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Dimensionality effects on the magnetization processes in arrays of superparamagnetic nanoparticles

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Abstract: We analyze the effect of inter-particle coupling and clustering on the zero-field cooled (ZFC) magnetization processes of superparamagnetic nanoparticles. The static interaction field in dense assemblies of nanoparticles of different dimensionalities (1D, 2D and 3D) is investigated by numerical micromagnetic simulations. An analytical model is then used in order to evaluate the influence of the dimensionality of these systems on the topology of zero-field cooled magnetization processes. Mutual static magnetic interactions are almost negligible in 3D arrays but become very important in systems of reduced dimensionality such as chains or layers of nanoparticles and they can account for up to 20% shift in the peak temperature of ZFC curves. The present results can be useful in evaluate the influence of interactions on the topology of ZFC magnetization curves.

Keywords: interaction field, superparamagnetic particles, Fast Fourier Transform (FFT), conjugate gradient (CG), zero-field cooled (ZFC) magnetization processes

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1 Introduction

Magnetic particles present a continuously increasing interest due to their potential applications ranging from magnetic recording to various biomedical applications [1, 2]. Contrary to ferromagnetic particles that have permanent magnetic moments even in the absence of any applied magnetic field, the superparamagnetic (SP) ones present the very interesting feature of acquiring non-zero magnetic moments only when submitted to magnetic fields. This feature solves many problems related to aggregation in the preparation of biological samples as well as to the re-dispersion of the particles after the magnetic capture. In order to reach this interesting superparamagnetic behavior, the dimensions of the particles have to go down below a certain critical size [3], usually from 5 to 15 nm, depending on the employed magnetic material. These small dimensions coupled with relatively low magnetization values and high saturation fields [4] make both manipulation and detection of individual SP particles very difficult [5]. An interesting approach to the synthesis of SP particles of larger diameters and increased magnetic moments, is to embed several small SP nanoparticles in a spherical non-magnetic matrix (see Refs. from 11 to 19 in [4]). The large surface of these beads coupled with stronger magnetic moments, as well as the possibility of bio-functionalization by coating biocompatible metals onto their surface make these aggregates very interesting from an application point of view. However, overall magnetic and hydrodynamic properties can not be obtained without accurate identification of their intrinsic physical parameters such as the magnetic susceptibility, saturation magnetization, coercive field, blocking temperature, magnetophoretic mobility, etc.

A technique able to provide rapid characterization of ensembles of superparamagnetic nanoparticles is the "zero-field cooled" (ZFC) method and it consists of the temperature dependence of the magnetic moment measured by cooling the sample in zero field, applying the field at low temperature and then measuring the magnetization while raising the temperature by steps. One important feature of this curve is its peak whose temperature is related to the anisotropy energy barrier in nanoparticles [6] and for most systems is expected to shift toward lower temperatures at increasing values of the applied field. Systematic changes in the topology of the ZFC curves obtained on superparamagnets of controlled-packing-ratios [7] or different dimensionalities [8] are usually attributed to magnetic interactions between particles. However, the complexity of the physical mechanisms involved in these processes have raised interesting debates on the role played by interactions in the dynamical behavior of the particle magnetization states [9-11]. Although simple analytical models [12, 13] can be employed for 1D or even 2D aggregates, several numerical approaches to the interaction field in 3D assemblies have also been proposed: the dipole-dipole interaction model [14-16] for explaining the chaining phenomenon [1, 14, 17-21] or the ZFC curves [15, 16], statistical Boltzmann approaches [9], fully 3D micromagnetic models [13, 22] as well as the phenomenological "T*" model [23] for accounting the dynamic interactions in ZFC modeling. In these approaches, especially the last one ([23]), the interaction field between

particles is considered to exert a disordering random torque to the magnetic moments and decrease the ordering effect played by the applied magnetic field (similar to the temperature effects). As a consequence, the ZFC magnetization curve is shifted toward lower temperatures by increasing the applied (static) field and toward higher temperatures by increasing mutual (dynamic) interactions between particles [24]. Thus, the two effects can somehow “weaken/cancel” each other in assemblies of close-packed nanoparticles (especially 1D and 2D) because of the geometry of the cluster that locally modifies the total applied magnetic field. In this paper we evaluate the influence of clustering and chaining on the static field in large ensembles of particles. We extend the micromagnetic approach proposed in Refs. [13, 22] by a coupled Fast Fourier Transform (FFT) + conjugate gradient (CG) algorithm. We then compute the dependence of the interaction field in clusters of different dimensionalities and evaluate the influence of this field on the peak temperature of simulated ZFC.

