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LETTER TO THE EDITOR

Orientation of particle attachment and local isotropy in diffusion limited aggregation (DLA)

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Abstract. We simulate 50 off-lattice diffusion limited aggregation (DLA) clusters of one million particles each. The probability distribution of the angle of attachment of arriving particles with respect to the local radial direction is obtained numerically. For increasing cluster size N, the distribution crosses over extremely accurately to a cosine with an amplitude which decreases towards zero as a power-law in N. From this specific viewpoint, asymptotically large DLA clusters are locally *isotropic*.

Diffusion limited aggregation (DLA) [1] is a fractal growth model exhibiting great complexity [2]. The properties of asymptotically large clusters are of fundamental importance. Some of these properties are independent of many of the details of aggregation rules and are expected to be shared by large clusters grown in various experimental or natural situations. Efficient algorithms and improved computational facilities have enabled the generation of very large off-lattice clusters with more than 100 million particles [3]. However, many asymptotic properties of DLA are still unclear. There have been many debates on the basic issues, including the scaling behaviour of large clusters and the multifractal properties of the growth probability measure [2].

This work concentrates on the local isotropy of large off-lattice DLA. Specifically, we investigate the orientation of particle attachment. For each new particle in the cluster, let $\boldsymbol{R}'$ and $\boldsymbol{R}$ be the position vectors of the new particle and its parent respectively, relative to the centre of the cluster. The parent is the particle in the aggregate upon which the new attachment is made and we define the centre of the cluster to be at the seed. The vector $\boldsymbol{R}$ characterizes the local radial direction of this sticking event and $\boldsymbol{r} = \boldsymbol{R}' - \boldsymbol{R}$ defines the sticking direction. The angle of attachment $\theta(-\pi < \theta \leqslant \pi)$ is defined as the angle measured counter-clockwise from $\boldsymbol{R}$ to $\boldsymbol{r}$. When $|\theta| < \pi/2$ the attachment can be described as forward. We focus on the probability distribution $P(\theta, N)$ of θ, which in general depends on the number of particles N in the cluster. If the DLA were compact with a nearly circular boundary, the above definition would imply that θ is always close to zero and a backward attachment is geometrically impossible. The distribution $P(\theta, N)$ should peak at $\theta = 0$. Although the DLA is far from being compact, at least for small clusters, forward attachment is favoured for similar geometrical reason.

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We compute the distribution $P(\theta, N)$ for $N = 150\times2^k$, where $k = 0$ to 12 corresponding to $N = 150$ to 614 400. For each k we form the histogram of the values of the sticking angle θ for the $(100 \times 2^k + 1)$th to the (200×2^k)th particles. Proper normalization gives $P(\theta, N)$ and the symmetry $P(\theta, N) = P(-\theta, N)$ is used. We averaged the results over 50 clusters. The statistical error is estimated from the sample to sample fluctuations.

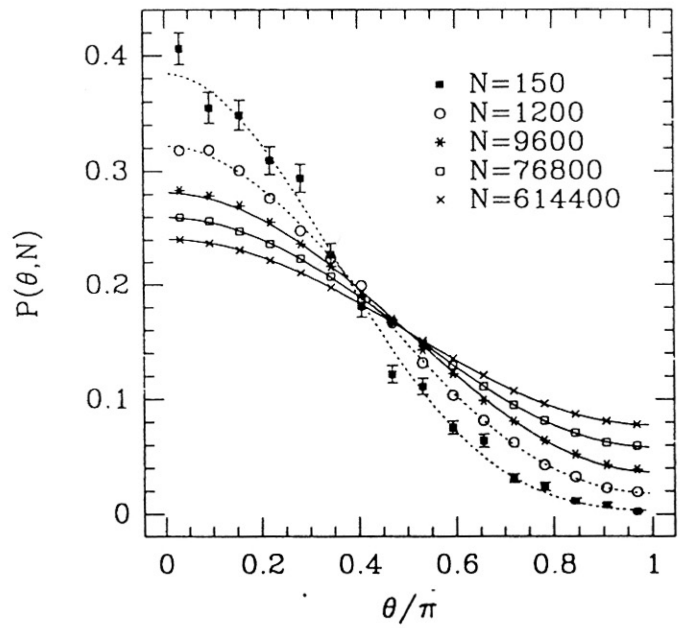

Figure 1. Probability distribution $P(\theta, N)$ of the angle of attachment θ for different cluster sizes N. Also shown are one parameter cosine fits for the data at $N = 9600$, 76 800 and 614 400 and two parameters fits for $N = 150$ and 1200.

