster mass) over the range $10^3 < M < 4\times10^6$. We find that the effective exponent $\nu_{\parallel} = d\ln(l)/d\ln(M)$ increases systematically from 0.585 to 0.61 at the highest mass. This may be consistent with a limiting value of $\frac{2}{3}$ (as found for uniaxially biased DLA in two dimensions) but only with large corrections to scaling in our range of M. The exponent $\nu_{\perp} = d\ln(w)/d\ln(M)$ *decreases* systematically, to about 0.48 at $M=4\times10^6$. Our results are consistent with an asymptotic (scaling) fractal geometry for square-lattice DLA but suggest that these fractals are neither self-similar nor homogeneous.

INTRODUCTION

The formation of fractal[1] aggregates by diffusion-limited aggregation (DLA) is a topic which has received considerable attention since the introduction of the Witten-Sander[2] model just over five years ago. Early, rather small scale, computer simulation results[2-5] indicated that DLA clusters are homogeneous, statistically self-similar fractals and that the fractal dimension which characterizes these clusters is insensitive to model details such as lattice type, sticking probabilities, etc. It is only within the past two years that this picture for the structure of DLA has begun to change. Meakin and Vicsek[6] and Kolb[7] found that (even for off-lattice DLA aggregates) the density-density correlations are different in the radial and tangential directions and Halsey and Meakin[8] showed that two-dimensional (2D) DLA aggregates do not have a homogeneous structure. At the same time, using improved algorithms, Ball and Brady[9] showed that DLA clusters grown on a square lattice have an overall "diamond"-like shape reflecting the structure of the lattice. This and similar observations[10] led Turkevich and Scher[11] to propose a theory for DLA which relates the fractal dimension to the lattice dimension to the lattice structure, assuming that the overall shape of the cluster reflects the structure of the underlying lattice. This theory predicts that the fractal dimension of DLA clusters is nonuniversal in the sense that it depends on lattice details.

In view of the rather robust universality which was believed to be a characteristic of DLA, it was generally assumed that anisotropy in the diffusion tensor or sticking probability would lead to the formation of clusters with an overall anisotropic shape but that no new scaling exponents would be generated. This idea was not tested in any of the earlier simulations. Ball *et al.*[12] found using both simulation results and theoretical arguments that any degree of uniaxial anisotropy (however small) will eventually cause the DLA aggregate to grow into a "needle"-like shape in which the length of the needle grows with the $\frac{2}{3}$ ($\nu_{\parallel}$) power of the mass and the width grows with the $\frac{1}{3}$ ($\nu_{\perp}$) power of the mass.

The results and ideas of Turkevich and Scher and Ball *et al.* have stimulated a renewed interest in nonequilibrium growth models more or less closely related to DLA,[13-17] of which this paper is a part. As a result of this more recent work, our picture for the structure of DLA aggregates is still evolving. At the present time it appears that for clusters of small sizes the radius of gyration (or other characteristic size of the cluster) grows with increasing cluster size according to the *universal* power law:

$$R_g \sim M^{\beta} \quad (\beta \simeq 0.585 \quad \text{for } d=2) . \tag{1}$$

For off-lattice DLA the effective value for the exponent β does not change with increasing cluster size[18] and is equal, within statistical uncertainties, to a recent theoretical suggestion[19] that $\beta = 2-\sqrt{2} = 0.5858\cdots$.

There is now strong evidence from a variety of models that anisotropy (even just that of the lattice) leads to a different value of β closer to $\frac{2}{3}$.[20] This is the theoretical maximum and it was found when a uniaxial bias (i.e., twofold symmetry) was applied.[12] With a threefold symmetric bias,[13] β also appears to approach $\frac{2}{3}$ and this consequently must be regarded as a leading possibility on the square lattice. The crossover from a value of about 0.585 to a value close to $\frac{2}{3}$ for the exponent β depends on both the structure of the lattice (the degree of anisotropy) and the amount of "randomness" in the model. Various methods have been devised for controling these parameters by averaging to reduce randomness in the growth process,[15,16] by examining the deterministic process on a lattice to eliminate noise,[17] or by increasing the anisotropy.[21–23] Kolb[24] has developed a model in which the growth is biased in a controlled way in the directions of the closest lattice axis passing through the origin of the cluster. Results from these models have been interpreted in terms of a limiting (large size) value of $\frac{2}{3}$ for the exponent β on lattices with two-, three-, four-, six-fold symmetry. However, the relationship between these models and the original square-lattice DLA model of Witten and Sander is not well understood at the present time. A characteristic feature of the modified (noise-reduced or anisotropy-enhanced) DLA models is that they lead to the formation of "star"-like clusters for which the length of the arms grows with increasing cluster mass according to

$$l \sim M^{\nu_{\|}} \quad (\nu_{\|}=\beta\simeq\tfrac{2}{3} \text{ for } M\rightarrow\infty)\ , \tag{2}$$

while the width appears to grow with a smaller exponent,

$$\omega \sim M^{\nu_{\perp}} \quad (\nu_{\perp}<\beta,\nu_{\|} \text{ for } M\rightarrow\infty)\ . \tag{3}$$

