Coupling of ferromagnetic nanoparticles through dipolar interactions

A. Weizenmann, M. Santos, W. Figueiredo *

Departamento de Física, Universidade Federal de Santa Catarina, 88040-900, Florianópolis, Santa Catarina, Brazil

A R T I C L E   I N F O

A B S T R A C T

We consider two ferromagnetic nanoparticles coupled via long-range dipolar interactions. We model each particle by a three-dimensional array of classical spin vectors, with a central spin surrounded by a variable number of shells. Within each particle only ferromagnetic coupling between nearest neighbor spins is considered. The interaction between particles is of the dipolar type and the magnetic properties of the system is studied as a function of temperature and distance between the centers of the particles. We perform Monte Carlo simulations for particles with different number of shells, and the magnetic properties are calculated via two routes concerning the dipolar contribution: one assuming a mean-field like coupling between effective magnetic moments at the center of the particles, and other one, where we take into account interactions among all the pairs of spins, one in each particle. We show that the dipolar coupling between the particles enhances the critical temperature of the system relative to the case in which the particles are very far apart. The dipolar energy between the particles is smaller when the assumption of effective magnetic moment of the particles is used in the calculations.

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1. Introduction

Magnetic nanostructured systems have been investigated in the last years, especially due to their interesting technological and biomedical properties [1–3]. The physics of a single domain particle is already very complex [4], where the reversal of the magnetic moment depends crucially on the magnitude of the uniaxial energy barrier, temperature and magnetic field. However, when a set of these particles are put close together, the long-range dipolar interaction between pairs of particles becomes important. Despite the dipolar energy between two spins be much weaker than the exchange interaction, it assumes a real importance in the case of the interaction between two single domain magnetic particles, because they contain hundreds or thousands of individual spins.

When we are in the regime of very high concentrations of magnetic nanoparticles, the purely uniaxial energy barrier seen by each nanoparticle is modified due to the presence of the long-range dipolar coupling, and the magnetic properties of the system, like coercive field, remanence and blocking temperature change accordingly [5–9]. Besides the concentration, the way the nanoparticles are arranged is also important to describe the ground state properties of the system [10–14]. For instance, in three dimensions, the lowest energy state of a collection of classical dipoles is ferromagnetic in a face centered cubic lattice whereas it is antiferromagnetic in a simple cubic lattice. In two dimensions, a square

lattice of classical point dipoles exhibits an antiferromagnetic arrangement of the moments while in the case of a triangular lattice the arrangement is ferromagnetic.

In order to understand the behavior of interacting magnetic nanoparticles, we investigate the magnetic properties of a pair of nanoparticles, each one with an internal structure where the individual spins interact through exchange coupling. In a previous paper [15] we studied a similar system of two identical particles, in two dimensions, each one formed by Ising spins. We have calculated the magnetization, susceptibility, and critical temperature as a function of the number of layers in each particle, temperature, as well as, distance between them. We have shown that the dipolar energy calculated by using the true distance between individual spins is lower than approximating it by the coupling between the effective magnetic moments of the particles.

In the present work our particles are formed by classical spin vectors interacting via nearest neighbor isotropic exchange coupling of the Heisenberg type. As before, the particles interact via the long-range dipolar interaction depending on the distance between their centers. Our system is similar to the two-particle model considered by Politi and Pini [13], however our particles are not single domain ones, and the total magnetic moment of each particle can change with temperature. In the last years we have considered the magnetic properties and phase diagram of single nanoparticles with a variable number of shells as a function of temperature and external magnetic field [16–19]. In all these studies, Ising or vector spins, are coupled by ferro- or antiferromagnetic interactions, and dipolar interactions were not considered. In the present study, we add dipolar interaction between the single

