## LETTER TO THE EDITOR

## Self-similarity of the loop structure of diffusion-limited aggregates

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Abstract. The structure of fjords in diffusion-limited aggregation (DLA) clusters can be described in terms of the loop size distribution  $n_R(x)$  which is the normalised number of loops with a neck to depth ratio x within a circle of radius R centred at the origin of the cluster. We find from the numerical study of very large off-lattice aggregates that  $n_R(x)$  converges quickly to a limiting distribution with a well defined smallest ratio  $x_{\min}$  larger than zero indicating the self-similarity of the loop structure. If the fjords are self-similar, i.e. they do not have long, tube-like structures, one does not expect a phase transition in the multifractal spectrum of growth probabilities of typical DLA clusters generated on the plain. Our study is essentially statistical and we cannot rule out the possibility of such 'rare events' (e.g. the occurrence of a few loops with anomalously small x) which may result in a qualitatively different behaviour concerning the multifractal spectrum.

The recent intense interest in the properties of diffusion-limited aggregation (DLA) clusters (Witten and Sander 1981) has been motivated by both the applicability of the DLA model to real physical phenomena (Meakin 1987, Stanley and Ostrowsky 1989, Vicsek 1989) and the rich scaling behaviour of the aggregates themselves. The related studies have revealed the complexity of the geometry of diffusion-limited aggregates which, in turn, is responsible for the complicated nature of such physical quantities as the distribution of the growth probabilities (see, e.g., Amitrano et al 1986, Hayakawa et al 1987).

In particular, it has been shown that the DLA clusters are fractals (Mandelbrot 1982) with anisotropic density correlations (Meakin and Vicsek 1985, Kolb 1985) expressing the geometrical constraint that a growing aggregate is bound to develop branches advancing preferably in the radial direction. Such a structure can be described in terms of a hierarchy of open loops which form deep fjord-like structures. The directedness of the branches does not violate self-similarity, however, it has relevant consequences concerning the growth probability distribution (Mandelbrot and Vicsek 1989).

The distribution of growth probabilities  $p_i$  is defined as the probability that the next particle is added at site *i*. The question whether the set of  $p_i$  represents a fractal measure has been addressed by many recent papers leading to a controversy concerning the possibility of a phase transition in the corresponding multifractal spectrum (in the following we shall consider the two-dimensional case without the loss of generality).

Early numerical work (Amitrano et al 1986, Hayakawa et al 1987) indicated the existence of a regular spectrum  $f(\alpha)$ , where  $f(\alpha)$  denotes the fractal dimension of the set of growth sites with growth probabilities  $p = L^{-\alpha}$ , where L is the linear size of the

aggregate. For a 'regular' multifractal spectrum there exists a largest  $\alpha$  such that  $f(\alpha) \leq 0$  for any singularity exponent larger than this  $\alpha_{\max}$ . This corresponds to a situation in which the smallest growth probability can be expressed in the form

$$p_{\min} \sim L^{-\alpha_{\max}}.\tag{1}$$

In the view of a few very recent studies, however, further possibilities leading to the breakdown of the above standard multifractal behaviour should be considered. In an exact enumeration study Lee and Stanley (1988) obtained a behaviour qualitatively different from (1) for a complete ensemble of small DLA clusters. Similar conclusions were made in the investigations of aggregate-like Julia sets (Bohr et al 1988) and typical diffusion-limited aggregates (Blumenfeld and Aharony 1989, Havlin et al 1989). If the growth sites are extremely strongly screened one can assume that their probabilities to grow tend to zero exponentially with the linear size of the aggregates

$$p_{\min} \simeq e^{-aL} \tag{2}$$

where a is some constant. The most recent numerical results (Schwarzer et al 1990), however, suggest that the scaling of the smallest growth probability is given by the expression

$$p_{\min} \simeq e^{-b(\ln L)^2} \tag{3}$$

with b being a constant. Before describing our results we discuss the geometrical pictures corresponding to the behaviours given by (1-3) and the consequence of (1-3) regarding the multifractal spectrum.

- (i)  $p_i$  scales as indicated by (1) if, as it has been argued (Mandelbrot and Vicsek 1989, Harris and Cohen 1990), the structure of the aggregates is such that there are always wedge- or cone-shaped major fjords which lead to the most screened sites.
- (ii) The growth probability becomes exponentially small (2) in clusters consisting of long 'tubes'. The main feature of these tubes needed for (2) is that the ratio x = w/l of their width w to their length l goes to zero with the size of the clusters as 1/L. In this case there is a phase transition in the multifractal spectrum at some  $\alpha$  where  $f(\alpha)$  becomes non-analytic. Since (2) corresponds to contributions coming from arbitrary large  $\alpha$ , and the multifractal spectrum is always convex, it follows from (2) that the shape of  $f(\alpha)$  is such that after reaching its maximum it remains constant as  $\alpha \to \infty$ .
- (iii) Interestingly, (3) results in a phase transition in  $f(\alpha)$  as well, although the growth probabilities go to zero much slower than in the case (ii). This behaviour was indicated by numerical determination of  $p_{\min}$  for clusters containing 2600 particles (Schwarzer *et al* 1990). It has been suggested that (3) corresponds to a hierarchy of open spaces (loops) whose linear size scales down according to some exponent.

Since it is the actual structure of the fjords in the aggregates which determines the scaling behaviour of the smallest growth probability we have undertaken a large scale, detailed study of the geometrical properties of these fjords. We have analysed the geometry of eight off-lattice DLA clusters each consisting of one million particles (the coordinates of the particles were given to us by P Meakin). The clusters were grown using an off-lattice algorithm (Tolman and Meakin 1989), but the configuration was stored by relaxing the particles to the closest grid points of an underlying square lattice. This means that the smallest possible distance between two particles in the clusters is equal to one particle diameter. The caliper diameter of the clusters was about 6000 lattice units.