The purpose of this paper is to explore the relationship between square-lattice DLA and these modified models using very large clusters in order to approach the $M\rightarrow\infty$ limit. Other aspects of the structure of square-lattice DLA aggregates are also presented.

COMPUTER SIMULATIONS

The efficiency of DLA simulations can be dramatically improved by allowing the random walkers to take large steps when they are a large distance from any occupied sites on the cluster.[9,10,25] Using this improvement clusters containing 10^5 occupied lattice sites can be generated in a few minutes on an IBM 3081 computer. To go beyond 10^5 sites an efficient procedure is also needed to store information concerning the location of the cluster. Using an algorithm developed earlier by Ball and Brady,[9] we have been able to generate clusters containing 4×10^6 occupied lattice sites using about 16 h of CPU time on an IBM 3081. The results obtained from 25 such simulations are described below.

The computer program used in this work was compared extensively with a more conventional (semilattice) DLA program which was developed completely independently. In this model[13] the random walker takes large off-lattice jumps when it is far from the cluster and moves by steps of one lattice unit on the lattice when it is within l lattice units from any of the occupied sites on the cluster. For $l\simeq6$ lattice units and clusters in the size range $5\times10^3-10^5$ occupied lattice sites, the program used in this work gave clusters with a radius of gyration about 1.5% smaller than those generated using the semilattice model. However, the effective radius of gyration exponent, β [Eq. (1)], differed by only 4×10^{-4} (an amount smaller than the statistical uncertainties.) For $l=0$ and 12 lattice units the two programs gave virtually identical results. The most probable explanation of these results is that there is a small bias introduced by the transition between on-lattice and off-lattice steps in the semilattice program. Tests were also carried out to determine the sensitivity of the simulation to the radius of the circle from which the random walkers are launched and the radius of the circle at which they are terminated if they move a very large distance from the cluster. Simulations

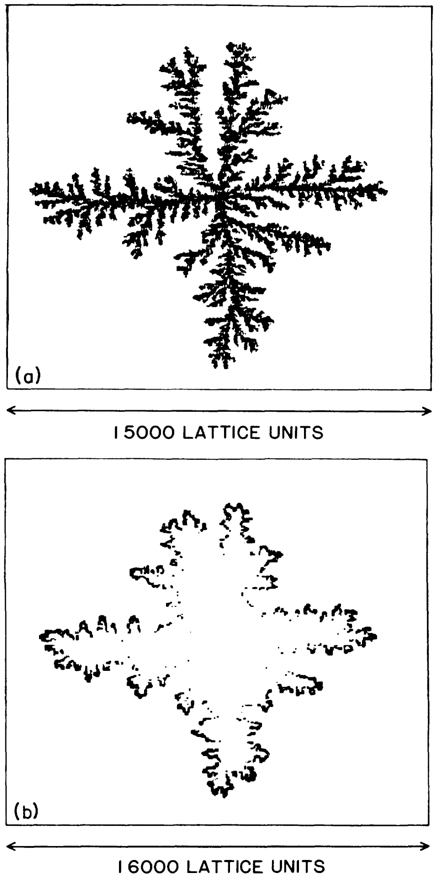

FIG. 1. (a) Shows a 4×10^6 site DLA cluster grown on a square lattice and (b) shows a map of the places where growth is occurring (the active zone) of the cluster. The active zone shown in (b) indicates those regions (50×50 blocks of lattice sites) in which growth has occurred during the addition of the last 5% of sites added. The active zone also represents a surface for the cluster which defines its overall shape.

were also carried out in which the circle at which trajectories are terminated was centered on both the original growth site and on the center of mass of the clusters. No indication was found that the procedures used in our simulations introduce a measurable distortion in the structure of the clusters.

