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LETTER TO THE EDITOR

Magnetization behaviour of small particle aggregates

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Abstract. We study the magnetization behaviour of small magnetic particle aggregates as a function of temperature and an external applied field. The Monte Carlo technique is used for the simulation of the aggregates and the magnetic particles interact via dipolar interactions. We show that there is a simple relation between the magnetization and the fractal dimensionality which characterizes the clusters produced in the aggregation process.

There has been interest for a number of years in small single domain magnetic particles. Their magnetic properties depend on the structure of an individual particle [1, 2]. However particles exist within clusters, and so interparticle interactions will also influence the magnetic behaviour of the clusters and also the aggregation process by which the clusters are formed. In practice, the small size of single domain particles (approximately a few nanometres), coupled with the wide distribution in particle radii [1], make a characterization of aggregation effects very difficult, although there have been attempts to measure fractal dimensionalities of magnetic particle clusters [3]. The clustering in an ideal experiment can be regarded as an example of diffusion limited cluster aggregation (DLCA). DLCA is rather well understood when just short range forces are present. A key question is how dipolar forces between magnetic particles will affect the process. This has been addressed by Helgesen *et al* [4] in an elegant table-top experiment with iron-oxide-coated colloidal microspheres in which scaling properties were studied. Related work on ferrofluids [5] and magnetic holes [6] has also been reported in the literature. The aggregation process studied is in a low particle density regime. At high particle density, the interaction of dipolar particles can result in a liquid–gas transition [7].

In this letter we consider an idealized, but experimentally realizable, model of a system of magnetic particles with equal size. We use Monte Carlo simulations to calculate the magnetization of the system as a function of temperature and an external magnetic field. We show that there is a simple power law relating the magnetization of a cluster to its fractal dimensionality. The latter is characteristic of the sample preparation conditions.

It is convenient to perform aggregation simulations on a lattice where possible. Usually in two dimensions, when a lattice can be used, a square one is employed. However a triangular one has certain advantages [7, 8], and that is what we implemented in our simulations.

The interaction energy between a pair of particles with magnetic moments $\boldsymbol{m}_i$ and $\boldsymbol{m}_j$ separated by distance $\boldsymbol{r}_{ij}$ is

$$E_{ij} = \left[\boldsymbol{m}_i \cdot \boldsymbol{m}_j - 3\left(\boldsymbol{m}_i \cdot \boldsymbol{u}_{ij}\right)\left(\boldsymbol{m}_j \cdot \boldsymbol{u}_{ij}\right)\right]/r_{ij}^3 \tag{1}$$

where $\boldsymbol{u}_{ij}$ is the unit vector along $\boldsymbol{r}_{ij}$. The dimensionless parameter which determines the effective strength of the dipole–dipole interaction relative to the disruptive thermal energy

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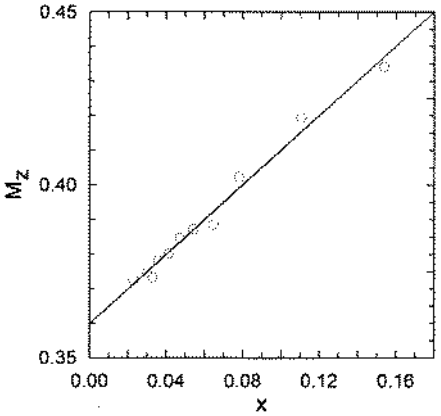

Figure 1. Zero temperature magnetization as a function of $x = N^{-1/D}$ (with $D = 1.23$) for an applied magnetic field $H_z = 2.5$ perpendicular to the substrate. Clusters with sizes between $N = 10$ and $N = 110$ particles have been used. The straight line is obtained from a least-squares fit.

is $\hat{K}_{dd} = m^2/d^3 k_B T$ and that which determines the strength of the dipole–field interaction is $\hat{K}_{df} = mH/k_B T$; m is the moment of the magnetic particle. In the dimensionless units used in our simulations, m, d and k_B are taken to be one. The dipole moments are free to reorient over three dimensions while the actual particle movements are confined to a two dimensional surface. Moments are allowed to relax after a cluster move. Simulations using a lattice of size $L = 60$, containing $N = 120$ particles at zero temperature, yielded a fractal dimensionality of 1.23 ± 0.04 [7]. This value is obtained from a least-squares straight-line fit to the radius of gyration, R_g, versus cluster size, N ($R_g \sim N^{1/D}$ where D is the fractal dimensionality). Our value for the fractal dimensionality agrees with Helgesen *et al* [4], demonstrating that simulations on a triangular lattice produce results in agreement with off-lattice models. This value of the fractal dimensionality shows the tendency of the particles to create linear chains. Within these chains the spins of the particles are aligned nose-to-tail on the plane.