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* Corresponding author. Tel.: +5548 37216834; fax: +5548 37219068.
  E-mail address: wagner@fisica.ufsc.br (W. Figueiredo).
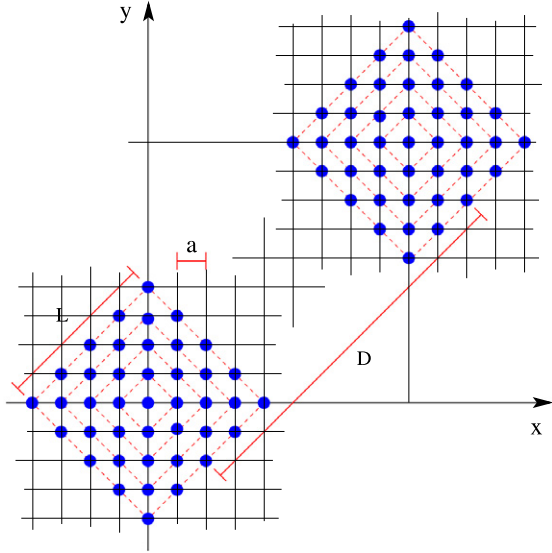
Fig. 1. Projection of the three-dimensional particles sites onto the x–y plane. The spins of the particles are placed on the sites of a simple cubic lattice with lattice parameter a, centered at the point $(0, 0, 0)$. We also show the distance between the particles' center, and the linear size of the lattice, D and L, respectively.

particles made of continuous spins. The magnetic properties are then considered as a function of the number of shells in each particle, distance between centers of the particles, and temperature. We employ in our calculations Monte Carlo simulations along with the Metropolis algorithm. Similarly, as for Ising spins, the difference in the values of the dipolar energy, calculated assuming effective spins in the center of the particles, and by directly computing the interactions among all the pairs of spins, one in each particle, presents the same dependence on distance as observed in the case of single domain interacting particles [13]. In Section 2, we describe the Hamiltonian model, the calculations based on Monte Carlo simulations, and present our results. Finally, in Section 3, we summarize the results of this work.

2. The model and results

The Hamiltonian of the model we consider in this work is the following:

$$\mathcal{H} = -J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j - J' \sum_{\langle k,l \rangle} \vec{S}_k \cdot \vec{S}_l$$
$$+ \frac{\Omega}{2} \sum_{(i,k)} \left[\frac{\vec{S}_i \cdot \vec{S}_k}{r_{ik}^3} - 3 \frac{(\vec{S}_i \cdot \vec{r}_{ik})(\vec{S}_j \cdot \vec{r}_{ik})}{r_{ik}^5} \right], \quad (1)$$

where $\vec{S}_i$ is the spin variable associated to the site i, with $|\vec{S}_i| = 1$, and $\vec{S}_i = (S_{ix}, S_{iy}, S_{iz})$. The first two sums represent the ferromagnetic exchange contributions calculated over all the distinct pairs of nearest neighbor spins of each particle, Ω is the magnitude of dipolar interaction between all the pairs of spins $\vec{S}_i$ and $\vec{S}_k$, one in each particle, and r_{ik} is the distance between them, measured in units of the lattice parameter a. One particle is centered at the origin of the coordinate system, and the spins are located at the sites of the cubic lattice built around the origin. The center of the other particle is located a distance D apart from the origin, and the spins are distributed on the sites of a simple cubic lattice. We show in Fig. 1, the projection on the x–y plane, of the sites of the particle, where the parameters a, D, and L are shown. In the first sum of Eq. (1) J is the exchange coupling between nearest neighbor spins in particle 1, and J' is the corresponding exchange coupling

between nearest neighbor spins in the particle 2. We have considered an equal number N of spins in each particle. The spins are disposed into shells, and in this work we have considered particles with up to $n = 10$ shells. In this particular case, the number of spins located in this shell ($n = 10$) is 402, and the total number of spins of the system is $N = 1561$, which means that around 26% of them are located at the surface of the particle. The dipolar parameter is $\Omega = (g\mu_B)^2/a^3$, where g is the gyromagnetic factor and μ_B is the Bohr magneton. In the ground state of the system, each particle behaves as a single domain particle, with spins of both particles pointing in the same direction. The dipolar interaction minimizes the energy of the two-particle system, and the ground state is ferromagnetic, with the total magnetic moment of the particles oriented along the line joining the particles' center. Even if the particles are very far apart, we have chosen this direction to break the rotational symmetry of the Hamiltonian of each particle. When they approach each other, the effective dipolar field of one particle on the other, minimizes the energy of the system for any values of n, D and L. Therefore, as the result of the dipolar interaction, each particle feels an effective magnetic field due to the presence of the other.