RESULTS

A typical (randomly selected) 4×10^6 square-lattice DLA cluster is shown in Fig. 1(a). Only 8×10^4 of the 4×10^6 occupied sites were actually used to construct the cluster shown in this figure. It is evident that the overall shape of the clusters has evolved beyond the diamondlike shape characteristic of clusters containing about 10^5 sites towards a crosslike shape. In Fig. 1(b) a map is shown of the places where growth occurred during the addition of the last 200 000 sites. Each element in the map consists of a 50×50 block of lattice sites and the element is filled if growth occurred in any of the lattice sites in that block. This figure shows the formation of an overall crosslike shape and side branching is also evident. It appears that as the DLA clusters become larger their shape becomes more similar to that associated with conventional dendritic growth.[26] Figure 1(b) can also be considered to be a map of the active zone (region where growth is occurring[27]) of the cluster shown in Fig. 1(a). To provide a more representative impression of the structure of large DLA clusters, the "active zones" from four randomly selected clusters are shown in Fig. 2. The active zones shown in Fig. 2 illustrate the fact that even for very large cluster sizes there is a considerable variability in the overall cluster shapes. For this reason it is necessary to generate the largest practical number of clusters in order to obtain reliable estimates of their ensemble scaling properties. Despite the variability in the cluster geometries exhibited in Fig. 2, it is clear that for all four clusters the overall shape has evolved well beyond the diamond-shape characteristic of clusters containing $\sim 10^5$ sites towards a crosslike shape.

To facilitate comparison with the results of other simulations, theory, and experiments, a more quantitative description of the structure of these clusters and its dependence on the cluster size is needed. Figure 3 shows the

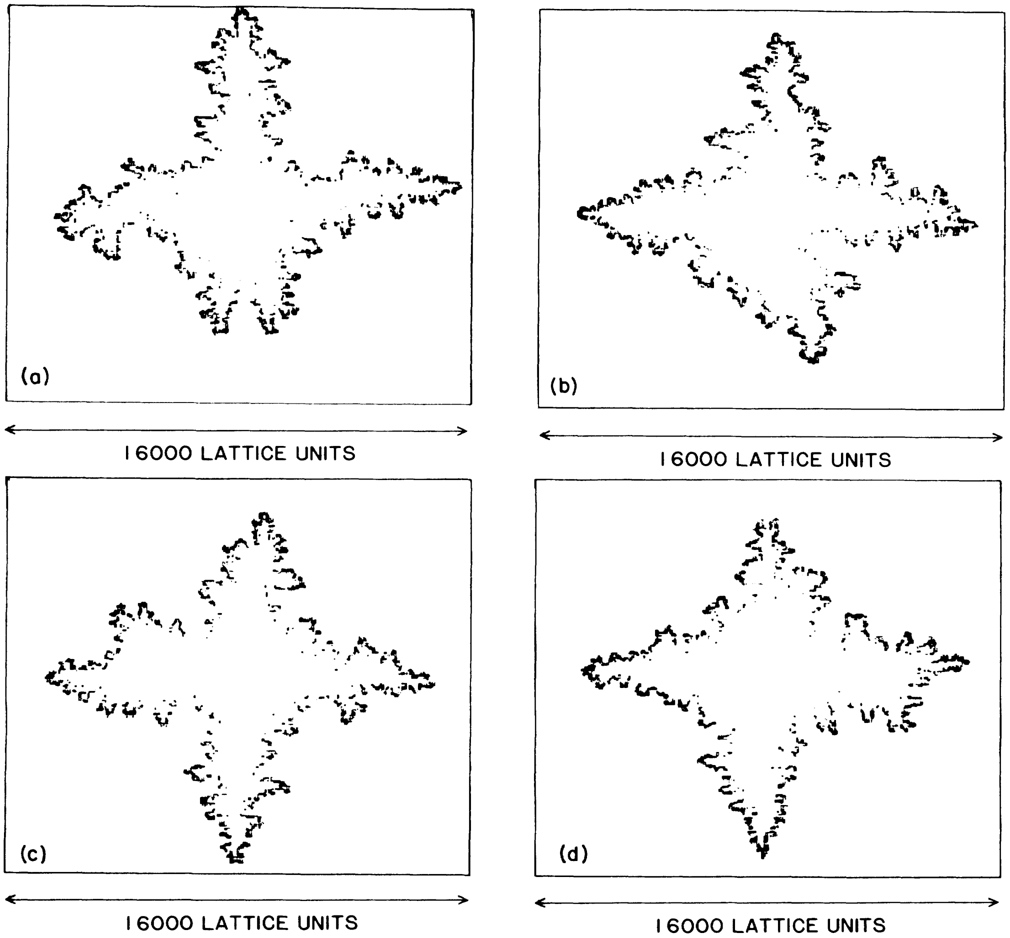

FIG. 2. Maps of the active zones taken from four randomly selected 4×10^6 site square-lattice DLA clusters. This figure illustrates the overall crosslike shape associated with these clusters.

