Phase diagram of uniaxial antiferromagnetic small particles: Monte Carlo calculations

V. S. Leite, B. C. S. Grandi, and W. Figueiredo
Departamento de Física, Universidade Federal de Santa Catarina 88040-900, Florianópolis, SC, Brazil
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We determined the phase diagram of a three-dimensional uniaxial antiferromagnetic particle in the presence of an external magnetic field parallel to its easy axis through Monte Carlo calculations. The structure of the particle is a simple cubic lattice, where its magnetic moments are distributed in spherical shells centered around a given site. The magnetic moments are continuous vectors that interact via a Heisenberg classical Hamiltonian. We consider particles with radii ranging from 3 to 12 spacing lattice units in order to determine their phase diagram in the plane field versus temperature and to get the explicit dependence of the transition fields on the diameter of the particle. The asymptotic low-temperature behavior of the transition fields is determined and, for particles with radii larger than three lattice spacings, we get good agreement with the predictions from the spin-wave theory.

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I. INTRODUCTION

The investigation of the phase diagram of uniaxial antiferromagnets has received a lot of attention since the prediction of Néel that phase transitions could be induced in these systems by strong magnetic fields [1]. In the microscopic description of Néel, below the transition field (H_{SF}), parallel to the easy axis of the particle, the magnetic moments are ordered into two sublattices with opposite magnetizations along the easy axis. However, increasing the magnetic field, we attain the transition field H_{SF}, where the sublattice magnetizations rotate, becoming almost perpendicular to the magnetic field, at the so-called spin-flop transition. Increasing even more the magnitude of the magnetic field, the sublattice magnetizations continue to rotate into the field direction until they reach a critical field, H_P, where the system enters into the paramagnetic phase. In this way, the very well-known phase diagram in the plane temperature versus magnetic field of a uniaxial antiferromagnet exhibits three distintic phases: an antiferromagnetic, a spin-flop, and a paramagnetic phase. Interestingly, due to the necessity of using very large magnetic fields, it took over 20 years before experiments confirmed the occurrence of spin-flop transitions.[2,3] Since then, these phase transitions have been intensively investigated both experimentally[4–6] and theoretically.[7–15]

The uniaxial antiferromagnetic structures in reduced dimensionalities have also received some attention since the prediction of a surface spin-flop state in a semi-infinite system.[16,17] After that, the magnetic measurements performed by Wang *et al.*[18] confirmed the presence of a spin-flop transition on magnetic superlattices. This type of investigation continued and other theoretical[19–21] and experimental[22–24] studies have appeared, focusing on the surface effects and size dependence of the phase diagram of antiferromagnetic systems. In particular, concerning the studies of antiferromagnetic small particles, we call attention to the recent work of Zysler *et al.*,[25] where they investigated the phase diagram obtained for hematite particles.

Finite-size and surface effects play a crucial role in the magnetic behavior of antiferromagnetic small particles. For instance, it is well known that, as the particle size decreases, a net magnetization moment appears due to the nonexact compensation of the magnetic sublattices—i.e., an imbalance in the number of *up* and *down* spins.[26] Besides, for the antiferromagnetic small particles, a superparamagnetic susceptibility, due to uncompensated spins, can dominate over the antiferromagnetic contribution itself. On the other hand, because of the structural disorder and broken bonds, the surface spin directions deviate from the normal antiferromagnetic alignment, inducing remarkable hysteresis at low temperatures and having noticeable coercivities and loop shifts.[27–30]

We consider in this work a three-dimensional antiferromagnetic small particle that has a single-ion uniaxial anisotropy. We perform Monte Carlo simulations to obtain the phase diagram of the system with the external magnetic field parallel to the easy axis of the particle. The magnetic moments of the particle are represented by continuous vectors, and they interact through a classical Heisenberg Hamiltonian. In order to investigate the dependence of the phase diagram on the size of the particle, we consider particles with radii from 3 up to 12 lattice spacings. We also determine the behavior of the phase boundaries as a function of temperature in the region of very low temperatures, and the results are compared with the well-known predictions based on spin-wave theory.

In the remainder of this work, in Sec. II we present the model and we describe the Monte Carlo simulations. Next, in Sec. III, we present our results and summarize our main conclusions.

