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1986 J. Phys. A: Math. Gen. 19 L257

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LETTER TO THE EDITOR

Diffusion-limited aggregation and regular patterns: fluctuations versus anisotropy

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Received 23 December 1985, in final form 23 January 1986

Abstract. We show that the patterns in diffusion-limited aggregation (DLA) on a lattice emerge from the interplay of lattice anisotropy and fluctuations. These fluctuations can be damped by Monte Carlo averaging. Increasing its amount, the effective anisotropy becomes larger and a crossover from tip splitting typical for continuum DLA to stable tips is observed, in analogy with a number of recent experiments. Our simulations suggest the following scenario for the transitions which take place as a function of the increasing effective anisotropy: disordered patterns \rightarrow dendritic structures \rightarrow needle crystals. It is shown that DLA clusters go through the same sequence of transitions as a function of their size. Therefore, diffusion-limited aggregates on a lattice are asymptotically not fractals.

Diffusion-limited aggregation (DLA) (Witten and Sander 1981, see also Herrmann 1986) has attracted much interest in recent years. The model shows very rich behaviour in spite of its simplicity. Particles are randomly diffusing from a distant circular (spherical) boundary in two (or higher) dimensions and they stick to the aggregate (initially a particle at the origin) if they hit. By this algorithm random fractal-like clusters are constructed.

Already Witten and Sander (1981), when introducing DLA, mentioned the connections of diffusion-limited processes to pattern formation phenomena like dendritic crystal growth (Langer 1980). Common to these processes is that the essential physics is described by a diffusion type field (the solution of a Laplace equation) with moving boundaries.

In this letter we would like to focus on the following problems.

(i) The lattice version of the Laplace equation with moving boundaries can be easily solved numerically (see Chen and Wilkinson 1985) and the observed shapes are far from that of random DLA clusters: in the case of square lattice a cross-shaped pattern grows along the axes. Why are the DLA clusters on a lattice like random fractals if the proper solution is a regular pattern?

(ii) On the other hand, as the size of lattice DLA clusters is increased, the effect of lattice anisotropy on their overall shape gradually becomes apparent (Meakin and Vicsek 1985, Meakin 1985a, Ball and Brady 1985). Does this mean that the anisotropy breaks through and finally even the fractal character of the clusters disappears? What is the characteristic size at which this happens and why are the fluctuations no longer important there?

(iii) The role of anisotropy in stabilising the tip of the dendrite was shown in the so-called local models (Brower *et al* 1983, Ben-Jacob *et al* 1983) and in the experiments by Ben-Jacob *et al* (1985), Grier *et al* (1985), Sawada *et al* (1985) and Chen and

Wilkinson (1985) where a transition between random DLA-like and dendritic patterns was observed. Since Monte Carlo (MC) averaging damps the fluctuations, the effective anisotropy can be tuned when growing DLA clusters. What can be learned about various growth regions from MC with variable averaging?

Up to medium sizes (10^4 particles) the two-dimensional DLA clusters seem to have a more or less circular envelope and can be considered as random fractals on lattices as well. This is in conflict with the experience that the patterns from the proper solution of the original problem on the lattice reflect the strong anisotropy of the grid (Kertész *et al* 1985, Chen and Wilkinson 1985). The reason for the discrepancy is that in the Monte Carlo solution of the Laplace equation one has to take an average over many random walkers. The relevance of this averaging on the patterns formed for related models has been pointed out by Tang (1985) and Szép *et al* (1985). (A different kind of averaging was used by Ball and Brady (1985) and Freche *et al* (1985) in order to obtain the average cluster envelope for the DLA and Eden models, respectively. However, our model enables us to tune the effective anisotropy by changing the amount of averaging, and in the limit of infinite averaging we recover the exact solution of the lattice Laplace problem with moving boundaries.)

The procedure is defined as follows. Similar to the usual DLA, particles are randomly walking from a distant circular boundary. However, in our case, they do not stick to the boundary of the aggregate if they hit it; instead, we count the number n_i of walkers terminating at site i of the boundary. As soon as one of the n_i values reaches a given cut-off m , site i is taken as a part of the aggregate (occupied) and new boundary sites i' are born with $n_{i'} = 0$ for each i' . The averaging is controlled by m ; the larger m is the more averaging is done and the smoother the solution. $m = 1$ is the usual DLA case.