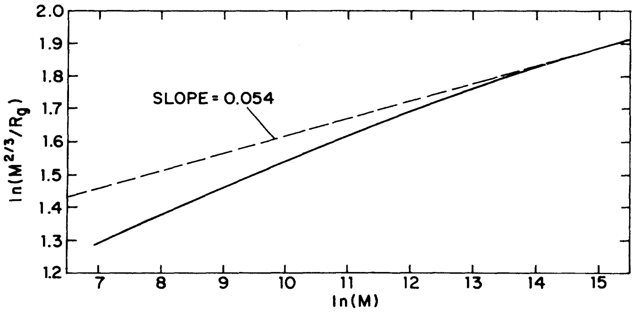

FIG. 3. Dependence of the radius of gyration (R_g) on cluster mass for clusters in the size range 10^3—4×10^6 sites.

dependence of the radius of gyration (R_g) on cluster mass (M) for clusters in the size range (M) of 10^3 to 4×10^6 occupied sites. In this figure $\ln(M^{2/3}/R_g)$ is plotted as a function of R_g. If the radius of gyration exponent (β) had a value of $\frac{2}{3}$ for all M this plot would be a horizontal line. Plotting $\ln(M^{2/3}/R_g)$ vs $\ln(M)$ rather than $\ln(R_g)$ vs $\ln(M)$ emphasizes the dependence of the effective exponent β on M. Figure 3 shows that β increases with increasing M. For very small clusters β has a value close to the universal value of 0.585 and for the largest cluster sizes ($M \simeq 4\times10^6$) β has a value of about 0.61. Although this might be interpreted in terms of a fractal dimension (D_β) of about 1/0.6 or $\frac{5}{3}$ it is clear that β is still increasing as the cluster mass increases. It is also apparent from Figs. 1 and 2 that at least two exponents describing the growth of the length and width of the cluster arms are needed to describe the structure.

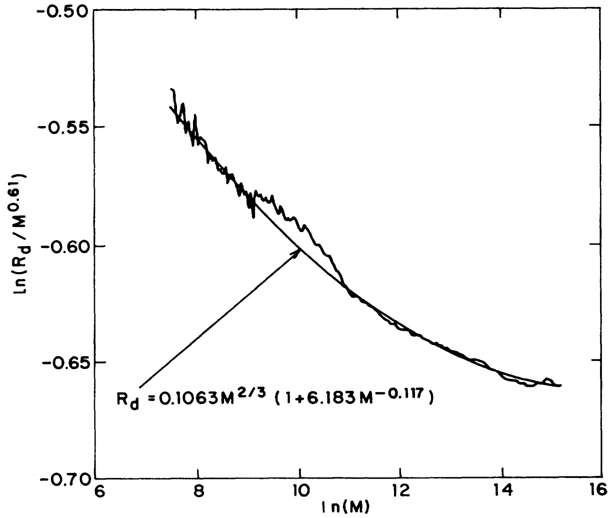

FIG. 4. Dependence of the mean deposition radius (R_d) on cluster mass for clusters in the size range 1800—4×10^6 occupied lattice sites.

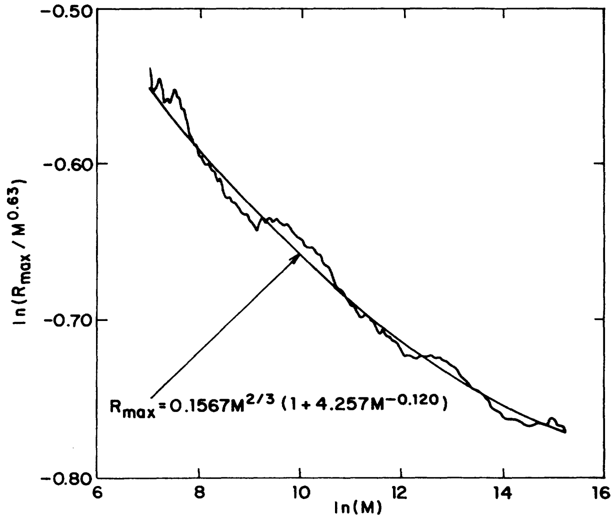

FIG. 5. Dependence of the maximum radius ($R_{\max}$) on cluster mass (M) for clusters in the size range $1100 < M < 4\times10^6$ occupied lattice sites.