2 Micromagnetic algorithm

We consider an assembly of n identical superparamagnetic spheres of unitary magnetic susceptibility $\chi = 1$ and located at the nodes of a regular cubic lattice of constant d and dimensions N_x , N_y and N_z . Each particle is labeled by a vector index $\vec{i} = (i_x, i_y, i_z)$ where $i_x = \overline{1, N_x}$, $i_y = \overline{1, N_y}$ and $i_z = \overline{1, N_z}$. The (static) equilibrium magnetization $\vec{M}_{\vec{i}}$ of the particle $\vec{i}$ is given by

$$\vec{M}_{\vec{i}} = \chi \left(\vec{H}_0 + \vec{H}_{\text{demag}, \vec{i}} + \sum_{\vec{j} \neq \vec{i}} \vec{H}_{\vec{j} \rightarrow \vec{i}} \right) \quad (1)$$

where the terms in the parenthesis correspond to the external applied field, the self-demagnetizing field and the interaction field, respectively. For spherical and uniformly magnetized particles, the self-demagnetizing field can be expressed as

$$\vec{H}_{\text{demag}, \vec{i}} = -\frac{1}{3} \vec{M}_{\vec{i}}. \quad (2)$$

The field $\vec{H}_{\vec{j} \rightarrow \vec{i}}$ generated by the particle $\vec{j}$ at the lattice point $\vec{i}$ will take the general form

$$\vec{H}_{\vec{j} \rightarrow \vec{i}} = \mathbf{a}_{\vec{i}, \vec{j}} \vec{M}_{\vec{j}} \quad (3)$$

where $\mathbf{a}$ is a 3×3 symmetric matrix whose diagonal and off-diagonal elements are given by respectively

$$(\mathbf{a}_{\vec{i}, \vec{j}})_{\alpha\alpha} = \frac{\nu}{4\pi d^3} \left[\frac{3(j_\alpha - i_\alpha)^2}{|\vec{j} - \vec{i}|^5} - \frac{1}{|\vec{j} - \vec{i}|^3} \right] \quad (4)$$

and

$$\left(\mathbf{a}_{\vec{i},\vec{j}}\right)_{\alpha\beta} = \frac{\nu}{4\pi d^3} \left[\frac{3(j_\alpha - i_\alpha)(j_\beta - i_\beta)}{|\vec{j} - \vec{i}|^5} \right], \quad (5)$$

with $\alpha, \beta = \overline{1,3}$ corresponding to the directions x , y and z . By rearranging the terms in Eq. (1) the problem of finding static magnetization values of individual particles can be easily reduced to a linear system of the form [13, 22]

$$\sum_{\vec{j}} \mathbf{A}_{\vec{i},\vec{j}} \vec{M}_{\vec{j}} = -\vec{H}_{0,\vec{i}} \quad (6)$$

with the magnetizations $\vec{M}_{\vec{j}}$ as unknowns. Since the particles are located at the nodes of a regular lattice, all the elements of the matrix $\mathbf{A}$ in Eq. (6) depend on the vector indices $\vec{i}$ and $\vec{j}$ through their difference $\vec{j} - \vec{i}$ only and consequently, the linear system can be rewritten as

$$\sum_{\vec{j}} \mathbf{A}_{\vec{j}-\vec{i}} \vec{M}_{\vec{j}} = -\vec{H}_{0,\vec{i}}. \quad (7)$$

Since the left side of the equation above is basically a convolution, the related matrix-to-vector products can be very efficiently evaluated by applying the convolution theorem and 3D Fast Fourier transforms [25] and the system solved iteratively by using a conjugate-gradient (CG) method [26, 27]. In our numerical implementation, we used the Fastest Fourier Transform in the West (FFTW) by Frigo and Johnson [28].

The system (7) was then solved for clusters of particles of different sizes and dimensionalities. In order to have an insight into the dependence of the magnetization state on the dimensionality of the assembly, the size of the system was varied in a systematic manner such that it gradually evolves from a single particle toward a cube of $N \times N \times N$ particles and back, such that planes and chains of particles of different orientations with respect to the applied field are considered (Figure 1). First we consider a uniform magnetic field along the vertical axis Oz and a single particle at the origin of the reference frame. The system is gradually extended on the Oz direction by adding particles until we obtain a linear chain of $N_z = N$ particles (so $N_x = N_y = 1$). We then gradually extend the system in 2D by adding chains of N particles along Oy until $N_y = N_z = N$ and further on in 3D by adding planes on Ox until we obtain a cube. In this way, the dimensionality of the cluster is systematically varied from 0 through 1D and 2D to 3D. Now we decrease the dimensionality of the cluster back to zero, by eliminating planes on Oz until we obtain a sheet of particles in the xOy plane and then by eliminating chains on Ox in order to get a simple chain along Oy . Thus, by gradually eliminating particles from this chain, we arrive back to the initial configuration of zero dimensionality (one single particle). The equilibrium magnetization state of all the particles in these clusters are computed for different values of N namely 5, 10 and 100 (for arrays of $100 \times 100 \times 100$ particles the amount of necessary memory is about 3 Gb but as we can see in the following, there is no need to go further with larger systems). For particles of 5 nm diameter and lattice constants of 10 nm, the above dimensions corresponds to respectively 50 nm, 100 nm and 1 μm for the size of the simulated cubes. The output quantity is the relative variation $\varepsilon_M = \Delta \bar{M}_z / M_0$ of the average magnetization

