The structure of two-dimensional Witten-Sander aggregates

1985 J. Phys. A: Math. Gen. 18 L661

(http://iopscience.iop.org/0305-4470/18/11/006)

J. Phys. A: Math. Gen. 18 (1985) L661-L666. Printed in Great Britain

LETTER TO THE EDITOR

The structure of two-dimensional Witten–Sander aggregates

Paul Meakin

Central Research and Development Department, E I du Pont de Nemours and Company, Experimental Station, Wilmington, Delaware 19898, USA

Received 12 March 1985, in final form 8 May 1985

Abstract. A relatively large number (10^2-10^3) of reasonably large (about 10^5 particles or sites) two-dimensional Witten-Sander aggregates have been simulated using new algorithms. This has enabled us to reduce statistical errors by an order of magnitude and has provided an opportunity to determine some new quantities describing in more detail the structure of Witten-Sander clusters. A quantitate description of the distortion of large lattice-based Witten-Sander clusters into a diamond-like shape is provided. The orientational correlation in the bond directions for off-lattice Witten-Sander clusters has also been measured. This quantity has a power law dependence on distance with an exponent of about -0.37.

In recent years a considerable interest has developed in a variety of non-equilibrium growth and aggregation models. The development of this area has as its genesis the discovery by Witten and Sander (1981) that a simple diffusion-limited aggregation model in which particles are added, one at a time, to a growing cluster or aggregate of particles via random walk trajectories, leads to structures with a fractal-like (Mandelbrot 1982) geometry. Despite the importance of this model in the development of this rapidly growing new area, there is still no rigorous quantitative theory for this simple growth process. In fact, there is relatively little known in quantitative terms about the structure of the aggregate formed by the Witten-Sander model. Most of our knowledge concerning the diffusion-limited aggregation process has resulted from computer simulations of a relatively small number (about 10) of rather small (approximately 10^4 lattice sites or particles) clusters (see, for example, Witten and Sander 1981, Meakin 1983). Here new algorithms are described for diffusion-limited aggregation which have enabled us to generate relatively large numbers (10^2-10^3) of somewhat larger (about 10^5 lattice sites or particles) clusters. This has resulted in a reduction of statistical uncertainties by an order of magnitude and permitted investigation of the correction to the simple scaling picture (Witten and Sander 1981, Meakin 1983, Sander 1984) for diffusion-limited aggregation.

One of the purposes of this paper is to describe the new algorithms which can be applied to a variety of other problems such as diffusion-limited cluster-cluster aggregation and the penetration of diffusing particles into complex structures. We also describe results for some new quantities which have not been previously reported for diffusion-limited aggregation.

An algorithm for the generation of Witten-Sander clusters has been described previously by Meakin (1983). Random walkers are released from a randomly chosen position on a circle which encloses the cluster. The particle then undergoes a random

0305-4470/85/110661+06$02.25

walk until it either contacts the cluster and the cluster grows or it wanders a large distance from the cluster (typically a distance of $3R_{max}$ from the centre of the cluster where R_{max} is the maximum radius of the cluster). In the latter event the random walk trajectory is terminated and a new walker is started off at a random position on the 'launching' circle which encloses the cluster. The procedure described above is repeated many times until a large cluster has been formed.

In the improved algorithm we start off by placing a particle at the centre of a lattice. Each of the elements in the lattice is given a rather arbitrary value of L_{max} ($L_{max} \simeq 30$) where L_{max} is the maximum step length which the random walker may take if it is in the vicinity of the cluster. For all lattice sites from which a random walker could contact the 'seed particle' by taking a step of length L' lattice units, the value of that lattice element is changed to $L'-1$ if $L'-1$ is smaller than L_{max}. Figure 1 depicts the early stages in a small scale off-lattice simulation carried out using this model. If the centre of the diffusing particle (random walker) lies on one of the lattice sites identified by a number (L) it can move by a distance of L particle diameters in any direction without contacting the cluster. The random walker is started off from a random position on the launching circle and travels by means of an *off-lattice* walk. The length of each step in the random walk is determined by the value of the lattice element in the lattice site which the random walker occupies. If the random walker is a distance R which is greater than $R_{max}+L_{max}$ lattice units from the centre of the cluster, it is allowed to jump by a distance which is a little smaller than $R-R_{max}$ lattice units. If the trajectories reaches a distance greater than $100R_{max}$ from the seed of the cluster, the trajectory is terminated and a new particle is started from a random position on the launching circle. After each new particle has been added to the cluster, the elements of the underlying lattice which are in its vicinity are updated so that the next particle cannot take a jump which would cause it to cross over any part of the cluster.