II. MODEL AND MONTE CARLO SIMULATIONS

To describe the antiferromagnetic particle, we consider a finite simple cubic structure, where we inscribe a spherical particle. Each site inside this sphere harbors a magnetic moment of the particle, which is represented by vectors of magnitude $|\vec{S}_i|=1$, where we write $\vec{S}_i=(S_{ix},S_{iy},S_{iz})$ for the components of the spin at the site i of the particle.

The classical Heisenberg Hamiltonian of the system is

$$\mathcal{H} = \frac{J}{2}\sum_{i=1}^{N}\sum_{j=1}^{q}(S_{ix}S_{jx}+S_{iy}S_{jy}+S_{iz}S_{jz}) - H\sum_{i=1}^{N}S_{iz} - k\sum_{i=1}^{N}S_{iz}^2, \tag{1}$$

where only the couplings among nearest neighbors are considered. N is the number of spins in the particle, q is the coordination number of the magnetic moments ($q=6$ for the internal spins), J is the exchange coupling, H represents the magnitude of the external magnetic field, and k is the single-ion uniaxial anisotropy constant. We assume that the magnetic field and the easy axis lie in the z direction and that the exchange couplings are of the antiferromagnetic type with the same value for all pairs of spins in the particle—that is, $J>0$.

The particle was simulated by employing the Metropolis algorithm.[31] In each Monte Carlo step (MCS), we performed N trials to change the state of the spins of the particle. To calculate the average magnetic properties we took in general 5×10^4 MCS, where the first 2×10^4 MCS were discarded due to the thermalization process.

In our algorithm we calculated the average and staggered magnetizations of the particle, as well as its components along the x, y, and z directions, as a function of temperature, external magnetic field, and anisotropy. These average values were first obtained by calculating the mean values of the magnetic moments inside the particle for each MCS after the thermalization:

$$m_x = \frac{1}{N}\sum_{i=1}^{N}S_{ix}, \tag{2}$$

$$m_y = \frac{1}{N}\sum_{i=1}^{N}S_{iy}, \tag{3}$$

$$m_z = \frac{1}{N}\sum_{i=1}^{N}S_{iz}, \tag{4}$$

$$m_{tot} = \sqrt{m_x^2+m_y^2+m_z^2}, \tag{5}$$

$$m_{sx} = \frac{1}{N}\sum_{i=1}^{N}(S_{ix}^{(a)}-S_{ix}^{(b)}), \tag{6}$$

$$m_{sy} = \frac{1}{N}\sum_{i=1}^{N}(S_{iy}^{(a)}-S_{iy}^{(b)}), \tag{7}$$

$$m_{sz} = \frac{1}{N}\sum_{i=1}^{N}(S_{iz}^{(a)}-S_{iz}^{(b)}), \tag{8}$$

$$m_s = \sqrt{m_{sx}^2+m_{sy}^2+m_{sz}^2}. \tag{9}$$

$S_{i\alpha}^{(a)}$ and $S_{i\alpha}^{(b)}$ are the components of the ith magnetic moment along the α ($\alpha=x,y,z$) direction in the (a) and (b) sublattices, respectively. Afterwards, the averages are taken by considering all Monte Carlo steps, after thermalization. Finally, we obtain the mean values of interest, $\langle m_x\rangle$ to $\langle m_s\rangle$, related to Eqs. (2)–(9), for each value of temperature, external magnetic field, and anisotropy. We also calculated the corresponding fluctuations of the magnetization, defined as

$$\Delta m_{tot} = |\langle m_{tot}^2\rangle - \langle m_{tot}\rangle^2| \tag{10}$$

and

$$\Delta m_s = |\langle m_s^2\rangle - \langle m_s\rangle^2|. \tag{11}$$

These fluctuations are used to determine the transition lines of the phase diagram as a function of temperature and magnetic field for different particle sizes.

