THE GEOMETRY OF DLA: DIFFERENT ASPECTS OF THE DEPARTURE FROM SELF-SIMILARITY

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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 numbers of very large off-lattice circular clusters. We inspected both dynamical and geometric asymptotic properties, namely the moments of the particle's sticking distances and the scaling behavior of the transverse growth crosscuts , i.e., the one dimensional cuts by circles. The emerging picture for radial DLA departs from simple self-similarity for any finite size. It corresponds qualitatively to the scenario of infinite drift starting from the familiar five armed shape for small sizes and proceeding to an increasingly tight multi-armed shape. We show quantitatively how the lacunarity of circular clusters becomes increasingly "compact" with size. Finally, we find agreement among transverse cuts dimensions for clusters grown in different geometries, suggesting that the question of universality is best addressed on the crosscut.

INTRODUCTION

The first model of irreversible growth that generates fractal structures was Diffusion Limited Aggregation (DLA) [1], followed few years later by the Dielectric Breakdown Model [2]. These models account for origin of fractal structure in a great variety of process: dendritic growth, viscous fingers in fluids, dielectric breakdown, electrochemical deposition etc. [3,4]. DLA's most characteristic feature is that it is intrinsically critical and gives rise spontaneosly to a fractal structure. However, as soon as one tries to make this statement precise and quantitative, diverse problems appear, many of which are still open. We tackle the question of asymptotic self-similarity, which has not to this day been resolved adequately. In fact, several clues suggests that asymptotic self-similarity is not the only possible scenario: conflicting values of the fractal dimension, slow cross-over to the asymptotic regime, weak universality with respect to the growth geometry and deviations from simple self-similarity such as lacunarity etc. [1,5,6,7]. To gain a deeper understanding of these problems we have undertaken a systematic study of the properties of large numbers of very large clusters of off-lattice circular DLA.

The construction of DLA clusters is very simple. It 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 to the cluster, at which point it becomes permanently stuck. A new particle is then added at random along the birth circle, and the process continues. Our study is based on 50 clusters of 1M perticles and 20 clusters of 10M particles, grown under meticulous control to avoid large scale instabilities due to approximations in the alghorithm [8]. We investigate the geometric structure of circular DLA from two points of view. A) Growth of various relative moments and of the maximum cluster radius. B) Structure of the transverse growth crosscuts, 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 moments of the distribution of the particle's sticking distance fail to cross over to the behavior characteristic of self-similarity, even for very large clusters. The measurement of the fractal dimension on the transverse cuts reveal a correction term that can be accounted for by postulating a power law behavior in the prefactor that measures the mass lacunarity. This "misbehavior" may help understand previous disagreements between estimates of the fractal dimension and suggests, moreover, that the question of universality is best addressed on the transverse-cuts. The analysis of gap also is fully compatible with this picture,

and the maximum gap behavior is compatible with a lacunarity effect revealing that the cluster drift to an increasingly tight multi-armed shape.
The conclusion is that, for radial geometry, the present results support Mandelbrot's scenario [6] in which, as samples grow, some properties of DLA drift without non trivial limit and with diverse departures from the simple self-similarity for any finite size cluster. This paper is a survey of various forms of analysis and the corresponding results. For the sake of clarity and concision, we avoid most of the technical details that will be reported elsewhere [9,10,11].

"MOMENTS" ANALYSIS USING NORMALIZED SCALE FACTORS

First we investigated the distributions we call radial density profiles [9]. Given a cluster of N particles let $F(R,N)$ be the number of particles at a distance $\leq R$ from the seed. The particles that become part of the cluster when cluster size ranged between $N-\Delta N$ and N are said to form a cluster shell. The radial cluster shell profile is therefore

$$\Delta F(R,N) = F(R,N) - F(R,N-\Delta N) \tag{1}$$

For $\Delta N = N$, the cluster shell is of course the entire cluster. The function $F(R,N)$ is a cumulative distribution relative to distances << R, and is often usefully replaced by a density-like function. For that, one divides $\log R$ into uniform bins and plots the number of atoms in each bin. Letting R_k be the distance from the seed to the k-th particle (in order of absorption into the cluster), the moment-based scale factors of the cluster shell are defined as

$$\sigma_q(N,\Delta N) = \left[\frac{1}{\Delta N} \sum_{k=N-\Delta N+1}^{N} \left| R_k - \frac{1}{\Delta N} \sum_{n=N-\Delta N+1}^{N} R_n \right|^q \right]^{1/q} \tag{2}$$