Two versions of the model have been developed. In the off-lattice version of the model particles which are close to the cluster (in the unoccupied region in figure 1) jump to a new position which is randomly selected from all possible positions within

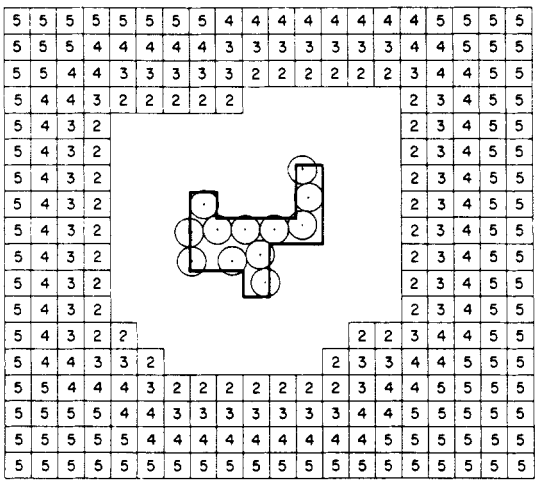

Figure 1. The early stages of a small scale off-lattice simulation. In this case L_{max} was set to a value of 5. Random walkers whose centres are on a lattice site identified by a number L can move by L particle diameters without contacting the cluster.

one lattice unit (the average step length is $1/\sqrt{2}$ lattice units). The particles, which are considered to have a diameter of 1 lattice unit, are stopped and added to the cluster at that point in their trajectory where they first contact another particle which has already been incorporated into the cluster. In this model the underlying lattice is also used to indicate to the random walker which particles are in its vicinity and must be examined for possible contact. After a particle has been added to the cluster, the lattice site which is occupied by its centre is assigned a number which indicates that it is occupied by this particle. In figure 1 the bold line encloses all of the occupied lattice sites at the stage of growth in this figure. The value assigned to the lattice element also indicates if it is occupied by more than one particle and if so provides a means of identifying all of the particles associated with that lattice site.

In the semi-lattice version of the model, the mobile particles undergo off-lattice random walks as before, but the aggregate is represented by filled lattice sites. In the vicinity of the growing aggregate, the particle which is considered to be of zero size jumps by a distance of 1 lattice unit. If the end of a step in the trajectory causes the particle to land on an unoccupied site adjacent to (nearest neighbour to) an occupied site the trajectory ends and the cluster grows by one lattice site into the unoccupied site. If the random walker jumps onto an occupied site in the cluster, the last step in the walk is reversed.

These simple algorithms can be used to generate an off-lattice aggregate containing 50 000 particles in about 7 min of CPU time on an IBM 3081 computer. A 100 000 site semi-lattice simulation requires about 11 min of CPU time. These algorithms are 100–1000 times faster than those described previously by Meakin (1983) and they should be capable of further improvements.

In terms of speed these algorithms seem to be comparable with the more complex methods developed very recently by Brady and Ball (unpublished). However, our methods do not make efficient use of storage capabilities and it would be difficult to generate clusters containing much more than 100 000 particles.

Figure 2(*a*) shows a 50 000-particle two-dimensional Witten–Sander cluster generated using the off-lattice model and figure 2(*b*) shows a 100 000-site cluster obtained from the semi-lattice model. Figure 2(*b*) shows quite clearly the anisotropy of the overall shape of the cluster. The formation of a diamond-like figure has been noticed previously for fully lattice model (on-lattice growth with on-lattice walks) simulations of diffusion-limited aggregation by Brady and Ball (unpublished) and Meakin (unpublished). This figure and many others like it demonstrate that the use of on-lattice walks is not responsible for the diamond shape. The formation of a diamond shape may be due simply to the fact that addition of a particle along the direction of one of the axes causes the structure to grow by 1 lattice unit in that direction whereas addition of a particle along one of the diagonals causes the aggregate to grow only by $1/\sqrt{2}$ lattice units in the diagonal direction.

In order to provide a quantitative measure of the diamond-like shape of two-dimensional semi-lattice Witten–Sander aggregates, the maximum projection of the coordinates of each particle onto the lattice axes and the diagonal axes (lattice axes rotated by 45°) have been determined for all of the occupied sites in the clusters. The parameter R defined by

$$R = \frac{\Sigma \text{ projections onto lattice axes}}{\Sigma \text{ projections onto diagonal axes}} \tag{1}$$

provide a simple quantitative description of the shape of the clusters. For a completely