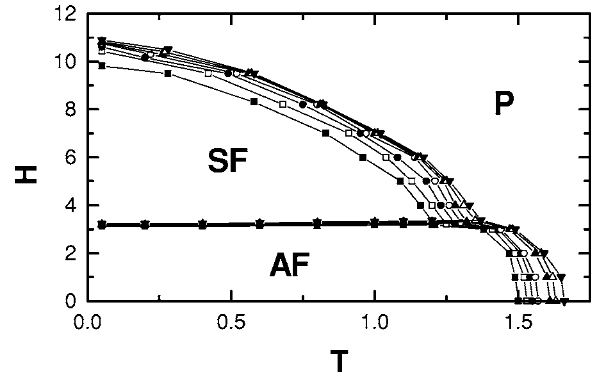

FIG. 1. Phase diagram in the plane temperature vs magnetic field for a uniaxial antiferromagnetic particle obtained through Monte Carlo simulations. From bottom to top the radius of the particle changes from three to nine lattice parameters.

III. RESULTS

By looking at the typical magnetization curves, as well as their corresponding fluctuations obtained through Monte Carlo simulations as a function of temperature and magnetic field, we can find the transition fields and, from them, we construct the phase diagram of the antiferromagnetic particles. The phase diagrams obtained for particles with radii ranging from three to nine spacing lattice units are shown in Fig. 1. In this figure, the symbols AF, SF, and P represent the antiferromagnetic, spin-flop, and paramagnetic phases, respectively. The thermodynamic limit of the system is already observed for a particle with radius $r=9a$ (a is the lattice parameter). As we can see, the Néel temperature and the transition fields increase with the size of the particle up to where we reach the thermodynamic limit of the particle. For a particle with radius $r=9a$, for example, we obtain for the Néel temperature $T_N=(1.70\pm0.05)J/k_B$. At $T=0$, the transition field between the antiferromagnetic and spin-flop phases is $H_{SF}=(3.22\pm0.05)J$, and the transition field between the spin-flop and paramagnetic phases is $H_P=(10.96\pm0.05)J$. These values are in very good agreement with the well-known results found in the literature for an infinite system.[7,9]

Indeed, we determine the boundaries of the phase diagram exhibited in Fig. 1 by looking at the total and staggered magnetizations of the particles and their fluctuations by both fixing the temperature and changing the magnetic field and by fixing the magnetic field and changing the temperature. The transition lines that separate the paramagnetic phase from the antiferromagnetic and spin-flop phases represent continuous phase transitions. On the other hand, the phase boundary between the antiferromagnetic and spin-flop phases denotes first-order phase transitions, where the total magnetization changes abruptlly from zero to a nonzero value. Also, at this transition, the staggered magnetization changes its direction from parallel to perpendicular to the magnetic field.

The two continuous transition lines and the line of first-order transitions meet at the bicritical point (T_b, H_b). We have found that both T_b and H_b increase with the size of the particle up to its thermodynamic limit. For instance, for a particle with radius $r=6a$ we obtain $T_b=(1.30\pm0.05)J/k_B$ and $H_b=(3.28\pm0.05)J$. Although we do not present here calculations based on mean-field theory, for this size of particle the coordinates of the bicritical point are given by $T_b=1.34J/k_B$ and $H_b=4.90J$. As is well known the mean-field calculations overestimate the values of the critical fields and temperatures, due to the long-range character of the interactions. In fact, the mean-field approach serves only to yield a rough description of the properties of the system.

By examining the values of H_{SF} and H_P at $T=0$ for different particle sizes, we found that both decrease with a $1/d^{\alpha}$ dependence, where d is the diameter of the particle. In the case of $H_{SF}(0)$ we obtain $\alpha=0.97\pm0.05$, which is the same behavior found for the hematite particles.[25] This appears to be a typical surface effect on a first-order transition. At the antiferromagnetic to spin-flop phase transition, the total magnetization changes suddenly from zero to a nonzero value. As the spins at the surface present a lower coordination number and they are less bounded than the core spins, they can rotate more easily into a direction almost perpendicular to the field. For the critical field between the spin-flop and paramagnetic phases, $H_P(0)$, we have $\alpha=2.54\pm0.05$.

