INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **17** (2005) 1625–1633 doi:10.1088/0953-8984/17/10/017

Geometric aspects of the dipolar interaction in lattices of small particles

Paola R Arias[1], D Altbir[1] and M Bahiana[2]

[1] Departamento de Física, Universidad de Santiago de Chile, USACH, Avenida Ecuador 3493, Santiago, Chile
[2] Instituto de Física, Universidade Federal do Rio de Janeiro, Caixa Postal 68528, Rio de Janeiro, RJ, 21941-972, Brazil

E-mail: monica@if.ufrj.br (M Bahiana)

Received 1 December 2004, in final form 10 February 2005
Published 25 February 2005
Online at stacks.iop.org/JPhysCM/17/1625

Abstract

The hysteresis curves of systems composed of small interacting magnetic particles, regularly placed on stacked layers, are obtained with Monte Carlo simulations. The remanence as a function of temperature, in interacting systems, presents a peak that separates two different magnetic states. At low temperatures, small values of remanence are a consequence of antiferromagnetic order due to the dipolar interaction. At higher values of temperature the increase of the component normal to the lattice plane is responsible for the small values of remanence. The effect of the number of layers, coordination number and distance between particles are investigated.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In the past decade the magnetism of fine particles embedded in a non-magnetic matrix has been a topic of interest because of their uses in chemical catalysis and magnetic recording [1]. The appearance of new experimental techniques capable of generating samples with controlled nanostructures [2, 3] has led to important advances in the preparation and understanding of the behaviour of granular materials. However, at the nanometre scale, magnetic systems are not easily reproduced and characterized, introducing difficulties for the investigation of these systems. Experimental and theoretical results obtained over the past few years show that there are clearly many factors that influence the magnetic and magnetotransport behaviour of these systems, such as the distribution of grain sizes, the average size and shape of the grains and the magnetic anisotropy of the individual grains. Also, the role of magnetic interactions among crystallites is a topic full of controversies, despite the intensive research on the subject. One of the most often used methods for investigating the role of interactions has been Monte Carlo simulations. Kechrakos and Trohidou [4] have investigated the remanence and coercivity of

an assembly of single-domain ferromagnetic interacting particles at very low temperatures using a relative scale formulation for dipolar, anisotropy and thermal energies. El-Hilo *et al* [5] had used it for determining the magnetoresistance dependence on the mean intergranular distance, or rather, the particle concentration, using a simple expression previously obtained by Gittleman *et al* [6]. Also Garcia-Otero *et al* [7] analysed the interplay between anisotropy and magnetic interactions and Chantrell *et al* [8] calculated the susceptibility and ZFC–FC (zero-field cooled–field cooled) magnetization curves for superparamagnetic particles. Some simple analytical models taking the dipolar interaction into account have been proposed. For example, Mørup and Tronc [9] have formulated a description for weakly interacting particles, and Dormann *et al* [10] have proposed the Dormann–Bessais–Fiorani (DBF) model, valid for weak and medium strength of the interactions. These two models lead to contradictory results, as analysed in [10]. The discrepancy arises when one tries to determine whether by increasing concentration, the interactions lead to an increase or a decrease of the energy barrier of the system. More recently, Allia *et al* [11] have proposed analytical models that take explicitly into account the correlation arising from the dipolar interactions in nearly superparamagnetic systems and Pike *et al* [12] investigated the role of magnetic interactions in low temperature saturation remanence of fine magnetic particles. From the experimental point of view, while dilute systems are well understood, results for denser ones, where the interactions between particles play an important role, are still not clear. The main reasons are the unavoidable particle size distribution and difficulties in controlling and replicating the geometrical arrangement and orientation of the easy axis for higher concentrations. In particular we can mention that for Fe particles embedded in an alumina matrix [13] and for γ-Fe_2O_3 [14] particles, an increase of the blocking temperature T_B with interaction strength is obtained. However, also for γ-Fe_2O_3 particles investigated by Mössbauer spectroscopy, T_B decreases with increasing concentration, as presented in [15]. Apart from the influence of concentration, or interparticle distance, in systems for which the dipolar interaction may not be neglected, it is important to consider the effect of dimensionality, especially when one deals with systems formed from sequential deposition of layers [2, 13, 16]. Luis *et al* [2] investigated the role of dipolar interactions in the magnetization of Co clusters grown in a quasi-ordered layered structure, and showed that the effective activation energy increases linearly with the number of nearest neighbour clusters. These results have been interpreted in terms of a transition from 2D to 3D collective dynamics in [16]. The characteristics of hysteresis curves have been related to various aspects of interacting systems. The remanence, or the value of the magnetization at zero field, for example, is a measure of the stability of the saturated state. A low value of remanence, in systems with an important dipolar interaction, may appear for different reasons, such that there is not a straightforward connection between its magnitude and amount of ordering in the system. Bahiana *et al* [17] investigated the effect of interactions on a 2D lattice of grains and found a peak in the remanence as a function of temperature, which can be interpreted as evidence of the existence of two low magnetization states: one due to an in-plane alignment perpendicular to the field direction at low temperatures, and a high temperature disordered state. In this paper we have extended these results to 3D systems, considering the effect of increasing the number of stacked layers, and varying lattice geometry. The hysteresis curves are obtained by Monte Carlo simulation, using a Metropolis algorithm with restricted dynamics.

