Magnetic behavior of thin rare-earth films

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We consider a thin rare-earth film with the *c* axis perpendicular to the surface, to study its nonuniform magnetic structure induced by the lower coordination near the surfaces. The influence of a dc magnetic field, applied perpendicular to the *c* axis, on the equilibrium configurations is also investigated. The spin-wave frequencies, in the $q=0$ limit, are calculated as a function of the strength of the external dc field. We use the bulk parameters of dysprosium and holmium to obtain the external field dependence of the equilibrium configuration, magnetization, and the lowest spin-wave modes of 30-monolayer-thick films. [S0163-1829(99)10209-1]

I. INTRODUCTION

The physical properties of ferromagnetic and antiferromagnetic films have been intensively studied for a long time.[1] The solutions of classical problems involving the magnetic behavior of these systems can now be found in several text books. However, the rapid development of the preparation techniques, which nowadays allow the growth of very thin magnetic metallic films of exceptional quality, opened up new areas of interest. In fact, the observation of new magnetic phenomena such as the giant magnetoresistance of transition metal trilayers,[2] and the perpendicular alignment of the thin film magnetizations also observed in transition-metal thin-film trilayers,[3,4] among others, revived the interest in the area of thin-film magnetism. The use of molecular-beam epitaxial (MBE) growth techniques, for example, has allowed the fabrication of single-crystal thin films with a well-defined number of atomic planes and allowed the test of fundamental theories of magnetism. Moreover, the need to prepare materials for several specific purposes has demanded a better understanding of the intrinsic characteristics of these systems, and motivated a considerable number of researchers to investigate details of their intriguing properties.

However, while ferromagnetic and antiferromagnetic films have received great attention and have been well studied theoretically and experimentally, the same cannot be said of rare-earth thin films. So far, most of the studies of geometrically limited rare-earth systems has been dedicated to multilayered systems and the main goal has been the understanding of the influence of the substrate or spacer on the thermodynamic behavior of the system.[5] The competition between the magnetic trends of the different layers[6,7] in multilayered systems has also been studied.

The magnetic properties of rare-earth (RE) superlattices result from the combined effect of the individual behavior of the constituent RE films and the features introduced by the periodicity of the multilayered structure. Therefore, local effects like the weaker exchange coupling of the moments near the surface of a film (due to the lack of coordination) can easily be masked by the overall behavior of the system. Under the influence of an external dc magnetic field a helimagnetic film should display a nonuniform distortion of the helix, with stronger field effects near the surfaces. The moments in the middle of thick films should display field effects as prescribed for the bulk. This fact must be reflected in the response of the system to an external input. However, in a periodic system, like superlattices, the response also contains the periodic character of the geometry artificially imposed and the surface effects may therefore be hidden. The aim of this paper is to study the influence of the lack of coordination of the magnetic moments localized near the surfaces on the magnetic behavior of thin rare-earth films. To avoid unnecessary complexity, we focus our discussion on the physical properties of RE films with the *c* axis perpendicular to the surface.

In the Sec. II of this paper we discuss the model used to describe the system and we study the equilibrium configuration (field induced magnetic phases) of a rare-earth system with a finite number of monolayers; Sec. III is devoted to a theoretical study of the $q=0$ spin waves and the relationship between their frequencies and the magnetic phases of the system; the conclusions as well as a discussion of selected features of rare-earth films is presented in Sec. IV.

II. THE EQUILIBRIUM CONFIGURATION

A basic feature of the known magnetic phases of bulk rare-earth metals, at any temperature and in presence of arbitrary dc fields, is that the moments in each basal plane are equivalent. Therefore the magnetic structure can be seen as a chain of spins, each representing the magnetic moment per lattice site in a basal plane.[8] The magnetic phases of a thin film with the *c* axis perpendicular to its surfaces can equally well be represented in this manner since the surfaces break the symmetry along the direction perpendicular to the basal plane but introduce no modification in the basic symmetry of the system. Spins in a given plane (parallel to the surface) are still equivalent.

The effective coupling between layers of equivalent spins is described by the conventional bilinear exchange. In equilibrium, the energy associated to the intraplane exchange is never modified since the moments in a given basal plane are always parallel to each other. Hence, this contribution to the magnetic energy does not need to be taken into account in

the magnetic Hamiltonian. Even the excitations associated with $q=0$ spin waves, or spin waves propagating along the c axis, do not modify this picture since the spins in any basal plane oscillate in phase.