Figure 1 shows the clusters obtained for several m values. Figure 1(a) was obtained with small averaging ($m = 2$) and the underlying lattice structure is not seen in the pattern. When m is increased (figure 1(b)), the pattern becomes quasi-regular with well defined main stems in the lattice directions. By further increasing m a needle crystal-like pattern emerges from the simulations (figure 1(c)). Such large averaging corresponds to a better approach to the solution of the discretised Laplace equation on the lattice mentioned where the motion of the boundary is performed in small time steps (see Chen and Wilkinson 1985).

Our pictures, which were obtained in the presence of MC noise, are very similar to those by Chen and Wilkinson (1985) where the randomness was introduced in the medium (the lattice consisted of tubes of cross sections with a random distribution). This shows that the origin of fluctuations is irrelevant; the main point is that they decrease the effective anisotropy.

The simulations presented in this letter suggest that the following scenario of transitions takes place in diffusion-controlled pattern formation as a function of the increasing effective anisotropy (cf Sander 1985). DLA-like patterns are formed (figure 1(a)) if the effective anisotropy is small. As the effective anisotropy is increased the random DLA geometry crosses over into a dendritic pattern with a regular anisotropic structure (figure 1(b)). Finally, as the effective anisotropy is further enhanced, another transition occurs: the most anisotropic pattern, that of needle crystals, grow (figure 1(c)). Such transitions have been observed in the beautiful experiment by Ben-Jacob *et al* (1985) in an anisotropically prepared Hele Shaw cell and in a series of related experiments (Chen and Wilkinson 1985, Grier *et al* 1985, Sawada *et al* 1985) as well as in computer simulations (Vicsek 1984, 1985, Kertész *et al* 1985, Chen and Wilkinson 1985). The reason for such a crossover is that the effective anisotropy stabilises the