2. Simulation conditions

A ferromagnetic particle becomes a monodomain when its linear size is below a critical value D_c determined by the minimization of the total energy, including magnetostatic, exchange and anisotropy contributions [18]. Such monodomain ferromagnetic particles can be viewed as

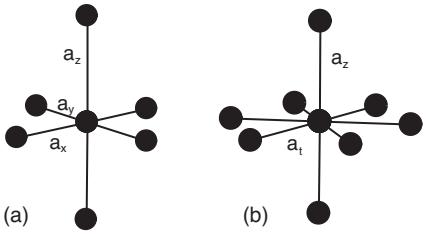

Figure 1. Different geometries considered in the simulations. (a) xy planes with rectangular arrangement. In this case the grains are separated by a_x and a_y along the x and y axis respectively, separated by a_z in the z direction. (b) xy planes with a triangular symmetry and nearest neighbour distance a_t are separated by a distance a_z.

large magnetic units, each one having a magnetic moment of thousands of Bohr magnetons. For neighbouring particles with surfaces separated by more than 1 nm, direct and indirect exchange (like RKKY) can be neglected [19]; thus, the magnetic properties of such an assembly of nanoparticles are determined by the dipolar interaction energy among the particles along with thermal and magnetic anisotropy energies. The magnetic irreversibility of an isolated nanomagnet is conventionally associated with the energy required for the particle moment reorientation, overcoming a barrier due to shape, magnetoelasticity and/or crystalline anisotropy [18]. In the presence of relevant dipolar interactions, this simplified picture no longer holds, as each particle is subject to a complicated energy landscape.

To investigate the magnetic behaviour of interacting grains we have examined two simple systems consisting of M layers of $N \times N$ magnetic 3D monodomain particles. Each layer is parallel to the xy plane and has free boundary conditions. Two nearest neighbour arrangements were considered, with square and triangular symmetries, as shown in figure 1. The distance between particles on each layer is defined by the lattice parameters a_x and a_y, for square lattices, and a_t, for triangular lattices; the layer separation is given by a_z. From now on the terms in-plane and out-of-plane refer to the xy plane. The particles have uniform magnetization, $m = 869\,\mu_B$, and anisotropy constant, $K = 1.32 \times 10^6$ erg cm^{-3}, corresponding to slightly elongated cobalt grains with about 511 atoms and linear dimensions of the order of 20 Å [20]. Each particle is described by the position of its centre of mass and the direction of the randomly chosen 3D easy magnetization axis, $\widehat{e_i}$, and they are coupled by means of dipolar interactions. Since we seek to understand the role of lattice geometry, we prefer to control the distance between grains through the parameters a_x, a_y, a_z and a_t, instead of concentration. The values of the lattice parameters are such that the particle surfaces are more than one cobalt lattice parameter apart [17]. The external field is always in the x direction.

