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LETTER TO THE EDITOR

Transparency effects in cluster–cluster aggregation with linear trajectories

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Abstract. Two hierarchical models of cluster–cluster aggregation whose aggregates follow random linear trajectories with (model I) or without (model II) random impact parameter are investigated numerically. The difference of the fractal dimension D of the clusters as a function of space dimension d is attributed to transparency effects which are less important in model II. A simple argument is given which suggests that, in contrast with model I, model II should not have an upper critical dimension.

A model of cluster–cluster aggregation (Meakin 1983a, Kolb *et al* 1983) has been proposed recently and appears to be quite appropriate to describe flocculation of aerosols or colloids (Weitz and Oliveria 1984, Schaefer *et al* 1984). In contrast with a previous model of particle–cluster aggregation (Witten and Sander 1981, Meakin 1983b, c), clusters of particles, as well as single particles, are allowed to diffuse together and sticking occurs mainly between clusters of almost the same size. Rediscovering an old idea by Sutherland and Goodarz-Nia (1971), Botet *et al* (1984a) introduced a simplified, hierarchical, version of this model, in which clusters of $2, 4, \ldots 2^k \ldots$ particles are built iteratively by always sticking together two independent clusters of strictly the same number of particles. The remarkable simplicity of the hierarchical formulation allows some results to be derived analytically, such as the upper critical dimension (Ball and Witten 1984).

In this letter, a further simplification of the cluster–cluster aggregation model is considered by using random linear trajectories, instead of Brownian trajectories. Two models, called I and II, with or without random impact parameters, respectively, are studied. The fractal dimension D of the clusters is estimated numerically for the first time, up to $d = 8$, in the case of model II. The difference between the curves $D(d)$ is attributed to transparency effects which in model I become more important, for high dimensions. Then to explain these results, I give a simple argument which predicts that, in contrast with model I, model II should not have an upper critical dimension.

The two models are motivated by completely different experimental situations. Model I still considers, as the original model, that the diffusion is of Brownian nature but with a mean free path larger than the largest size of the clusters. Thus the relative trajectory is an arbitrary straight line, completely random in space. On the other hand, model II corresponds to a situation where, in addition to the short ranged interactions, which ensure the irreversible sticking, there exists some other, strong, long ranged interactions which forces the two coalescing clusters to follow a linear trajectory going through their centres of mass. However, in both cases, I assume that the clusters stay

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rigid and do not rotate during their relative motion. Both models have already been studied in the context of particle–cluster aggregation (Bensimon *et al* 1983, Meakin 1983d, for model I; Meakin 1983e, Jullien *et al* 1984, for model II) leading always to almost compact structures ($D \sim d$). In the context of cluster–cluster aggregation, model I, already introduced by Sutherland and Goodarz-Nia (1971) has been also investigated recently (Meakin 1984, Ball and Jullien 1984) and some preliminary results on model II have been reported (Botet *et al* 1984b).

In both models, successive collections of clusters are built iteratively, starting from P_0 individual spherical particles. Let us assume that, at step k, a collection of $P_0/2^k$ clusters, of $N = 2^k$ particles each, is available. These clusters are grouped into $P_0/2^{k+1}$ pairs of clusters. Then one considers each pair of clusters successively. Let us call (1) and (2) the two clusters of the pair, whose centres of mass are denoted G_1 and G_2, respectively. This pair generate a new cluster of $2N = 2^{k+1}$ particles as follows.

(1) The two clusters are independently, randomly rotated around their respective centres of mass, in the whole d-dimensional space.

(2) Cluster (1) is considered as fixed with G_1 positioned at the origin.

(3) One chooses a point, say H, randomly located on a large sphere centred around G_1. This defines a random direction in space: $\mathrm{H}G_1$.

(4) Then the two models differ in the way one chooses the initial position G_2^0 of cluster (2). In model II, cluster (2) is initially positioned such that G_2^0 is exactly at H. In model I instead, one chooses G_2^0 randomly located in a $(d-1)$-dimensioned hyperplane, perpendicular to the random direction $\mathrm{H}G_1$ at the point H. The vector $\mathrm{H}G_2^0$ then represents the random impact parameter (which is strictly zero in model II).

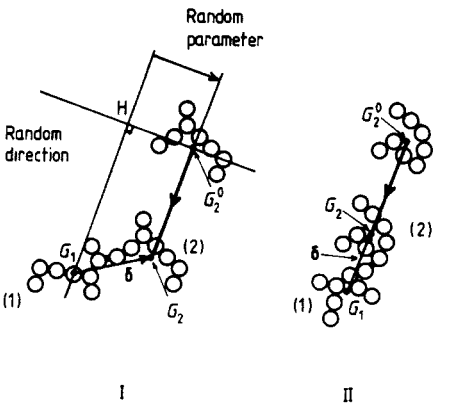

Figure 1. Sketch of the different collision processes of model I and II.