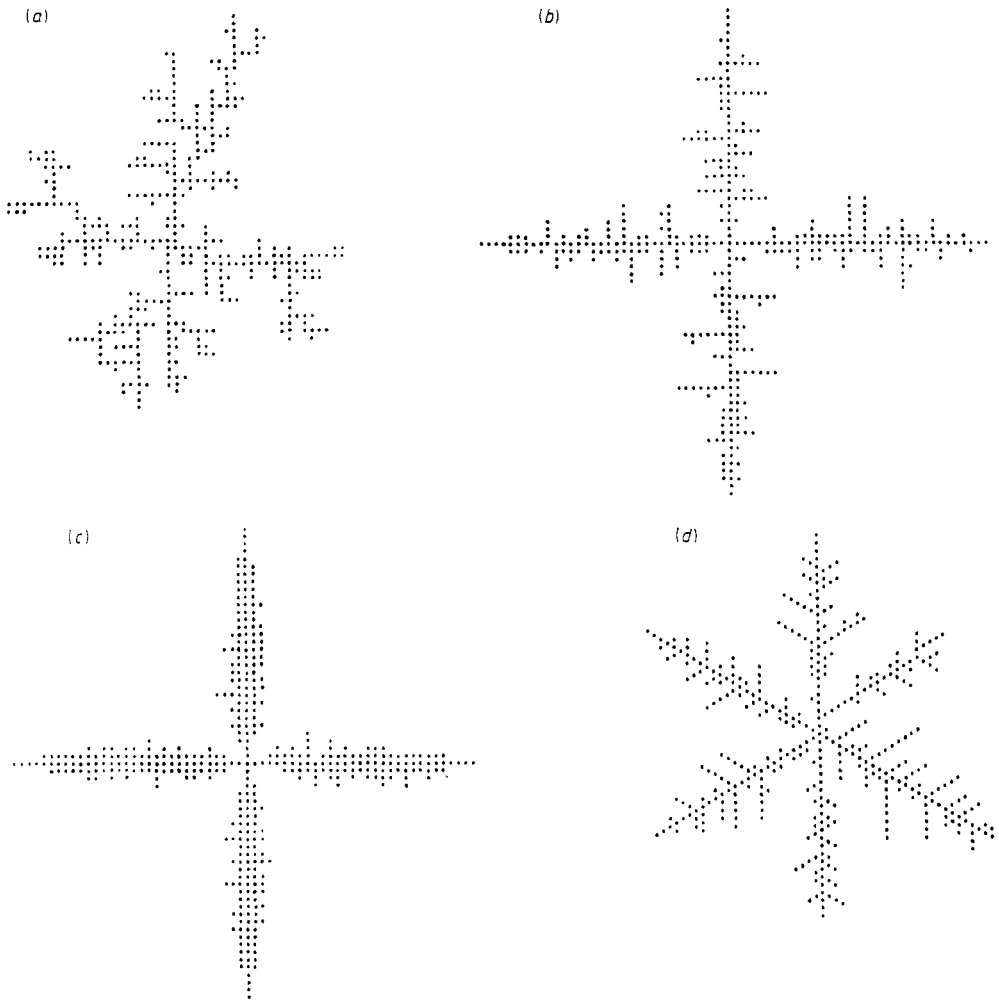


Figure 1. DLA clusters containing $N = 400$ particles; patterns grown on the square lattice with different averaging. (a) $m = 2$, random fractal; (b) $m = 20$, dendritic growth; (c) $m = 400$, needle crystals; (d) as an illustration a snowflake-like cluster of $N = 400$ particles grown on the triangular lattice with $m = 40$ is also shown.

tip of the dendrites against tip splitting if it becomes large enough (local models; Brower *et al* 1983, Ben-Jacob *et al* 1983).

The way by which we controlled the effective anisotropy smears out these transitions, but the regions can clearly be distinguished from each other. The role of averaging was to damp fluctuations and thus the effect of lattice anisotropy could become stronger.

As we shall show below, the role of fluctuations in DLA clusters without averaging decreases with growing N . Therefore the scenario described above should be observed as a function of increasing size. The active zone (Rácz and Plischke 1985) of the DLA cluster is a good measure of the fluctuations. The strong corrections to the fractal dimension D due to the inherent anisotropy of the clusters (Meakin and Vicsek 1985) have the effect that the active zone scales with a different effective exponent, ν' , from $\nu = 1/D$ (Plischke and Rácz 1985) and only asymptotically $\nu' = \nu$ even in the off-lattice

case (Meakin and Sander 1985). As a consequence of the fact that the relative strength of the fluctuations is decreasing with $N(\nu' < \nu)$ the fluctuations are more dominant at early stages. This is the reason why, in the case of lattice DLA, these fluctuations determine the shape of clusters at the beginning and hinder the appearance of the lattice anisotropy. With growing size, the relative strength of fluctuations decays and the anisotropy of the grid appears in the pattern. This effect has been seen in the recent large-scale computer simulations of the square lattice case (Meakin 1985a, Ball and Brady 1985) where clusters with diamond-shaped envelopes were observed at sizes with 10^4 - 10^5 particles.

The following rough calculation illustrates the above ideas. First, we assume that the active zone ξ scales differently from the radius r :

$$\xi \approx 0.266N^{0.484} \quad \text{and} \quad r \approx 0.693N^{0.585} \quad (1)$$

(Rácz and Plischke 1985). Second, we suppose that the limiting form of the envelope is a perfect diamond. The first assumption was shown to be asymptotically incorrect (Meakin and Sander 1985) and later we will argue that the second is so too, but we do not want to apply them for very large clusters. Furthermore we assume that the envelopes can be constructed from circular and straight parts (figure 2).