In the presence of an external magnetic field $\vec{H}$, the total energy of the system is written as

$$\mathcal{E} = \sum_i \left[-\vec{m}_i \cdot \vec{H} - \kappa \left(\frac{\vec{m}_i \cdot \widehat{e_i}}{m_i} \right)^2 + \frac{1}{2} \sum_{j \neq i} E_{ij} \right], \tag{1}$$

where $\kappa = KV$, V being the volume of each grain. E_{ij} is the classical dipolar energy between grains i and j given by

$$E_{ij} = \frac{\vec{m}_i \cdot \vec{m}_j - 3(\vec{m}_i \cdot \widehat{n}_{ij})(\vec{m}_j \cdot \widehat{n}_{ij})}{r_{ij}^3}.$$

Here r_{ij} is the distance between the centres of particles i and j, and $\widehat{n}_{ij}$ is the unit vector along the direction that connects them. Using this expression for the energy we have simulated hysteresis

curves for different values of temperature and lattice symmetries. Hysteresis curves correspond to sequences of non-equilibrium states of the system, and therefore depend on the field variation rate. In terms of a Monte Carlo simulation this means that we have to avoid equilibrium by means of a sufficiently fast variation of the field, and the usual mechanism of time averaging instead of ensemble averaging is not valid. Monte Carlo simulations were carried out using the Metropolis algorithm with local dynamics in which the new orientation of the magnetic moment was chosen within a solid angle around the previous moment direction, with aperture $d\theta = d\phi = 0.1$. This method was studied by Pereira Nunes *et al* [21], applied to the simulation of ZFC–FC magnetization curves, which are also a collection of non-equilibrium states, and it is a reasonable approach for a qualitative analysis. Although the Monte Carlo method has been devised for the study of systems in thermodynamic equilibrium, it has long been used to simulate the dynamics of unstable and metastable phases [22–24], providing a valuable tool whenever the traditional partial differential equation description leads to numerically unstable discretization schemes. The method has also been used to study ageing in spin-glass models [25], an essentially off-equilibrium problem. In the specific case of hysteresis curves, Monte Carlo simulations have been used in a variety of magnetic systems with good qualitative agreement with experimental data [7, 8, 26], so we believe that the method is adequate for studying the dynamics of complex systems.

For simulating hysteresis, we started at a fixed temperature from a configuration in which the magnetic moment directions were randomly chosen. An external magnetic field $H = 0.25$ kOe in the x direction was turned on; one of the grains was randomly chosen, and had its magnetic moment rotated by an angle restricted to a cone, as explained above. The change in energy ($\Delta\mathcal{E}$) was calculated and the rotation accepted with probability $p = \min[1, \exp(-\Delta\mathcal{E}/k_B T)]$. This procedure was repeated $N^2 \times M$ times, comprising one Monte Carlo step. The number of Monte Carlo steps in non-equilibrium simulations is a rather arbitrary choice, as explained by Pereira Nunes *et al* [21]. Actually, the variation rate of the external field is the important quantity. In this case, we first fixed the value of the variation step for the external field, dH, with the objective of having enough points in the region of interest, near $H = 0$, but still being able to bring the system to saturation. We found that 200 Monte Carlo steps was a good choice for $dH = 0.25$ kOe, since the system shows hysteresis for some values of temperature, and the area of the hysteresis cycle shows sensitivity to temperature variation. The virgin curve was then obtained by increasing the field until the magnetization reached at least 99.995% of its saturation value, m_s. Starting from this last value, the field was decreased to negative values at the same rate (200 Monte Carlo steps per $dH = 0.25$ kOe). The whole procedure was repeated 5–20 times, depending on the system size, for different random choices of easy magnetization axis directions, the averaged hysteresis curve was calculated and the remanence, m_r, was determined as the x component of the magnetization for $H = 0$. The graphs below show the behaviour of the reduced remanence, defined as m_r/m_s, with typical error bar values in the range 0.01–0.005, the larger ones being for points in the peak region.

3. Results

3.1. Square lattices

We start our calculations considering lattices with M layers of 8^2 particles and $a_x = a_y = a_z = 3.098$ nm, as defined in figure 1. These values of the lattice parameter would correspond to a 20% concentration in a simple cubic system. Figure 2 shows the thermal variation of the reduced remanence as a function of M. One can see from this figure that the remanence of interacting systems exhibits a maximum at low temperatures, which is not present in the

