Anisotropy and Cluster Growth by Diffusion-Limited Aggregation

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We describe a simple theory of diffusion-limited–aggregation cluster growth which relates the large-scale shape of the cluster to its fractal dimension. We present results of computer simulation for DLA clusters grown with anisotropic sticking rules which provide strong confirmation of our model in two dimensions. New universal exponents are predicted and found. We are also able to obtain good estimates for the fractal dimension of ordinary DLA.

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Kinetic growth models describing the formation of large aggregates from small subunits have recently attracted considerable interest. Especially studied has been the diffusion-limited–aggregation (DLA) model of Witten and Sander.[1] This model has been used to describe such diverse phenomena as dendritic electrodeposition,[2] sputter deposition of thin films of $NbGe_2$,[3] and the viscous fingering of water displacing oil in a porous medium.[4] Many variants of DLA have been studied by computer simulation and many inconclusive attempts have been made to calculate analytically the fractal dimension, D, of DLA clusters as a function of the dimension, d, of the space in which they were grown. At present the one exact analytic result for DLA clusters grown with d finite is the inequality $D \geqslant d-1$ which was obtained by a causality bound argument.[5]

In this Letter we propose a simple analytic model for the growth of DLA clusters in two dimensions. We relate the large-scale diamond shape of the cluster to its rate of growth in the x and y directions and to its fractal dimension. For ordinary DLA clusters grown in two dimensions the model predicts $D=\frac{5}{3}$ and for DLA clusters grown with anisotropic sticking probabilities it indicates (i) that any sticking anisotropy will force the cluster to grow into a rodlike object and (ii) that $X_1 \sim N^{2/3}$ and $X_2 \sim N^{1/3}$ in the limit $N \rightarrow \infty$, where X_1 and X_2 are the dimensions of the cluster in the "easy" and "hard" directions of growth, and N is the number of particles in the cluster. We believe that these anisotropic exponents are exact, whereas the result for the isotropic case may only be approximate. However, it is not only the predictive power of our model that is important; at least equally so is the new simple and intuitive picture of DLA growth that it provides.

We have confirmed the predictions of our model by growing (by computer simulation) large ($\sim 10^5$ particles) DLA clusters with various anisotropic sticking probabilities on a square lattice. No results for such clusters are available in the literature; this is surprising because they are a simple variant of ordinary DLA and it is easy to imagine physical situations where they will occur. We grew these clusters as follows: Each particle is launched from a randomly chosen place on a circle enclosing the cluster and centered on the cluster center of mass. It moves diffusively (by use of the algorithm of Ball and Brady[6]) until it is either two hundred cluster radii from the cluster, in which case it is "killed" and another particle is started from the launch circle, or until it is adjacent to the cluster. If there is an occupied nearest-neighbor cluster site to the right or the left of the particle it sticks and another particle is launched. If the only occupied nearest-neighbor site is above or below the particle it sticks with probability p and continues its random walk (with the constraint that it cannot move onto the cluster) with probability $1-p$.

We start from the growth rate of the cluster. By estimating both the rate of gain of particles, dN/dt, and the increase of cluster radius, dR/dt, we can eliminate time to find dR/dN and hence the scaling of R with N Following Ref. 5 we study the extremal radius of the cluster so that dR/dt is given by the rate of deposition on the most extreme tips. The crucial issue, then, is the disposition of the absorbing cluster boundary relative to these tips. We model this by a cone of exterior half angle β. There are three motivations for this: (i) The cone angle is directly related to the singularity of the deposition rate onto the tip; (ii) this is the only simple scale-invariant form; and (iii) large anisotropic DLA clusters are strikingly diamond shaped as shown in Fig. 1(a). For an infinite cone the appropriate solution to $\nabla^2\Phi=0$ with $\Phi=0$ on the boundary $\theta \pm \beta$ is

$$\Phi(r,\theta)=Cr^{\pi/2\beta}\cos(\pi\theta/2\beta), \tag{1}$$

where C is a normalization factor. Thus the steady-state flux of random walkers onto the cone edge at a distance ρ from its tip [see Fig. 1(b)] is

$$u(\rho)=(\pi/2\beta)C\rho^{(\pi/2\beta)-1}. \tag{2}$$

To find the growth rate of the cluster we introduce a large cutoff at $\rho \sim R$ and a small cutoff at $\rho \sim a$, where a is the tip radius or lattice spacing (which we take to be unity). Thus from (2), by integration, we find $dN/dt \simeq CR^{\pi/2\beta}$ and $dR/dt \simeq C$ so that $dR/dN \simeq R^{-\pi/2\beta}$.

