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LETTER TO THE EDITOR

Intrinsic anisotropy of clusters in cluster–cluster aggregation

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Abstract. The anisotropy ratio between the largest and the smallest eigenvalues of the radius of gyration tensor R_{ij}^2 has been analytically investigated and numerically estimated for three different cluster-cluster aggregation processes. This ratio is of the order of 4.5 for a two-dimensional projection of a three-dimensional cluster grown by cluster-cluster processes and is quite independent of the precise nature of the cluster trajectories.

In the last five years, two main classes of models for the aggregation of particles have been introduced and numerically investigated: particle-cluster (PC) (Witten and Sander 1981) and cluster-cluster (CC) (Meakin 1983a, b, Kolb *et al* 1983) models. They successfully explain the tenuous *fractal* structure of aggregates which shows up in several condensation experiments (Landau and Family 1984, Boccara and Daoud 1985, Stanley and Ostrowski 1985, Pietronero and Tosatti 1986, Herrmann 1986). In many cases, the unique parameter for quantitative comparisons between simulations and experiments has been the so-called fractal dimension, D, which tells how the mean radius, i.e. the radius of gyration, R_g, varies with the number of particles, N, in the aggregate:

$$R_g^2 \sim N^{2\nu} \qquad \text{with } \nu = 1/D. \tag{1}$$

While D is a very interesting global quantity which can be directly measured in many experiments, it ignores some intrinsic details, one of which is the intrinsic anisotropy of the clusters, i.e. the possible deviation from a spherical shape, which appears in some specific experiments (transport properties, induced polarisation, etc). Here we do not consider the induced anisotropy which is due to the subjacent lattice (Ball and Brady 1985, Kolb 1985, Meakin 1986).

The anisotropy properties of clusters of particles have been recently studied in some models: random walks (Kuhn 1934, Solc 1971, Bishop and Michels 1985), self-avoiding walks (Rudnick and Gaspari 1986) and lattice animals and percolation (Freche *et al* 1985, Family *et al* 1985, Garik 1985, Lam 1986). Since the very beginning of the CC model, it has been recognised that clusters grown by this process were anisotropic (Hentschel 1984, see also Allain and Jouhier 1983), taking into account some very old results (Medalia 1967, 1970, Medalia and Heckman 1971, Sutherland and Goodarz-Nia 1971, Ravey 1975). In this letter, we investigate both numerically for $d = 2, 3, 4$ and analytically for any d the anisotropy of clusters grown with the different known CC models: the original version (Meakin 1983a, Kolb *et al* 1983) which considers Brownian diffusion (BCC), the one which considers linear trajectories (LCC) (Ball and Jullien 1984, Meakin 1984) and the chemical model (CCC) (Jullien and Kolb

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1984, Kolb and Jullien 1984, Brown and Ball 1985) which corresponds to considering a vanishing sticking probability. In all these calculations the 'hierarchical' procedure (Botet *et al* 1984) is systematically used.

As in Family *et al* (1985), we define the radius of gyration tensor R_{ij}^2, where the indices i and j refer to the i and j coordinates ($1 \leq i, j \leq d$ in a d-dimensional space). Diagonalising this tensor gives the principal radii of gyration, that we denote by R_i^2, where the single index i now labels the eigenvalues ($1 \leq i \leq d$) that we put in the following conventional order:

$$R_1^2 \geq R_2^2 \geq \ldots \geq R_d^2. \tag{2}$$

Due to the invariance of the trace of the tensor R_{ij}^2, we have

$$\sum_{i=1}^{d} R_{ij}^2 = R_g^2. \tag{3}$$

Moreover, since we consider self-similar objects, we will assume that all the radii R_{ij}^2 and R_i^2 scale asymptotically with N as $N^{2\nu}$ as for R_g^2 (see (1)).

Now we would like to show that the clusters are anisotropic in any dimension d and find a lower bound for the largest anisotropy ratio:

$$A_1 = R_1^2 / R_d^2.$$

This comes directly from the hierarchical process of aggregation. When two clusters of the same number of particles stick together, the averaged square distance $\langle \Delta^2 \rangle$ between their centres of mass can be derived when calculating the radius of gyration of the final cluster as a function of the radii of gyration of the two colliding clusters (Ball and Witten 1984, Jullien 1984, Botet 1986):

$$\langle \Delta^2 \rangle = 4(R_g^2(2N) - R_g^2(N)).$$

Using (1), this can be written as

$$\langle \Delta^2 \rangle = 4(2^{2\nu} - 1) R_g^2(N). \tag{4}$$

Let us call $\boldsymbol{a}$ the axis which goes through the two centres of mass and express R_{aa}^2 as a function of the R_i^2 for the two clusters:

