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Crosscut Analysis of Large Radial DLA: Departures from Self-Similarity and Lacunarity Effects.

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Abstract. – In order to understand better the morphology and the asymptotic behavior in Diffusion-Limited Aggregation (DLA), we studied a large number of very large off-lattice circular clusters. We inspected both dynamical and geometric asymptotic properties via the scaling behavior of the transverse growth crosscuts, *i.e.* the one-dimensional cuts by circles. The emerging picture corresponds qualitatively and quantitatively to the scenario of infinite drift that starts from the familiar five-armed shape for small sizes and proceeds through increasingly tight multi-armed shapes. The transverse crosscuts show quantitatively how the lacunarity of circular clusters becomes increasingly «compact» with size. Finally, we find the transverse-cut dimensions to be in agreement for clusters grown in circular and cylindrical geometry, suggesting that the question of universality is best addressed on the crosscut.

Many fractal growth processes observed in nature [1] can be modeled using the so-called Laplacian models such as Diffusion-Limited Aggregation (DLA) [2] and Dielectric-Breakdown Model (DBM) [3]. The rules governing the growth of DLA and DBM are simple. The structures generated, however, are very complex fractal patterns which result from a self-organizing stochastic process based on the Laplace equation. Even to this day basic questions remain open and several clues suggest that simple self-similarity is not the only possible scenario: conflicting values of the fractal dimension, failure of the asymptotic regime to be observed, weak universality with respect to the growth geometry and deviations from

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self-similarity [1, 4-10]. Thus, while DLA is a critical phenomenon with no characteristic length, it differs in diverse ways from ordinary critical phenomena (such as percolation or Ising clusters) [5, 10].

Given the richness of DLA growth and structure, it stimulated a huge number of investigations [1,4], and our systematic study of the properties of very large DLA clusters uses improvements in earlier-generation algorithms [4, 10, 11]. For radial DLA we found in a previous paper [10] that the active region shows deviations from simple self-similarity that decay extremely slowly and in a non-trivial way (no observed finite crossover). This «infinite drift» scenario [8] could account for several previously proposed corrections to scaling, non-trivially related one to each other, and it calls for other quantitative characterizations of the clusters such as, for example, fractal lacunarity [12-14]. Lacunarity is indeed related to the size distribution of the holes and characterizes fractal sets beyond their fractal dimension (1).

To seek a deeper understanding of this scenario and its implications, this paper investigates the geometric structure of circular DLA via the transverse growth crosscuts of clusters, *i.e.* the one-dimensional cuts by circles. On these cuts, we study the fractal dimension, the gap distribution, the behavior of the maximum gap and other morphological properties. We find that the fractal dimension from transverse cuts is $D \approx 1.65$ instead of the expected value $D \approx 1.71$. The difference is a correction term that can be accounted for by a power law behavior in the prefactor of the mass-radius relation. This prefactor is a measure of mass lacunarity and it may explain reported disagreements between different measurements of the fractal dimension. The analysis of the distribution of gaps is also fully in agreement with this picture, and the maximum gap behavior is compatible with a lacunarity effect. In conclusion, our results support the «infinite drift» scenario [8] in which, as samples grow, some properties of radial DLA have diverse departures from self-similarity and the cluster drifts toward an increasingly tight multi-armed shape. The crosscut analysis helps to understand the relations between some scaling corrections and lacunarity, and gives some insight on the «weak» universality of DLA.

Our study is based on 50 clusters of 1 M particles and 20 clusters of 10 M particles, grown under meticulous control to avoid large-scale instabilities due to approximations in the off-lattice algorithm [15]. The construction of radial DLA clusters begins with a particle at a random location on a "birth" circle at some distance to the existing cluster. The new particle undergoes Brownian motion until it comes in contact with the cluster, at which point it becomes permanently stuck. A new particle is then added at random along a revised birth circle, and the process continues.

We analyzed the DLA clusters via one-dimensional cuts by circles of varying radius R. Each of these «crosscuts» is on average transverse to the growth direction of the aggregate. This kind of analysis has been already used in ref. [8, 16]. In practical terms, we define an annulus whose thickness is one particle radius on either side of the circle of radius R. All particles whose center falls inside this annulus intersect the target crosscut circle (and vice versa). «Generically», a one-dimensional cross-section (slice) of a fractal of dimension D is a fractal dust of dimension D-1 [12]. For DLA the widely accepted value D=1.71 obtained from the mass-radius relation yielded the heuristic expectation that the fractal dimension of the crosscut is $D_c=0.71$.