In all cases, the $q \leq 0$ values of σ_q are without interest, being dominated by the inner cutoff due to the atoms' positive size. It is obvious indeed, that $\sigma_0 = 1$ and it is easy to show that $q < 0$ yields $\sigma_q \propto N^{1/q}$. To the contrary, when $q>0$ and $N >> 1$, the atom' size no longer matters. As $q \to \infty$, σ_q converges to $R_{max}(N)$, the largest distance from the seed to a particle in a cluster. It is important to notice that our shell analysis focuses particularly on the active region of the DLA cluster, where new particles become part of the aggregate. If, beyond atom size, the cluster had been statistically self-similar, it would have been characterized by a single well-defined D, and all the quantities $\sigma_q(N,\Delta N)$ would have been proportional (for large N) to $N^{1/D}$ and to each other.
The first inference from our data is a very familiar one. For $q=1$ we find, as previous authors observed again and again, that $\sigma_1(N) = 1/N \sum_{k=1}^{N} R_k \propto N^{1/D}$, where $D=1.71$. The study of the cluster shell moment continues more easily by normalizing and writing

$$\sigma_q(N) = \sigma_1(N) \cdot \lambda_q(N); \text{ and } R_{max} = \lim_{q \to \infty} \sigma_1(N) \cdot \lambda_q(N) . \tag{3}$$

For given N, the factors $\lambda_q(N)$ necessarily increase with q. A necessary but not sufficient condition for the clusters' being self-similar is that each $\lambda_q(N)$ tends to a limit as $N \to \infty$. To test this inference, we evaluated these quantities for logarithmically spaced values of N, and plotted them in doubly logarithmic coordinates (see fig.1). Our most striking observation is that