The deviation from the circular shape is characterised by A , the relative amount of the largest distance of the actual envelope from the circle. Then, in order to see the relative deviation, A , the fluctuations must obey the following inequality:

$$\xi \leq Cr(1 - A)$$

where $C = 1 - \sqrt{2}/2$ (see figure 2). Using (1) for ξ and r we get the following expression:

$$A(N) \leq 1 - N^{-0.101}/1.31 \quad (2)$$

showing that in this approximation which deviation belongs to a given size. According

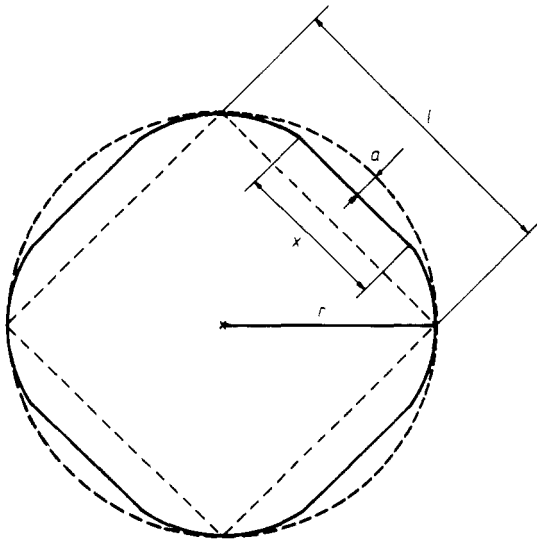


Figure 2. Schematic shape of the envelope (full line) of DLA clusters of radius r grown on the square lattice. The deviation a from the spherical shape becomes visible if the fluctuations satisfy (4), where $A = a/(1 - \sqrt{2}/2)r$ parametrises the effective anisotropy. According to our criterion incision starts if $x/l \sim 0.5$.

to this criterion, an almost unrecognisable small distortion corresponding to a deviation $A = \frac{1}{3}$ appears for $N \approx 800$. It is clear that there is no sharp transition from circular to diamond shapes. This finding is in full agreement with Meakin's (1985a) computer experiments. Moreover the shape of the function of $A(N)$ is very similar to his $R(N)$ which characterises the deviations from circular patterns in his case.

Our assumptions have only limited applicability. First, (1) was obtained for relatively small clusters and it is known that the effective exponent ν' increases (Meakin and Sander 1985), shifting A downwards for a given N . On the other hand, it is not to be expected that the asymptotic shape of the envelope is the diamond on the square lattice. If the straight part of figure 2 is long enough, it becomes incised and the diamond is unstable. Such a tendency was observed by Vicsek (1984) in a DLA-related model.

According to earlier experience (Vicsek 1984), this incision happens if the straight part x of figure 2 becomes approximately half of the distance l . Using this criterion and (2) we get $N \sim 15\,000$ for the size of DLA clusters on the square lattice, where the incision first appears. This is in good agreement with the computer experiments of Meakin and Vicsek (1985), Meakin (1985a) and Ball and Brady (1985) where anisotropic cluster shapes, and main stems characteristic for dendritic growth were observed in very large ($N \sim 10^4$ - 10^5) clusters. Of course the above value of the characteristic cluster size should be regarded as a rough estimate, since, for example, the N value calculated from (2) is very sensitive to small changes in A and the other parameters of the problem.

The recent results about the envelope of large DLA clusters approaching the diamond shape (Meakin and Vicsek 1985, Meakin 1985a, Ball and Brady 1985) are the first manifestations of the mechanism described above. However, according to our proposed scenario, the asymptotic envelope is not the diamond, but dendritic forms should appear and, finally, the coarse-grained DLA clusters become like needle crystals. The most serious consequence of this picture is that DLA clusters on lattices are asymptotically not objects with fractal dimensionality. The fact that Meakin's (1985a) anisotropy parameter R in simulations on the square lattice goes beyond the value characteristic for the diamond is already a sign of the effect described. In the latest calculations by Meakin (1985b) the DLA clusters on the square lattice tend to be in the shape of a cross, already showing the final stage of the above scenario.

In conclusion, we have shown that as a function of effective anisotropy a scenario of transitions should take place in the processes we have considered. This effective anisotropy can be tuned in DLA either by the introduction of an averaging procedure or by going to larger size, while in the dendritic growth it is controlled by the ratio of the anisotropy in the surface tension to the driving force.

The authors are grateful to P Meakin for communicating his results about the cross-like shape of very large DLA clusters prior to publication as well as to L Sander for informative correspondence. We thank the referee for sending us a preprint of a related paper submitted to *Nature* by J Nittmann and H E Stanley.

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