Magnetic structures in ultra-thin Holmium films: Influence of external magnetic field

L.J. Rodrigues [a,b], V.D. Mello [b], D.H.A.L. Anselmo [a], M.S. Vasconcelos [c,*]

[a] Departamento de Física Teórica e Experimental, Universidade Federal do Rio Grande do Norte, Natal 59600-900, RN, Brazil
[b] Departamento de Física, Universidade do Estado do Rio Grande do Norte, Mossoró 59625-620, RN, Brazil
[c] Escola de Ciência e Tecnologia, Universidade Federal do Rio Grande do Norte, 59072-970 Natal, RN, Brazil

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ABSTRACT

We address the magnetic phases in very thin Ho films at the temperature interval between 20 K and 132 K. We show that slab size, surface effects and magnetic field due to spin ordering impact significantly the magnetic phase diagram. Also we report that there is a relevant reduction of the external field strength required to saturate the magnetization and for ultra-thin films the helical state does not form. We explore the specific heat and the susceptibility as auxiliary tools to discuss the nature of the phase transitions, when in the presence of an external magnetic field and temperature effects. The presence of an external field gives rise to the magnetic phase Fan and the spin-slip structures.

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1. Introduction

The magnetic properties of rare-earth materials were the subject of intensive studies in the sixties years (for major reviews, see Refs. [1,2]). With the success of modern crystal growth techniques to produce rare-earth multilayers, the interest in this subject is currently undergoing a fast resurgence, with exciting new perspectives for the future [3].

On the other hand, magnetic ordering in the heavy rare-earth lanthanides [4] is mediated by the RKKY interaction in which the polarization of conduction electron yields an indirect exchange between localized 4f moments on neighboring lattice sites [5]. The interplay between this long-range interaction and anisotropic and magneto-crystalline effects results in complex magnetic ordering and the possibility of a variety of magnetic structures. As stated before, understanding the magnetism of thin films is of general importance in a technological context, and rare-earth metals are used in a wide range of applications, and have attained global economics importance [6,7]. One of the most recent purposes is the use of these materials in solid-state rare-earth-ion-doped systems, which justifies their status as very strong candidate to a long-lived quantum memory system [8]. Also, very recently, Lovric et al. [9] have reported a high fidelity optical memory, made of a rare earth doped crystal, in which dynamical decoupling is used to extend the storage time. The present work intends to contribute to the study of the magnetic phases of rare-earth thin films, aiming further applications.

Among the rare-earths, Holmium in particular displays unusual behavior which, coupled with the large temperature stability of the intermediate helimagnetic phase when compared to Dy and Tb, has led to it being treated as a model system in a number of recent experimental [10] and theoretical [11,12] studies. In the bulk, Holmium metal orders magnetically below $T_N = 132$ K (Néel temperature) and the moments in consecutive neighboring basal planes rotate to give a long-period helical phase. At temperatures below $T_C = 20$ K (Curie temperature) a second first-order phase transition to a bulk FM state occurs driven by competition between magnetoelastic and exchange energies. It arranges as a cone with angle of $80.5° \leq \theta \leq 90°$, giving rise to a conical phase [13]. When a magnetic field is applied in the basal plane other magnetic structures are observed including a basal plane ferromagnetic phase, fan phase and helifan phase [14,15]. In thin films when the thickness is comparable with the periodicity of the ordered structure, it is expected that even the magnetic arrangement itself can be strongly modified.

For ultrathin films, however, as the number of spin layers is reduced by decreasing the film thickness, additional structures are established including a fan and spin-slip [16,17] structures, highlighting the importance of surface and interfacial effects. This finite-size effect is caused by the reduced number of atoms in the direction perpendicular to the film plane that leads to a decrease of the total magnetic exchange energy. Rare-earth helimagnets such as Ho, Dy, and Tb (for a review see [18]) represent the best candidates to put into evidence such finite-size effects.

* Corresponding author.
E-mail address: mvasconcelos@ect.ufrn.br (M.S. Vasconcelos).

The magnetic phases in ultrathin helimagnetic holmium films were recently studied by using of Monte Carlo simulations [11], where the authors have described the system by a Heisenberg model with easy-plane single-ion anisotropy and seven coupling constants (obtained by experimental neutron scattering measurements). They have observed that for the analyzed thicknesses of 8–16 monolayers, it is seen that by increasing the temperature the films show, after the distorted helical configuration, a temperature range where disordered inner planes coexist with surface ones in a block ordered state (spin-slip). Also, it is observed, at low temperatures, that the progressive reduction in the number of layers does not imply a sudden helical order suppression but rather, as a consequence of surface effects, a gradual passage to a Fan-like order, accompanied by the helix distortion.

In this paper we investigate a c-axis thin film, consisting of a stacking of atomic layers with equivalent spins, infinitely extended in the $x - y$ directions. We consider that the spins in each monolayer are exchange coupled with the spins in the first and second neighbor monolayers. The anisotropy is uniform throughout the film and the near-surface spins have reduced exchange energy. To study the magnetic phases as functions of the applied magnetic field, we use a self-consistent local field model which incorporates the surface modifications in the exchange field and the thermal average values ($\langle J(n) \rangle$; $n = 1; ...N$) and the orientation of the spins in each layer ($\langle \phi_n \rangle$; $n = 1; ...N$). Our main purpose is to study the magnetic phases of very thin Ho films in the temperature interval between 20 K and 132 K. Specifically, we will investigate the influence of the surface and thickness of a thin film, associated with the existing competition between the energies of exchange and magneto-crystalline anisotropy, when in the presence of an external magnetic field and temperature.