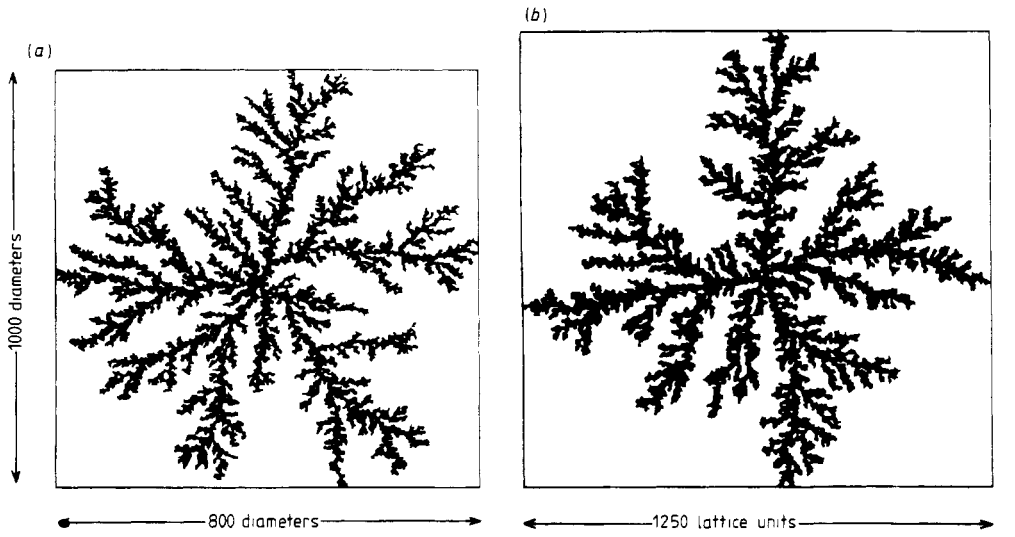

Figure 2. (*a*) A typical 50 000-particle off-lattice two-dimensional Witten-Sander aggregate. (*b*) A typical 100 000-site semi-lattice two-dimensional Witten-Sander aggregate. The preferential growth along the axes of the lattice (characteristic of this model) can easily be seen in this figure.

isotropic structure, R has a value of 1.0 and for a uniform diamond shape R has a value of $1.5/\sqrt{2}$ or 1.061. Figure 3 shows the dependence of R on cluster size for semi-lattice Witten-Sander aggregates. The results shown in figure 3 were obtained from 234 clusters and the statistical uncertainties (95% confidence limits) are less than ± 0.01 for the larger cluster sizes. It is clear from figure 3 that R has not reached its limiting (large N) value even for clusters of 100 000 sites and that R has a value larger than that of a uniform diamond shape.

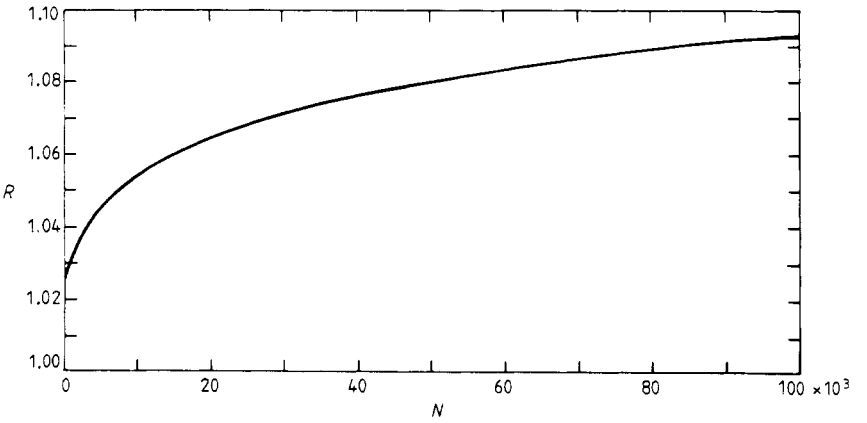

Figure 3. This figure shows how the parameter R (the ratio of the sum of the maximum projections of the site coordinates along the natural lattice axes divided by the sum of the projections along the diagonal (45°) axes) depends on cluster size (N) for two-dimensional semi-lattice aggregates. This curve consists of 500 linear segments joining adjacent data points.

In the off-lattice model for diffusion-limited aggregation, a bond vector ($\boldsymbol{b}$) can be associated with each of the particles in the aggregate. The vector $\boldsymbol{b}_i$ is a unit vector in the direction of the bond which is formed when the ith particle is added to the cluster. The bond is considered to join the centre of the contacted particle in the cluster to the centre of the new particle. The ordinary two-point density-density correlation function for an N particle aggregate is given by

$$C(r)=\frac{1}{2\pi rN\delta r}\sum_{|\boldsymbol{r}'-\boldsymbol{r}''|=r\pm\frac{1}{2}\delta r}\rho(\boldsymbol{r}')\rho(\boldsymbol{r}'') \tag{2}$$

where $\rho(\boldsymbol{r})$ is 1 if there is a particle at position $\boldsymbol{r}$ and 0 otherwise. The bond vectors can be used to define similar correlation functions which depend on the relative directions of the bonds associated with pairs of particles separated by a distance δ. For example