Therefore, we model the rare earth film as a stack of atomic monolayers, infinitely extended in the x-y directions, which are coupled through exchange interaction with the nearest- and the next-nearest-neighbor monolayers. Each monolayer also feels an anisotropy that is responsible for the helimagnetic or conic behavior of the entire system. We assume the c axis of the rare-earth material is perpendicular to the surfaces of the film. Therefore the c axis is along the z direction. In a helimagnetic configuration the magnetic moments are in the basal plane (x-y plane) and the direction of the moments of consecutive monolayers are turned by a constant angle if the thickness of the film goes to infinite. In a conical configuration the moments have a finite component parallel to the c axis while the moment projections in the basal plane follow a helimagnetic structure. The cone angle is the equilibrium configuration resulting from the competition between the anisotropy and interplane exchange energies. A complete discussion of these features can be found in the papers listed in Ref. 8.

In our model, the spins in the surface region have the same anisotropy as the rest of the system but have a weaker exchange coupling due to the lack of coordination. In the equilibrium configuration each spin is aligned with the local effective field at any layer.[9] Therefore surface effects are not restricted to the surface layers. A few layers near the surface are also affected by the lack of surface coordination. The actual number of layers modified by the surface effects depends on the way the effective local field relaxes towards the bulk pattern in the middle of the film.

To obtain the equilibrium configuration of the system, we use the model described above to write the magnetic energy as

$$E=J_1\sum_{n=1}^{N-1}\mathbf{S}(n)\cdot\mathbf{S}(n+1)+J_2\sum_{n=1}^{N-2}\mathbf{S}(n)\cdot\mathbf{S}(n+2)+\sum_{n=1}^{N}[K_2S_z^2(n)+K_4S_z^4(n)-\gamma\mathbf{S}(n)\cdot\mathbf{H}_o]. \quad (1)$$

Here J_1 and J_2 are the parameters that describe the exchange interaction between the nearest and next-nearest monolayers, respectively, $\mathbf{S}(n)$ denotes spins in the nth monolayer, K_2 and K_4 are the parameters that specify the anisotropies of the system, $\gamma=g\mu_B$, and $\mathbf{H}_o$ represents the externally applied dc field. Notice that surface effects are incorporated in Eq. (1) since the spins near the surfaces have the exchange energy reduced by the absence of nearest and/or second-nearest neighbors. As mentioned before, the lack of coordination affects directly only the spins of the first two layers near the surfaces ($n=1$, 2, $N-1$, and N) but, as will be seen later, the effect may be felt by spins at monolayers several atomic planes away from the borders of the film.

Since $|\mathbf{S}(n)|=S$ is a constant, we can write the components of $\mathbf{S}(n)$ as $S_x(n)=S\sin(\theta_n)\cos(\varphi_n)$, $S_y(n)=S\sin(\theta_n)\sin(\varphi_n)$, and $S_x(n)=S\cos(\theta_n)$, where the angles θ_n and φ_n determine the direction of $\mathbf{S}(n)$ with respect to the z axis and x axis, respectively. The equilibrium configuration is obtained from the calculation of the angles θ_n and φ_n that minimize the magnetic energy given by Eq. (1). This procedure is equivalent to looking for values of θ_n and φ_n that make the torque on every spin $\mathbf{S}(n)$ equal to zero. Therefore, to find out the equilibrium configuration we should obtain the profile $\{\theta_n,\varphi_n;n=1,\ldots,N\}$, that makes $\mathbf{S}(n)[\partial E/\partial\mathbf{S}(n)]=0$ for all spins and also has the minimum energy. For fixed values of the parameters and external dc field, there are several profiles that make the torque on every spin equal to zero. The equilibrium configuration is the profile that gives the lowest energy. We use the numerical method described in more detail in Ref. 9 to obtain the magnetic structure for films of dysprosium and holmium. At low temperatures, these films are described by the following parameters: $S(\mathrm{Dy})=5/2$, $J_1(\mathrm{Dy})=-6.08\times10^{-15}$ ergs/ion, $J_2(\mathrm{Dy})=2.07\times10^{-15}$ ergs/ion, $K_2(\mathrm{Dy})=1.33\times10^{-15}$ ergs/ion, $K_4(\mathrm{Dy})=-0.13\times10^{-15}$ ergs/ion, $S(\mathrm{Ho})=2$, $J_1(\mathrm{Ho})=-7.31\times10^{-15}$ ergs/ion, $J_2(\mathrm{Ho})=2.11\times10^{-15}$ ergs/ion, $K_2(\mathrm{Ho})=-0.44\times10^{-15}$ ergs/ion, $K_4(\mathrm{Ho})=1.24\times10^{-15}$ ergs/ion. The anisotropy parameters above are related to the experimental values of K_2^0, K_4^0, and K_6^0, given in Ref. 10, by