(5) Then cluster (2) is moved from its initial position along a trajectory parallel to $\mathrm{H}G_1$ until a collision occurs, i.e. until the minimum distance between the centre of any particle of (1) and the centre of any particle of (2) becomes equal to the sphere diameter, which is taken here as the unit length. In both models, if cluster (2) does not collide with cluster (1), the trial is abandoned and another trajectory is chosen, independently from the previous trial (i.e. we go back to step 3) and this is repeated as often as it is necessary until a collision occurs.

(6) The reunion of the two clusters, just after the collision, then forms a cluster of the new collection.

The radius of gyration of each cluster is defined as:

$$R_N^2 = \frac{1}{2N^2} \sum_{i,j} (\boldsymbol{r}_i - \boldsymbol{r}_j)^2 \tag{1}$$

where $\boldsymbol{r}_i$ locates the ith particle inside the cluster and where the double sum runs over all particles. Numerically, R_N^2 is estimated, for size N, by averaging R_N^2 over all the clusters of the kth collection. The fractal dimension of the clusters, defined by:

$$R_N \sim N^{1/D}, \qquad N \to \infty$$

can be estimated by extrapolating to $N \to \infty$, an effective N-dependent fractal dimension $D_{\text{eff}}^{(1)}(N)$, obtained when comparing two successive collections:

$$D_{\text{eff}}^{(1)}(N) = \ln 2 / \ln(R_{2N}/R_N).$$

I have also calculated another finite-size estimate:

$$D_{\text{eff}}^{(2)}(N) = \ln 4 / \ln\{(R_{2N}^2 - 1/4)/R_N^2\}.$$

Obviously this estimate has the same limit for $N \to \infty$ but, as shown by Ball and Jullien (1984), its size variations are considerably smaller, leading to better extrapolations, as shown by numerical results on Brownian and linear (model I) trajectories.

The numerical results for $D_{\text{eff}}^{(1)}(N)$ and $D_{\text{eff}}^{(2)}(N)$ as a function of N^{-1} are shown in figure 2 for model II with space dimension d ranging from 2 to 8. I have considered

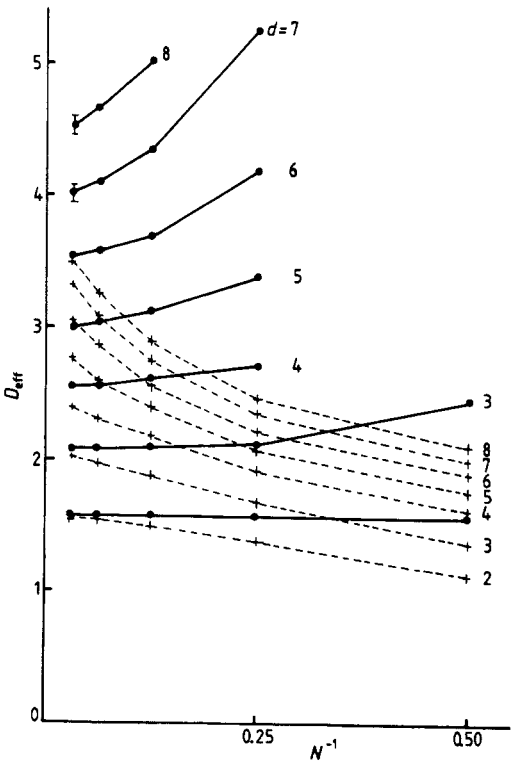

Figure 2. Plot of $D_{\text{eff}}^{(1)}$ (broken curves) and $D_{\text{eff}}^{(2)}$ (full lines) as a function of N^{-1} for model II.

2048 trials for all dimensions, starting with $P_0 = 512$ particles, stopping after 6 iterations, so that 16 384 independent clusters of 64 particles were reached.

It can be seen from figure 2 that $D_{\text{eff}}^{(2)}(N)$ allows a better extrapolation to $N \to \infty$ than $D_{\text{eff}}^{(1)}(N)$. However, in contrast with model I (Ball and Jullien 1984), one observes some size dependence which increases with d. This is a first indication that the main hypothesis used to introduce $D_{\text{eff}}^{(2)}(N)$ is Ball and Jullien (1984), which is the existence of an upper critical dimension above which $D_{\text{eff}}^{(2)}(N)$ would become size independent, might be wrong for model II.

