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LETTER TO THE EDITOR

Large scale lattice effect in diffusion-limited aggregation

R C Ball and R M Brady

Cavendish Laboratory, Madingley Road, Cambridge CB3 0HE, UK

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Abstract. Several diffusion-limited aggregates were computed according to the classical two-dimensional square-lattice model of Witten and Sander. A new algorithm enabled large clusters of 10^5 particles to be investigated with moderate computing resources. At large radius (and in contrast to previous work on the same model) the aggregates were observed to be anisotropic, their overall shape depending on the lattice of deposition.

There has recently been interest in non-equilibrium growth processes, particularly those governed by diffusion where protrusions grow more rapidly. Dendritic and often chaotic patterns result. Measurements have been reported on: exothermic crystal growth governed by heat diffusion [1], diffusing copper ions depositing on a growing cathode [2]; and on systems governed by analogous or related equations: electrical breakdown [3], anodic etching of silicon [4], the moving interface between liquids of different viscosities [5], and diffusing colloid particles sticking together [6]. Interest has centred on the (statistical) *scaling* properties [7] of these patterns. If a pattern is *self-similar* then all relevant lengths (e.g., mean radius, standard deviation in radius) are proportional to one another. If it is *fractal* the mean number of particles N within radius R of a particle shows $N \propto R^D$ where D (the Hausdorff dimension) need not be an integer. Many of the experiments indicate self-similar fractal behaviour.

These experiments have been related to the model for diffusion-limited aggregation proposed by Witten and Sander [8] and further investigated on computer by Meakin [9, 10] and others. Particles diffused one at a time until they wandered far away (in our simulations to 100 cluster radii) or reached 'sticky' sites adjacent to the cluster, where they deposited and made adjacent unoccupied sites sticky. For simplicity a square lattice was used, the sticky sites of a deposited particle were in a square pattern (deposition habit), and surface tension was neglected. It was suggested [8, 9] that random noise would smear out these microscopic details at sufficiently large radii, leaving universal macroscopic behaviour. Indeed the Hausdorff dimension D of this model has been confirmed experimentally [2, 3, 5].

Recently published computations [10, 11] on clusters of 2500 particles grown with a square deposition habit showed an unexpected departure from self-similarity in measurements comparing the deposition radius R_d with the standard deviation in R_d and the seed to centre-of-mass (SCM) distance, but it was unclear if this represented the universal macroscopic behaviour. The principal problems with growing large clusters to check this have been the computer time (order R^2 per particle) and store (order R^2 in two dimensions) required.

There is an established method to speed up diffusion simulations [12, 13]. If a large circle can be drawn which is centred on the diffuser and which contains no

obstacles or sticky sites, then the diffuser will first cross this circle at random angle, by symmetry. Thus the several small steps to reach the boundary of the circle can be replaced by a single choice of random angle. The complicated shapes of diffusion-limited aggregates make it difficult to determine large empty circles; so we adapted this method to use large empty squares centred on the diffuser instead. A lookup table (calculated analytically using Laplace's equation for the diffuser's probability density) was used with a random number to compute the first crossing of the square. The distribution of first crossings was compared with a conventional stepping algorithm and found to be in agreement to better than 1%.

To help determine large empty squares, information was maintained as each particle was deposited. Each new sticky site was entered on a conventional lattice grid map, and also on a hierarchy of coarser grid maps (boxes of 2×2 lattice spacings, 4×4 and so on). An entry was zero only if it contained no sticky sites. In a refinement, information was omitted on a fine scale about empty coarse-scale boxes. Occupied boxes then contained the memory addresses of their finer-scale information. This, together with bit-maps on the finest scale, reduced the store to 1.3 bytes per deposited particle and overcame the problem of storing large clusters.

Clusters of 10^5 particles required about ten minutes of CPU time on an IBM 3081. This represents approximately a tenfold improvement over conventional algorithms. Figure 1 shows that CPU time per particle increased as $(\log R)^2$ where R is the maximum cluster radius. This may be understood qualitatively by noting that, if diffusion were by small steps of constant distance, the time for diffusion would increase as R^2 (the usual time requirement). However, each step was on average proportional to the distance to the cluster, so on a logarithmic distance scale they were constant, giving the $(\log R)^2$ dependence.