$$K_2S^2=\frac{3}{2}K_2^0-\frac{15}{4}K_4^0+\frac{1575}{240}K_6^0 \quad (2a)$$

and

$$K_4S^4=\frac{35}{8}K_4^0-\frac{4725}{240}K_6^0. \quad (2b)$$

By imposing cyclic boundary conditions on a film containing complete helices we have reproduced the main results of bulk materials. For low dc fields applied parallel to the basal plane, the magnetization increases linearly with the strength of the magnetic field, reflecting weak distortions in the helix. The distortions in the helical pattern become larger as the strength of the external field is increased. By further increasing the external field strength one reaches a field where a first-order transition to the fan phase occurs. In this fan phase the directions of the equilibrium positions of the moments oscillate around the direction of the applied field. The amplitude of these oscillations gradually decreases, as the external field is increased, until saturation is reached. These results are reported in a number of review articles.[10–12]

We have studied surface and field effects for films of various thicknesses. As could be expected, we found the distortions imposed on the helix by the combined action of the low surface coordination and the applied field are stronger near the surface region. In the absence of external field, the two layers of spins near the surfaces are directly affected by the drop in coordination and the rest of the film is only indirectly affected by the surface effects. We have found that, in very thin films (12 layers, for example), practically all the layers are affected by the surface induced distortions. On the other hand, in thick films (a few hundreds of planes) only a minor fraction of the film (near the surfaces) is affected and, similar to what is found in the unbounded RE

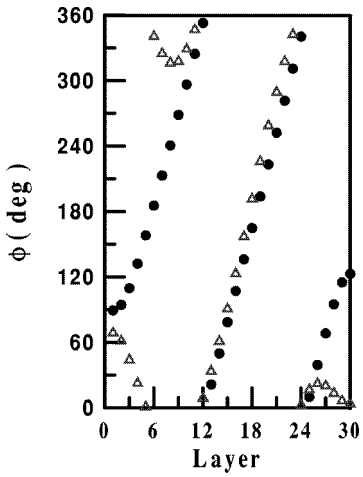

FIG. 1. Turn angles of an holmium film, 30-monolayers thick, in the absence of field (dots) and when a dc field of 2 T is applied parallel to the x direction (triangles).

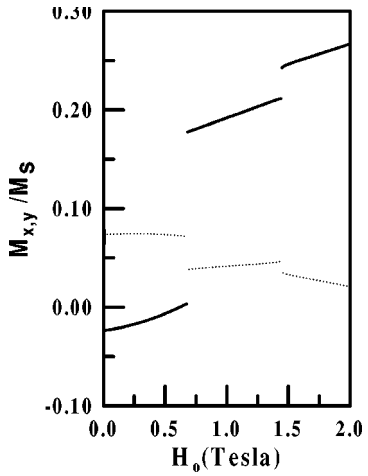

FIG. 2. Field dependence of the x (full lines) and y (dot lines) components of the magnetization of the holmium film with 30 monolayers. The magnetization is shown in unit of the saturation magnetization.

materials, the magnetic pattern is mainly determined by the intrinsic competition between first- and second-neighbor exchange.

The effect of an applied field on the magnetic structure of thin RE films is by no means obvious. The spins near the surfaces are softer and consequently respond more easily to the external dc field. One might then think that the field should induce stronger alignment of moments near both surfaces. However, the magnetization pattern is controlled by the surface effects and also by the twist imposed in the central part of the film. The field induces unequal surface regions. The restriction of equivalent surface regions, as might be expected at first glance, leads to higher energy.

In Fig. 1 we display the turn angles of the spins in the layers of a 30-monolayers-thick (≈ 170 Å) Holmium film in the absence of field (dots) and when a dc field $H_o = 2$ T is applied perpendicular to c axis and parallel to x direction (triangles). In this picture, we can see that, in the absence of external field, the helix is deformed only in the two monolayers near the surfaces. On the other hand, in the presence of an applied field of $H_o = 2$ T, only a small fraction of spins, near the middle of the film, preserves the helix character. In this case near one of the surfaces, a fan phase is produced with an amplitude around $\pi/6$, while near the other surface a few spins are aligned with the applied field. Upon further increase of the applied field (results not shown here) the fan phase covers the whole film.