Figure 1 shows $P(\theta, N)$ for five values of N. We used 16 histogram bins. The error bars are smaller than the symbols except for $N = 150$. The errors are as small as about 0.2% at $N = 614\,400$ where our data are most accurate. For small N forward sticking dominates so that $P(\theta, N)$ peaks at $\theta = 0$ as expected; backward stickings are rare. The peak broadens as N increases. At $N = 614\,400$ the ratio $P(\pm\pi, N)/P(0, N)$ between perfectly backward and forward attachments becomes about 0.31. Figure 1 shows the least-squares fits to

$$P(\theta, N) = 1/2\pi + a_1(N)\cos(\theta) \tag{1}$$

for the three larger values of N where $1/2\pi$ ensures normalization. At $N = 614\,400$ the quality of the fit is excellent. There is, apparently, no systematic deviation trend, as the data points scatter around the fitted curve by amounts comparable to the statistical errors. The fit is still good at $N = 76\,800$, but at $N = 9600$ there are noticeable systematic deviations. A good fit is obtained if we take the first three terms of the cosine expansion:

$$P(\theta, N) = \frac{1}{2\pi} + \sum_{n=1}^{\infty} a_n(N)\cos(n\theta). \tag{2}$$

Figure 1 shows the two-parameter fits for $N = 150$ and 1200. More generally, we fitted our complete data set of $P(\theta, N)$ with the first four cosine terms. Figure 2(a) shows the

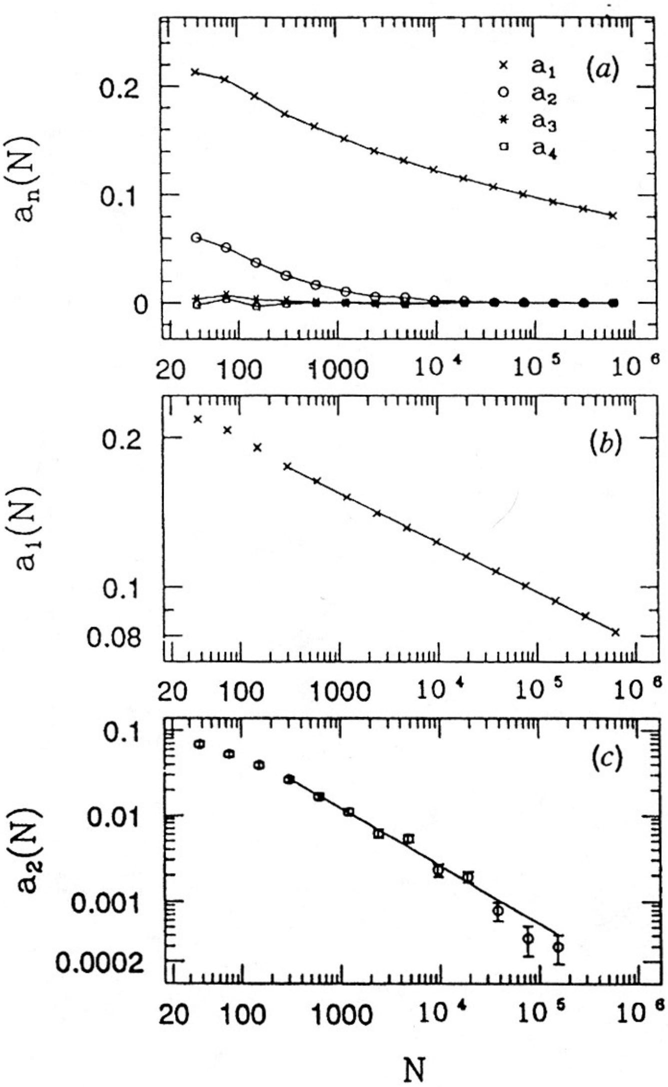

Figure 2. Amplitudes $a_n(N)$ of the cosine terms in the expansion of $P(\theta, N)$ as a function of cluster size N: (a) a_1 and a_4 in a semi-log plot; (b) a_1 and (c) a_2 in log–log plots respectively.

values of the amplitudes $a_n(N)$ in a semi-log plot. We used histograms of 64 bins and the result agrees with those of 16 bins. We also computed $a_n(N)$ by Fourier transform and obtained the same results again. Figures 2(b) and (c) show, respectively, $a_1(N)$ and $a_2(N)$ in log–log plots. The linearity at $N \gtrsim 300$ implies

$$a_n(N) \simeq A_n N^{-\gamma_n} \tag{3}$$

for $n = 1$ and 2, where $\gamma_1 = 0.0997(3)$, $A_1 = 0.309(2)$, $\gamma_2 = 0.67(3)$ and $A_2 = 1.2(3)$. The bracketed values are the fitting errors. For $n \geqslant 3$ the measured $a_n(N)$ is not precise enough for a test of the above algebraic decay.

We also studied the closely related problem of the branch orientation of DLA. We adopt the Horton–Strahler scheme of branch ordering. The smallest branches without a side-branch are assigned order one and the main stems have the highest order. Other authors have used slightly different schemes [4, 5] which should give similar results. From visual examinations of the branches shown in [4], it is evident that those with the highest order are nearly radial and the directedness decreases for lower orders. Quantitatively, we define