in the direction of the applied field ($\overline{M_z}$) with respect to the magnetization of isolated (non-interacting) particles (M_0). The dimensionality of the cluster is evaluated by using the expression

$$\zeta = \frac{\ln n}{\ln N} \quad (8)$$

where n is the total number of particle in the cluster and N the dimension of the considered cube. For a single particle $n=1$ and $\zeta=0$, for a square sheet of $N \times N$ particles $\zeta=2$ whereas for $N \times N \times N$ cubes of identical particles ζ is obviously 3.

3 Interaction field and dimensionality effects

The dependence $\varepsilon_M(\zeta)$ obtained by solving the system (7) for different configurations of particles is shown in Figure 2. The average magnetization of the cluster is enhanced ($\varepsilon_M > 0$) for 1D and 2D configurations parallel to the applied field and decreased in perpendicular fields. The relative variation of the magnetization in chains of SP particles parallel to the applied field reaches a maximum of $\varepsilon_M \cong 17.2\%$ for $N \rightarrow \infty$ but is reduced by about 7% when the chains and the applied field are perpendicular. Similarly, magnetization values of 2D distributions of particles are enhanced in parallel configurations and inhibited in perpendicular ones. However, while the enhancement of magnetization in 1D parallel configurations surpass the attenuation in perpendicular ones, for 2D distributions the situation is reversed: average magnetization values of perpendicularly polarized sheets of particles drop to about 20% with respect to isolated particles whereas parallel polarized configurations are comparable with parallel linear chains. The fact that the magnetization is enhanced in parallel chains ($1D_{\parallel}$) more than it is reduced in parallel configurations ($1D_{\perp}$) means that the chaining of particles will always enhance the magnetization in a sample unless the most chains are perpendicular to the applied magnetic field. 2D layers of particles will have a reversed effect since perpendicular configurations ($2D_{\perp}$) reduce the magnetization more that it is enhanced in the parallel ones ($2D_{\parallel}$). However, if this difference is about 10% in the one-dimensional case, it accounts for 5% or even less in the two-dimensional one.

It is very interesting to observe on Figure 2 that the average magnetization in 3D assemblies is almost identical to isolated (non-interacting) particles thus the effect of clustering in these ensembles is not present. We can safely consider the dynamic effects [23] as the only ones responsible for the interparticle interactions in these systems. Moreover, the influence of the dimensionality on the total magnetic moment of the cluster becomes important for relatively small clusters: chains of 5 particles oriented parallel to the applied field correspond already to about 12% enhancement of their magnetization whereas the limit for infinite chains goes to 17% (Figure 2) that is a difference of 5% only.

4 ZFC magnetization processes

We consider a ZFC analytical model with a monotonic field dependence of the blocking temperature, as presented in Ref. [6]. According to this model, a ZFC magnetization process is well described by

$$m_{ZFC}(T; H) = \frac{\mu_0 M_s^2}{3K_{eff}} H \Gamma_3|_{D_b}^\infty + \frac{\mu_0 \pi M_s^2}{18k_B T} H \Gamma_6|_0^{D_b} \quad (9)$$

where μ_0 and k_B are respectively the vacuum magnetic permeability and the Boltzmann constant, M_s the saturation magnetization, K_{eff} the effective anisotropy constant of the particles, H the applied magnetic field and T the sample temperature. D_b is the diameter of the particles (of volume V and coercive field H_c) that corresponds to a given blocking temperature

$$T_b = \frac{K_{eff} V (1 - H/H_c)^\alpha}{28k_B}. \quad (10)$$

The coefficient α in the equation above is usually taken 1.5, this value corresponding to a random orientation of the particles anisotropy axes. Moreover, we employed the notation

$$\Gamma_n|_a^b = \int_a^b D^n f(D) dD \bigg/ \int_0^\infty D^3 f(D) dD \quad (11)$$

with D the diameter of particles and $f(D)$ the well known log-normal distribution function.