We have also measured the dependence of the mean deposition radius (radius of the active zone[24]), R_d, on the cluster mass. The dependence of $\ln(M^{2/3}/R_d)$ on $\ln(M)$ looks similar to Fig. 3. The effective value of the exponent ν defined by

$$R_d \sim M^\nu \tag{4}$$

has a value of about 0.61 for $M = 4\times10^4$. Figure 4 shows the dependence of $\ln(R_d/M^{0.61})$ on $\ln(M)$ obtained from

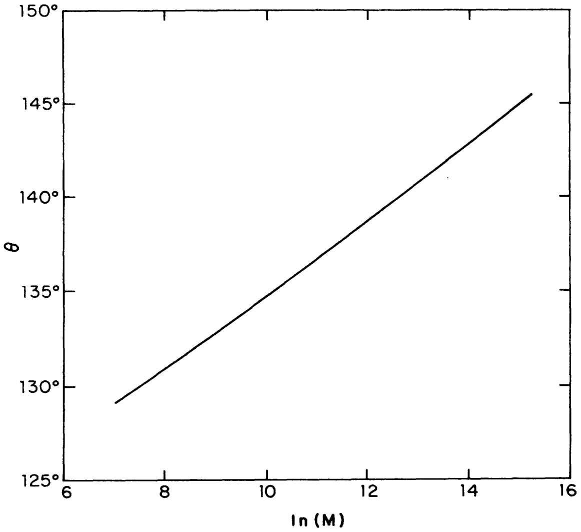

FIG. 6. Dependence of the effective cone angle for the arms of the clusters on the cluster size (M). An angle of 135° would correspond to a diamond shape and an angle of 180° would correspond to a needle. The angle θ was obtained from the rate of growth of the maximum radius with increasing cluster mass. The angle $\pi-\theta$ is one half of the tip angle describing the shape of the ends of the cluster arms.

all of the clusters. The smooth curve in Fig. 4 corresponds to the functions

$$R_d = aN^{\nu}(1+bN^{-\delta}) \tag{5}$$

with the parameters $\nu=\frac{2}{3}$, $a=0.1063$, $b=6.183$, and $\delta=0.117$. The parameters a, b, and δ are varied such that the functional form given in Eq. (5) best fits the data. Figure 4 shows that the dependence of R_d on M is consistent with an asymptotic value of $\frac{2}{3}$ for the exponent ν. Figure 5 shows similar results for the maximum radius of the cluster. In this case the smooth curve corresponds to the function

$$R_{\max} = aM^{\nu_m}(1+bM^{-\delta_m}) , \tag{6}$$

where $a=0.157$, $\nu_m=\frac{2}{3}$, $b=4.257$, and $\delta_m=0.120$. The theoretical models of Ball *et al.*[12] and Turkevich and Scher[11] can be used to analyze the growth of the maximum radius in terms of a "tip" angle. According to this theory[12] the rate of growth of the maximum radius (dR/dM) is given by $(dR/dM) \sim R^{-\pi/2\theta}$, where $\pi-\theta$ is half of the tip angle. Figure 6 shows the value of the angle θ obtained from the growth of the maximum radius. An angle of 135° would correspond to a diamond shape $(2\pi-\theta)=90°$. For $M \simeq 4\times10^6$ we find that $\theta \geq 145°$ and there is no indication that θ will saturate below the limiting value of π corresponding to a cross shape with a diverging length to width ratio for its arms. The dependence of θ on $\ln(m)$ is surprisingly linear over the range of cluster masses shown. However, it is not inconsistent with limiting values of $\leq 180°$ for large (M) and $\simeq 127°$ $(\nu_{\parallel} \sim 0.585)$ for small M.

Figure 7 shows the dependence of the width of the active zone (ξ) on the cluster mass in the form of a plot of $\ln(M^{2/3}/\xi)$ vs $\ln(M)$. For the larger cluster masses this curve is almost horizontal indicating that

$$\xi \sim M^{2/3} . \tag{7}$$

Taken together the results shown in Figs. 4—7 are consistent with the idea that the exponent describing the growth in the length of the cluster arms with increasing mass has a value of $\frac{2}{3}$ $(\nu=\nu_m=\nu_{\parallel}=\frac{2}{3})$, but only with a very slow crossover which is not nearly complete at $M=4\times10^6$.