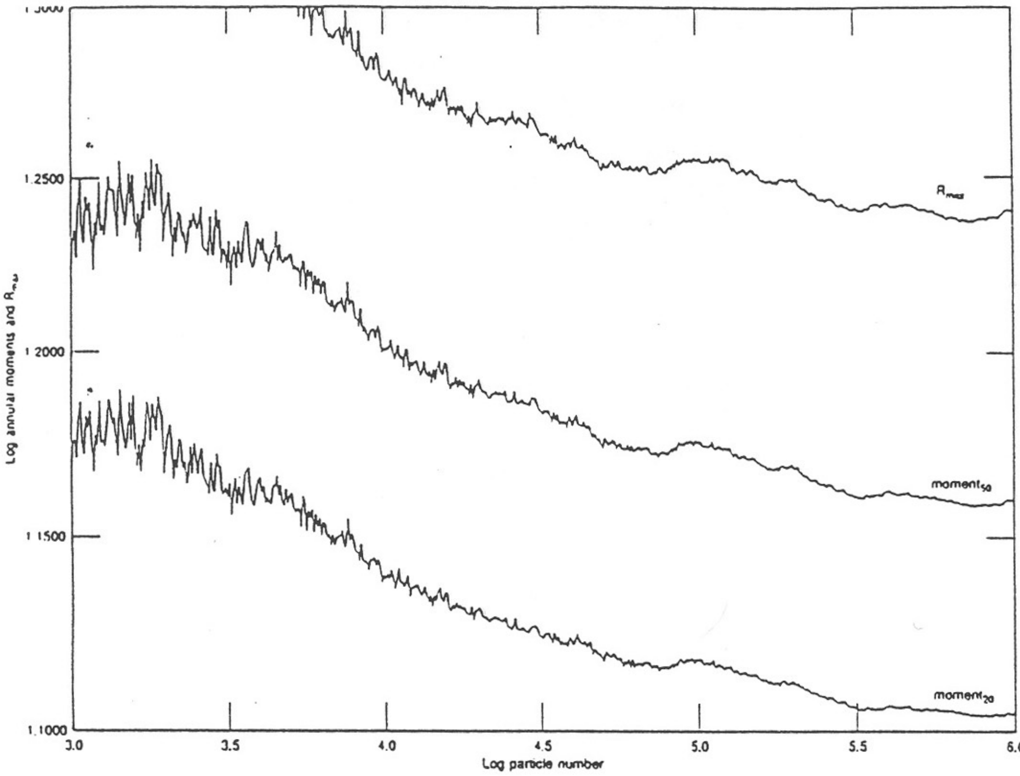

Fig.1) Analysis of DLA circular clusters based on the annular moments behavior. The various moments are normalized with respect to the first one and averaged over 50 one million particle clusters.

$\lambda_q(N)$ continually decrease for every value of $q>2$, including $q \to \infty$. In fig.1, it is easy to note the curvature of the $\lambda_q(N)$ up to the limit sample size. In order to visualize this behavior, we perform a local slope analysis of $\lambda_q(N)$ (fig.2). The local slope is definitely different from zero even for $N \cong 10^6$, and approaches zero only for very large size clusters. The cross-over from fast to slow decrease depends on q. For large q, $\lambda_q(N)$ continues to decrease rapidly, even in the range where, in absence of other information, it might have been argued that $\lambda_2(N)$ has finally settled down. Thus, $\lambda_q(N)$ continues to decrease non trivially up to the size of our simulations. Moreover, accurate analysis shows that a logarithmic behavior of the $\lambda_q(N)$ curves seems consistent up to $N \cong 10^4$ but not for larger clusters. Therefore, these results support the scenario of an "infinite drift" with persistent deviations from self-similarity for arbitrary large finite size samples and an unusual approach to the thermodynamic limit.

ANALYSIS OF THE TRANSVERSE GROWTH CROSSCUTS

The moments-based analysis in the preceding section is two dimensional, but can also be performed geometric and quantitative analysis on various one-dimensional sub sets of the cluster. In this spirit, we analyzed DLA via transverse growth crosscuts, namely one-dimensional cuts by circles of different radius R. Each is roughly transverse to the growth direction of the aggregate. "Generically" a one-dimensional cross-section (slice) of a fractal of dimension D is a fractal dust of dimension $D-1$. For DLA the widely accepted value D=1.71

