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LETTER TO THE EDITOR

Diffusion-limited aggregation with radial bias

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Abstract. We introduce radial bias into off-lattice diffusion-limited aggregation in order to simulate radial anisotropy present in experiments on viscous fingering in liquid crystals. For large cluster sizes the model is also expected to provide information about the asymptotic shape of the ordinary off-lattice diffusion-limited aggregates. We find that for large enough radial bias the overall shape of the clusters becomes star-like with a spontaneously selected number of main arms which is either four, or less frequently five. The crossover to this structure is accompanied by a change in the radius of gyration exponent which approaches the value 1.5 previously observed for systems with axial anisotropy or very large clusters grown on the square lattice.

Many of the complex geometrical patterns in nature are produced by processes in which the velocity of the interface is determined by a diffusion-type field. In order to simulate the growth of such patterns Witten and Sander [1] introduced a model called diffusion-limited aggregation (DLA) which was later shown to lead to clusters strikingly similar to the patterns found in the related experiments [2].

Recent studies of DLA have revealed that diffusion-limited aggregation and its generalisations exhibit a much richer behaviour than originally expected. For moderate cluster sizes DLA clusters were found to have, on average, a nearly circular envelope and the power law decay of the density correlations within these clusters was shown to correspond to a fractal dimension independent of the details of the lattice [1, 3, 4] on which the growth was simulated. However, as more effective methods for generating diffusion-limited aggregates were developed [5, 6] it became clear that the anisotropy of the underlying lattice has an important effect on the overall shape of very large clusters. At sizes $M \sim 10^5$ (where M is the number of particles in the cluster) the aggregates grown on the square lattice attain a diamond shape. It has very recently been shown that this overall shape is not stable and for $M > 10^6$ it crosses over into a dendritic structure with four major arms along the axes of the lattice [7].

Simulations of aggregation models closely related to the original version of DLA have resulted in similar conclusions [8-15]. Introduction of a curvature-dependent sticking probability resulted in a dendritic growth along the axes of the square lattice [8]. Very recently it has been argued that taking an average over the random walkers [9, 10] may reveal the asymptotic behaviour of clusters using considerably smaller M [11]. In this method a particle is added to the cluster only if the perimeter site it

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touches has been previously visited by other random walkers (or chosen in the dielectric breakdown model) a given number of times. DLA clusters grown on the square and triangular lattices using averaging were found to reflect the symmetry of the lattice already for $M \sim 10^3$ [11]. Analogous results were obtained by Nittmann and Stanley [12] who applied averaging to the dielectric breakdown model. The prevailing anisotropy in these cases changes the radius of gyration exponent of the clusters from 1.7 to about 1.5, indicating a crossover in the global behaviour of the aggregates as a function of M. The same exponent (1.5) was shown to apply for DLA clusters grown with a uniaxial bias of the sticking probability [14]. Deterministic models of DLA also led to clusters reflecting the lattice structure [15].

The question of the asymptotic shape of the off-lattice aggregates, however, is still open. Does the envelope of off-lattice DLA clusters remain nearly isotropic in the $M \to \infty$ limit? As an alternative, one cannot exclude the possibility that, with the number of particles in the clusters going to infinity, the aggregates develop elongated needle-like arms as a result of the dominance of singularities at some of the most advanced tips. Up to now the largest off-lattice clusters which have been generated ($M \sim 10^5$) do not show the signs of such a crossover. On the other hand, some growth models have been shown to exhibit extremely slow crossovers [7, 11, 16] as a result of the interplay between the fluctuations and some of the regular features of the aggregation process. A simple model which is expected to be related to the asymptotic behaviour of off-lattice DLA clusters (but at much smaller sizes) should be helpful in our understanding the shape of very large off-lattice aggregates.

In this letter we investigate the following simple modification of the two-dimensional off-lattice diffusion-limited aggregation process. As in its original form, the particles released far from the aggregate undergo a diffusional motion during which the centre of a circular particle is moved with the same probability to any point on the perimeter of a circle centred on the position of the particle. If the particle is far from the cluster the radius of this circle is a few particle diameters smaller than the distance to the nearest particle in the cluster. In the vicinity of the cluster the particle is moved by 0.7 particle diameters in a randomly selected direction during each step of the random walk. If a particle is found to overlap with the aggregate, it is moved back to the position where it first touched the cluster and becomes part of the growing aggregate.