⁽¹⁾ The properties of a fractal set are not completely determined by the fractal dimension D. In ref. [13,14], fig. 1 shows a stack of Cantor sets that share the same fractal dimension but are indeed very different. To face the need to distinguish between those fractals, it is worth introducing the concept of lacunarity.

We measured the fractal dimension of the crosscuts of 20 clusters of 10 M particles by intersecting circles with radii ranging from R=200 to R=5000 particle diameters. The radius did not exceed 3/4 of the radius of gyration of the clusters to ensure that our study concerns the frozen part of the aggregates that is extraordinarily unlikely to change with further growth. To obtain the fractal dimension on the crosscut our first method is box-counting. We rescale each crosscut to the unit circle and then coarse grain it with one-dimensional segments of different lengths. Our second method uses the mass-radius relation in one dimension: the variation of the total number of particles within an increasing distance from an occupied point of the crosscut. The fractal dimension is obtained from the log-log plots of the number of occupied boxes or cluster points vs, the scaling size. To eliminate noise, we always average the data over several clusters of the same size. Both methods are clearly affected by finite size effects for very small sizes (particle radius) and very large sizes (the entire crosscut). In our experience a reliable result is possible only with very large samples that ensure enough statistics on the crosscuts.

Strikingly, the fractal dimension obtained in this way is lower than the expected value 0.71. In fact, both methods agree and give a fractal dimension that is practically constant from $R \simeq 1500$, its average value being

$$D_c = 0.65 \pm 0.01. \tag{1}$$

This result differs strongly from what was expected for simple self-similarity. As already done in [8], it can be related to a mass-lacunarity effect in the measurement of the fractal dimension. In fact, we can write the exact scaling relation on a crosscut as

$$N_{\rm c}\left(\frac{R}{l}\right) = \lambda(R)\left(\frac{R}{l}\right)^{D_{\rm c}},\tag{2}$$

where N_c is the number of particles and l is the scaling length. Along a crosscut, R is a constant (the radius of the intersection), therefore $\lambda(R)$ is a constant prefactor. The fractal dimension of the crosscut is obtained from the scaling with respect to the size l and with our method we have the result of eq. (1). For the whole cluster, the mass-radius fractal dimension, *i.e.* scaling behavior in the two-dimensional embedding space, is related to the scaling on the crosscuts by the formula

$$N_{\rm t}(R) = \int_{0}^{R} \lambda(r) \left(\frac{r}{l}\right)^{D_{\rm c}} \mathrm{d}r, \qquad (3)$$

where $N_{\rm t}$ is the total number of particles in a cluster of radius R and l is fixed and corresponds to the resolution scale. As mentioned above, the scaling behavior $N_{\rm t}(R) \sim R^{\rm D}$ with D=1.71 is confirmed over several decades. If $\lambda(R)$ changes gradually, the only one way for log $N_{\rm t}$ to be represented by a straight line is to have $\lambda(R) \sim R^{\delta D}$. It follows, therefore, that eq. (3) gives $N_{\rm t}(R) \sim R^{1+D_{\rm c}+\delta D}$, and comparing this relation with the mass-radius behavior yields $\delta D=D-1-D_{\rm c}$. By substituting the numerical values obtained for the crosscut fractal dimension it suggests for the lacunarity prefactor bias the value

$$\delta D = 0.06 \pm 0.01 \,. \tag{4}$$

The prefactor $\lambda(R)$, characteristic of the clusters, is a numerical rate that measures one aspect of lacunarity. Therefore, our results indicate that the lacunarity decreases relative to the radius as the DLA cluster grows and becomes increasingly compact.

Accepted values of D range from $D \approx 1.65$ for clusters grown in cylindrical geometry, to

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 $D \simeq 1.71$ when the fractal dimension is calculated using the mass-radius relation in radial geometry. Thus the lacunarity effect present in DLA grown in circular geometry could explain the discrepancies among various measurements of the fractal dimension. Moreover, the value of the fractal dimension is the same for circular crosscuts and for crosscuts of clusters grown in cylindrical geometry, and does not depend upon the measurement technique. This seems to suggest that the question of the universality of DLA is best addressed on the crosscuts. In fact, this is the only set that possesses an important geometric characteristic that is independent of the growth boundary conditions: it is always transverse to the overall growth direction. Therefore it is possible that transverse-crosscut measurements take into account the intrinsic growth dynamics of the phenomenon but leave aside the effects induced by the geometry of the boundary conditions.

