Finite-size effects and compensation temperature of a ferrimagnetic small particle

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(Received 19 August 2004; revised manuscript received 5 November 2004; published 30 March 2005)

In this work we study the magnetic properties of a ferrimagnetic small particle on a hexagonal substrate. The particle is described by a mixed-spin Ising model in which the $\sigma=1/2$ and $S=1$ spins are distributed in concentric and alternate hexagonal rings. We consider particles with different number of rings and show that particles with more than 11 shells can be considered as infinite systems. For a particle in which the finite-size effect is relevant, we investigated the role of the different parameters of the Hamiltonian in the appearance of a compensation temperature. As the model incorporates σ-S, σ-σ and S-S nearest-neighbor interactions, we observe the existence of a compensation point without the necessity of any next-nearest-neighbor interaction. The appearance of a compensation point depends only on the value of the σ and S intrasublattice couplings. The σ intrasublattice interaction should be ferromagnetic and above a threshold value. On the other hand, the S intrasublattice interaction should be mostly antiferromagnetic and restricted to a narrow range of values.

DOI: 10.1103/PhysRevB.71.094427 PACS number(s): 05.50.+q, 75.10.Hk, 75.40.Mg, 75.50.Gg

I. INTRODUCTION

The appearance of a compensation point near room temperature in some ferrimagnetic materials, has a crucial importance in the area of the thermomagnetic recording devices.[1–3] The compensation temperature (T_{comp}) appears due to the different temperature dependences of the sublattice magnetizations that form the ferrimagnetic material. At T_{comp}, the sublattice magnetizations cancel each other and the total magnetization vanishes.[4] In addition at this point it has been found that some physical properties present a peculiar behavior. For instance, the coercive field (H_c) is strongly temperature dependent only in the vicinity of T_{comp}:[1,5,6] it is maximum at T_{comp}, falling to a minimum below the compensation temperature, before rising again at low temperatures. This peculiar temperature dependence of H_c, together with a local heating by a focused laser beam, can be applied to attain a direct overwrite capability in magneto-optical recording media.

Mixed-spin Ising systems were introduced as the simplest models that exhibit a ferrimagnetic behavior. For an infinite system, formed by two interpenetrating sublattices of $\sigma=\pm1/2$ and $S=\pm1$, 0 spins, many studies have been performed to describe the appearance of a compensation point. The existence of T_{comp} in this system was already investigated by mean-field,[7] renormalization-group calculations,[8] Monte Carlo simulations,[9,10] and numerical transfer-matrix techniques.[10] The mean-field calculations show the appearance of a compensation point considering a model with only nearest-neighbor couplings and a crystal field interaction. On the other hand, in Refs. 8–10 was shown that a compensation temperature appears only when the model incorporates ferromagnetic next-nearest-neighbor interactions between σ spins. The consideration of next-nearest-neighbor interactions in the case of interpenetrating sublattices is the only way to take into account the interactions between σ spins in a square lattice, where the coordination number is $z=4$. The σ-σ exchange coupling enhances the critical temperature of the system and therefore can give rise a compensation point. In a recent work,[11,12] we considered a ferrimagnetic model on a hexagonal lattice ($z=6$) formed by alternate layers of σ and S spins. For this spin arrangement, the intrasublattice interactions are always between nearest-neighboring spins and the compensation point appears by taking into account suitable range of values for the σ-σ and S-S exchange couplings.

In this work we consider a two-dimensional ferrimagnetic small particle described by a spin arrangement similar to that studied in Ref. 11. This particle is formed by a central spin, surrounded by alternate rings of σ and S spins. If we imagine this model extended to three dimensions it could describe some properties of real ferrimagnetic materials. For instance, the work of Chern *et al.*[13] reports some measurements of the compensation point and phase diagram of Fe_3O_4/Mn_3O_4 superlattices, which is a system grown by a deposition of alternate layers of Fe_3O_4 and Mn_3O_4 coupled antiferromagnetically. If these layers were grown cilindrically our model could be seen as a perpendicular cut to the axis of the cilinder.

As already have been observed, the effects of the surface and size of the particle are manifested through a wide variety of anomalous magnetic properties with respect to those of bulk material. These small systems are prototype of magnetic nanoparticles and they have been intensively studied in the recent years.[14–17] In the specific case of a ferrimagnetic small particle, some studies have been performed through the micromagnetic formalism[18] and by Monte Carlo simulations.[19]