The above argument gives no indication as to the

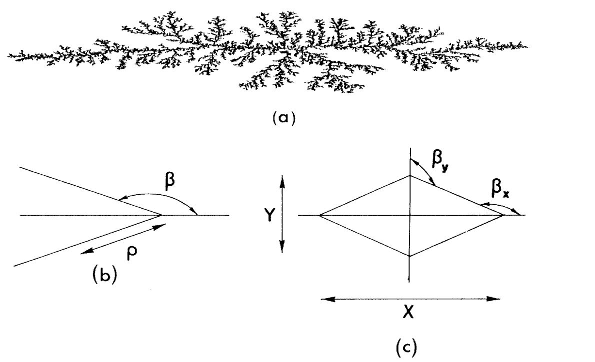

FIG. 1. (a) A 50 000-particle DLA cluster grown with $p=\frac{1}{5}$. (b) Model cone geometry. (c) Model diamond geometry.

value of β; to find how β evolves we must consider the overall shape of the cluster and not just the rate of deposition onto a single extremal tip. To do this we model a DLA cluster as a perfectly absorbing diamond [see Fig. 1(c)]. We find the steady-state particle flux onto it by using a Schwarz-Christoffel mapping to solve $\nabla^2\Phi=0$ with $\Phi=0$ on the diamond boundary. The flux onto the diamond near its extremal points (tips) along the x and y axes, u_x and u_y, satisfy

$$u_x \sim C(1/R)(\rho_x/R)^{(\pi/2\beta_x)-1}, \tag{3}$$

$$u_y \sim C(1/R)(\rho_y/R)^{(\pi/2\beta_y)-1}. \tag{4}$$

Here $R=\frac{1}{2}(X^2+Y^2)^{1/2}$ is the length of each side of the diamond, X and Y are the tip-to-tip dimensions, $\beta_x=\pi-\arctan(Y/X)$, $\beta_y=\pi/2+\arctan(Y/X)$, and ρ_x and ρ_y are the distances along the edge of the diamond from its x and y tips. We also find $dN/dt \sim C$ independent of R by Gauss's theorem. Again, using a short-distance cutoff to eliminate the divergence in the flux at the tips of the diamond, and identifying dX/dt with u_x and dY/dt with u_y, we have

$$dX/dN=(dX/dt)(dt/dN)=AR^{-\pi/2\beta_x}, \tag{5}$$

$$dY/dN=(dY/dt)(dt/dN)=BR^{-\pi/2\beta_y}. \tag{6}$$

The coefficients A and B are bounded of order 1 but may be slowly varying functions of X and Y and of p.

It should be understood that a number of approximations are implied in these equations. (i) Modeling a DLA cluster as a perfectly absorbing diamond (or cone) means neglecting fluctuations in the shape of a single DLA cluster. (ii) We have assumed that DLA clusters grown with anisotropic sticking probabilities can be modeled as perfect absorbers although particles do not necessarily stick on first contact. This is because any particle will stick near its first contact point with the cluster.[7] The average distance between the first and final contact points is independent of the size of the cluster and thus sticking is local on the scale where our equations are valid. (iii) We approximated the exact solution for the exterior of a perfectly absorbing diamond (however, we retained the correctly scaling divergences of the steady-state particle flux at the corners of the diamond). (iv) It should finally be stressed that these are continuum equations; they apply to the mean growth rates of the length and width of a DLA cluster.

For an ordinary DLA cluster ($p=1$) grown from a right-angled diamond profile ($X_0=Y_0$) the two functions A and B must be equal and Eqs. (5) and (6) imply that $X=Y$ throughout the growth of the cluster. Hence $\beta_x=\beta_y=3\pi/4$ at all times, and so by integrating (5) and (6) one has $R\sim N^{3/5}$. Thus our model yields a fractal dimension of $D=\frac{5}{3}$ for ordinary DLA grown in two dimensions from a right-angled diamond profile. The cone model also gives $D=\frac{5}{3}$ if one inputs by hand the requirement that the cone be right angled.[8] This result compares satisfactorily with the simulation result of Meakin,[9] $D=1.71\pm0.01$.

A DLA cluster grown with anisotropic sticking probabilities ($p<1$) from a right-angled diamond profile should be described by Eqs. (5) and (6) with $A>B$ initially. Thus X at first will grow faster than Y with the result $B_x>B_y$. This will make dX/dN greater and greater than dY/dN causing the diamond to grow ever more elongated in the x direction. Eventually the limiting behavior $X\sim N^{2/3}$, $Y\sim N^{1/3}$ is approached and the cluster shape becomes rodlike. It should be noted that it is not necessary to have $A>B$ throughout the growth to obtain this limiting behavior.

It should be remarked that the arguments leading to our result for the exponents do not involve the type of lattice on which the clusters are grown. While we have made appeal to the fact that our anisotropic clusters are roughly diamond shaped, we expect this feature to be a result of the uniaxial anisotropy and to be realized to the same extent for clusters grown off lattice or on a triangular lattice. Ordinary DLA clusters can hardly be considered diamond shaped and the prediction $D=\frac{5}{3}$ should be looked upon only as the extrapolation of our arguments to the case of zero anisotropy.