$$R_{aa}^2 = \tfrac{1}{2}(R_1^2 \cos^2\theta_1 \cos^2\theta_2 \ldots \cos^2\theta_{d-1} + R_2^2 \cos^2\theta_1 \cos^2\theta_2 \ldots \cos^2\theta_{d-2} \sin^2\theta_{d-1} + \ldots$$
$$+ R_d^2 \sin^2\theta_1 + \Delta^2/4) + \mathrm{CT}$$

where the θ_i are the angles between the $\boldsymbol{a}$ axis and the principal axes of one cluster and where CT denotes the corresponding terms written for the other cluster. Now we must average over all the angles $\theta_1, \theta_2, \ldots, \theta_{d-1}$ for both clusters, without knowing their precise probability distribution. Let us define $\alpha_i = \tfrac{1}{2}\langle \cos^2\theta_1 \ldots \cos^2\theta_i \rangle$; one has

$$R_{aa}^2 = (R_1^2 - R_2^2)\alpha_{d-1} + (R_2^2 - R_3^2)\alpha_{d-2} + \ldots + (R_{d-1}^2 - R_d^2)\alpha_1 + R_d^2/2 + \langle \Delta^2 \rangle / 8 + \mathrm{CT}.$$

Since, from (2), all the quantities $R_i^2 - R_{i+1}^2$ are positive as well as the α_i, one deduces the inequality

$$R_{aa}^2 \geq R_d^2 + \langle \Delta^2 \rangle / 4.$$

Using the fact that the largest eigenvalue of the tensor of radii of gyration is obviously not smaller than R_{aa}^2 and using the scaling properties, we find

$$2^{2\nu} R_1^2(N) = R_1^2(2N) \geq R_{aa}^2(2N) \geq R_d^2(N) + \langle \Delta^2 \rangle / 4$$

from which we extract a lower bound for the anisotropy:

$$A_1 \geqslant 2^{-2\nu}(1+\langle\Delta^2\rangle/(4R_d^2)).$$

Combining (2)–(4), we get

$$\langle\Delta^2\rangle/(4R_d^2) = (2^{2\nu}-1)R_g^2(N)/R_d^2(N) \geqslant d(2^{2\nu}-1)$$

which allows us to derive the following inequality:

$$A_1 \geqslant d(1-2^{-2\nu})+2^{-2\nu}. \tag{5}$$

We can also find a lower limit independent of the fractal dimension. Since we know (Ball and Witten 1984, Botet 1985) that $D \leqslant D_c = \ln 4/\ln(3/2)$ for any space dimension, we find

$$A_1 \geqslant (d+2)/3.$$

As a direct consequence, for any hierarchical cluster-cluster aggregation process and for any space dimension (different from one) the resulting clusters are anisotropic and the anisotropy ratio A_1 between the largest and the smallest eigenvalues must tend to infinity, at least linearly in d, when d tends to infinity. This lower bound is, however, certainly greatly underestimated and a numerical calculation is needed to estimate reasonable values for the A_i.

Using the hierarchical procedure previously described (Botet *et al* 1984), we have built, in two dimensions, up to 100 independent clusters of 1024 particles in the three models BCC (on lattice), LCC (off lattice) and CCC (on lattice). The eigenvalues R_1^2 and R_2^2, calculated for each cluster separately, have been averaged over the whole collection of clusters of 2^k particles at each step k of the iterative procedure. The results for the ratio $A_1 = \langle R_1^2\rangle/\langle R_2^2\rangle$ are given in figure 1, as a plot of A_1 against $1/N$. Within the rather large fluctuations, A_1 is roughly size independent, yielding good confidence on its extrapolation to infinite size. A_1 is estimated to be 5.7 ± 0.2, 5.2 ± 0.2 and 4.7 ± 0.2 in BCC, LCC and CCC, respectively. Although slightly larger, the value

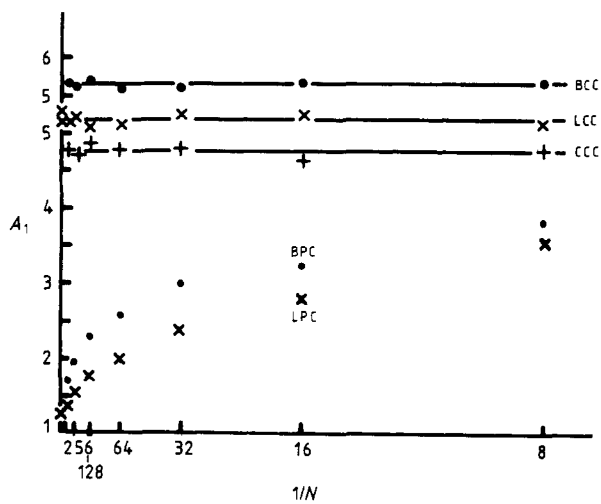

Figure 1. Plot of the anisotropy ratio A_1 as a function of $1/N$ for three different cluster–cluster aggregation processes (BCC, LCC and CCC, see text) and for two different particle–cluster processes (BPC and LPC).