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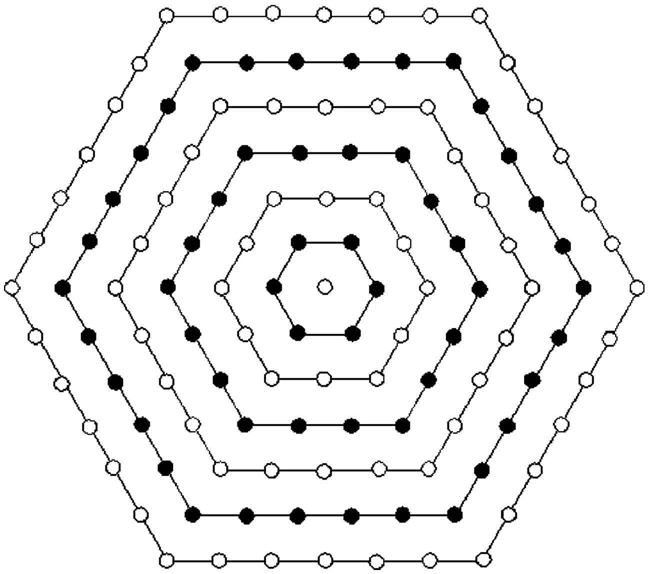

FIG. 1. Schematic representation of a hexagonal ferrimagnetic particle with six shells. The particle is formed by alternate shells of σ (open circles) and S (solid circles) spins.

Up to now, no model was studied in order to investigate the role of the finite-size effect on the compensation point of a ferrimagnetic small particle. In this work we consider a particle with perfect surface, and we investigate the effects of the reduced coordination number of the surface spins on the determination of the compensation temperature. Particularly, we find the threshold parameters of the Hamiltonian to give a compensation point in a ferrimagnetic small particle.

This paper is organized as follows. In Sec. II the model and the dynamic equations of motion for the magnetizations are determined in the mean-field approximation. In Sec. III, we describe the Monte Carlo simulations. In Sec. IV we present our results, and finally, we draw our conclusions in Sec. V.

II. THE MODEL

The ferrimagnetic small particle is described by a mixed-spin Ising model on a hexagonal substrate. As we can see in Fig. 1, the different layers of spins are disposed in alternate rings, that is, if the central spin is of the σ type, the first ring is of the S type, the second is of the σ type, etc. The spins are described by Ising variables, which can take the values $\sigma = \pm 1/2$ and $S = \pm 1$, 0. With this particular arrangement of spins, the model presents intersublattice and intrasublattice nearest-neighbor interactions of the type σ-S, σ-σ, and S-S. We also take into account a crystal field contribution associated with the S spins. The Hamiltonian of the model is

$$\mathcal{H} = -J_1 \sum_{\langle ij \rangle} S_i \sigma_j - J_2 \sum_{\langle ij \rangle} \sigma_i \sigma_j - J_3 \sum_{\langle ij \rangle} S_i S_j - D \sum_i S_i^2, \quad (1)$$

where J_1, J_2, and J_3 are the exchange couplings between nearest-neighbor pairs of spins σ-S, σ-σ, and S-S, respectively. D is the crystal field parameter. The exchange parameter J_1 will be taken negative in all the subsequent analyses, that is, the intersublattice coupling is antiferromagnetic. In order to study this model in the mean-field approximation, we consider the dynamic equations for the average spin magnetizations

$$m_{\sigma_i}(t) = \sum_{\langle \sigma, S \rangle} \sigma_i P(\sigma, S; t) \quad (2)$$

and

$$m_{S_i}(t) = \sum_{\langle \sigma, S \rangle} S_i P(\sigma, S; t), \quad (3)$$

where the sums are over all the possible spin configurations, and $P(\sigma, S; t)$ is the probability to find the system in a given state (σ, S) at time t. In this case, as we are considering a finite system, we must compute the average magnetization for each spin (this is the reason for the index i in $m_{\sigma i}$ and in m_{Si}).

The calculation of these averages is straightforward in the dynamic equations of motion for the site approximation, where we have

$$\frac{d}{dt} m_{\sigma_i} = -m_{\sigma_i} + \frac{1}{2} \tanh\left[\frac{\beta}{2}\left(J_1 \sum_j m_{S_j} + J_2 \sum_j m_{\sigma_j}\right)\right] \quad (4)$$

and

$$\frac{d}{dt} m_{S_j} = -m_{S_j} + \frac{2 \sinh\left[\beta\left(J_1 \sum_k m_{\sigma_k} + J_3 \sum_k m_{S_k}\right)\right]}{2 \cosh[\beta\left(J_1 \sum_k m_k + J_3 \sum_k m_{S_k}\right)] + \exp(-\beta D)}, \quad (5)$$

where the sums in Eqs. (4) and (5) are over the nearest-neighbor of spins σ_i and S_j, respectively. For instance, if the central spin is of the type σ, the dynamic equation for σ_0 can be written as

$$\frac{d}{dt} m_{\sigma_0} = -m_{\sigma_0} + \frac{1}{2} \tanh\left[\frac{\beta}{2}\left(J_1 \sum_j m_{S_j}\right)\right], \quad (6)$$