Table 1. Numerical results for the factal dimension.

d		2	3	4	5	6	7	8
D	model I	1.51 ± 0.03	1.91 ± 0.03	2.22 ± 0.04	2.47 ± 0.05	2.7 ± 0.3		
	model II	1.56 ± 0.03	2.06 ± 0.04	2.53 ± 0.05	2.97 ± 0.06	3.46 ± 0.08	3.92 ± 0.1	4.35 ± 0.15

The extrapolated D values for model II, with error bars, are listed in table 1, and plotted against d in figure 3, where they are compared with the previous results for model I (Ball and Jullien 1984) up to $d = 5$. Another point, for $d = 6$, with a larger error bar, obtained with $P_0 = 512$ but with only 20 trials, has been added. The broken line on the same figure indicates the condition of transparency between a fixed cluster, of fractal dimension D, and a moving cluster, of fractal dimension $D + d_w$, where d_w is the fractal dimension of the trajectory ($d_w = 1$, here). This condition is written as $D + (D + 1) = d$, i.e.

$$D = \tfrac{1}{2}(d - 1)$$

(Ball and Witten 1984). As long as the clusters have a non-zero probability of sticking, their fractal dimension must stay above this broken line. The numerical results on

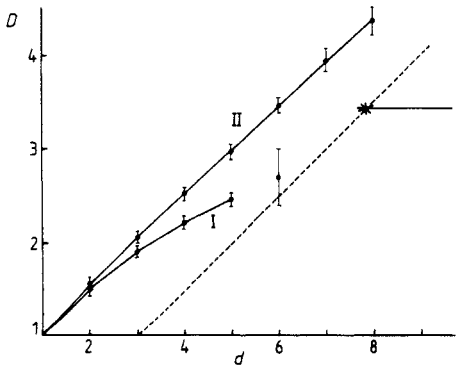

Figure 3. Curve $D(d)$ for model I and II. The broken line indicates the condition of transparency $D = \frac{1}{2}(d - 1)$. The star locates the upper critical dimension for model I, expected from the Sutherland's ghost model (Ball and Witten 1984). Above d_c, D must be constant and equal to $D_g = \ln 4 / \ln \frac{3}{2} = 3.42$.

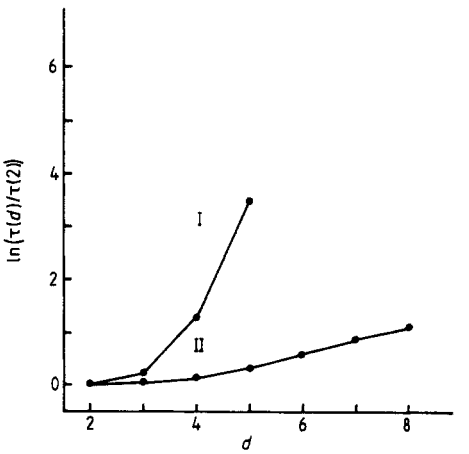

Figure 4. Plot of the logarithm of the CPU time $\tau(d)$ needed to obtain a given number of successful collisions, up to $N = 64$ (reduced by the corresponding time for $d = 2$) as a function of d.

model I strongly support the existence of a characteristic dimension d_c, above which the clusters will interpenetrate freely. Then if one assumes that the 'Sutherland's ghost' model proposed by Ball and Witten (1984) and Kolb (1984) whose fractal dimension is $D_g = \ln 4/\ln(3/2) \simeq 3.42$, is valid above d_c, $D(d)$ must reach the broken line at $d_c = 2D_g + 1 \simeq 7.84$. This is not inconsistent with the numerical results, if one assumes some sigmoidal form for $D(d)$. Unfortunately, in the case of model I, it is difficult to obtain numerical points for larger d since, due to increasing transparency effects, many more trials become necessary until a collision occurs. This is clearly shown in figure 4 where we have plotted the logarithm of the CPU time as a function of d. The dramatic increase observed in model I must be attributed to these transparency effects.

Such effects are considerably less pronounced in model II where $d = 8$ can be reached without difficulty. The curve $D(d)$ stays above and roughly parallel to the broken line, suggesting that there is no upper critical dimension. Note that the numerical results for D are near $D = \frac{1}{2}(d+1)$, although I have no justification for such a simple formula.