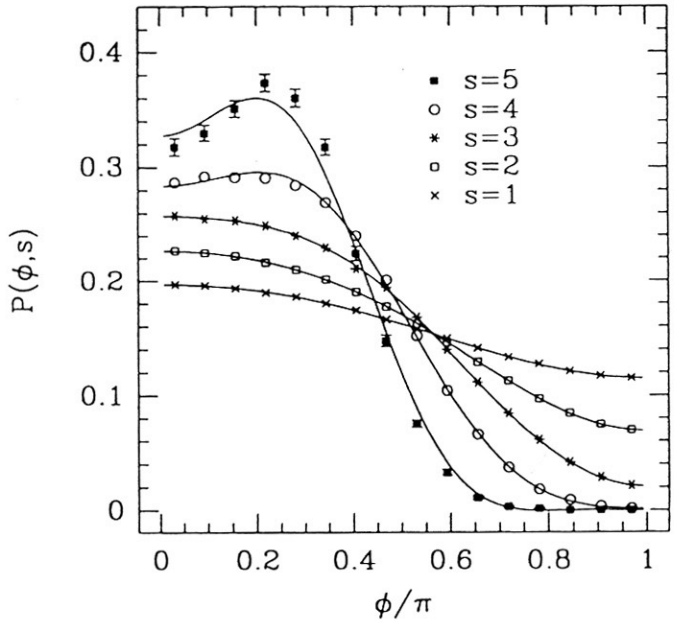

Figure 3. Probability distribution $P(\phi, s)$ of the branch orientation angle ϕ for branches of order s and fits with four cosine terms.

the branch orientation angle ϕ to be measured counter-clockwise from the position vector of the base of the branch to the branch orientation vector pointing from the base to the tip. Figure 3 plots the probability distribution $P(\phi, s)$ of ϕ for the branch order $s = 1$ to 5. The data were averaged over 50 clusters of one million particles each. The distribution $P(\phi, s)$ can be approximated reasonably well by a cosine curve for n close to one. In the cosine expansion of $P(\phi, s)$, the coefficient of the $\cos(\phi)$ term decreases rather quickly with s, while that of $\cos(2\phi)$ is negative and rising up towards zero. The coefficients as functions of s are not well described by simple functional forms.

The local anisotropy of DLA is far from being stabilized for clusters of size $N \lesssim 10^6$. If we extrapolate equation (3) to $N \to \infty$ we would get $\lim_{N\to\infty} P(\theta, N) = 1/2\pi$, meaning that DLA is locally *isotropic* asymptotically. The reliability of this extrapolation deserves special attention due to its simplicity and excellent agreement with simulations for $300 \lesssim N \lesssim 10^6$. At finite N, the anisotropy can quantitatively be expressed by the proportion P_F of forward sticking events given by $P_F \simeq 0.5 + 2a_1 N^{-\gamma_1}$ from equation (1). For $N = 10^3$ to 10^6, P_F decreases from 80% to 65%. The anisotropy is still strong. Achieving approximate isotropy with, for example, $P_F \lesssim 55\%$ requires $N \gtrsim 10^{11}$!

The distribution $P(\phi, s)$ for the branch orientation also tends to be flattened for decreasing s, which corresponds to a deeper level of side-branching. As the cluster size increases, the branches for any fixed s correspond to increasingly deep side-branches, thus we expect $P(\phi, s)$ to converge to the constant $1/2\pi$. This asymptotic isotropy is closely related to that of the sticking angle, since the manner in which the particles stick directly determines the orientation of the branches. It may appear that the asymptotic local isotropy of attachment contradicts the radial geometry of the first order ($s = 1$) branches. The resolution lies in the fact that the branch order is defined only *after* growth is complete. The scheme of ordering is such that it effectively groups only the forwardly attached particles selectively into the $s = 1$ branches. In fact, these main stems have asymptotically vanishing weight compared to the whole cluster and their anisotropy does not generalize to the distribution of all the attachment processes.

Similar asymptotic local isotropy applies to many other deterministic fractals. Take the Koch curve as an example. For the pre-fractal at early generations, the distribution of the orientation of the line segments is anisotropic and depends on the initial generator. For higher generations the orientation randomizes and local isotropy is approached asymptotically.

In conclusion, we have demonstrated numerically that the structure of DLA is locally isotropic in asymptotically large clusters. The computation of the distribution of the angle of attachment is one of the most accurate non-trivial measurements ever done on DLA. The statistical errors of our data points are as small as 0.2% at cluster size $N = 614\,400$. The excellent one parameter fit of the corresponding distribution to the very simple analytic form of a cosine is rare and deserves special attention. A similar approach may also be attempted for the dielectric breakdown model [2].

After this letter was written, Hegger and Grassberger [6] published very similar numerical measurements on the angle of attachment for diffusion limited deposition. Their result indicates the same asymptotic local isotropy and is in full agreement with ours. However, the functional form of the probability distribution of the angle was not examined.

References

[1] Witten T A and Sander L M 1981 *Phys. Rev. Lett.* **47** 1400; 1983 *Phys. Rev.* B **27** 2586
[2] Vicsek T 1992 *Fractal Growth Phenomena* 2nd edn (Singapore: World Scientific)
[3] Kaufman H unpublished
[4] Feder J, Hinrichsen E L, Måløy K J and Jøssang T 1989 *Physica* **38D** 104
[5] Ossadnik P 1992 *Phys. Rev.* A **45** 1058
[6] Hegger R and Grassberger P 1994 *Phys. Rev. Lett.* **73** 1672