and the sum is over the six spins S_j at the first shell of the particle. On the other hand, if the central spin is of the type S, we have for S_0

$$\frac{d}{dt} m_{S_0} = -m_{S_0} + \frac{2 \sinh\left[\beta\left(J_1 \sum_k m_{\sigma_k}\right)\right]}{2 \cosh\left[\beta\left(J_1 \sum_k m_{\sigma_k}\right)\right] + \exp(-\beta D)} \quad (7)$$

and now, the sum is over the six spins σ_k at the first shell of the particle. Then the equation of motion of a given spin localized between the first and last shell of the particle, takes in account four nearest-neigbor spins of the same type of the spin considered and two of the other type. At the surface, because of the lower coordination number, each spin has only three (spin at the corner of the particle) or four nearest-neighbors spins.

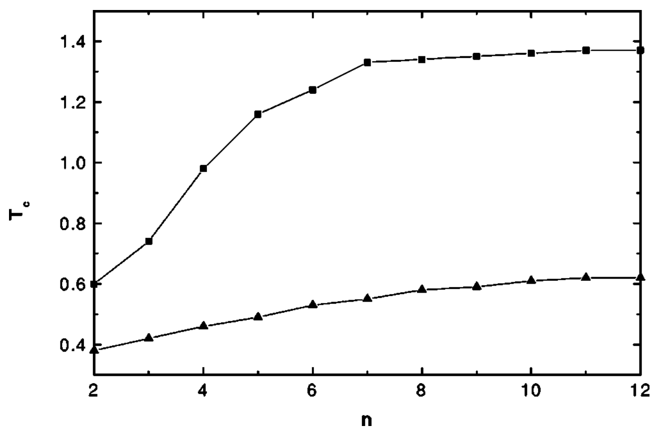

FIG. 2. Critical temperature obtained as a function of the number of shells of the particle (n). The mean-field results are shown by the line with squares and the Monte Carlo simulations are represented by the line with triangles. For particles with $n \geqslant 11$ shells, the critical temperature reaches the expected value for the corresponding infinite system. The parameters used are $J_2 = -J_1$, $J_3 = J_1$, and $D = -0.75|J_1|$. Temperature is measured in units of $|J_1|/k_B$.

We considered in our calculations, particles with a number of shells ranging from 2 to 12. In any case, due to the hexagonal symmetry of the particle, we had to solve a system of $P < N$ coupled equations, where N is the total number of spins of the particle. For example, we have $P=4$ and $N=19$ in a particle with 2 shells, and $P=49$ and $N=469$ in a particle with 12 shells. To solve these equations we considered only the equilibrium states, and the magnetizations were found as a function of the temperature, for different values of the Hamiltonian parameters.

Despite the mean-field calculations give only a crude evaluation of the critical and compensation temperatures of this model, as we will see next, they still exhibit the same essential features observed in the Monte Carlo simulations. As in this study we are not interested in the calculation of critical exponents, the mean-field approximation is the simplest analytical method that can be used to extract the general qualitative behavior of the system, and as expected, it establishes the upper bound values to the critical and compensation temperatures.

III. MONTE CARLO SIMULATIONS

The model described in the last section was simulated by using the heat-bath algorithm.[20] In each Monte Carlo step (MCS), we performed N trials to flip the spins. We performed around 6000 MCS, where the first 1000 were discarded for the thermalization process. In order to get reliable results, we also considered averages over 100 different samples in our calculations. Although not shown in the figures, the error bars are smaller than the symbol sizes.

Our algorithm calculates the magnetizations σ and S, defined as

$$m_\sigma = \frac{1}{N_\sigma}\left\langle \sum_{i=1}^{N_\sigma} \sigma_i \right\rangle, \tag{8}$$