The field induced nonsymmetrical modification of the magnetic pattern reported here is analogous to what has been reported for other systems of current interest. This is the case, for instance, of the low field surface induced instability of the antiferromagnetic phase of antiferromagnetic multilayers[13] with finite number of layers.

The analysis of the energy of the system, as a function of the applied dc field, shows that the system has two phase transitions in this field interval, which correspond to different kinds of distortion of the helical projection in the basal plane. These phase transitions can be seen in Fig. 2 where we display the x (full lines) and y (dot lines) components of the magnetization as a function of the dc field, applied parallel to the x direction. In Fig. 2 the jumps in the magnetization reflect major changes in spin arrangements near the surfaces. From $H_o = 0$ to $H_o = 0.75$ T the helix deforms continuously as the field is increased. The first magnetization jump reflects the onset of a large amplitude ($\pi/3$) fan phase at one surface (involving ten spins), which gradually aligns with the field. The second magnetization jump, at $H_o = 1.5$ T, results from a sudden reduction of the amplitude of the oscillations, decreasing to nearly $\pi/6$.

III. SPIN WAVES

The magnetic behavior of the system can also be studied through the analysis of its spin waves. Once the equilibrium configuration is known, for a given value of the applied dc field, the frequencies of the $q = 0$ spin waves can be found from the equations of motion, $[d\mathbf{S}(n)/dt] = -\mathbf{S}(n) \times (-[\partial E/\partial \mathbf{S}(n)])$. Following the standard procedure, we write

$$\mathbf{S}(n) = \mathbf{S}_o(n) + \mathbf{s}(n,t) \tag{3a}$$

and

$$-\frac{\partial E}{\partial \mathbf{S}(n)} = \mathbf{H}^o_{\text{eff}} + \mathbf{h}(n,t), \tag{3b}$$

where $\mathbf{S}_o(n)$ and $\mathbf{s}(n,t)$ denote the equilibrium and the oscillation of the spins of the nth monolayer, respectively. $\mathrm{H}^o_{\text{eff}}$ is $-[\partial E/\partial \mathbf{S}(n)]$ calculated in the equilibrium, and $\mathbf{h}(n,t)$ represents the time dependent part of the effective field introduced by $\mathbf{s}(n,t)$. Then, we linearize the equations of motion with respect to $\mathbf{s}(n,t)$ and $\mathrm{h}(n,t)$ and assume a harmonic dependence for all dynamical quantities $[\mathbf{s}(n,t) = \mathbf{s}(n) \exp(-i\Omega t)]$. Thus the coupled equations of motion can be written in the matrix form:

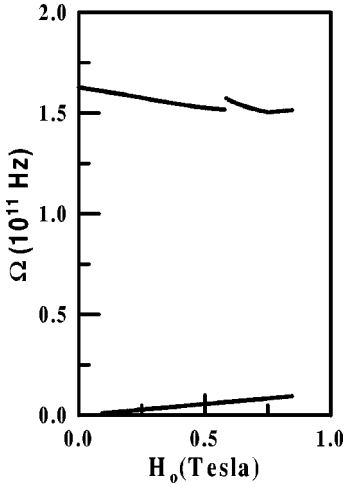

FIG. 3. Dispersion relation of the two lowest frequency modes of the $q=0$ spin waves in a 30-monolayers-thick dysprosium film.

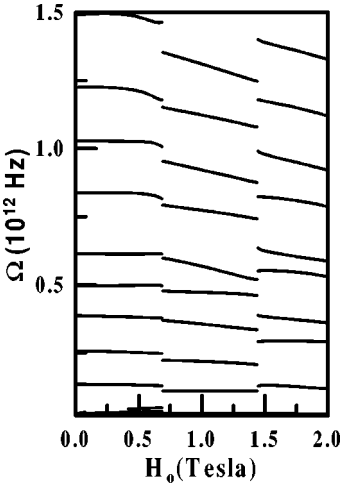

FIG. 4. Dispersion relation of several modes of the $q=0$ spin waves in a 30-monolayers-thick holmium film.