found for A_1 in the case of linear trajectories corresponds to the one reported previously (Sutherland and Goodarz-Nia 1971). When comparing the three models studied here one observes a systematic decrease of A_1 when increasing the fractal dimension. The anisotropy, as well as the fractal dimension, is however only slightly dependent on the nature of the trajectory and the quantitative differences are only slightly larger than the error bars. The fact that the anisotropy ratio A_1 is quite independent of cluster size can be related to the fact that the correction term to the scaling equation (1) appears to be a constant (Ball and Jullien 1984). If this can be generalised to all eigenvalues R_i^2, anisotropy ratios must behave as

$$A(N) \sim A(\infty) + O(1/R_g^2)$$

so that the corrective term should be about two orders of magnitude smaller than A for all the sizes reported in figure 1.

For comparison, we have also shown in figure 1 the values of A_1 calculated for two particle-cluster aggregation processes with linear (LPC) and Brownain (BPC) trajectories (which is the original DLA model of Witten and Sander (1981)). As expected, the limiting value for the anisotropy is 1. This means that the PC clusters have equal principal radii of gyration in the thermodynamic limit. We must note that the diamond shape recently observed for some PC clusters on a square lattice (Ball and Brady 1985, Freche *et al* 1985, Meakin 1986) is another kind of anisotropy which is induced by the subjacent lattice and which is not in contradiction with equal eigenvalues along the orthogonal principal axes. In figure 1, we observe strong size dependence for the anisotropy of PC models, which, in the Brownian case, can be estimated to be

$$A_1 \sim 1 + aN^{-b}$$

with $a = 6.3 \pm 0.2$ and $b = 0.32 \pm 0.04$. This scaling is reminiscent of the correction terms to (1) for the DLA process (Kolb 1985). The case of linear trajectories is more complicated and no power law dependence is found for A_1 against N. There could be perhaps some logarithmic corrections as already proposed for this model (Meakin 1983b).

To summarise the results displayed in figure 1 and table 1, one can say that in 2D cluster-cluster aggregates are anisotropic and that their anisotropy ratio ($A_1 \sim 5$) does not depend too much on the particular version used. They are completely different from the particle-cluster models for which the anisotropy ratio tends to 1 for large cluster sizes, but with strong size dependence.

In table 2, we have reported the results for the $d-1$ anisotropy ratios $A_i = \langle R_i^2 \rangle / \langle R_d^2 \rangle$ for space dimensions $d = 2, 3, 4$. The calculations have been performed on the LCC model only since small differences between the models exist (quantitative differences are then of the order of the error bars). Here also we have built clusters up to 1024

Table 1. Anisotropy ratio between the largest and the smallest eigenvalues of the radius of gyration tensor, in dimension 2, estimated from finite-size extrapolations (figure 1) in the case of three cluster-cluster aggregation models.

	Brownian	Linear	Chemical
A_1	5.7 ± 0.2	5.2 ± 0.2	4.7 ± 0.2
D	1.42 ± 0.003	1.51 ± 0.03	1.56 ± 0.03

Table 2. Anisotropy ratios $A_i = \langle R_i^2 \rangle / \langle R_d^2 \rangle$ between the ith and the lowest eigenvalues of the radius of gyration tensor, estimated from finite-size extrapolations in the case of the cluster-cluster aggregration model with linear trajectories, for space dimensions up to $d = 4$. The anisotropy ratio A_1' for a two-dimensional projection of a three-dimensional cluster is also given.

d	A_1	A_2	A_3	A_1'
2	5.2 ± 0.02			
3	10.0 ± 0.3	2.5 ± 0.3		4.5 ± 0.3
4	14.0 ± 0.5	4.2 ± 0.4	2.0 ± 0.3	

particles but the number of generated clusters descreases when increasing space dimension (100, 20 and 4 for $d = 2, 3, 4$, respectively). One observes that, for a given value of d, all the eigenvalues are different. Moreover the value of A_1 seems to grow linearly with d, as in (5), but with a considerably large slope. From an experimental point of view, it is of interest to have information about the projection of three-dimensional clusters onto a plane. The coefficient A_1' for a such a projection has been numerically calculated and is also reported in table 2. A_1' is of the order of 4.5, slightly smaller than for a two-dimensional process. To conclude, we hope that the present work will stimulate further experimental investigations.

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Note added in proof. After this letter was accepted for publication we heard of an earlier (unpublished) similar work by P Meakin, F Family and T Vicsek who found exactly the same numerical results as us, in two dimensions. They also calculated the ratio $A_1' = \langle R_1^2 / R_d^2 \rangle$ that they always found to be smaller than $A_1 = \langle R_1^2 \rangle / \langle R_d^2 \rangle$. $A_1' \simeq 4.4$ and 4.0 respectively for Brownian and linear trajectories as compared with $A_1 = 5.7$ and 5.2. We acknowledge P Meakin for providing us with these results.

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