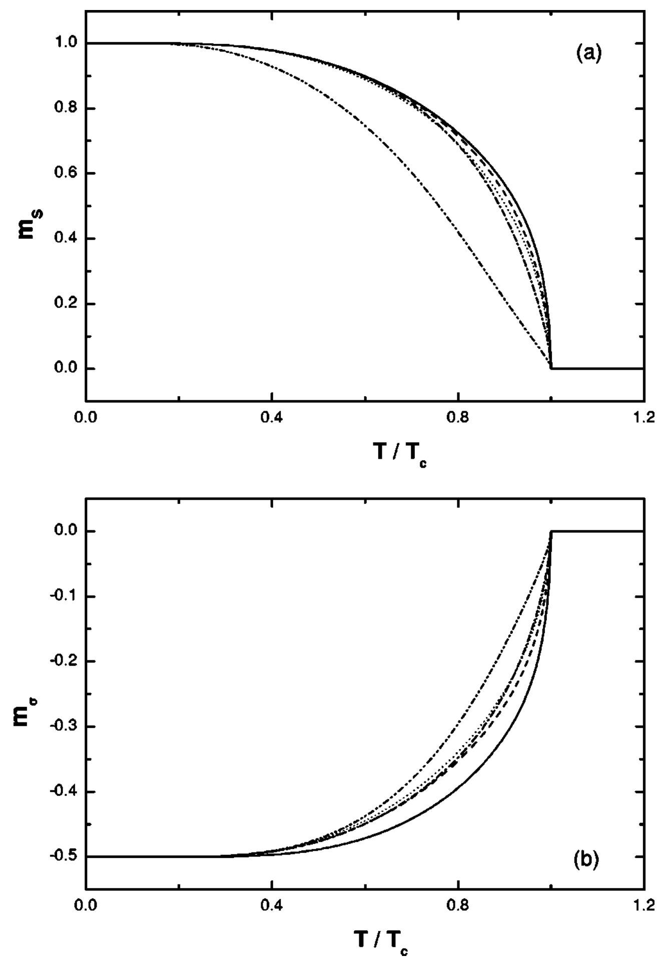

FIG. 3. Shell magnetizations of a particle with a central spin σ and seven shells, for $J_2 = -J_1$, $J_3 = -J_1$, and $D = 0$, obtained through mean-field calculations. (a) From top to bottom: bulk, shell 1, 3, 5, and 7. (b) From bottom to top: bulk, central spin, shell 2, 4, and 6.

$$m_S = \frac{1}{N_S}\left\langle \sum_{i=1}^{N_S} S_i \right\rangle, \tag{9}$$

and the total magnetization

$$m_{\mathrm{tot}} = \frac{N_\sigma}{N} m_\sigma + \frac{N_S}{N} m_S, \tag{10}$$

where N_σ and N_S are the number of spins σ and S, respectively. In addition we also evaluate the shell magnetizations of the particle.

At the compensation point the total magnetization must vanishe. Then, the compensation temperature can be determined by the crossing point between the absolute values of the magnetizations σ and S. Therefore, at the compensation point, we must have

$$|N_\sigma m_\sigma(T_{\mathrm{comp}})| = |N_S m_S(T_{\mathrm{comp}})|, \tag{11}$$

and

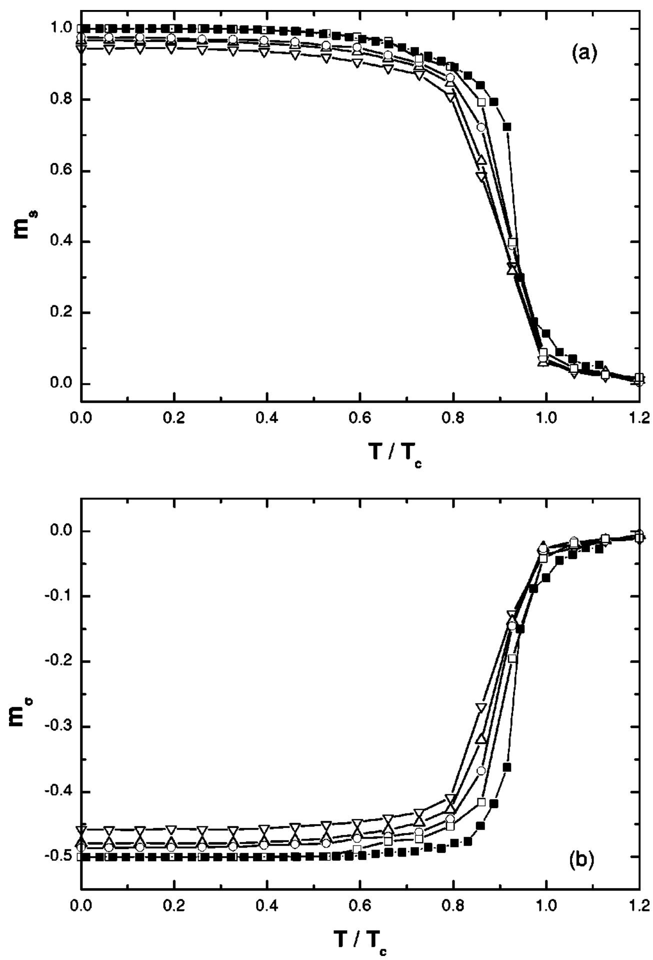

FIG. 4. The same legend as in Fig. 3 but for Monte Carlo simulations.

$$\mathrm{sgn}[m_\sigma(T_{\mathrm{comp}})] = -\,\mathrm{sgn}[m_S(T_{\mathrm{comp}})]. \qquad (12)$$

We also require that $T_{\mathrm{comp}} < T_c$, where T_c is the critical temperature.