We assumed that the structure of the clusters can be described in terms of a hierarchy of open loops each having a neck of linear size w and a depth l (where l is the distance from the neck to the point which is closest to the origin and belongs to the same loop). w and l were determined in the following way. A walker was allowed to move along the perimeter sites of the cluster, and its position was recorded. Then, each time the walker moved towards the centre crossed circles concentric to the origin and of given radii  $R_j$ , we stored its position. The neck size was given by the distance of this point from the position where the walker crossed the same circle next (moving outward). Simultaneously, we recorded the coordinates of the point which was visited by the walker between the two crossings and was closest to the origin. In fact, to account for overhangs, w was determined taking into account the positions of the walker (moving outward) in the layers preceding and exceeding  $R_j$  by k lattice units, where k was typically equal to 25.

The structure of loops was investigated by three methods. In the first approach we determined the loop size distribution function  $n_R(x)$  which is the normalised number of loops with a neck to depth ratio x = w/l up to the radius R. The data were collected for increasing radii separated by 50 lattice units.  $n_R(x)$  was normalised in such a way that it could be considered as a probability distribution. The results are displayed in figure 1. The main conclusion which can be drawn from this figure is that the normalised loop size distribution falls onto the same curve for various R; its convergence to a limiting distribution is apparent for the sizes considered in our simulations. As one can see from this figure 1 there is no tendency for the maximum to be shifted towards the smaller ratio values as R is increased which indicates that the average neck to depth ratio remains constant. The fact that  $n_R(x)$  becomes stationary is very important; it supports the assumption that the above discussed behaviour is expected to hold in the asymptotic regime as well.

The next figure (figure 2) shows how  $x_{\min}$ , the smallest neck to depth ratio, changes as R is increased. This figure suggests the saturation of  $x_{\min}$  for large R. The data are shown up to the radius of 1900 particle diameters, because this is the region within which the structure of the cluster can be considered as almost entirely completed.

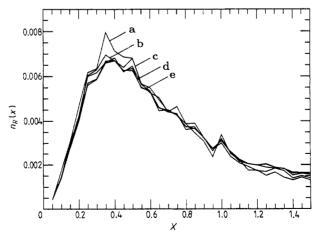


Figure 1. The normalised distribution  $n_R(x)$  of the number of loops with a neck to depth ratio x within a circle of radius R. The curves a-e were obtained for R = 500, 1000, 1500, 2000 and 2500, respectively.

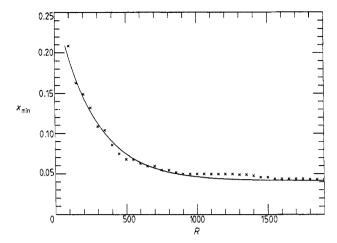


Figure 2. This figure demonstrates how the smallest observed ratio  $x_{\min}$  saturates as loops within circles of increasing radii are checked. The continuous curve is a fit of the form  $0.222 \exp(-0.0038R) + 0.0415$  to the data obtained by averaging over eight clusters.

To indicate saturation, an exponentially decaying function of the form  $0.222 \exp(-0.0038R) + 0.0415$  is fitted to the results. Because of the nature of our algorithm, it is possible that some of the anomalous places in the cluster are not located in our approach. Therefore, the data in this figure should be regarded as corresponding to an overall behaviour rather than showing the rare events.

Finally, we determined for various values of R the average size  $w_0$  of those necks which belonged to large loops. In these calculations only those necks were considered from which the perimeter walk directly reached sites being within a circle of radius r=30 centred at the origin (i.e., the depth of these necks was R-r). The middle curve in figure 3 shows the data obtained by averaging the size of such necks, first, over a single cluster, and then over eight clusters. The lower plot corresponds to the minimum neck sizes for each R averaged over the clusters. The upper plot shows similar data

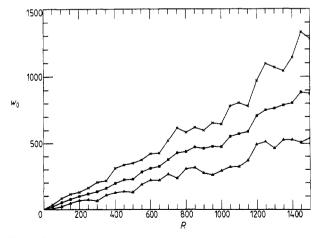


Figure 3. Results obtained by calculating the average neck sizes  $w_0$  (see the text) for various radii R. The three curves correspond to the behaviour of the largest  $(\times)$ , average  $(\square)$  and the smallest  $(\triangle)$  necks.

for the largest necks. Again, this figure is consistent with the existence of fjords having a finite opening angle directly related to the slope of the lower curve.

In conclusion, we have carried out a large scale study of the geometrical structure of fjords in diffusion-limited aggregates. Our results demonstrate that these fjords do not contain long, tube-like structures of length proportional to the linear size of the aggregates. This information of geometrical origin can be used to make the following statement about the growth probability distribution. Since the wedge-shaped structures with a finite opening angle result in a power-law scaling of the smallest growth probability with the cluster size, our data suggest the absence of a phase transition in the multifractal spectrum of DLA clusters grown in two dimensions. However, our study is inherently statistical and we cannot exclude the possibility of configurations which locally violate overall self-similarity. Such rare events as the occurrence of loops with an anomalously small neck to depth ratios (Schwarzer et al 1990) may lead to a behaviour (e.g. phase transition) qualitatively different from that implied by the overall self-similarity of the fjords. Finally, our findings are consistent with the self-similarity of the aggregates themselves, while other types of fjord shapes would correspond to a considerably more complicated picture.

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