We present here a representative sample of results of an analysis of DLA clusters grown with anisotropic sticking rules. Full details of this study will appear in a separate publication. For each value $p=\frac{1}{50}$, $\frac{1}{20}$, $\frac{1}{10}$, $\frac{1}{5}$, $\frac{1}{3}$, $\frac{1}{2}$, and $\frac{2}{3}$ of the sticking probability in the y direction we grew at least seven clusters of 5×10^4 particles (10^5 particles for $p=\frac{1}{2},\frac{2}{3}$). It should be noted at this point that Jullien, Kolb, and Botet[10] have considered the growth of DLA clusters from anisotropically diffusing particles on a plane strip geometry. They find $D=\frac{3}{2}$ in the extreme asymptotic case which should correspond to our p going to zero. This agrees with our results.

In each cluster the tip-to-tip distances X and Y (i.e., the difference in lattice units between the maximum and minimum abscissae and ordinates) were measured as a function of the number of particles N in the growing cluster. We also measured the corresponding radii of gyration X_g and Y_g in the x and y directions.

The average aspect ratio $\langle Y/X \rangle$ for each set of clusters with a given p is shown in Fig. 2 as a function of N. this quantity does indeed decrease as N increases and it is seen that the asymptotic behavior expected from Eqs. (5) and (6) $\langle Y/X \rangle \sim N^{-1/3}$ is reached already at $N \sim O(10^4)$ for the lower values of p considered. This asymptotic behavior is approached more slowly for values of p closer to 1.

For each cluster we measured the exponents $\mu_x(N,p)$ and $\mu_y(N,p)$ [defined by $X_g(N,p) \sim N^{1/\mu_x}$ and $Y_g(N,p) \sim N^{1/\mu_y}$]. We obtained the average values $\bar{\mu}_x$ and $\bar{\mu}_y$ of the exponents for each set of clusters grown with the same p. One can combine these with the results for the average aspect ratio $\langle Y/X \rangle$ as a function of N in order to obtain $\bar{\mu}_x$ and $\bar{\mu}_y$ in terms of $\langle Y/X \rangle$ and p. Figure 3 collects the results of this procedure for all values of p considered here.

In our equations (5) and (6) the dependence on p is confined to the prefactors A and B. The results of Fig. 3 are, indeed, strongly suggestive of universal behavior with small p-dependent corrections (complete p independence would cause all the measured values of $\bar{\mu}_x$ and $\bar{\mu}_y$ to lie on the same curve). The continuous curves plotted in Fig. 3 indicate the behavior predicted by a zeroth-order iteration of Eqs. (5) and (6) with A and B taken to be constants. Consider Eq. (5) in the neighborhood of a value $\langle Y_0/X_0 \rangle$ of $\langle Y/X \rangle$: Expanding the exponent $\pi/2\beta_x$ in a Taylor series one has

$$dN/dX \cong AX^{\nu_0}(1 + \epsilon\nu_0' \ln X + \ldots), \tag{7}$$

where ν_0 and ν_0' are respectively the values of the exponent $(\pi/2\beta_x)$ and of its derivative at $\langle X/Y \rangle = \langle X_0/Y_0 \rangle$. Neglecting terms in $\epsilon = \langle Y/X \rangle - \langle Y_0/X_0 \rangle$ one has

$$\mu_x = \pi/2\beta_x + 1 \tag{8}$$

and a similar treatment of Eq. (6) gives

$$\mu_y = \frac{\pi/2\beta_x + 1}{1 + \pi/2\beta_x - \pi/2\beta_y}.$$

These approximate values of μ_x and μ_y are the continuous curves plotted in Fig. 3 (note that they do not depend on the values of A and B). They are in remarkable agreement with the simulation data. It should be noted that the sum of the exponents $1/\mu_x$ and $1/\mu_y$ gives an indication of the compactness of the cluster: When the sum approaches 1 the clusters become compact.

The above results suggest the possibility that for any $p < 1$ a DLA cluster will eventually (no matter how small the anisotropy) grow into a compact rodlike object, its exponents evolving along curves close to those shown in Fig. 3 as the cluster mass increases. A first attempt to explore this issue is shown in Fig. 4: Each of the curves represents the measured aspect ratio $\langle Y/X \rangle$ as a function of p at fixed values of N. One sees that if these curves are extrapolated towards $p = 1$ the value of $\langle Y/X \rangle$ remains smaller than 1. This

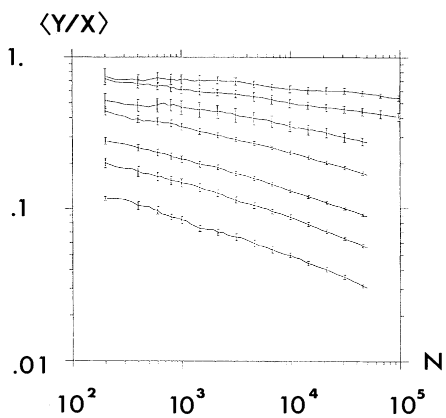

FIG. 2. Average measured aspect ratio $\langle Y/X \rangle$ as a function of N. Error bars are shown at selected data points. The curves refer to $p = \frac{2}{3}, \frac{1}{2}, \frac{1}{3}, \frac{1}{5}, \frac{1}{10}, \frac{1}{20}$, and $\frac{1}{50}$ going from top to bottom.