These conditions show that at T_{comp}, the magnetizations σ and S cancel each other, whereas at T_c both are zero. As we have seen for an infinite system,[11] at the compensation point the model does not present any critical phenomenon; only at T_c the critical behavior is really observed. For instance, while at T_c the susceptibility and specific heat are singular functions of temperature, at the compensation point these functions are regular.

IV. RESULTS

First, let us consider the finite-size effects on the equilibrium magnetic properties of the ferrimagnetic particle. In Fig. 2 we show the critical temperature obtained for a particle with the number of shells ranging from 2 to 12, for the parameters $J_2=-J_1$, $J_3=J_1$ and $D=-0.75|J_1|$. The critical temperatures obtained by mean-field approximation and Monte Carlo simulations are drawn in the same plot as a matter of comparison. As expected, the critical temperature found in the mean-field calculations are higher than those

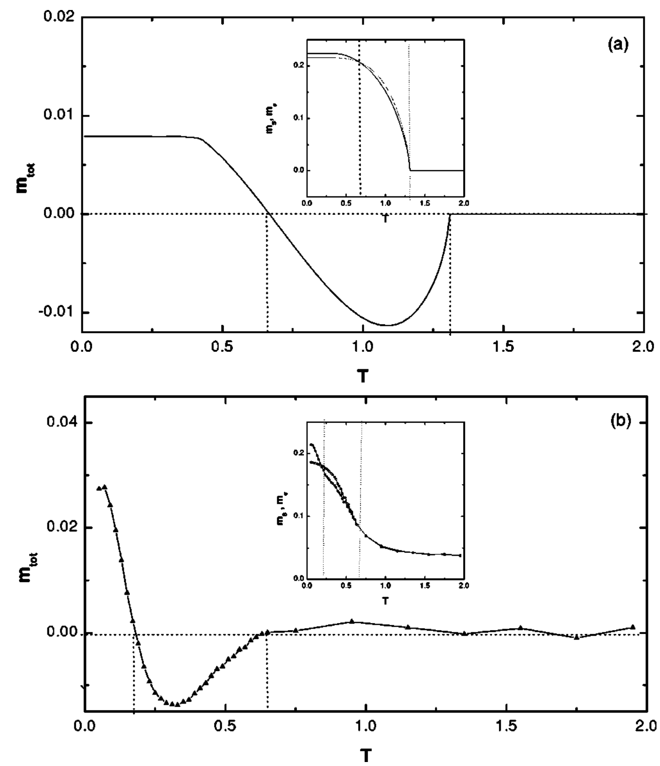

FIG. 5. Sublattice and total magnetizations as a function of temperature. (a) Mean-field calculcations for $J_2=-J_1$, $J_3=J_1$, and $D=-0.9|J_1|$ and (b) Monte Carlo simulations for $J_2=-J_1$, $J_3=0.2J_1$, and $D=-2.0|J_1|$. In the insets we show the crossing of the sublattice magnetizations. T_{comp} and T_c are shown in the figure. Temperature is measured in units of $|J_1|/k_B$.

obtained through Monte Carlo simulations. The results indicate an increase in the critical temperature of the particle with the number of shells considered. When the number of shells of the particle increases, the ratio between surface and volume of the particle decreases and, the finite-size effects become negligible. For the particle we are studying, in the case of $n \geqslant 11$ shells, the critical temperature reaches the expected value of the corresponding infinite system,[11,12] which are $T_c=1.37|J_1|/k_B$ and $T_c=0.62\pm0.02|J_1|/k_B$ by mean-field calculations and Monte Carlo simulations, respectively.

It is important to stress that the critical temperature obtained in the Monte Carlo simulations for the finite ferrimagnetic particle is not a true critical temperature, but a pseudocritical one. As is well known, a finite system cannot exhibit a true singularity at a nonzero temperature, but a pseudocritical temperature can be related to the sharp peak in the susceptibility and specific heat.[21]

Figures 3 and 4 show the shell magnetizations for a particle with a central σ spin and seven shells, for the particular set of parameters $J_2=-J_1$, $J_3=-J_1$, and $D=0$. As we can see in this plot, the magnetizations of the more internal shells have a behavior closer to the infinite system than the shells near to the surface. For this particle with seven shells, the ratio surface/volume is near 0.25, and as can be seen, it still exhibits some of the effects due to its finite size. The surface spins have a lower coordination number and therefore expe-