We have employed Monte Carlo simulations for the two-particle ferromagnetic system through the Metropolis algorithm [20]. In each Monte Carlo step (MCs) we performed ($2N$) random trials to change the spins of both particles. The procedure we used to determine the equilibrium states of the system is based on the minimization of the free energy. In each trial, a given spin is selected at random, in particle 1 or particle 2, and we try to move it to a new position in such way that the deviation from the old state is random, but within a maximum solid angle. Then, we calculate the change in energy of the system (ΔE), and if $\Delta E \leqslant 0$, the transition to a different configuration is accepted. On the other hand, if $\Delta E > 0$, the transition to a different configuration is made with probability $\exp(-\frac{\Delta E}{k_B T})$. To determine the average magnetic properties, we considered 3.3×10^5 MCs, where the first 3.0×10^4 MCs were discarded due to the thermalization process. This number of MCs to reach the equilibrium state was chosen by taking the maximum solid angle variation to be 0.1π, where approximately 50% of the attempted moves were successful. We have taken 100 independent samples to determine the thermal properties. We calculated the average magnetization, as well as its components in the x, y and z directions as a function of temperature. These average values were obtained, first, by calculating the mean values of the magnetization and susceptibility of the system for each MCs after the thermalization process. Afterward, an average is performed over all the selected samples. The expressions we used are the following:

$$m_x = \frac{\frac{1}{N}\sum_i S_{ix} + \frac{1}{N}\sum_k S_{kx}}{2}, \quad (2)$$

$$m_y = \frac{\frac{1}{N}\sum_i S_{iy} + \frac{1}{N}\sum_k S_{ky}}{2}, \quad (3)$$

$$m_z = \frac{\frac{1}{N}\sum_i S_{iz} + \frac{1}{N}\sum_k S_{kz}}{2}, \quad (4)$$

$$m_t = \sqrt{m_x^2 + m_y^2 + m_z^2}, \quad (5)$$

$$\chi = 2\beta N \left[\langle m_t^2 \rangle - \langle m_t \rangle^2 \right]. \quad (6)$$

In these expressions, the indices i and k run over the sites of the particles 1 and 2, respectively. The components of magnetization per spin are m_x, m_y and m_z, m_t is the total magnetization per spin, and χ is the susceptibility per spin. For simplicity, we assume the same value for the exchange coupling between nearest neighbor spins inside the particles, $J = J'$. We also use in our calculations arbitrary units, $\Omega = 1$ and $J = 1$. We did not consider in this work
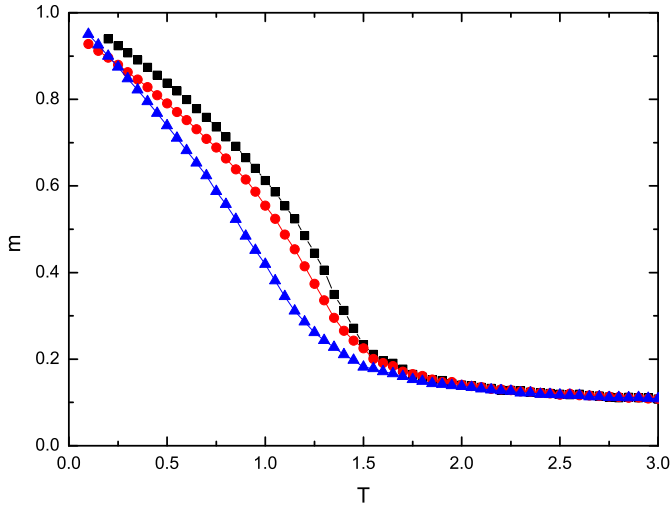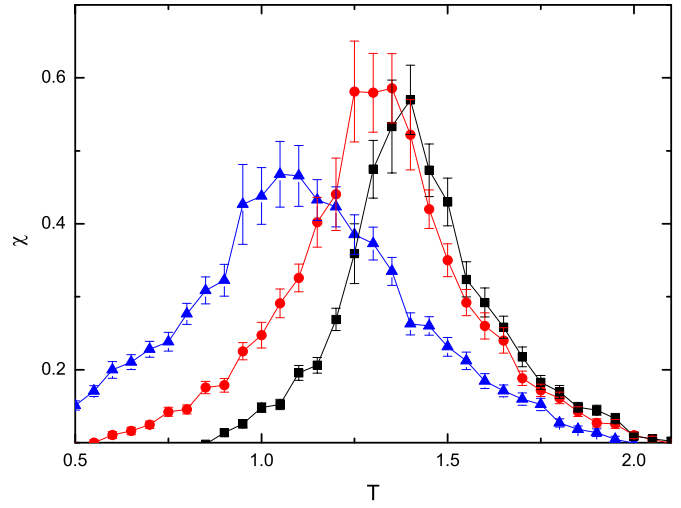
Fig. 2. Total magnetization as a function of temperature for particles with 4 shells. Triangles, noninteracting particles $(D/L \gg 1)$, circles and squares, spin–spin and particle–particle calculations, respectively, for $(D/L = 1.2)$. Magnetization and temperature are in arbitrary units.