For a finite cluster containing N magnetic particles and having integer dimensionality ($D = 1, 2, 3$), one can easily show that the magnetization as a function of temperature T and the applied magnetic field H obeys the power law

$$M = a + bN^{-1/D}. \tag{2}$$

Physically, a can be interpreted as the bulk contribution to the magnetization and $bN^{1/D}$ the contribution due to the surface of the cluster. The parameters a and b are functions of temperature and the applied magnetic field.

It is anticipated that the same power law will be obeyed in the case of clusters with non-integer (fractal) dimensionality. Our simulations have demonstrated the validity of this power law for magnetic clusters with fractal dimensionality $D = 1.23$. This can be seen in figure 1, where we plot the magnetization at zero temperature as a function of x (where $x = N^{-1/D} \sim 1/R_g$) for an applied field $H_z = 2.5$ perpendicular to the substrate. For a fractal object, bulk and surface are not so clearly distinct and so equation (2) should perhaps be better regarded as a convenient way of expressing finite-size effects.

A practical implication of this simple power law is that knowing the fractal dimensionality of an aggregate, which depends on the preparation conditions, one can

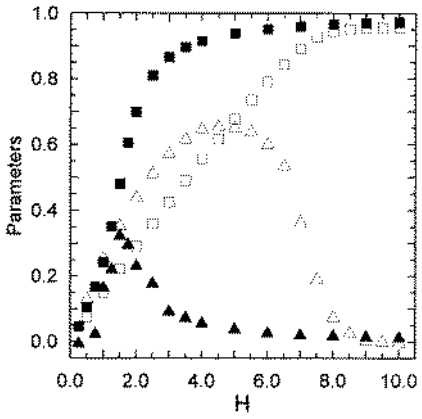

Figure 2. Field dependence of the parameters a (squares) and b (triangles) at zero temperature for an in-plane applied field (full symbols) and a field normal to the surface (open symbols).

estimate its magnetization behaviour.

Let us now examine the field dependence of the parameters a and b at zero temperature. In figure 2 we plot a and b versus field in the case of an in-plane applied field and for a field applied normal to the surface. In both cases the parameters a and b initially increase with increasing field. There is a crossover value of the field and after this the parameter a increases further up to its saturation value while the parameter b decreases to zero. This behaviour confirms the origin of the two contributions to the magnetization as the bulk (first term) and the surface (second term) contribution in equation (2). We might expect that the cross-over field will be lower for the in-plane case since, as we showed above, the spins in the clusters tend to align in the plane; this also results in the earlier elimination of surface effects reflected in the low values of b in this case.

The cross-over field in both cases occurs around the point at which we start to have deviations in the magnetization from a linear (paramagnetic) behaviour.

In figure 3 we demonstrate the temperature dependence of the parameters a and b for a fixed applied field. We choose the cross-over field values for both cases, namely $H_z = 5.0$ for the normal-to-surface field and $H_{xy} = 2.0$ for the in-plane field.

Again the behaviour of the parameters reflects the character of their origin. The surface contribution is more temperature dependent than the bulk one, due to the fact that the surface spins, having less neighbours than the bulk spins, experience a weaker local field and they respond better to the thermal effects.

Usually the physical systems under examination have an intrinsic anisotropy. In our work we have ignored the effect of anisotropy during the aggregation process and in the study of the magnetization behaviour of the clusters. In a previous work [9] we have examined the interplay of uniaxial anisotropy and dipolar interactions in a system composed of small magnetic particles and we have shown that for anisotropy energy up to about the dipolar energy, the behaviour of the magnetization is mainly determined by the dipolar interactions. We therefore expect that inclusion of a weak anisotropy in our present study will affect the aggregation process, resulting in a different fractal dimensionality, but the scaling law, equation (2), will still hold, with modified values of the parameters.

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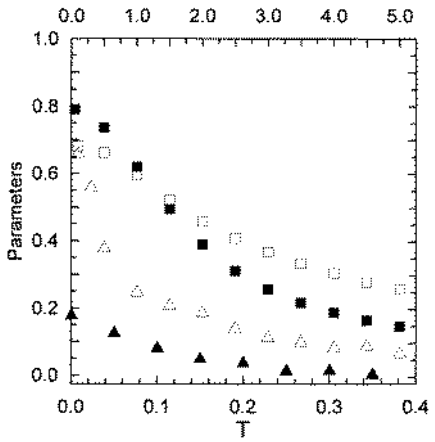

Figure 3. Temperature dependence of the parameters a (squares) and b (triangles) for an in-plane applied field $H_{xy} = 2.0$ (full symbols) and a normal-to-the-plane field $H_z = 5.0$ (open symbols). Full triangles refer to the lowest temperature axis, and all other symbols refer to the upper temperature axis.

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