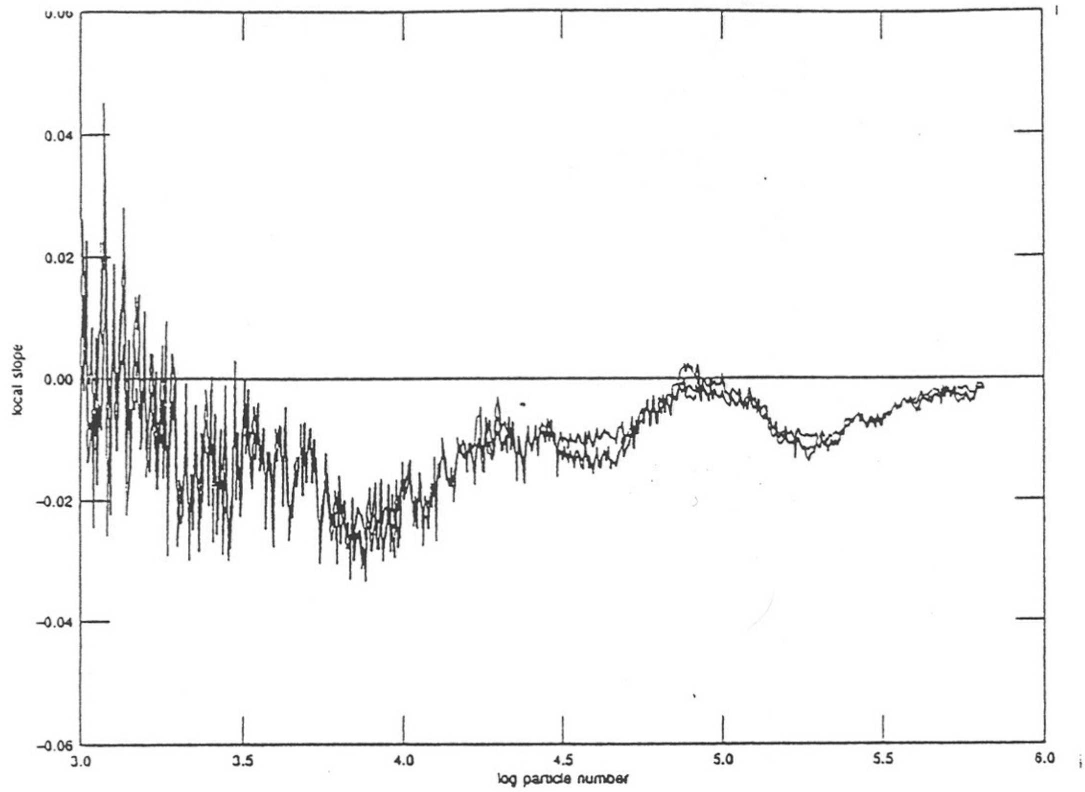

Fig.2) Local slope analysis of the moments behavior. Excluding the initial region (small sizes effect), the local slope is appreciably and definitely lower than zero.

obtained from the mass-radius relation yielded the heuristic expectation that the fractal dimension of the crosscuts is $D_c = 0.71$.

We measured the fractal dimension of the crosscuts of 20 10M particles clusters and intersecting circles with a radius 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 in the further growth.

Our first method to obtain the fractal dimension on the crosscut set is box-counting. We rescale each crosscut in the unit circle and then we coarse grain with boxes of different lengths. Our second method uses the mass-radius relation in one dimension. It measures the total number of particles within an increasing distance from an occupied point of the crosscut. The scaling behavior corresponding to the values of the fractal dimension is obtained from the log-log plots of the number of occupied boxes or cluster's points versus 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 corresponding to very small sizes (particle radius) and very large sizes (the entire crosscut), therefore, as the radius of intersection for the crosscut increases the fractal dimension is measured more precisely. 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 thus agree the fractal dimension is practically constant from $R = 1500$, its average value being

$$D_c = 0.65 \pm 0.01 \tag{4}$$

This result differs strongly from the expected one and can be related to a mass-lacunarity effect in the measurement of the fractal dimension. In fact, as already done in [6], we can write the exact scaling relation on a crosscut as

$$N_c\left(\frac{R}{\ell}\right)=\lambda(R)\cdot\left(\frac{R}{\ell}\right)^{D_c} \tag{5}$$

where N is the number of particles and ℓ is the scaling length. For each crosscut R is a constant, namely the radius of the intersection, and $\lambda(R)$ is therefore 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.(4).
For the whole cluster, we can relate the mass-radius fractal dimension, i.e., scaling behavior in the two-dimensional embedding space, to the scaling on the crosscuts by using the following formula

$$N_t(R)=\int_0^R \lambda(R)\cdot\left(\frac{R}{\ell}\right)^{D_c} dR \tag{6}$$

where N_t is the total number of particles in a cluster of radius R and ℓ is fixed and corresponds to the resolution scale. We know that

$$N_t(R)\propto R^D \tag{7}$$

with D=1.71 with a scaling behavior confirmed on several decades. If $\lambda(R)$ changes gradually, there is only one way for $\log(N_t)$ to be represented by a straight line; one must have $\lambda(R)\propto R^{\delta D}$ from which we have

$$N_t(R)\propto\int_0^R R^{\delta D}\cdot\left(\frac{R}{\ell}\right)^{D_c} dR\propto R^{1+D_c+\delta D} \tag{8}$$