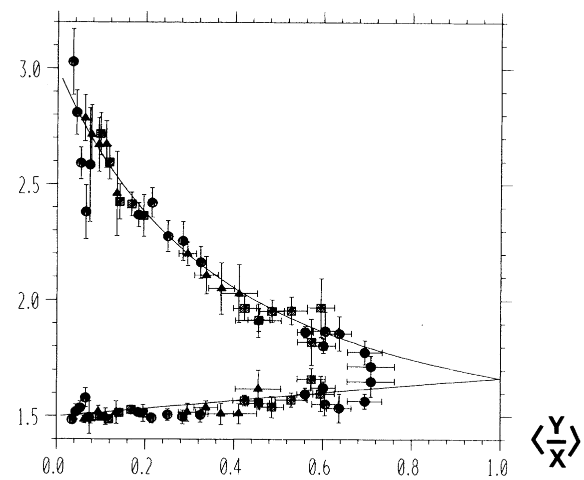

FIG. 3. Measured values of μ_x (lower set of data) and μ_y (upper set) vs $\langle Y/X \rangle$. Different groups of symbols correspond to different p: from left to right $p = \frac{1}{50}$ (circles), $p = \frac{1}{20}$ (triangles), $p = \frac{1}{10}$ (squares), $p = \frac{1}{5}$ (circles), $p = \frac{1}{3}$ (triangles), $p = \frac{1}{2}$ (squares), and $p = \frac{2}{3}$ (circles). Data for different p represented by the same symbols do not overlap.

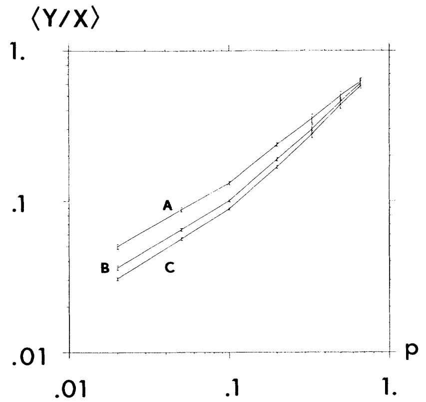

FIG. 4. Average measured aspect ratio $\langle Y/X \rangle$ as a function of p at fixed values of N. Curves A, B, and C correspond to $N = 10\,000$, $N = 30\,000$, and $N = 50\,000$.

agrees with a picture in which any cluster (with any $p < 1$) will as $N \rightarrow \infty$ reach the fixed-point behavior corresponding to $\mu = \frac{3}{2}$ and $\mu_y = 3$.

In conclusion, the main difference between the model presented here and previous continuous models is the emphasis on the overall average geometrical shape of the cluster. Using our model we are able to predict the effect of sticking anisotropies in DLA cluster growth and to produce values for the fractal dimension of ordinary DLA which agree well with computer simulation results. We have presented numerical evidence that any anisotropy causes DLA clusters to grow into compact rodlike objects characterized by the scaling relations $X \sim N^{2/3}$, $Y \sim N^{1/3}$. Our model predicts these relations and appears to describe very well the approach to the scaling regime. Although our treatment is approximate we hope that it will provide a useful starting point towards a more complete understanding of DLA. We also hope that our results on anisotropic DLA will stimulate further research in this direction.[11]

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[7]Note that independently of the value of p a particle will stick on first contact to any site adjacent to the cluster in the x direction.

[8]For $d > 2$, by performing a cylindrical average we model a DLA cluster by two back-to-back right-angled d-dimensional cones. Proceeding as before we find that for ordinary DLA $D(d) = d - 1 + \nu$, where $C_\nu^{(d-2)/2}(-1\sqrt{2}) = 0$. Here $C_\nu^\lambda(x)$ is a Gegenbauer function of degree ν. In particular, one has $D = 2.463$ in three dimensions (the Gegenbauer function becomes an ordinary Legendre function P_ν) and $D = \frac{10}{3}$ in four dimensions.

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[11]After completion of this work we received a preprint by L. A. Turkevich and H. Scher [Phys. Rev. Lett. **55**, 1026 (1985)], where ordinary DLA in two dimensions is treated along lines partly similar to ours.