Figure 2 shows three representative clusters, and also the superposition of sixteen clusters, all grown with the conventional square habit. The superposition shows a striking diamond profile (this was also observed in superpositions of clusters as small as 500 particles), indicating that fingers extended faster in radius along axial directions. The angular distribution of particles is shown quantitatively in the upper part of figure

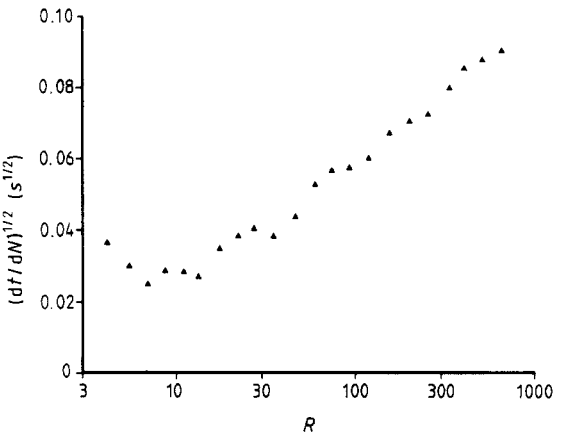

Figure 1. Plot of computer time per particle dt/dN against maximum cluster radius R (shown in lattice units). The plot indicates that $dt/dN \propto (\log R)^2$. Particles accreted at a rate of about 100 per second at $R = 1000$ lattice units.

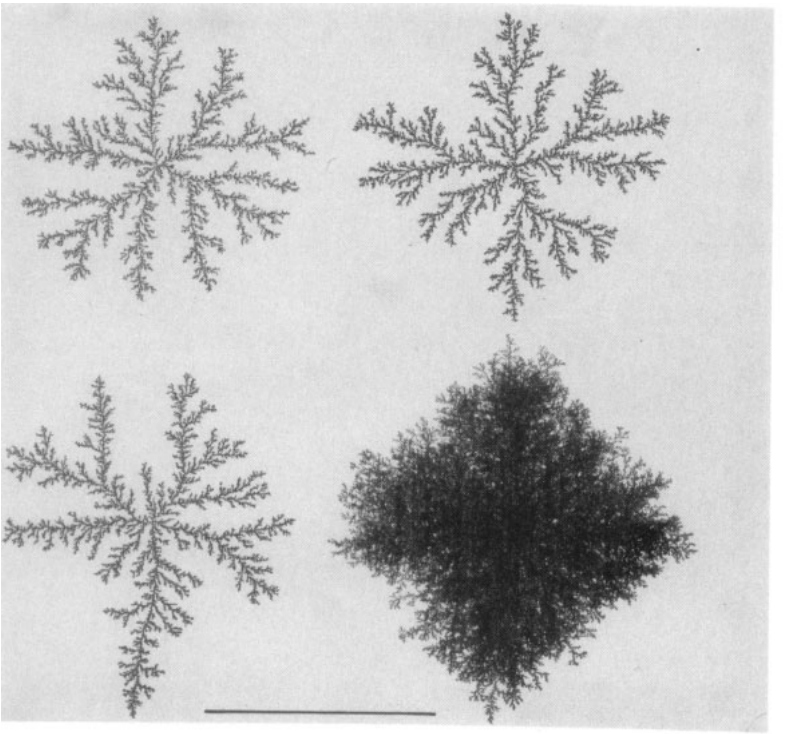

Figure 2. Three representative clusters, and the superposition of sixteen clusters, each of 10^5 particles and grown on the standard DLA model with a square deposition habit. Scale bar is 1000 lattice units.

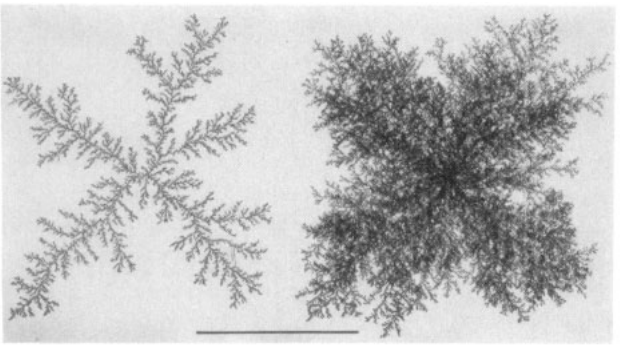

Figure 4. One representative cluster, and the superposition of eight clusters, each of 10^5 particles and grown with a diagonal deposition habit. Scale bar is 1000 lattice units.