Fig. 3. Susceptibility as a function of temperature for particles with 4 shells. Triangles, noninteracting particles $(D/L \gg 1)$, circles and squares, spin–spin and particle–particle calculations, respectively, for $(D/L = 1.2)$.

Fig. 4. Critical temperature as a function of the ratio D/a, where D is the distance between the particles' center, and a is the lattice parameter, for particles with 4 shells, and spin–spin calculation.

the dipolar interaction between spins inside the same particle because the dipolar coupling is hundreds or thousands weaker than the exchange coupling. If we had used $\Omega = 1$ and $J = 1$ inside the particles, the ground state would be very complicated with a non-uniform distribution of spins, and internal fields would need to be taken into account due to the cubic symmetry of our particles [21].

The dipolar interaction between the particles 1 and 2 is calculated in two different manners: one assuming effective magnetic moments in the centers of the particles, which we call particle–particle interaction, and the other directly computing the interactions among all the pairs of spins, one in each particle, which we call spin–spin interaction. Although the calculation based on the spin–spin interaction is more accurate, we will see next that the particle–particle calculation is a good approximation for the magnetic properties of the pair of interacting particles. When the two particles are very close, the dipolar interaction between pairs of spins in the spin–spin calculation, is locally minimized for spins oriented parallel and along the direction of their relative positions. However, due to the cubic symmetry of our particles, the overall orientation is not uniform inside each particle, and this is more pronounced for spins on the boundaries, where the coordination number is also reduced.

We show in Fig. 2, the magnetization as a function of temperature for particles with $n = 4$ shells, each one containing 129 spins. We compare the cases where the particles are very close $D = 1.2L$, and when they are far apart $(D/L \gg 1)$, for which particles are essentially noninteracting. D and L are the distance between the centers of the particles and the linear size of the cubic lattice, respectively. Note the difference between the two types of calculations at low temperatures: the magnetization is lower when the calculation of the dipolar contribution is made via spin–spin interaction. Fig. 3 exhibits the susceptibility as a function of temperature for the same particles of Fig. 1. We observe that, due to the dipolar interaction, the peak of susceptibility moves to high temperatures. The peaks in these curves indicate the critical temperature of the system. It is important to mention that the critical temperatures obtained in these Monte Carlo simulations for small particles are, actually, pseudocritical temperatures. On physical grounds, a finite system cannot display a true singularity at non-zero temperatures, but the pseudocritical temperature, as usual, can be associated with the peak in the susceptibility. It is also interesting to note that the critical temperature determined from the spin–spin calculation is smaller than the one calculated
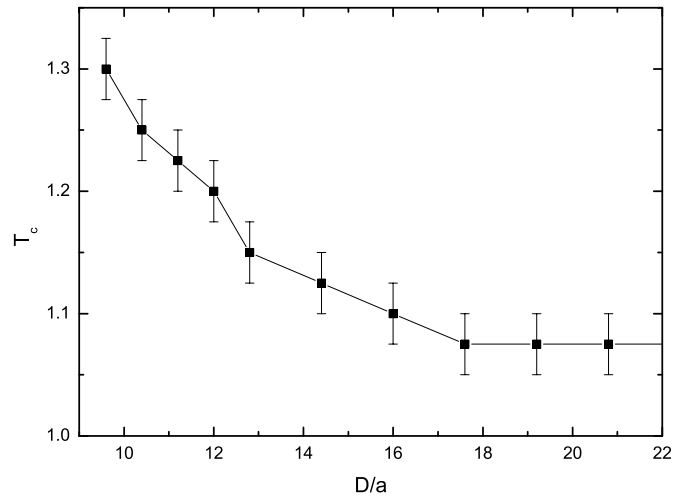
via particle–particle approximation. This is somehow expected, because the particle–particle calculation is a kind of mean field approximation that, as it is well known, overestimates the critical temperature of the magnetic systems. Fig. 4 displays the plot of the critical temperature as a function of distance between the centers of the particles for the same pair of particles considered in Fig. 2. As to be expected, the critical temperature decreases with distance between the centers, and for $D/a \approx 18$, the effects of the dipolar interaction are negligible. Fig. 5 shows the critical temperature as a function of the number of shells for two values of the relative distance between the particles, one for noninteracting particles $(D/L \gg 1)$ and the other for $D = 1.5L$. We would like to make another comment regarding the arbitrary units we have used in this work. In any real system the ratio between the exchange and the dipolar couplings (J/Ω) is very large. For instance, if we had used a value like 100 for this ratio in our calculations, the only effect would be a reduction in the difference between temperatures, for each value of n of Fig. 5, by an equivalent amount.