To explain these results, I now present a simple argument which assumes spherical symmetry and self-similarity of the clusters, for any d. Let me consider two colliding clusters whose centres of mass are G_1 and G_2 just after the collision (see figure 1). The radius of gyration of the new cluster is

$$8N^2R_{2N}^2 = \sum_{i,j} (\boldsymbol{r}_i - \boldsymbol{r}_j)^2.$$

The sum can be divided in four parts according to the fact that i and j can belong to the same old cluster, (1) or (2), or not:

$$8N^2R_{2N}^2 = 2N^2R_N^2 + 2N^2R_N^2 + 2\sum_{i_1,j_2} (\boldsymbol{r}_{i_1} - \boldsymbol{r}_{j_2})^2.$$

I use the equality

$$\boldsymbol{r}_{i_1} - \boldsymbol{r}_{j_2} = \boldsymbol{r}_{i_1} - \boldsymbol{r}_{G_1} - (\boldsymbol{r}_{j_2} - \boldsymbol{r}_{G_2}) - \boldsymbol{\delta}$$

where

$$\boldsymbol{\delta} = \boldsymbol{r}_{G_2} - \boldsymbol{r}_{G_1}$$

is the vector joining the centres of mass after the collision. Then, using the spherical symmetry hypothesis to calculate the sum and averaging over all the different ways to collide for a given direction of the trajectory, one gets

$$R_{2N}^2 = R_N^2 + \tfrac{1}{4}\langle\delta^2\rangle.$$

As any characteristic length, δ must scale like the radius of gyration R, for sufficiently large clusters. This is a direct consequence of the self-similarity hypothesis. In model I, the average is taken over all directions and one has, simply

$$\langle\delta^2\rangle \sim A_1 R_N^2 \qquad \text{(model I)}.$$

In model II, the average must be done along a single direction in space, say x, and one has instead

$$\langle\delta^2\rangle \sim A_2 R_{N_x}^2 = (A_2/d) R_n^2 \qquad \text{(model II)}.$$

In these formulae A_1 and A_2 are some unknown numbers which, *a priori*, depend on d. However, assuming that self similarity holds up to $d \to \infty$, we will suppose that

these constants stay finite (different from 0 or ∞) when $d \to \infty$. This leads to:

$$D \sim \frac{\ln 4}{\ln(1+A_1/4)} \underset{d\to\infty}{\sim} \text{constant} \qquad \text{(model I)}$$

$$D \sim \frac{\ln 4}{\ln(1+A_2/4d)} \underset{d\to\infty}{\sim} \frac{4\ln 4}{A_2} d \qquad \text{(model II).}$$

Thus, while D is expected to saturate in high dimension for model I (as is confirmed by the Sutherland ghost model with $A_1 \equiv 2$ for $d > d_c$), D must increase indefinitely with d, without an upper critical dimension, in model II.

In conclusion the existence, or not, of an impact parameter in cluster–cluster aggregation with linear trajectory have important consequences in high dimensions. Both numerical results and a simple argument suggest that the model without impact parameter should not have an upper critical dimension. Thus the consideration of long range interactions between clusters affect their fractal dimension. Note however that this change remains small in $3d$ ($D \sim 2.06$, instead of 1.91). I hope that this work will suggest some further analytical investigations on such models.

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References

Ball R C and Jullien R 1984 *Preprint*
Ball R C and Witten T A 1984 *J. Stat. Phys.* to be published
Bensimon D, Domany E and Aharony A 1983 *Phys. Rev. Lett.* **51** 1394
Botet R, Jullien R and Kolb M 1984a *J. Phys. A: Math. Gen.* **17** L75
—— 1984b *Preprint*
Jullien R, Kolb M and Botet R 1984 *J. Physique* **45** 395
Kolb M 1984 *Preprint*
Kolb M, Botet R and Jullien R 1983 *Phys. Rev. Lett.* **51** 1123
Meakin P 1983a *Phys. Rev. Lett.* **51** 1119
—— 1983b *Phys. Rev.* A **27** 604
—— 1983c *Phys. Rev.* A **27** 1495
—— 1983d *Phys. Rev.* A **27** 2616
—— 1983e *J. Colloid. Interface Sci.* **96** 415
—— 1984 *Phys. Rev.* A **29** 997
Schaefer D W, Martin J E, Wiltzius P and Cannel D S 1984 *Phys. Rev. Lett.* **52** 2371
Sutherland D N and Goodarz-Nia I 1971 *Chem. Eng. Sci.* **26** 2071
Weitz D A and Oliveria M 1984 *Phys. Rev. Lett.* **52** 1433
Witten T A and Sander M 1981 *Phys. Rev. Lett.* **47** 1400