$$C'(r)=\frac{1}{2\pi rN\delta r}\sum_{|\boldsymbol{r}'-\boldsymbol{r}''|=r\pm\frac{1}{2}\delta r}|\boldsymbol{b}(\boldsymbol{r}')\cdot\boldsymbol{b}(\boldsymbol{r}'')| \tag{3a}$$

$$C''(r)=\frac{1}{2\pi rN\delta r}\sum_{|\boldsymbol{r}'-\boldsymbol{r}''|=r\pm\frac{1}{2}\delta r}\boldsymbol{b}(\boldsymbol{r}')\cdot\boldsymbol{b}(\boldsymbol{r}''). \tag{3b}$$

Here $\boldsymbol{b}(\boldsymbol{r})$ is the bond vector associated with the particle at position $\boldsymbol{r}$ if there is a particle at this position and 0 otherwise.

Figure 4 shows the correlation functions $C(r)$, $C'(r)$ and $C''(r)$ obtained from 34 50 000-site off-lattice Witten-Sander aggregates. As expected, $C(r)$ exhibits the same scaling behaviour with distance (r) as the ordinary two-point density-density correlation function ($C'(r)\sim r^{-\alpha}\alpha=d-D_\alpha$). However, $C''(r)$ shows a quite different behaviour, $C''(r)\sim r^{-\eta}$, where the exponent η has a value of about 0.66. Part of the power law decay for $C''(r)$ can be attributed to the power law decay of the density-density correlations. However, the additional contribution of the decay of angular correlation can be described by an exponent ϕ having a value of about -0.37. This means that angular correlations in diffusion-limited aggregation are long range.

Relatively efficient off-lattice and semi-lattice algorithms for diffusion-limited aggregation have allowed us to reduce statistical uncertainties by an order of magnitude.

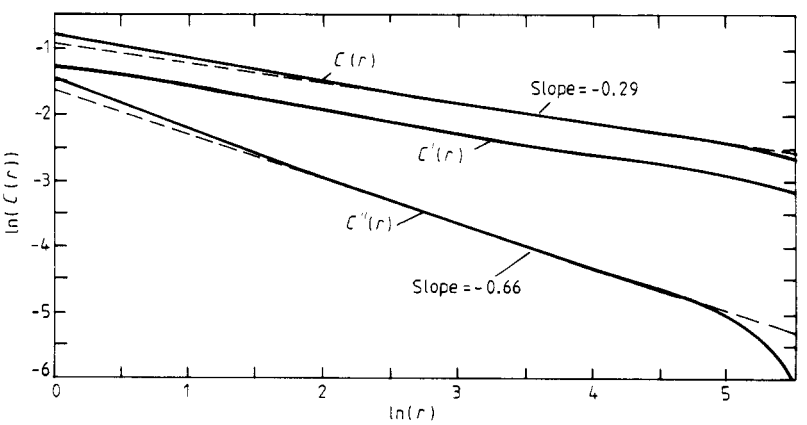

Figure 4. Comparison of the ordinary two-point density-density correlation function ($C(r)$) with the orientational correlation functions ($C'(r)$ and $C''(r)$) defined in the text.

By measuring the maximum projections of particle coordinates along the natural lattice axes and the diagonals, a simple quantitative measure (as expressed by the parameter R above) of the tendency of lattice-based aggregates to grow preferentially along the directions of the lattice axes has been obtained. Our results indicate that this tendency for preferential growth along the lattice directions is larger than that found in a simple diamond shape. The growth of the parameter R to the value characteristic of a diamond shape can be understood on the basis of simple geometric arguments. The additional preferential growth along the axes must be due to screening of the edges of the diamond shape by the more exposed corners.

Perhaps the most interesting result of this work is the measurement of the bond orientation correlation function in off-lattice aggregates. Our results indicate that orientational correlations decay only slowly with increasing distance (with a power law whose exponent is about -0.37. This suggests a high degree of 'directionality' in diffusion-limited aggregation and may be related to the observation that the minimum path-distance between two points on the aggregate scales linearly with their separation in the embedding space or lattice (Meakin *et al* 1984).

References

Hentschel H G E 1984 *Phys. Rev. Lett.* **52** 212
Mandelbrot B B 1982 *The Fractal Geometry of Nature* (San Francisco: Freeman)
Meakin P 1983 *Phys. Rev.* A **27** 604, 1495
Meakin P, Majid I, Havlin S and Stanley H E 1984 *J. Phys. A: Math. Gen.* **17** L975
Sander L M 1984 *Kinetics of Aggregation and Gelation* ed F Family and D P Landau (Amsterdam: North-Holland) p 13
Witten T A and Sander L M 1981 *Phys. Rev. Lett.* **47** 1400