Fig. 6 displays the dipolar contribution to the total energy of the system for a particle with 4 shells as a function of the distance between the centers of the two particles, and at very low tem-
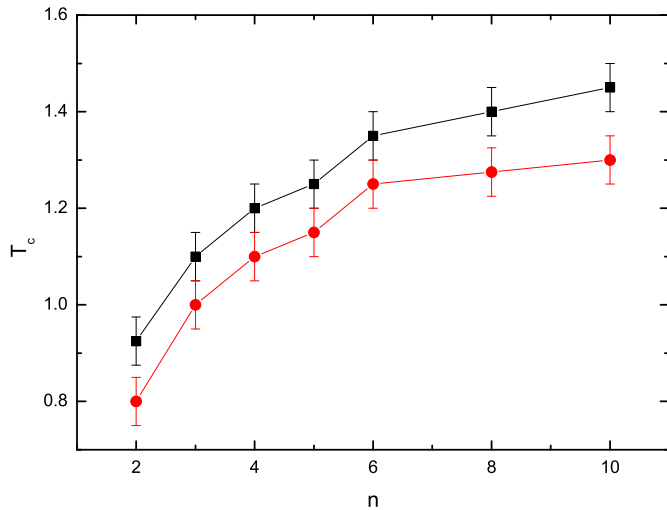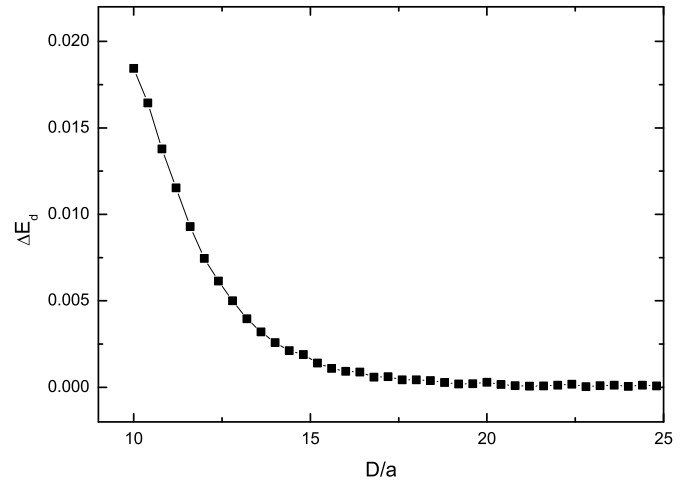
Fig. 5. Critical temperature as a function of the number of shells, for noninteracting (circles) and interacting (squares) particles, for which $D/L = 1.5$, and spin–spin calculation.
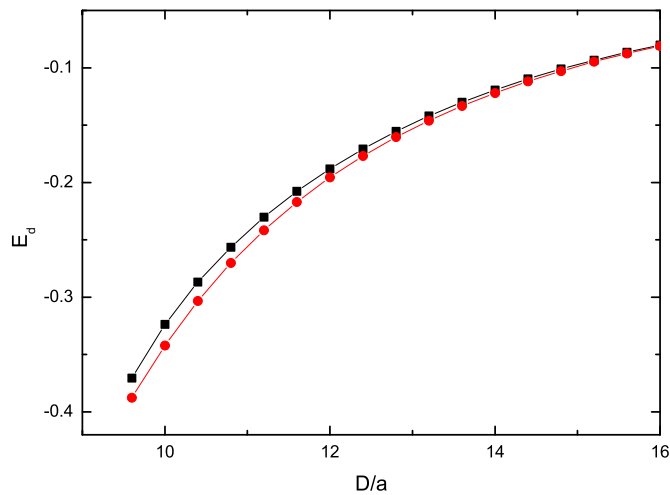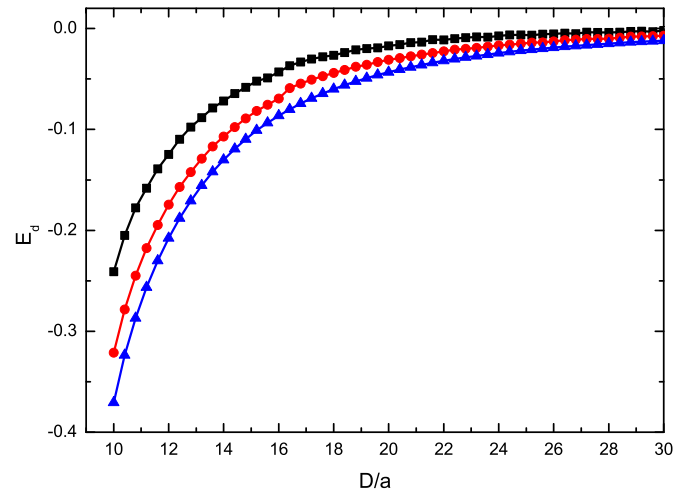