This paper is organized as follows. In Section 2 we provide our theoretical framework for the Hamiltonian, which takes into account the hexagonal anisotropy. A discussion about our model and comparison with other works are done in Section 3. Finally, we summarize our main conclusions in Section 4.

2. Theoretical model

We investigate a c-axis thin film, consisting of a stacking of atomic layers with equivalent spins, infinitely extended in the x–y directions. The spins in each monolayer are exchange coupled with the spins in the first and second neighbour monolayers. The anisotropy is uniform throughout the film and the near surface spins have reduced exchange energy. The magnetic Hamiltonian is given by

$$H = J_1(g - 1)^2 \sum_{n=1}^{N-1} \vec{J}(n) \cdot \vec{J}(n+1) + J_2(g-1)^2 \sum_{n=1}^{N-2} \vec{J}(n) \cdot \vec{J}(n+2)$$
$$+ \sum_{n=1}^{N} \left[K_6^6(T)\cos(6\phi_n) - g\mu_B \vec{J}(n) \cdot \vec{H} \right] \tag{1}$$

In Eq. (1), J_1 and J_2 describe the exchange interaction between the nearest and next nearest monolayers respectively, $\vec{J}(n)$ denotes the total angular momentum per atom in the n-th monolayer, $K_6^6(T)$ describes the hexagonal anisotropy and the last term is the Zeeman Energy, where the external field $\vec{H}$ is applied in one easy direction in the hexagonal plane, making an angle of 30° with the x-axis and $g = 5/4$ is the Landé factor, corresponding to a saturation magnetic moment per atom of 9.5 μ_B.

We use a self-consistent local field model that incorporates the surface modifications in the exchange field and the thermal average values ($\langle J(n) \rangle$; $n = 1; ...N$) and the orientation of the spins

in each layer ($\langle \phi_n \rangle$; $n = 1; ...N$) [19,20]. With our model, one can reproduce (as a way of checking the model), the bulk phases in Holmium, observed experimentally [21]. Therefore only two exchange constants are enough to our intent.

We consider the Ho bulk energy parameters [22], where $J = 8$, $J_1 = 47k_B$, $J_2 = -J_1/4\cos\phi(T)$, where $\phi(T)$ is the temperature dependent helix turn angle [23]. Also, $K_6^6(T)$ is adjusted so as to reproduce the temperature dependence [24] of the hexagonal anisotropy energy.

Surface effects are incorporated in Eq. (1) since the spins near the surfaces have the exchange energy reduced by the absence of nearest and second nearest neighbors. Therefore only the spins of the first two planes near the surfaces ($n = 1$, 2, $N - 1$ and N) are directly affected by surface effects. However the lack of coordination near the surfaces may be felt by spins deep inside the film. The helix as a whole accommodates to the combined effects of applied field, temperature and the reduction of the exchange near the surfaces.

The equilibrium configuration is obtained from the angles $[(\phi_n), n = 1, ...N]$ which minimize the magnetic energy given by Eq. (1). The numeric algorithm is equivalent to looking for values of ϕ_n that make the torque on every spin $\vec{J}(n)$ equal to zero. Therefore, to find out the equilibrium configuration we obtain the profile $\{\phi_n; n = 1, ...N\}$, which makes $\vec{J}(n) \times \partial\vec{H}/\partial\vec{J}(n)| = 0$ for all spins. The numerical method is described in more detail in [25,26].

3. Results and discussion

We concentrate our analysis on the phase diagrams H–T obtained for Holmium bulk and for thin and ultra-thin films composed by $n = 24$, 10 and 7 monolayers. We present the H–T diagram in the temperature interval from 20 K to 132 K. The magnetic phase transitions are functions of the external magnetic field H, temperature T, and thickness N. We use the specific heat (magnetic contribution only), and the magnetic susceptibility to identify the nature of the magnetic phase transition.

In Fig. 1 we show the H–T diagram of Ho bulk in the temperature range from 20 K to 132 K. When the field is increased on the isothermal process, there is a phase transition from helimagnetic phase to fan phase, going to ferromagnetic phase (FM): Helix → Fan → FM. For a heating process (isofield process), we can see up to four transitions. For example, at 8 kOe: FM → Fan → Helix → Fan → PM.

In Fig. 2 we show the dependence of isothermal magnetization with applied magnetic field, for both $T = 50$ K and 70 K. The magnetic susceptibility (inset) for selected values of magnetic field and temperatures is also presented. We can identify that in the phase diagram, for $T = 50$ K and $T = 70$ K, there are two magnetic phase transitions: Helix → Fan → FM. For $T = 50$ K we can see a peak in the magnetic susceptibility at $H = 5$ kOe marking the magnetic phase transition, Helix → Fan and a Fan → FM transition at $H = 7.5$ kOe.

The specific heat is also investigated, as shown in Fig. 3. This contribution was numerically calculated from the difference in the magnetic energy of Eq. (1), where the energy parameters are temperature dependent, for T and $T + \delta T$, and δT is a small increment in the value of the temperature [27]. Namely,

$$C(T, H) = \left(\frac{\partial E}{\partial T} \right)_H \tag{2}$$

We can see that for $H = 6$ kOe, the magnetic phase transitions are marked by the specific heat. The FM → Fan phase magnetic transition is marked by a deep peak. However, the Fan → Helix transition is not marked, because the temperature interval which