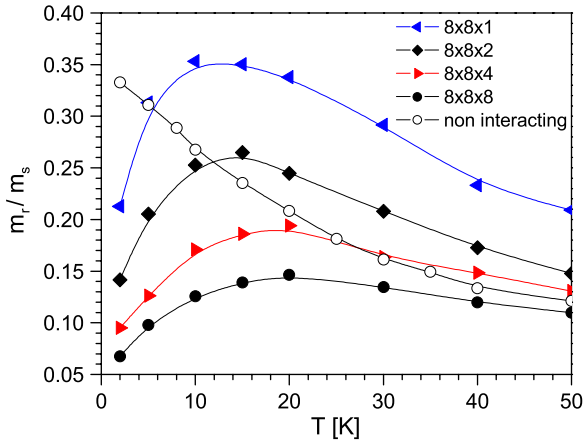

Figure 2. Remanence as a function of temperature for interacting systems with M layers (full symbols), and for a non-interacting system (open symbols). Each layer is composed by 8^2 particles, placed on a square lattice. The interlayer and intralayer nearest neighbour distance is 3.098 nm. The curves are guides to the eye. Error bars are of the order of the single size of the symbols.

non-interacting particle curve. As shown in [17] for the $M = 1$ system, the remanence peak separates distinct states of low magnetization. At low temperatures the absolute value of the dipolar energy is much larger than the thermal energy and an ordered state, due to the dipolar interaction, appears. In this case, magnetization patterns at $H = 0$ show a very small contribution of z magnetization and antiparallel alignment between lines normal to the previous field direction, the y axis in this case. The energy required to satisfy the dipolar coupling between lines parallel to the x axis would be too large and an antiparallel alignment along the y direction is favoured. At the maximum, the two energies are closer, and thermal fluctuations provide enough energy to destroy the low temperature y dipolar order. At higher temperatures, the thermal energy dominates, leading to a significant increase of the z component of the magnetization, also resulting in a low remanence value. This behaviour is numerically well described by the average values of $|m_x|$, $|m_y|$ and $|m_z|$, defined as μ_x, μ_y and μ_z, respectively. At 3 K, averaging over 10 samples, we have the values $\mu_x = 503\ \mu_B$, $\mu_y = 540\ \mu_B$ and $\mu_z = 255\ \mu_B$, compatible with a mainly in-plane magnetization, with an important y contribution due to the antiparallel alignment. In the high temperature region, for example at 70 K, the average values are $\mu_x = 472\ \mu_B$, $\mu_y = 479\ \mu_B$ and $\mu_z = 351\ \mu_B$, showing an increase of the out-of-plane component favoured by thermal energy.

The remanence curves for $M > 1$ are similar to the $M = 1$ curve, with two low magnetization regions, one at low temperatures dominated by the dipolar coupling, in which antiparallel alignment in the directions normal to the applied field is present, and a high temperature disordered state with magnetic moments randomly aligned. It is clear from figure 2 that, as M increases, for a given temperature, the remanence decreases, and the peak slightly shifts to higher temperatures, the interaction with out-of-plane neighbours being the main cause of this behaviour. Regarding the ordered low temperature state, the presence of layers below and above a given plane offers an alternative direction for antiparallel alignment leading to a decrease of the remanence. Due to the range of the interaction, the dipolar energy per particle increases with system size up to a saturation value; therefore, it is natural to expect that on increasing the number of layers, the low temperature ordered state becomes more stable, reflecting in a shift of the peak position to higher temperatures.

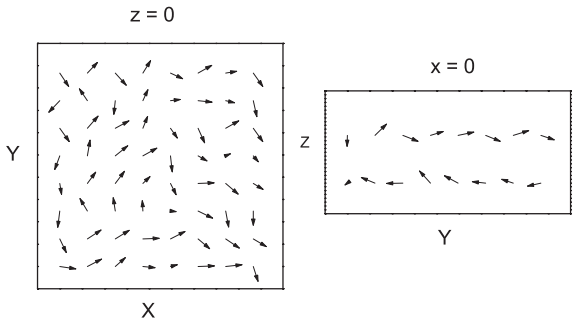

Figure 3. Snapshots of the magnetic moments at surface layers of a $8 \times 8 \times 2$ system.

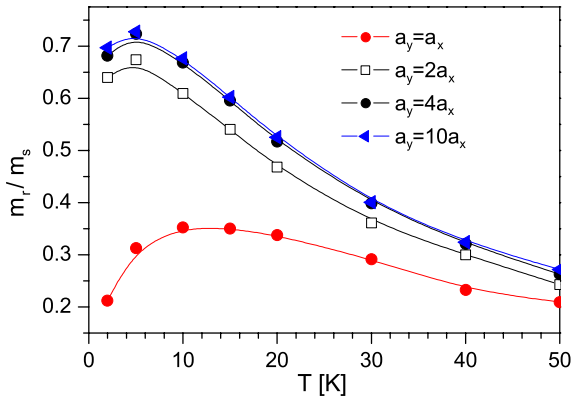

Figure 4. Remanence as a function of temperature for different values of a_y in $8 \times 8 \times 1$ lattices. Error bars are of the order of the single size of the symbols.