Fig. 6. Dipolar energy as a function of the distance between the centers of the two particles, each one with 4 shells, and temperature $T = 0.1$. Circles and squares are based on the particle–particle and spin–spin calculations, respectively.

Fig. 7. Difference between the values of the spin–spin and particle–particle dipolar energies as a function of the distance between the particles' center. The fit to the data points is the power law $(D/a)^{-5}$.

Fig. 8. Spin–spin dipolar energy as a function of the distance between the particles' center for three different temperatures, and particles with 4 shells. $T = 0.1$ (triangles), $T = 0.3$ (circles) and $T = 0.6$ (squares).

peratures. The calculation, made within particle–particle procedure, gives slightly lower values than that employing the spin–spin method. In the particle–particle calculation, dipolar contribution to the energy is determined assuming that the distance between any two spins is approximated by the distance between the centers of the particles, while in the spin–spin calculation we take into consideration the true distance between all the pairs of spins, one in each particle. When the distance between the particles is large enough, there is no difference between both calculations. However, when the particles are very close, the particle–particle dipolar energy is smaller than the spin–spin dipolar energy. In Fig. 7, we plot the difference between these two values of the dipolar energy as a function of the distance between the centers of the particles. The fit to these points is consistent with a dependence $(D/a)^{-5}$. Note that this result is similar to the one obtained earlier by Politi and Pini [13] on the dipolar interaction between two-dimensional magnetic particles, where they showed that the first correction term to the dipolar coupling between particles of finite sizes decays as $(D/a)^{-2}$. We also show in Fig. 8 the behavior of the dipolar energy as a function of the distance between the centers of the particles, each one with 4 shells, for three different temperatures below the

critical temperature, and within the spin–spin calculation. As we can see from these plots, for temperatures below the critical one the dipolar energy between particles does not change significantly, and becomes independent of temperature for distances larger than $D/a \approx 30$.

3. Conclusions

In this Letter, we have investigated the magnetic properties of two small ferromagnetic particles that are coupled through long-range dipolar interaction. The particles present an internal structure, cubic symmetry, and are arranged into a variable number of shells. The sites of the particles are occupied by continuous spin vectors, and we assume a ferromagnetic coupling between nearest neighbor spins. We take into consideration a dipolar coupling between the particles, and we calculate this energy by two different procedures. In the first one, which we call particle–particle interaction, the effective dipolar contribution is calculated by assuming that the distance between any two spins is equal to the distance between the particles' centers. In the second one, called spin–spin interaction, the true distance between all the pairs of spins, one in each particle, is considered. We employed Monte Carlo simulations in our calculations, and we determined the magnetization

and susceptibility as a function of temperature, number of particle shells, and distance between the particles. In this work we did not investigate the super-paramagnetic behavior because we have disregarded any type of anisotropy and demagnetizing fields, for which the orientation of x, y and z axes in the particles would change the magnetic state of the system. Our particles are not single domain ones, and the concept of blocking temperature is meaningless in our case. We focused our attention only on the critical properties of the system. Our results show that the dipolar coupling between the particles enhances the critical temperature of the system relative to the case in which the particles are far apart. The dipolar energy between the particles is smaller when the assumption of particle–particle interaction is taken into account. This contribution represents a first-order term to the dipolar coupling between the particles. The difference in energies, calculated employing the spin–spin and particle–particle procedures behaves as $(D/a)^{-5}$, which is in agreement with the results of Politi and Pini [13] for a pair of two-dimensional single domain particles.

Acknowledgements

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