We also have determined the coordinates of the bicritical point as a function of the particle size. We have seen that H_b and T_b increase with the size of the particle exhibiting the same $1/d^{\alpha}$ behavior, with $\alpha=1.98\pm0.05$ and $\alpha=1.97\pm0.05$, respectively. The Néel temperature ($H=0$) also decreases with decreasing diameter following a law of the type $1/d^{\alpha}$ with $\alpha=1.51\pm0.05$. The α exponent in our model is the inverse of the critical exponent ν. For a three-dimensional model this exponent assumes very close values for the Ising and isotropic Heisenberg models: $\nu\approx0.64$ and $\nu\approx0.70$, respectively. Then, within the error bars, the value we found for the uniaxial Heisenberg model, $\nu\approx0.66$, is in reasonable agreement with this picture.

We have also investigated through Monte Carlo simulations the dependence of the phase boundaries on temperature in the asymptotic region of very low temperatures (see Fig. 2). We have seen that for particles with radius $r>3a$, the critical field between the spin-flop and paramagnetic phases decreases with temperature following a $T^{3/2}$ law, which agrees with the predictions of spin-wave theory for uniaxial antiferromagnetic systems.[7,9,14] The critical field is given by $H_P(T)=H_P(0)-\alpha(r)T^{3/2}$. Although the dependence on temperature is already seen for small particles, the coefficient α depends on the size of the particle. For instance, α decreases with the size of the particle: $\alpha(r=3a)=7.96$, $\alpha(r=6a)=1.55$, and $\alpha(r=9a)=1.31$. That is, when the ratio between the number of spins at the surface and at the volume is large, the finite-size effects become important. On the other hand, the transition field separating the antiferromagnetic and spin-flop phases increases with temperature according to a $T^{7/2}$ law, which is in agreement with the spin-wave calculations for the thermodynamic boundary between these two phases.[15]

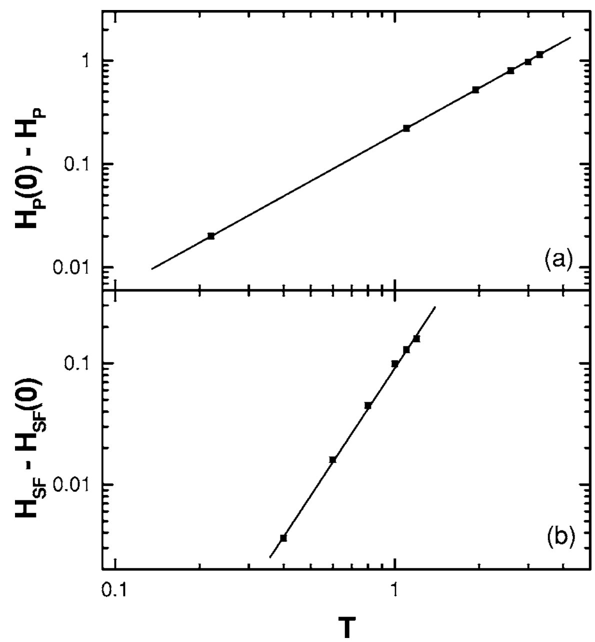

FIG. 2. Low-temperature behavior of the (a) phase boundary between the spin-flop and paramagnetic phases and (b) phase boundary between the antiferromagnetic and spin-flop phases. $H_P(0)$ and $H_{SF}(0)$ are the corresponding transition fields at $T=0$.

Summarizing, in this work we have considered Monte Carlo simulations to study the phase diagram of uniaxial antiferromagnetic spherical particles. We have investigated the magnetic behavior of the particle as a function of temperature, external magnetic field, and size of the particle. We have shown that particles with sizes as large as nine lattice parameters already behave as a bulk system. For particles with radii $3a\leq r\leq12a$, we have obtained the phase diagram in the plane field versus temperature and we have calculated the asymptotic values of the transition fields as a function of their diameter. We have shown that the critical field separating the spin-flop and paramagnetic phases decreases with temperature according to a $T^{3/2}$ law. On the other hand, the transition field between the antiferromagnetic and spin-flop phases increases with temperature following a $T^{7/2}$ behavior in the region of very low temperatures. Although these asymptotic low-temperature behaviors are well established by spin-wave calculations for uniaxial antiferromagnets, we have seen that they are already observed for small spherical particles with radius equal to three lattice parameters.

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