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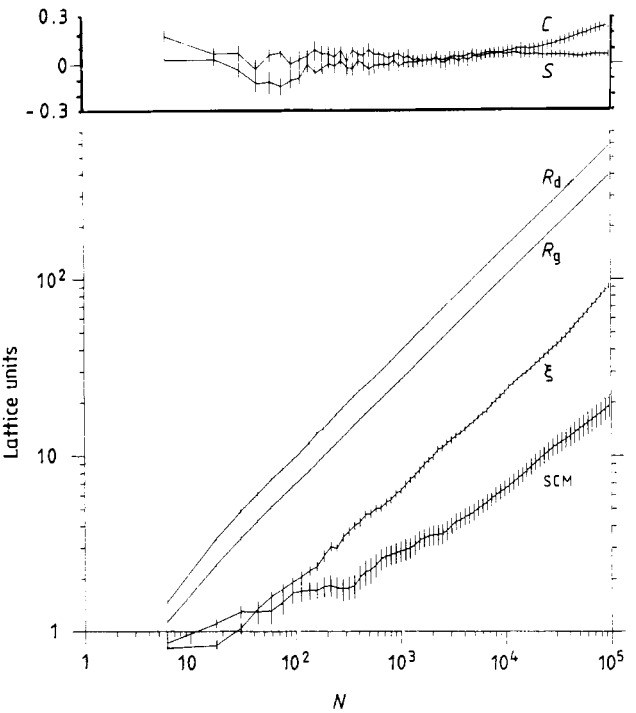

Figure 3. Plot of mean deposition radius R_d, radius of gyration R_g, penetration depth ξ, seed centre-of-mass SCM, anisotropy of depositing particles $C = \langle \cos 4\theta \rangle$, $S = \langle \sin 4\theta \rangle$ against number of particles N.

3, and shows no significant dependence on angle from the seed up to 30 000 particles, indicating that axial fingers did not grow significantly faster in mass. Beyond 30 000 particles the fourth circular harmonic $C = \langle \cos 4\theta \rangle$ began to appear above noise, perhaps as a result of dendritic competition favouring the more extended axial growths, although this was a weak effect which required averaging over a large number of growths in order to be seen clearly. Figure 4 and the upper part of figure 5 provide evidence that this anisotropy resulted primarily from the deposition habit rather than the square lattice for diffusion. The deposition rule is rotated through 45 degrees, and the anisotropy was correspondingly rotated. (The two habits were not strictly comparable because multiple deposition was possible in the second habit.) Anisotropy has independently been noted [14] in smaller growths on a different model where surface tension processes were simulated and fractal behaviour may have been absent, and it was attributed to the initial square-habit growth pattern becoming amplified by the growth process.

The simplest interpretation of the straight-sided growth profile is in terms of growth paths directed away from the seed having equal numbers of steps. Suppose that the path has N_x, N_y steps in the $\pm x$, $\pm y$ directions, where $N_x + N_y = N$. If the steps were uncorrelated, as in a random walk, there would be $\langle x^2 \rangle = N_x$ and $\langle y^2 \rangle = N_y$, giving $\langle x^2 + y^2 \rangle = N$ and a circular profile. If on the other hand all the steps were directed away from the seed point, there would be $|x| = N_x$ and $|y| = N_y$ and the equation $|x| + |y| = N$ which yields a diamond profile similar to that observed in our growths.