In further support of the previous numerical results we performed a detailed analysis of the gap distribution on the crosscuts. In this kind of analysis (introduced in ref. [8]) a gap is defined as an interval whose end points belong to the fractal set but whose interior points do not. We rescale each intersection by the radius R, obtaining angular crosscut gaps. In the case of fractal sets the probability of finding an angular crosscut gap of length $> \gamma$ follows the gap number rule $P(\Gamma > \gamma) \sim \gamma^{-D_c}$. We tested this scaling behavior by plotting the graph of log $P(\Gamma > \gamma)$ vs. log γ , providing another numerical estimate of D_c . Figure 1 shows the doubly logarithmic plot of this gap distribution. The expected two cut-offs are given by the particle size and the maximum value of the gap. The slope of the straight part between these cut-offs is found to be $D_c = 0.65$, in agreement with our previous measurements.

In addition, a careful look at distributions obtained from crosscuts with increasing radius R shows an unexpected behavior: the upper cut-off, *i.e.* the largest angular gap, seems to decrease as the cluster grows. This is confirmed by following directly the size of the largest gaps as a function of the size of the clusters. Figure 2 plots on a doubly logarithmic scale the average angles of the six largest gaps with respect to the radius of intersection of the crosscut. The usual picture is that DLA evolves rapidly towards a five-fold symmetry (five main branches), suggesting that largest gaps fluctuate around the constant value $\gamma = 2\pi/5$. In sharp contrast, our data show that the largest gaps continually decrease as R increases and the value of $\gamma_{\rm max}$ soon becomes definitely $< 2\pi/5$. The fact that the few largest gaps become continually smaller as the cluster grows indicates that the structure drifts toward a tighter multi-armed shape. It is simple to recognize that this manifests a decreasing lacunarity in the crosscuts. In fact, the narrowing of the larger fjords implies the presence of an increasing

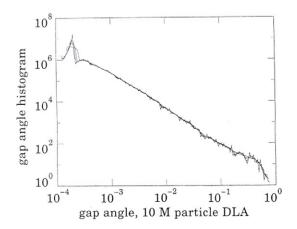


Fig. 1. - Plot of the crosscut gap distribution.

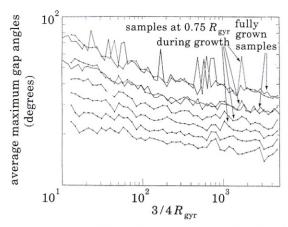


Fig. 2. – Behavior of the average angles of the six largest gaps as a function of the crosscut radius. The plot is on a doubly logarithmic scale.

number of main branches, and therefore a less lacunar structure even if the scaling properties on the crosscut remain the same.

In order to visualize this effect, we introduced a novel graphical way to see the structure of DLA clusters, namely polar coordinates. Figure 3 shows a circular DLA cluster in which the abscissa is the angle on the circumference and the coordinate is the radial distance from the seed. The top of the figure begins at 3/4 the radius of gyration of the DLA. At this radius, the 12 largest gaps are plotted as line segments. These gaps are then tracked inward, toward the center of the DLA, as they form the fjords. We found it graphically effective to measure and display the fjord morphology (the negative space) as opposed to the particle morphology. This figure is the pictorial counterpart of the quantitative analysis shown so far. The fjords' geometric behavior is especially well represented and confirms the previous analysis: the fjord widths tend, on the average, to decrease. Evidently, fjord widths correspond in this representation to angular gaps on the crosscuts and the perception that they "shrink" confirms pictorially the lacunarity effect on the structure's morphology.

In conclusion, our analysis supports the scenario in which radial DLA does not show finite crossover to self-similarity, presenting scaling corrections that are related to the non-equilibrium dynamical aspects of the phenomenon. In fact, the drift of the corrections to

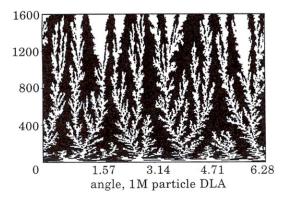


Fig. 3. – Polar-coordinates representation of a radial DLA cluster. Each point is identified by the radial distance from the seed and the corresponding angle on the circumference (see text for details).

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