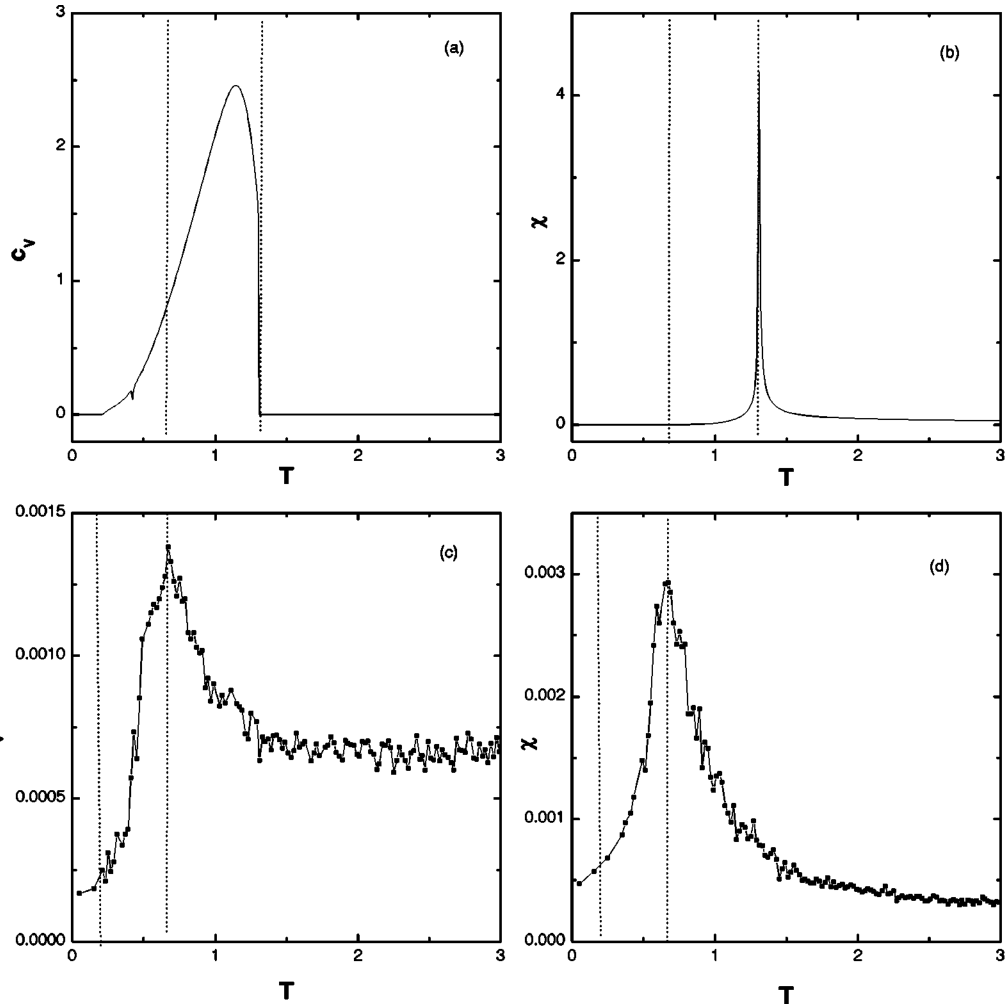

FIG. 6. Specific heat and susceptibility. (a) and (b) mean-field calculations, (c) and (d) Monte Carlo simulations. We used the same parameters as in Fig. 5. T_{comp} and T_c are shown in the figure. Temperature is measured in units of $|J_1|/k_B$, the specific heat in units of k_B, and the susceptibility in units of $|J_1|^{-1}$.

rience a reduced mean field. This is the only surface effect considered in this work, and it is responsible for the different values we found for the layer magnetizations. Another scenario would arise if we had chosen exchange couplings at the surface layer different from those of the core, or considered some kind of disorder at the topmost layers. To stress the surface effects in our model, in what follows, we will refer to a particle composed of seven shells.

To see the presence of a compensation point in this ferrimagnetic small particle, we show in Fig. 5 the total magnetization and the magnetizations σ and S as a function of the temperature, for selected values of the Hamiltonian parameters. In this figure, we have $J_2=-J_1$, $J_3=J_1$, and $D=-0.9|J_1|$, in the mean-field calculations, and $J_2=-J_1$, $J_3=0.2J_1$, and $D=-2.0|J_1|$, in the Monte Carlo simulations. As we will explain below we had to take different values of the parameters in the mean-field and Monte Carlo analyses in order to see clearly the occurence of a compensation point in each method. From Fig. 5(a), from the mean-field calculations, we find $T_{\mathrm{comp}}=0.67|J_1|/k_B$ and $T_c=1.31|J_1|/k_B$, for the compensation and critical temperatures, respectively. On the other hand, in Fig. 5(b), Monte Carlo simulations gives $T_{\mathrm{comp}}=(0.19\pm0.02)|J_1|/k_B$ and $T_c=(0.66\pm0.02)|J_1|/k_B$. If we would have applied the same set of parameters employed in Fig. 5(a), which gives rise to a compensation point in the mean-field approximation, to the case of Monte Carlo simulations, we would get a configuration where $m_\sigma > m_S$ for any temperature below the critical. Figure 6 shows the susceptibility and specific heat for the seven shell particle as a function of temperature, for the same set of parameters as in Fig. 5. This figure serves to illustrate the monotonic behavior of these thermodynamic functions at T_{comp} compared with those observed at T_c, where they present their maximum values.