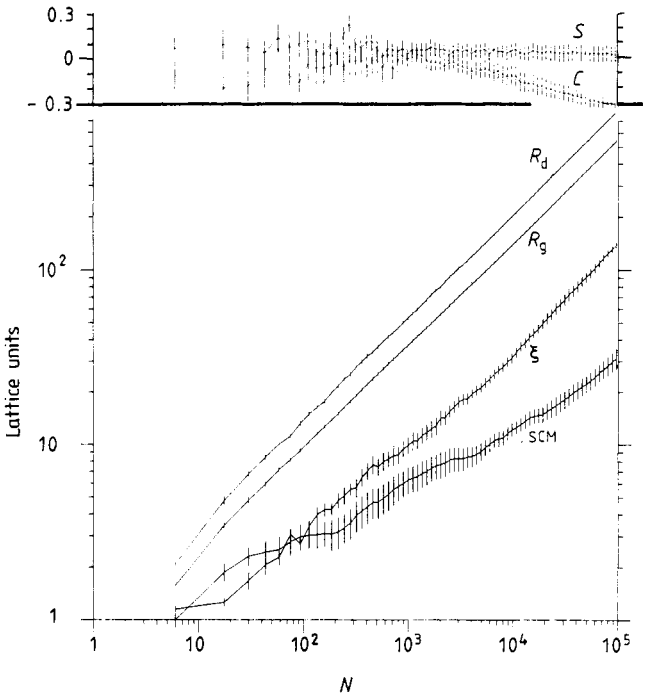

Figure 5. As for figure 3, but for a diagonal deposition habit.

Note the strikingly straight edges in the superposition of figure 2, which support this interpretation. Also, recent computer measurements by Meakin (private communication) indicate that the fingers of growth are one-dimensional (like a directed walk); indeed visual inspection of figure 2 does suggest directed growth (contrary to the observations in [8]). Natural snowflakes also exhibit directed growth with a crystalline habit, and similar interpretation may account for the accurately straight-sided hexagonal profiles which can result (see for example figure 1 of [1]).

The above interpretation neglects such complications as splitting or backtracking of paths; and, more importantly, dendritic competition between different fingers of growth. The diamond profile may well not represent the final limit of cluster anisotropy: note in figure 3 that the anisotropy appeared to be increasing even for our largest growths, and compare figures 4 and 5 for the diagonal habit clusters, which have clearly become more anisotropic. More generally we can conclude that the *local* perturbation of restricting the growth to a square lattice becomes increasingly important at *large* length scales.

Figure 3 also shows the mean and its standard error, averaged over sixteen clusters of 10^5 particles, for: deposition radius R_d, radius of gyration R_g, penetration depth ξ and SCM distance. The data are in good agreement with the equivalent experiment (to clusters of only 2500 particles) reported in [11]. (We measured the penetration depth ξ over the last $P = 10\%$ of deposited particles, during which the mean deposition radius was increasing. Our formula differs from that of [11] by $P R_d/(2D\sqrt{3})$, or less than $0.02 R_g$ in our case.) The power laws which best fit the data in the extended range $N = 3000$ to $N = 10^5$ particles are: $R_d \propto N^{1.69}$, $R_g \propto N^{1.71}$, $\xi \propto N^{1.73}$, and SCM $\propto N^{(2.2\pm0.4)}$. We conclude that these results are consistent with simple scaling: $R_d =$

1.48 R_g, $\xi = 0.22\ R_g$, but their interpretation is complicated by the anisotropy (see discussion below). The SCM distance reached a maximum of only 20 lattice units, so it may have been still settling down to scaling over the range reported, and we can only extrapolate SCM $< 0.05\ R_g$. For comparison, figure 5 shows corresponding plots for the diagonal deposition habit, but note with this habit approximately 8% of particles were able to bypass sticky sites and reach already occupied sites, where they deposited.

If large square-lattice DLA clusters are not statistically isotropic, it is no surprise that their penetration depth (which measures the departure of deposition from a circle) scales proportionately to cluster radius. To obtain a true measure of penetration one must work relative to the actual average cluster shape, itself varying with N, which would require many more clusters than we were able to generate. An apparent alternative is to study DLA off-lattice, where the clusters are statistically isotropic by symmetry, but again the observation $\xi \propto R_g$ would only prove that the clusters were not circular.

Several important question are raised. Within our range (to 10^5 particles) the angular distribution of particles has only begun to show significant anisotropy: does the effect saturate, and if so what is the limiting behaviour? Do the exponents such as the fractal dimension D depend on the lattice for very large aggregates? Analogous measurements might be made in higher dimension or with other lattices (e.g. triangular lattices). Finally one must question to what extent large off-lattice DLA clusters develop a characteristic shape. This last question might be answered by measuring the power spectrum of the angular distribution of particles for very large clusters.

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