By comparing this relation with eq.(7) we have

$$\delta D=D-1-D_c \tag{9}$$

and substituting the numerical values obtained for the crosscuts fractal dimension we can estimate the bias due to the lacunarity prefactor as :

$$\delta D\cong 0.05 \tag{10}$$

It easy to recognize that the prefactor $\lambda(R)$ is a characteristic of the clusters. It is a numerical rate that contribute to measuring lacunarity. Therefore, our results indicate that the lacunarity decreases as the DLA cluster grows and becomes increasingly compact. This lacunarity effect is specific to DLA grown in circular geometry and could explain the discrepancies among various measurements of the fractal dimension. The discrepancies among DLA clusters growing with different boundary conditions indicate a sort of "weak" universality of this phenomenon. However, it is worth observing that the value of the fractal dimension we measure for circular crosscuts is the same as that obtained for crosscuts of cluster grown in cylindrical geometry. 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 growth direction. Therefore it is possible that measurements on this set takes into account only the intrinsic growth dynamics of the phenomenon leaving apart the effects induced by the geometry of the boundary conditions.

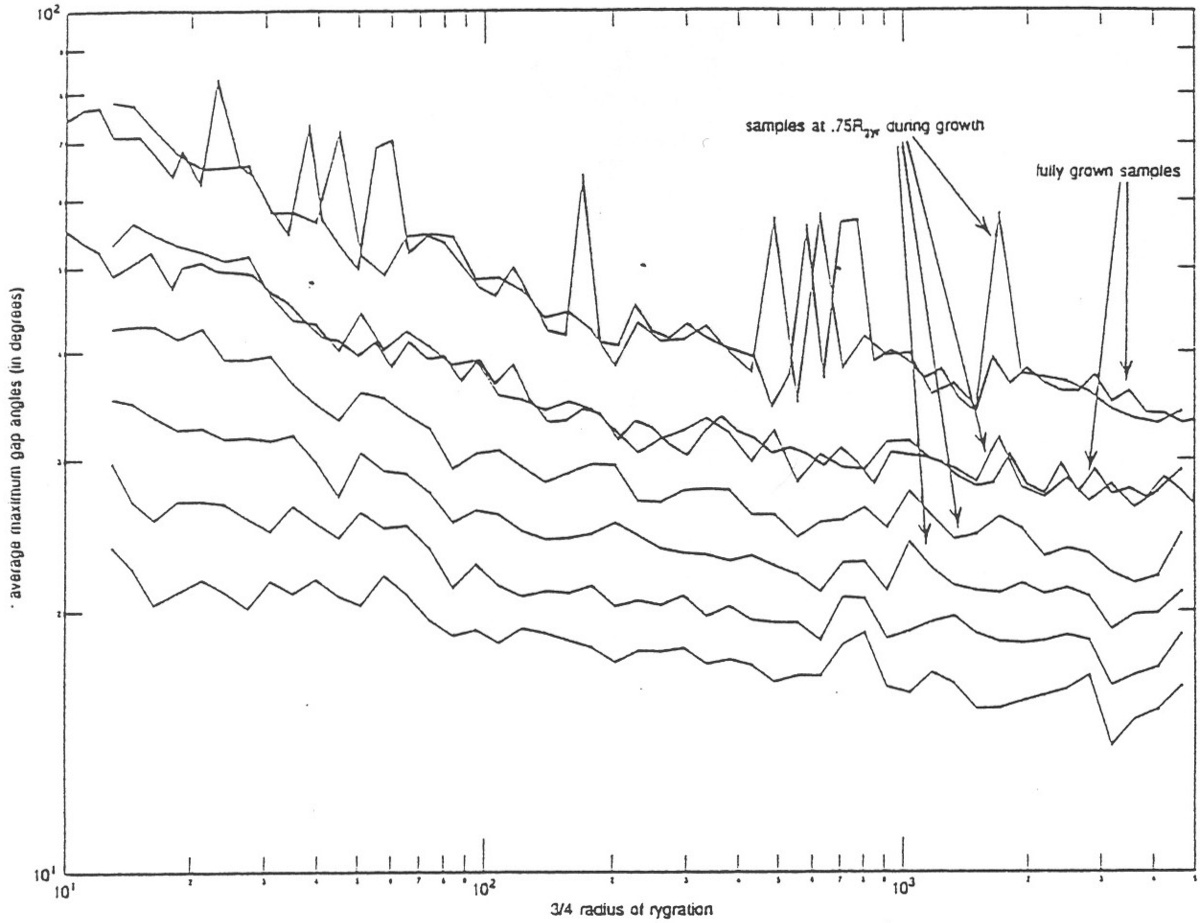

Fig.3) Average angles of the six largest gaps on the crosscuts with respect to the radius of intersection. The fact that the gaps become smaller corresponds to a decreasing lacunarity on the crosscuts.

In support of the previous numerical result we performed a detailed analysis of the gap lengths on the crosscuts. This kind of analysis was introduced in Ref [6]. A gap is defined as an interval whose end points belong to the fractal set but whose interior points do not. Denote by γ a possible value of the gap on the crosscut. To compare the gap length distribution for different crosscuts of different radius we rescale each intersection by the radius R, so that gaps are measured in angles. In fig. 3 we plot on a doubly logarithmic scale the average angles of the six largest gaps with respect to the radius of intersection of the crosscut. In the usual picture of DLA the structure 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 continuously decrease as R increases and the value of γ_{max} is definitely $< 2\pi/5$. The fact that the few largest gaps become smaller as the cluster grows indicates that the structure is drifts to a tighter multi-armed shape. It is simple to recognize that this corresponds to a decreasing lacunarity on the crosscuts. In fact, the narrowing of the larger fjords implies the presence of an increasing number of main branches, and therefore a less lacunar structure even if the scaling properties on the crosscut remain the same.