In order to investigate the low temperature dependence on M, we have examined the magnetization patterns for systems with $M = 2$ and 8. Figure 3 illustrates the behaviour of the individual magnetic moments at $T = 3$ K, for surface layers of a $M = 2$ system. In this case, there are two alternative directions for the antiparallel coupling, y and z, but, as explained above, there is a predominance of the y component since the number of neighbours in this direction is larger. This can be confirmed by looking at the average values of the magnetization components in the x, y and z directions. At $T = 3$ K, considering 10 samples, the average values for the whole lattice are $\langle m_x \rangle = 403\ \mu_B$ and $\mu_x = 458\ \mu_B$, $\langle m_y \rangle = -9.47\ \mu_B$ and $\mu_y = 504\ \mu_B$, $\langle m_z \rangle = 1.85\ \mu_B$ and $\mu_z = 339\ \mu_B$. Clearly an antiparallel alignment on the yz plane occurs, which is stronger along the y direction.

For the $M = 8$ system the y and z are equivalent directions for the antiparallel coupling. The amount of z alignment increases at the expense of the x contribution as can be seen from the average values for the lattice, $\langle m_x \rangle = 246\ \mu_B$ and $\mu_x = 388\ \mu_B$, $\langle m_y \rangle = 14.3\ \mu_B$ and $\mu_y = 517\ \mu_B$, $\langle m_z \rangle = -5.78\ \mu_B$ and $\mu_z = 398\ \mu_B$.

It is interesting to investigate the behaviour of the remanence as the values of a_y and a_z are varied. Figure 4 shows the remanence curves for $M = 1$ systems with $a_y = a_x, 2a_x, 4a_x$ and $10a_x$. Since the antiparallel order in the y direction is the main reason for the low value of the remanence at low temperatures, as the y distance is increased, the dipolar coupling decreases and the magnetic moments can follow the field more easily, giving rise to an increase of the remanence.

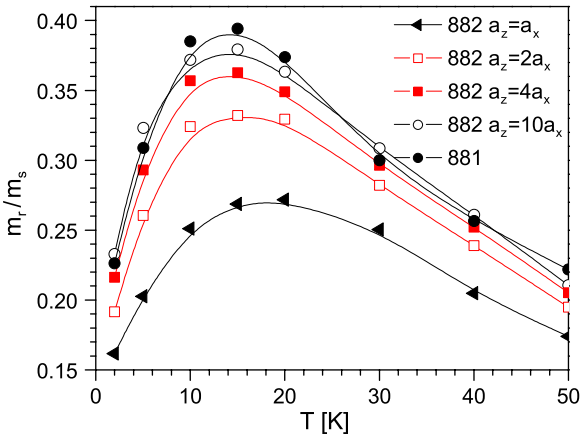

Figure 5. Remanence as a function of temperature for different values of a_z in $8 \times 8 \times 2$ lattices. The curve for $8 \times 8 \times 1$ is also shown as a reference. Error bars are of the order of the single size of the symbols.

The effect of varying a_z is evident when we compare systems with $M = 2$ and $a_z = a_x$, $2a_x$, $4a_x$ and $10a_x$ with an $M = 1$ system. Figure 5 illustrates the remanence curves for such systems, showing that, as the layers become more separated, the interlayer coupling decreases and the system approaches the $M = 1$ behaviour.