Let us consider now for what values of the Hamiltonian couplings a compensation point is possible. Figure 7 shows the threshold of the ferromagnetic σ-σ interaction J_2 as a function of D, in the mean-field approximation and Monte Carlo simulations for $J_3=J_1$. For values of J_2 below the threshold, the magnetization S is always larger than the magnetization σ for any temperature for which $T<T_c$. This threshold depends on the crystal-field intensity and, as we can see, it is an increasing function of D. As D decreases, the sublattice magnetization m_S decays faster, and the crossing point between the two sublattice magnetizations moves to

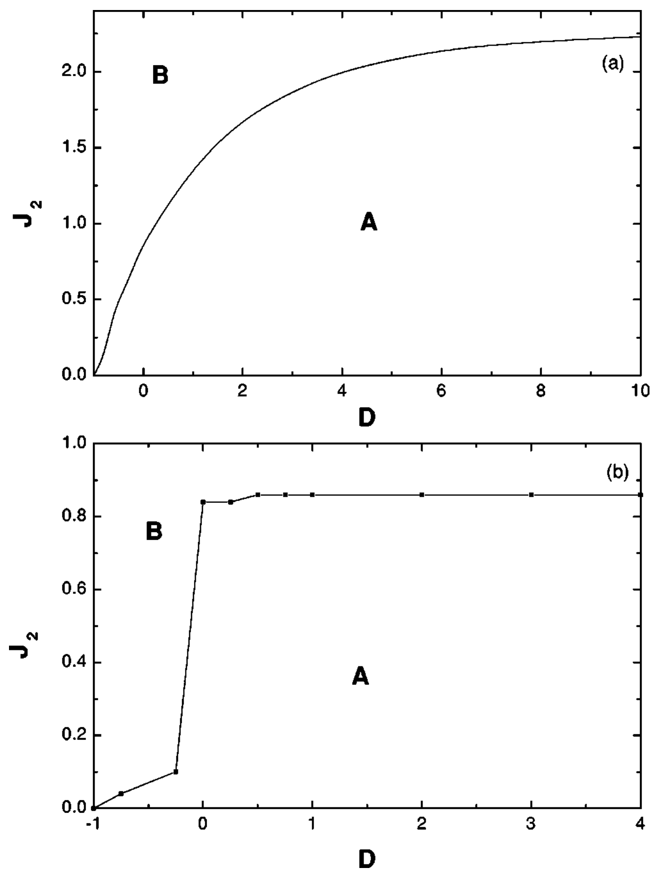

FIG. 7. Threshold values of J_2 as a function of D, to find a compensation point. (a) Mean-field calculations for $J_3 = J_1$ and (b) Monte Carlo simulations for $J_3 = 0.9J_1$. In the region A, $m_S > m_\sigma$ for any temperature $T < T_c$. B is the region where the compensation point can appear. J_2 and D are measured in units of $|J_1|$.