According to our present model, however, after having been attached to the cluster, the particles are relaxed to their final position in a way which is biased in the direction of the radial vector, $\boldsymbol{r}$, pointing from the seed particle (centre of the cluster) to the particle to which the newly added particle has just been attached (figure 1). In order to achieve this goal we determine the angle ϕ between $\boldsymbol{r}$ and the vector $\boldsymbol{r}_l$ which connects the centres of the newly attached particles. Then the new particle is rotated so that the value of ϕ becomes $\phi' = \phi/(1+B)$ (figure 1), where B is a parameter corresponding to the level of the radial bias. In short, the particles tend to move closer to the radial direction (away from the centre) once they become stuck to the cluster.

The simple rule introduced above is motivated by two different but related considerations. First, it simulates a particular kind of anisotropy which prefers the growth in the radial direction instead of the direction of previously fixed axes. Such a type of anisotropy is not very common, but, for example, it is likely to dominate the formation of patterns in the experiments on viscous fingering in the radial Hele-Shaw cell with a nematic liquid crystal [17], where the local orientation of the *director* was found to be *radial*. Second, the rotation of the particles to a direction closer to the radial one

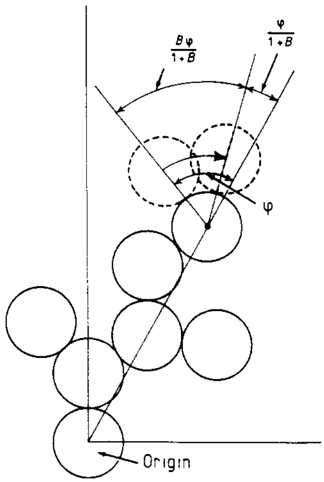

Figure 1. Schematic illustration of the model studied in this letter. The position of the particle when it first contacted the cluster is denoted by a broken line. The final position (●) is obtained by rotating the particle closer to the radial direction. B is a parameter corresponding to the strength of the bias and θ denotes the angle between the radial direction and the direction of the vector connecting the centres of the newly added particle and the particle it has become attached to.

can be considered as an *approximation to an averaging* over the directions along which the particles made their last move before touching the aggregate.

We have carried out simulations of two-dimensional off-lattice diffusion-limited aggregation with radial bias for various values of B. Two typical clusters (for $B=1$ (a) and $B=10$ (b)) are shown in figure 2. 100 clusters were generated for each B up to a linear size of about 1000 particle diameters. The quantity $\ln(M^{0.585}/R_g)$ was

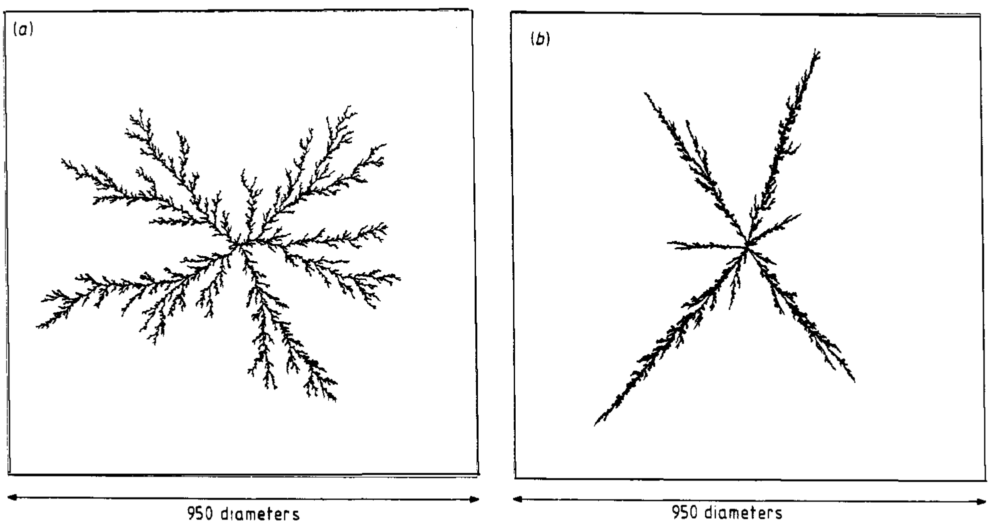

Figure 2. Two clusters having a typical structure for the corresponding two values of the radial bias $B=1$ (a) and $B=10$ (b). As B is increased the radial anisotropy results in the appearance of a star-like shape with four or five main arms.

calculated for these clusters (where R_g is the radius of gyration) and plotted against $\ln(M)$ (figure 3) in order to see the deviations from the ordinary off-lattice DLA. A horizontal line in figure 3 would correspond to a radius of gyration exponent $\beta = 1/D = 0.585$, where $D \simeq 1.7$ is the fractal dimension of the two-dimensional diffusion-limited aggregates. The curves we obtained, however, are not horizontal but seem to have a continuously changing slope presumably converging to the value -0.072 corresponding to a radius of gyration exponent close to $\frac{2}{3}$.