3.2. Triangular lattice

For layers with triangular symmetry, the number of in-plane neighbours is higher, so that any effects related to confinement of the magnetic moments in the xy plane are enhanced. Figure 6 shows the remanence curves for triangular lattices with $a_t = 3.098$ nm, obtained under the same conditions as the curves in figure 2. The remanence values are considerably larger than the ones in figure 2, which is compatible with the picture of a smaller z component. We also analysed the magnetization pattern for typical configurations of these triangular systems. Figure 7 shows snapshots of the x and y components of the individual magnetic moments at $T = 3$ K for $M = 1$. From this figure we can see that, at low temperature, the small value of the remanence is mainly caused by in-plane alignment along the directions connecting the nearest neighbour clusters of the system. This effect, due to the predominance of the dipolar interaction, is stronger than in the square lattice since the number of in-plane nearest neighbours, six in this lattice, is higher, as compared to four in the square lattice. This behaviour is numerically well described by the average values of magnetization components at $T = 3$ K: $\langle m_x \rangle = 571\ \mu_B$, $\langle m_y \rangle = -88.4\ \mu_B$, $\langle m_z \rangle = -25.9\ \mu_B$, $\mu_x = 586\ \mu_B$, $\mu_y = 433\ \mu_B$ and $\mu_z = 285\ \mu_B$, compatible with a mainly in-plane magnetization. In the high temperature region, the system is disordered and the z component of the magnetization increases, leading to a low remanence region.

4. Discussion and conclusions

The above results confirm the existence of two low remanence yet distinct behaviours, one at low temperature, where the dipolar energy dominates, and another, at higher temperatures, where the thermal energy is responsible for the magnetic disorder. The position and height

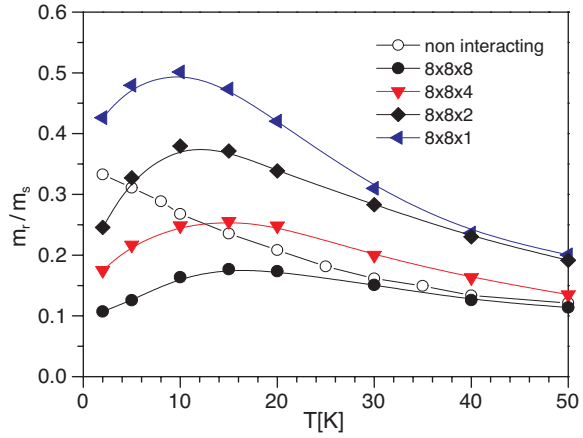

Figure 6. Remanence as a function of temperature for interacting systems with M layers (full symbols), and for a non-interacting system (open symbols). Each layer is composed by 8^2 particles placed on a triangular lattice. Error bars are of the order of the single size of the symbols.

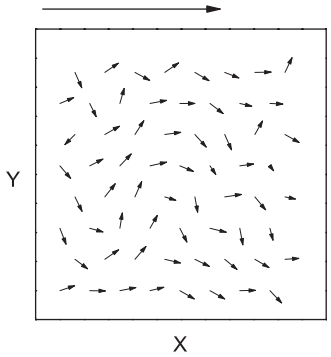

Figure 7. Snapshot of the in-plane magnetization at $H = 0$ and $T = 3$ K, for a system of $8 \times 8 \times 1$ particles and triangular symmetry. The previous direction of the field is indicated by the arrow.

of the peak separating those regimes is related to the strength of the dipolar interactions, and depends on the lattice geometry. As is known, dipolar interactions favour closed circuit alignment, which may result in antiparallel alignments between lines of magnetic moments parallel to a given direction, provided, for example, by a weak external field. At low temperatures a small external magnetic field provides a preferred direction for the parallel coupling, leading to two possibilities of antiparallel alignment in 3D systems. On the other hand, for a 2D system, there is only one possible direction for the antiparallel alignment. Figure 2 shows that for systems formed by sequential deposition of layers it is possible to observe a transition from the 2D to the 3D behaviour as the number of layers increases. As an alternative direction for antiparallel alignment appears, the height of the remanence peak decreases reflecting the decrease of the magnetization along the field direction. Also, for a given number of layers, the variation of the distance between magnetic particles and coordination number can drastically change the effective dimensionality, as the dipolar interaction is enhanced along certain directions. Figures 4 and 5 show this dimensionality transition caused by variation of the lattice parameter, and figure 6 that caused by the change in coordination

number. These aspects are responsible for difficulties in the analysis of the hysteresis cycle, especially if one associates the area of the cycle with the stability of the magnetization moment. Larger values of remanence are basically a consequence of a decrease in the number of degrees of freedom, due to the confinement of the magnetic moment to a plane or to a line.

Acknowledgments

In Chile the authors received financial support from FONDECYT under grants no. 1010127 and 7010127 and Millennium Science Nucleus 'Condensed Matter Physics' P02-054F. Paola R Arias and M Bahiana acknowledge MECESUP support under grant USA0108. In Brazil, the authors acknowledge the support from FAPERJ, CNPq and Instituto de Nanociências/MCT.

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