$$\left[i\Omega \begin{pmatrix} 1 & 0 & 0 & . & . & . \\ 0 & 1 & 0 & . & . & . \\ 0 & .0 & 1. & . & . & . \\ . & . & . & . & . & . \\ . & . & . & . & . & . \\ . & . & . & . & . & 1 \end{pmatrix} - \begin{pmatrix} M_{1,1} & M_{1,2} & . & . & . \\ M_{2,1} & M_{2,2} & . & . & . \\ M_{3,1} & . & . & . & . \\ . & . & . & . & . \\ . & . & . & . & . \\ . & . & . & . & M_{3N,3N} \end{pmatrix} \right] \begin{pmatrix} s_x(1) \\ s_y(1) \\ s_z(1) \\ . \\ . \\ s_z(N) \end{pmatrix} = 0, \tag{4}$$

where the $3N$ column vector is constructed with the three components of the N vectors that denote the oscillation of the spins and $M_{i,j}$ are the elements of the $3N \times 3N$ matrix $\overleftrightarrow{M}$ constructed from the 3N linearized equations of motion, which depends on θ_n and ϕ_n (angles that give the equilibrium configuration). The computation of the matrix elements $M_{i,j}$ is straightforward but the final expressions are quite long and we do not show here. The frequencies of the spin waves are the imaginary part of the eigenvalues of $\overleftrightarrow{M}$. Each eigenvector of $\overleftrightarrow{M}$, associated with a given frequency, describes the characteristics of the oscillation in that frequency.

In Fig. 3 we show the frequencies of the two lowest frequency spin waves in a dysprosium film, 30-monolayers thick, for different values of the dc magnetic field applied parallel to the x direction. For the lowest frequency mode there is a linear dependence of the frequency on the strength of the magnetic field. Moreover, the eigenvector associated with this mode shows that it corresponds all spins oscillating in phase and with the same amplitude. So we can say that this mode is similar to the acoustic modes observed in multilayered systems. The second lowest frequency mode has a different character. In this mode the film is divided in two parts oscillating with opposite phases. A spin near the center of the film, with zero amplitude of oscillation, separates the two parts. When the strength of the dc field is increased, the nonoscillating spin is displaced in the direction of the surface.

In Fig. 4 we display several modes of the holmium film described earlier. In this picture, in addition to the H_o dependence of the low-frequency modes, one can observe that the $q=0$ spin waves also give information about the phase transitions of the film. In Fig. 2 we showed that the magnetization is discontinuous at fields where the system has a phase transition. In Fig. 4 we can see that these phase transitions are also characterized by discontinuity in the dispersion relation of the spin waves.

IV. FINAL REMARKS

The diversity of the magnetic behavior of bulk rare-earth materials attracted a great deal of attention in the past. This happened mainly due the rich variety of magnetic phases of these materials and the intriguing effects produced by dc magnetic fields.[8,12] More recently, nonsymmetrical modification induced by externally applied magnetic field on finite transition-metal multilayered systems, with even number of magnetic layers, was reported[13] and motivated several authors to investigate this unusual characteristic. We have demonstrated here that RE films may have not only the regular bulk RE behavior but also surface features rather similar to antiferromagnetic multilayers. The common feature is the modification in the magnetic equilibrium profile which results from soft spins near the surfaces. In antiferromagnetic multilayers composed of thin transition-metal films[13] each layer corresponds to a thin transition-metal film which can be represented by a single spin variable. In the RE film each magnetic layer corresponds to an atomic plane.

One of the greatest motivations for the study of new magnetic systems is the high demand for new materials to fit specific technological applications. To meet this goal the magnetic system has to be well characterized in order to have a reliable value. In this paper we studied the physical behavior of RE thin films and directed our calculation to obtain quantities that can be used to characterize them. Among

other techniques the magnetic optical Kerr effect can be used to ''see'' the magnetic phases of the system. Furthermore the measurement of the frequencies of the excitations of the system may also give relevant information about the magnetic phases.

In our numerical calculation, we have used parameters that describe bulk RE materials. The symmetry break, imposed by the surfaces of the films, may modify the intrinsic physical properties of the system and, as a consequence, modify the parameters that describe them. However, we do not expect qualitative changes of the physical behavior reported here. In fact, despite the simple model used to describe RE films, we expect that the prediction that these films have modes with acoustical and optical character (so far not observed in magnetic films) can be experimentally investigated. We are sure that an experimental study of RE thin films would reveal the unknown features of these systems.

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