To describe the growth in the width of the cluster arms with increasing cluster mass we have examined the mean deposition distance from the nearest axis of the cluster (the x or y axis). Figure 8 shows the dependence of this quantity (Y) on the cluster mass in the form of a plot of $\ln(M^{2/3}/Y)$ vs $\ln(M)$. The slope of this curve (about 0.19) for clusters containing more than 10^5 sites corresponds to an exponent $(\nu_{\perp})$ of 0.67—0.19 or 0.48.

Some other aspects of the structure of square-lattice DLA have also been investigated. Figure 9 shows the angular distribution of mass obtained from 14 clusters. The angular mass distribution was averaged for all 14 clusters and then all of the octants in the polar mass distribution were averaged to construct Fig. 9. Figure 9(a) shows the mass distribution (for 5% increments in the total mass M) for clusters of sizes up to 1.19×10^4 lattice sites. Even for clusters of this small size (which could easily be generated 3—4 years ago with crude algorithms[4,5]) the angular mass distribution has evolved beyond that expected for a homogeneous diamond shape.

Only at very small cluster sizes is the mass distribution circular (apart from local lattice effects). For clusters containing more than 10^6 sites [Fig. 8(c)] the mass is very strongly concentrated along the lattice axis. We expect that as the cluster grows larger and larger the angular mass distribution will become more and more strongly concentrated along the lattice axis as is required if the two exponents describing the length and width of the arms are different.

Garik[28] has found for 2D off-lattice DLA that the ratio of the radii of gyration (R_x and R_y where $R_y > R_x$), measured about the principal axis of the inertial tensor, appears to approach a limiting (large cluster size) value of unity. For off-lattice clusters containing 50 000 particles an average value for this ratio (R_x/R_y) was found to be

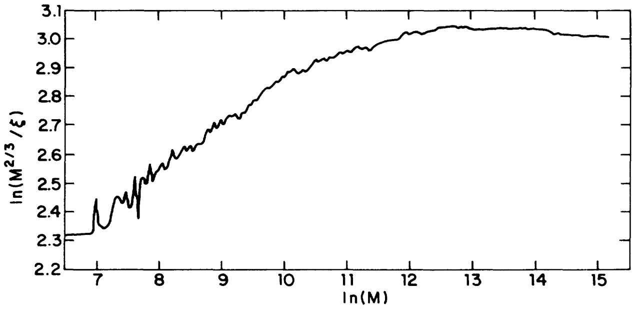

FIG. 7. Dependence of the width of the active zone (variance of the deposition radius) ξ on cluster mass for clusters in the size range $10^3 < M < 4\times10^6$ occupied sites. The observation that the curve is almost flat for $M > 10^5$ indicates that $\xi \sim M^{2/3}$ for large M.

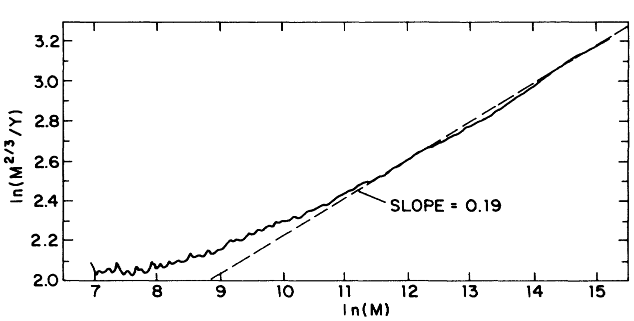

FIG. 8. This figure shows how the quantity Y depends on the cluster mass (M) for clusters in the size range $10^3 < M < 4\times10^6$ sites. Y is the mean deposition distance measured from the closest axis of the coordinate system used in the square-lattice DLA system. This quantity provides a measure of the way in which the width of the cluster arms increases with increasing cluster mass.