We also determined the density correlations, $c_R(\theta)$, in a layer at a distance R from the centre of the clusters as a function of the angle θ measured from the centre, with θR being the distance separating two particles in the layer. According to our results the tangential correlations for $\theta \ll 1$ and $B < 1$ decay algebraically as a function of θ with an exponent $\alpha_\perp$ depending on B. This is demonstrated in figure 4, where $c_R(\theta)$ is shown for $B = 1$. The θ dependence of the correlations of the form $c_R(\theta) \sim \theta^{-\alpha_\perp}$ is indicated by the approximately straight parts of the curves in this log-log plot.

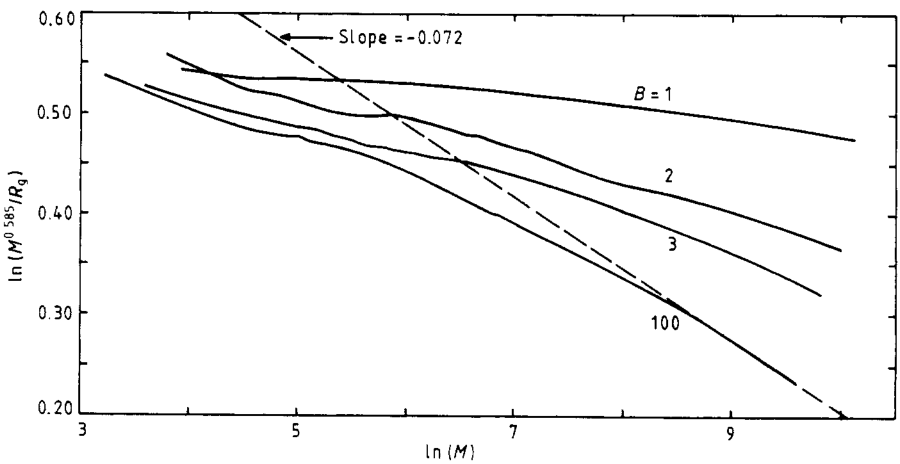

Figure 3. The dependence of the quantity $\ln(M^{0.585}/R_g)$ on $\ln(M)$, where M is the number of particles in the cluster and R_g is the radius of gyration. For $B \gg 1$ the slope of the curves seems to approach a limiting value -0.72 which corresponds to a radius of gyration exponent of about 1.5.

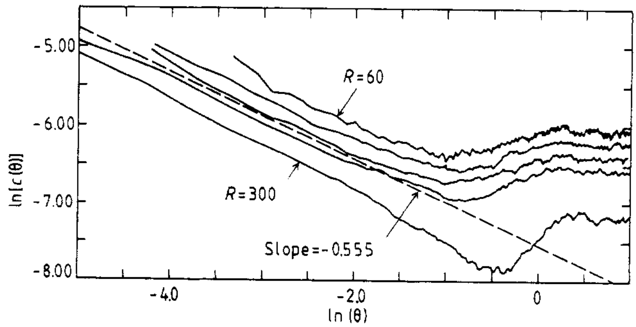

Figure 4. The logarithm of the tangential correlation function $c_R(\theta)$ (see text) plotted against the logarithm of the angle θ. The slope of the curves for $\theta \ll 1$ indicates that the tangential correlations decay faster than the correlations in the radial direction.

The tangential correlation function in most of the cases has a broad second maximum for θ values in the region 0.35–0.45 which can be associated with the existence of a few major arms growing out from the centre of the clusters. In fact, the visual appearance of the aggregates for $b > 2$ clearly shows that, as the radial bias is increased, the shape of the clusters crosses over from an irregular branched structure into an n-fold star with typically $n = 4$, or less frequently with $n = 5$. Naturally, the direction of the arms is different in each realisation and putting the clusters over each other would result in a circular shape.

In conclusion, we have shown that introduction of a simple radial bias results in a non-trivial crossover in the structure of off-lattice diffusion-limited aggregates. The model we introduced corresponds to a locally uniaxial anisotropy, however, without any fixed axes. As in the experiments on viscous fingering in the Hele-Shaw cell, the radial anisotropy resulted in a growth which can be associated with stable tips. Since during DLA the particles, on average, arrive at the surface of the cluster from a radial direction, our results also suggest that the asymptotic shape of the ordinary off-lattice diffusion-limited aggregates may be similar to an n-fold star with $n = 4$ or 5. This conclusion, however, should be tested in the future by independent approaches.

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