In the following we consider nanoparticles of mean sizes between 5 and 10 nm, $K_{eff} = 10^5 \text{ J/m}^3$, $H_c = 4200 \text{ A/m}$ and different standard distributions (from 0.1 to 0.3). ZFC magnetization curves are simulated by using Eq. (9) and the peak position of these curves evaluated at different sizes, distributions and dimensionalities. The effect of the dimensionality is included in this model by adding an interaction field as given by (7) to the external applied magnetic field H . In this way, changes in magnetization of particles due to the dimensionality are considered equivalent with variations of the applied field H in non-interacting systems. The overall effect of the clusters of particles on these processes can be seen in Figure 3.a) and is related to Eq. (10). The 3D configuration corresponds to the non-interacting case, where the total field is given by the external applied magnetic field only (2100 A/m). An important shift of the ZFC peak to lower temperatures is observed for $1D_{||}$ and $2D_{||}$. The shift is almost the same for these configurations since the interaction field is almost the same (as seen on Figure 2). On the contrary, perpendicular configurations ($1D_{\perp}$ and $2D_{\perp}$) shift the ZFC peak in the opposite direction (toward higher temperatures) but with very different quantities since the interactions in these systems are stronger in the two-dimensional case. As we can see in Figure 3.b), between the two extreme situations ($1D_{||}$ and $2D_{\perp}$) we can have a shift in the ZFC peak of about 20%.

5 Discussion

The numerical calculations presented in the previous section deal with relative magnetization variations ε_M of interacting nanoparticles with respect to isolated (non-interacting) ones. This quantity may easily be related to the static interaction field since the variations of the magnetization of the particles in interacting clusters originate in their mutual interactions. Consequently, any enhancement of the average magnetization values of the particles means a “positive” interaction field whereas the decrease of this quantity will stand for a “negative” one. It is very clear from Figure 2 that low-dimensional clusters of particles present positive (enhancing) interactions in parallel field and negative interactions for perpendicular applied fields. In 3D configurations, the interaction field is the result of the competition between dipolar axial and lateral interactions that are canceling each other, thus diminishing the influence of mutual interactions on the equilibrium states of the assembly. It is very interesting to observe from Figure 2 that the transition from 2D configurations with parallel magnetic field toward 3D (by successively adding more and more layers) is accompanied by a decrease of the static interaction field, due to the additional lateral interactions induced by the neighbor layers.

An interesting part of this calculation is that when particles are chaining, the magnetization is enhanced even in the case of a uniform angular distribution of the chains. This is due to the fact that there is a difference of 10% between parallel and perpendicular 1D configurations. Similar effects, but responsible for lower magnetization can be obtained with 2D clusters where the difference in the two directions is about 5% only. However, since the most probable configurations in real samples are the linear chains of particles, it is expected that the interactions be positive and enhance the overall magnetic moment. This will correspond with a shift of the ZFC peak toward lower temperatures.

Even if the calculation is done for regular grids, the presented algorithm can easily be applied for relatively disordered systems by considering some “void particles” at different points of a regular lattice. The results (interaction field or peak temperature) on such systems will always be in the interval between the extreme configurations $1D_{\parallel}$ and $2D_{\perp}$. However, the use of completely irregular grids will seriously affect the size of system under consideration (total number of particles) since the left hand of Eq. (6) can not be evaluated by the convolution theorem and the matrix $\mathbf{A}$ has to be explicitly stored in the computer memory.

6 Conclusions

In this paper we have presented a numerical approach to the static interaction field in clusters of superparamagnetic particles. We analyzed the influence of size, dimensions and interparticle spacings on the equilibrium magnetization states of individual interacting nanoparticles. The average magnetizations of interacting SP particles are enhanced in low-dimensional clusters parallel to the excitation field and decreased in perpendicular fields. A maximum difference of about 35% in the relative variation of the interaction field is observed between parallel and perpendicular polarized 2D configurations. Static interaction fields in 3D assemblies of SP particles can safely be neglected. The static field associated with the presence of low-dimensional clusters of particles can also influence the position of the peaks in ZFC measurements.

Figure 1. Schematic assemblies of superparamagnetic particles of various dimensionalities for $N=5$. Similar configuration were employed for systems of higher dimension (up to $N=100$). The applied magnetic field $\vec{H}$ is assumed to be uniform.

Figure 2. Relative variations of magnetization for assemblies of different sizes (N) and dimensionalities (ζ). Each point on this diagram has been obtained by solving (7) with the described FFT-CG algorithm.

Figure 3. a) Influence of the static field on simulated zero-field cooled magnetization processes for superparamagnetic particles assembled in clusters of different dimensionalities ($H = 0.5 H_c$); b) blocking temperatures at different applied magnetic fields and different clustering states. Different colors correspond to different mean diameters and different standard size distributions of the particles.

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