J Mater Sci (2010) 45:5036–5039
DOI 10.1007/s10853-010-4467-7

ICAM 2009

Surface spin slips in thin dysprosium films

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Received: 29 November 2009 / Accepted: 3 April 2010 / Published online: 17 April 2010

Abstract We report a theoretical investigation of new magnetic phases of thin dysprosium films. At the surfaces, the balance between exchange and anisotropy energies favors the alignment of spins along the basal plane easy axis directions. Near the Curie temperature, the helimagnetic phase is strongly modified, allowing surface spins to lock into the basal plane anisotropy easy directions. As the temperature raises, we find transitions to other surface lockin patterns, which are tunable by external field strengths of a few hundred milli-Oersteds.

Introduction

Considerable research effort has been dedicated to investigate the impact of confinement and surface effects on the equilibrium phases of magnetic systems with size comparable to fundamental magnetic lengths.

Particularly interesting are magnetic systems in which the bulk magnetic phases consist of a periodical repetition of units containing a finite number of spins. This is the case of the helical structure of rare earth metals, and the helix period is a valuable length scale to evaluate confinement effects.

The rare-earth (RE) elements exhibit a reach variety of magnetic phases, either induced by external magnetic fields or by thermal effects. Finite-size and surface effects add new features leading to new magnetic phases of thin films [1–4].

In this paper, we present a theoretical discussion of new magnetic phases of thin dysprosium films. These phases are associated to the lockin of surface spins to the easy axis directions in the basal plane (see Fig. 1 for typical patterns). They resemble the spin slip phases of Ho, in the sense that they originate from the prevalence of the anisotropy energy over the exchange energy. Also, as the temperature raises there is a sequence of surface lockin phases, each within a finite temperature interval, with surface pinning to different easy directions in the basal plane.

Recent reports on new phases of thin film helimagnetic systems include a strong thickness dependence of the Neel temperature of Ho thin films, indicating that the helical state does not form below ten atomic layers [1]; large thermal hysteresis of Dy thin films, induced by an alternate helicity phase associated to the locking of surface spins to the external field direction [2, 3]; and a giant magnetocaloric effect of Dy ultra-thin films, due to the absence of the helical phase and the abrupt drop of the thermal average value of the magnetization at the Neel point [4].

At low temperatures, there are prominent anisotropy effects for low temperature helimagnetic RE metals. For holmium the basal-plane moments bunch strongly around the easy *b* directions, leading to a magnetic pattern commensurable with the lattice. The modifications in this basic figure as the temperature raises, lead to various spin slip patterns at certain temperature intervals [5, 6].

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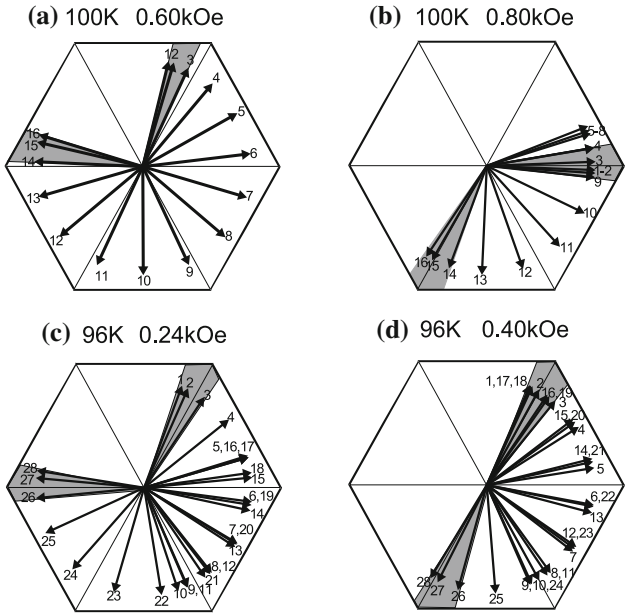