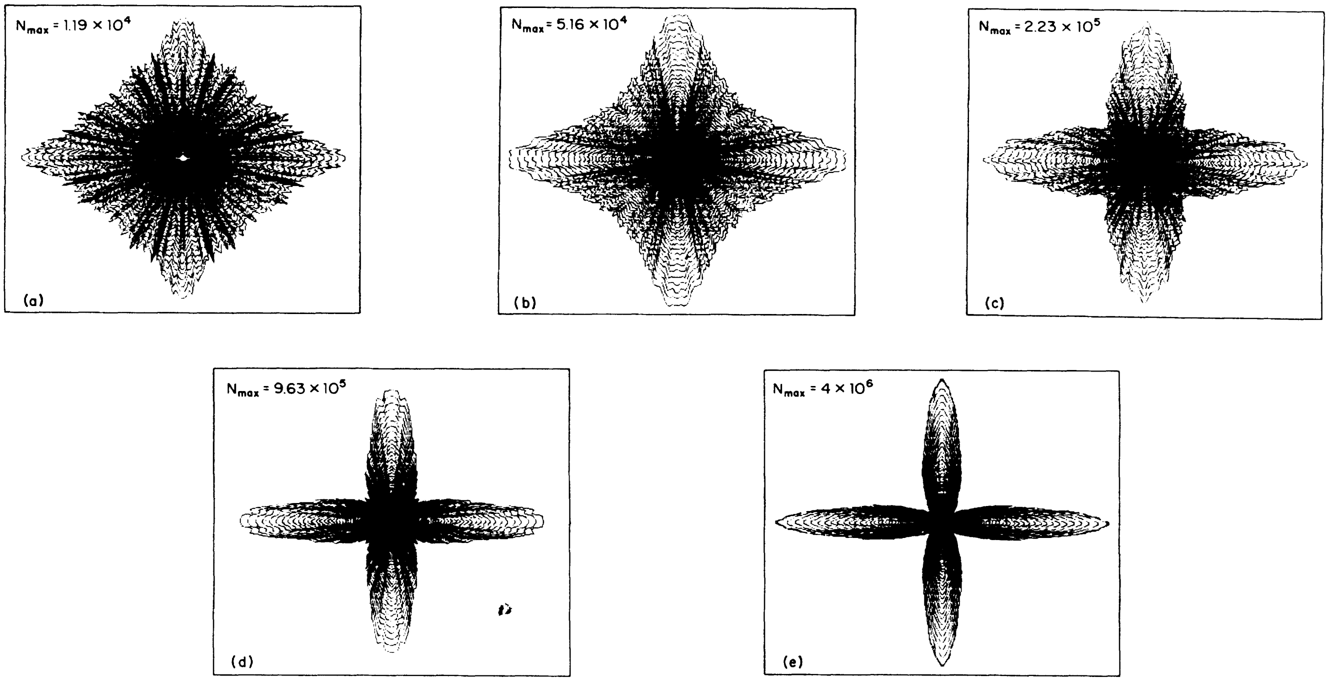

FIG. 9. Angular distribution of mass in square-lattice DLA clusters. (a)—(e) show the angular mass distribution for clusters containing up to 1.2×10^4, 5.2×10^4, 2.2×10^5, 9.6×10^5, and 4.0×10^6 sites, respectively. The "contours" show the angular mass distribution for smaller clusters at 5% intervals in the mass.

0.87 ± 0.07 and Garik estimated that clusters containing about 1.5×10^6 particles would be needed to see asymptotic behavior. Figure 10 shows the dependence of R_x/R_y on cluster sizes obtained from our square-lattice simulations. For $M=4\times10^6$ this ratio has a value of about 0.925. Our results are consistent with a limiting value of unity but again with a very slow approach to this limiting value.

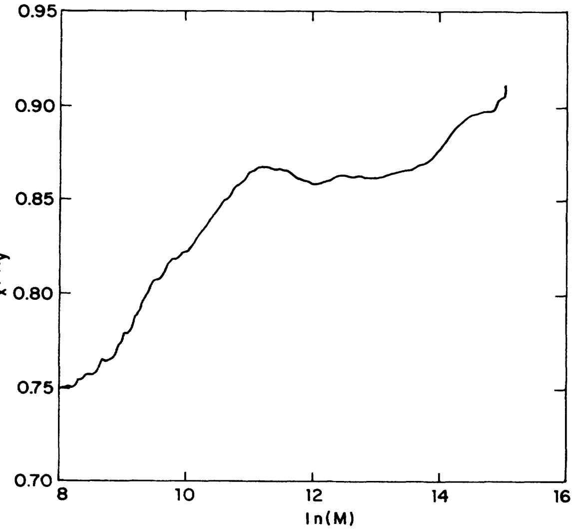

FIG. 10. Ratio of the radii of gyration (R_x and R_y) about the principal axes of the inertial tensor for 2D aggregates in the size range $5\times10^3<M<4\times10^6$ occupied lattice sites. This figure shows the average value for R_x/R_y where $R_x<R_y$.