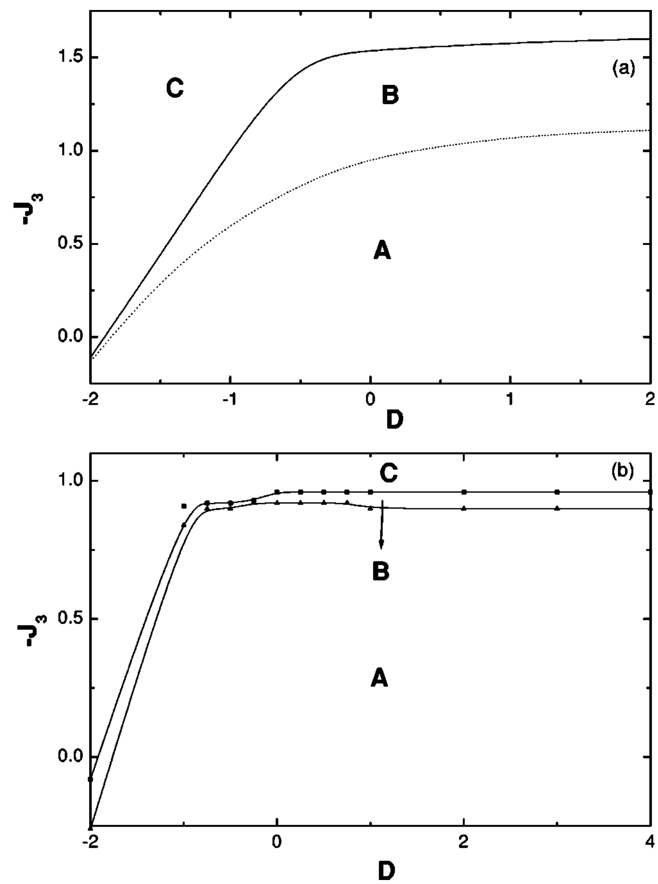

FIG. 8. Range of values of J_3 giving rise to compensation points, as a function of D, for $J_2 = -J_1$. (a) Mean-field calculations, (b) Monte Carlo simulations. For the regions A and C, we always have $m_S > m_\sigma$ and $m_\sigma > m_s$, respectively. B is the region where we can have compensation points. J_3 is measured in units of J_1 and D in units of $|J_1|$.

lower temperatures. For the set of parameters considered in these plots, the model does not exhibit any compensation point for $D < -1.0|J_1|$, in both, mean-field and Monte Carlo calculations. For this range of values of D, the magnetization S is lower than the magnetization σ for any value of the intrasublattice σ-σ interaction.

Finally, as can be seen in Fig. 8, the mean-field and Monte Carlo results, predict a small range of values for the antiferromagnetic intrasublattice S-S interaction, J_3, in order to appear a compensation point for $J_2 = -J_1$. Below the lower bound curve we have $m_S > m_\sigma$ for any temperature below the critical, because the $|J_3|$ is not large enough to decrease the sublattice magnetization S sufficiently. On the other hand, above the upper bound curve, the antiferromagnetic interaction between spins of the S sublattice is so large that m_S is always lower than m_σ for any temperature below the critical. Only in the intermediate region between these two boundaries, a compensation point can appear. It is worthwhile to stress that the Monte Carlo simulations give a much more narrow range of values of J_3 that the corresponding range found in the mean-fiel approximation.

In spite of Figs. 7 and 8 being related to a particle with seven shells, the same kind of picture is also observed for other particle sizes. The location of the threshold curves changes but the qualitative behavior remains the same for the appearance of the interesting region B, where the compensation points can occur. For example, with the parameters used in Figs. 5(a) we cannot observe a compensation point for particles with $n \leqslant 6$ shells in the mean-field calculations. On the other hand, with the parameters used in Fig. 5(b) the compensation point is not present for particles with $n \leqslant 5$ shells throught Monte Carlo simulations. In both cases, these sets of parameters would represent points in the corresponding regions C.

V. CONCLUSIONS

In this work we have considered mean-field calculations and Monte Carlo simulations to study the finite-size effects of a ferrimagnetic small particle on its compensation point. The particle is described by a mixed-spin Ising system, where the $\sigma = 1/2$ and $S = 1$ spins occupy alternate rings of a hexagonal lattice. The Hamiltonian of the model includes intersublattice (σ-S), intrasublattice (σ-σ, S-S), and crystal-field (D) interactions for the particle. In order to have a ferrimagnetic behavior, the intersublattice interaction must be antiferromagnetic.

We focused our attention on the role played by the different couplings in the Hamiltonian to predict a compensation point for a small particle. Our results show that, as it happens for the infinite system, the compensation point appears only when the intrasublattice interaction between σ spins is ferromagnetic. There is a minimum value for this coupling, which depends on the other Hamiltonian parameters, for the occurence of a compensation point. On the other hand, the sublattice magnetization S must decrease enough in order to cross the curve of the sublattice magnetization σ at a finite temperature below the critical. This can be achieved by two different ways: decreasing the crystal-field coupling D or increasing the antiferromagnetic coupling between spins of the sublattice S. We have shown that there is a very narrow range of values of this intrasublattice coupling in order to find a compensation point. Finally, we have seen as in the case of the hexagonal infinite system, we need only nearest-neighbor exchange interactions to find a compensation point in this ferrimagnetic small particle.

Our results indicate that a particle with more than eleven shells can be assumed to be in the thermodynamic limit. As we have seen in Fig. 2 it is easy to drawn the curve of the critical temperature as a function of the particle size for a fixed set of Hamiltonian parameters. The same task is not so easy for the compensation temperature, as we have discussed in the final paragraph of the last section. As we move along the size axis with the same set of parameters we can get out of the region B of the possible compensation points. The region of the parameters where the compensation point can appear changes with the size of the particle. For the smaller particles the ferromagnetic interaction between σ spins should be larger and the antiferromagnetic coupling between S spins should not be very strong. These are the appropriated conditions to have a high critical temperature and at the same time a compensation point.

ACKNOWLEDGMENTS

The authors acknowledge the financial support given by the Brazilian agency CNPq, Collaboration Inter-American in Materials (Grant No. 690052/03-0) and by FUNCITEC(SC).

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