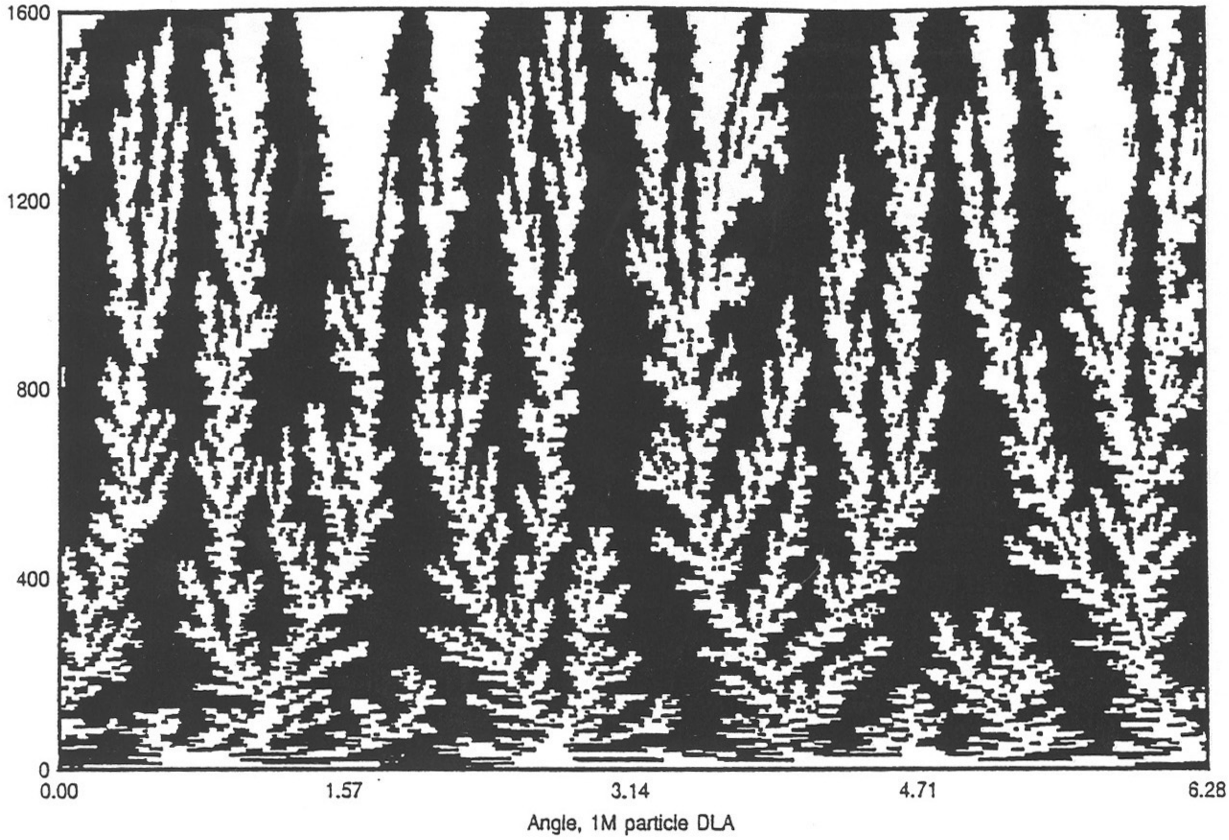

Fig4) Plot of a 1M particle DLA circular cluster in polar coordinates. The coordinates of each particle represent the radial distance from the seed and the angle on the circumference.

In order to visualize this effect we introduced a novel graphical way to seek on the structure of DLA clusters, namely polar coordinates. Fig.4 shows a circular DLA cluster in which each point is identified by the radial distance from the seed and the corresponding angle on the circumference. This figure is the pictorial counterpart of the quantitative analysis shown so far. The fjords' geometric behavior is particularly well represented and, confirming the previous analysis, we observe that the fjord widths tend, on the average, to decrease. Evidently fjords widths correspond to angular gaps on the crosscuts and the perception that they "shrink" is indeed the pictorial confirmation of the lacunarity effect on the structure morphology.

DISCUSSION AND CONCLUSION

Our analysis of very large DLA clusters suggests a new picture of this phenomenon, and likely of other irreversible growth phenomena. In fact, DLA is mainly a non-equilibrium phenomenon in which the dynamical aspects play a major role. We can identify two different cluster regions. The first is a fully grown core that will not be further modified by growth. The second region is the active zone where the growth process continues. This situation differs strongly from the usual equilibrium problems, such as percolation and the Ising clusters, where there is no distinction between cluster regions. Moreover, in these problems the deviations from the asymptotic behavior is solely due to finite size effects. Growth phenomena, specifically DLA, also raise the problem of the internal boundary conditions, i.e., the growing structure itself and the irreversible nature of the process. This means that the "old" part of the cluster had been "young" and preserves a memory of the dynamical process by which it has been generated.

This effect is emphasized in the circular DLA, in which it is the growing structure itself that defines the boundary conditions size. That dynamical aspects are present in this geometry from the early stages of the growth process up to the very large size we have analyzed and suggest the infinite drift scenario supplied previously [6]. This scenario allows persistent deviations from self-similarity for arbitrary large finite size samples and therefore calls for other quantitative characterization of the clusters such as lacunarity.

The present picture it is also compatible with a "weak" universality of DLA. In fact, the dynamical process generates different transient effects depending upon the growth geometries. For example there are numerical indications that large DLA clusters grown in cylindrical geometry do not present deviations from self-similarity. This is probably due to the fact that the cylinder size is defined externally and it does not depend upon the growth process. In this sense we believe that keeping in mind the dynamical nature and the "weak" universality of the DLA phenomenon helps discriminate among the quantities and features that are really universal and thus which can be considered as clues or elements for a theory of fractal growth [12,13].

Acknowledgments

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