DISCUSSION

The main result of our work is that 2D square-lattice DLA asymptotically evolves into cross shapes with apparently different exponents describing the growth of the length and width of the arms. All of our results indicate that the exponent associated with the growth of the length of the arms (with increasing cluster mass) has a value greater than 0.6; they are also consistent with a limiting value $\nu_{\parallel}=\frac{2}{3}$. We found an effective value of $\nu_{\perp}=0.48$ for the exponent describing the growth of the width of the arms with mass but two reservations should be noted concerning our measurement of the arm width. The first is that we used the mean absolute distance to the nearest axis. But as can be seen from the cluster in Fig. 1 even at $M=4\times10^6$ the quadrant in which a particle lies is not a totally reliable criterion for which arm of the cluster it belongs to. The second is that if major arms grow parallel to but offset from the coordinate axes then that offset makes a contribution to our measurement of the arm width. Both of these difficulties recede as the cross shape becomes better developed, but this in turn implies a transient suppression of the increase of the width w with mass and hence a transient lowering of $\nu_{\perp}$. Very similar results have been obtained by Kolb[24] ($\nu_{\parallel}=\frac{2}{3}$, $\nu_{\perp}=\frac{1}{2}$) using a semilattice (off-lattice walks with on-lattice growth) model in which the growth direction is biased towards the direction of the closest lattice axis passing through the origin of the cluster. However, the effects of this locally uniaxial bias on the DLA process are not well understood and this model may have different asymptotic behavior from ordinary square-lattice DLA. Finally, we note that very recent results for the square-lattice DLA model but with reduced noise (Thompson[29] and Meakin[30]) show transient behavior similar to our results ($\nu_{\parallel} \sim 0.61$, $\nu_{\perp} \sim 0.48$). However, they clearly indicate that the aspect ratio of the arms ultimately saturates when the noise is reduced so that $\nu_{\parallel} = \nu_{\perp} = \nu$. The limiting common value of these exponents is close to but definitely less than $\frac{2}{3}$. Although our data are quite consistent with this as the ultimate asymptotic behavior, even without noise reduction, the cluster sizes needed to see this appear almost prohibitive with any known algorithms and current computers. We have grown square-lattice DLA clusters to 40 times more particles than previously reported but at least a further factor of 40 would appear to be required.

P. Ramanlal and L. M. Sander were supported by the U.S. Department of Energy, Grant No. DE-FG02-85ER45189.

---

[1] B. B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, San Francisco, 1982).

[2] T. A. Witten and L. M. Sander, Phys. Rev. Lett. **47**, 1400 (1981).

[3] T. A. Witten and L. M. Sander, Phys. Rev. B **27**, 5686 (1983).

[4] P. Meakin, Phys. Rev. A **27**, 604 (1983).

[5] P. Meakin, Phys. Rev. A **27**, 1495 (1983).

[6] P. Meakin and T. Vicsek, Phys. Rev. A **32**, 685 (1985).

[7] M. Kolb, J. Phys. Lett. **46**, L631 (1985).

[8] T. C. Halsey and P. Meakin, Phys. Rev. A **32**, 2546 (1985).

[9] R. C. Ball and R. M. Brady, J. Phys. A **18**, L809 (1985).

[10] P. Meakin, J. Phys. A **18**, L661 (1985).

[11] L. Turkevich and H. Scher, Phys. Rev. Lett. **55**, 1026 (1985).

[12] R. C. Ball, R. M. Brady, G. Rossi, and B. R. Thompson, Phys. Rev. Lett. **55**, 1406 (1985).

[13] P. Meakin, Phys. Rev. A **33**, 3371 (1986).

[14] P. Meakin, Phys. Rev. A **33**, 4199 (1986).

[15] J. Kertesz and T. Vicsek, J. Phys. A **19**, L257 (1986).

[16] J. Nittmann and H. E. Stanley, Nature **321**, 663 (1986).

[17] P. Garik, R. Richter, J. Hautman, and P. Ramanlal, Phys. Rev. A **32**, 3156 (1985).

[18] P. Meakin and L. M. Sander, Phys. Rev. Lett. **54**, 2053 (1985).

[19] R. C. Ball, Physica **104A**, 62 (1986).

[20] F. Family and H. G. E. Hentschel (unpublished).

[21] J. D. Chen and D. Wilkinson, Phys. Rev. Lett. **55**, 1892 (1985).

[22] M. Matsushita and H. Kondo, J. Phys. Soc. Jpn. **55**, 2483 (1986).

[23] P. Meakin, Z. Y. Chen, and P. Evesqne (unpublished).

[24] M. Kolb (unpublished).

[25] M. Kolb (private communication).

[26] J. S. Langer, Rev. Mod. Phys. **52**, 1 (1980).

[27] M. Plischke and Z. Racz, Phys. Rev. Lett. **53**, 415 (1984).

[28] P. Garik, Phys. Rev. A **32**, 1275 (1985).

[29] B. R. Thompson (unpublished).

[30] P. Meakin (unpublished).