Fig. 1 Schematic representations of surface lock in phases of a 16 layers dysprosium film (**a** and **b**) and a 28 layers thick film (**c** and **d**) at selected values of temperature and external field strengths. The numbering in the panels indicate the atomic layers

Similar spin slip structures in bulk dysprosium have not been reported so far. Dysprosium is helimagnetic between the Curie temperature, around 80 K, to the Neel temperature of 180 K. Therefore, the anisotropy effects in Dy are much weaker. For Ho the hexagonal anisotropy $\left(K_6^6\right)$ at $T = 4.6$ K is 8.34×10^{-16} ergs/atom [7], whereas for Dy at $T = 80$ K the anisotropy is of the order of 4.42×10^{-17} ergs/atom [7]. At low temperatures, these materials are described by the following exchange energy parameters: for Dy, the nearest neighbor exchange energy is $J_1(Dy) = 6.072 \times 10^{-15}$ ergs/atom and the second neighbor exchange energy is $J_2(Dy) = -1.75 \times 10^{-15}$ ergs/atom [8], while for Ho the corresponding values are $J_1(Ho) = 1.602 \times 10^{-16}$ ergs/atom and $J_2(Ho) = -4.65 \times 10^{-17}$ ergs/atom [1]. Thus, Ho have anisotropy and exchange energies of the same order of magnitude at low temperatures, $K_6^6/J_1 = 5.3$. For Dy, at low temperatures, the anisotropy energy is three orders of magnitude smaller than the exchange energy, $K_6^6/J_1 = 7.3 \times 10^{-3}$.

The existence of commensurate phases in Holmium bulk samples, in which the magnetic structure has the same periodicity of the lattice, fulfills the requirement imposed by the hexagonal anisotropy. The exchange energy alone would lead to a regularly spiraling helix in the basal plane. The strength of the commensurable effect depends on how well the gain in anisotropy energy, with spins locked to near easy axis directions in the basal plane, counterbalances the increase in the exchange energy.

In thin rare earth helimagnetic films, the energy balance may be significantly modified due to the lower coordination of spins in the near surface region. The lack of second neighbors leads to a nearly ferromagnetic arrangement of the spins, with smaller values of the turn angles in the surface region. As a result, the alignment of surface spins with the anisotropy easy axis directions is favored.

In this paper, we report a theoretical study of the low-temperature phases of dysprosium thin films with thickness ranging from below to a few helical periods, showing that: (1) near the Curie temperature the helimagnetic phase is strongly modified, originating new phases with surface spins locked into the easy axis directions; (2) by raising the temperature up to about 120 K, new surface lockin patterns are formed, leading to plateaus in the magnetization curve; (3) the surface lockin patterns are tunable by modest external magnetic field values (a few hundred milli-oersteds); and (4) the number of surface lockin phases depend on thickness of the film.

Theoretical model

We investigate the magnetic phases in the temperature range from 80 to 200 K. The magnetic energy per unit area is given by:

$$\begin{aligned}\mathcal{H} &= J_1(g-1)^2 \sum_{n=1}^{N-1} \mathbf{J}(n)\cdot\mathbf{J}(n+1) \\ &+ J_2(g-1)^2 \sum_{n=1}^{N-2} \mathbf{J}(n)\cdot\mathbf{J}(n+2) \\ &+ \sum_{n=1}^{N} \left\{K_6^6 \cos(6\varphi_n) - g\mu_B \mathbf{J}(n)\cdot\mathbf{H}\right\}.\end{aligned} \tag{1}$$

where the magnetic moment per atom in the nth atomic layer is represented by a spin $\mathbf{S}(n) = (g-1)\mathbf{J}(n)$, where $\mathbf{J}(n)$ is the total angular momentum per atom. The spins are kept in the basal plane and φ_n is the angle with the easy axis. Each layer is exchange coupled with the first and second neighbor layers, and in Eq. 1 the first two terms represent the exchange energy, and the third term is the hexagonal anisotropy energy, whose temperature dependence is represented by fitting the experimental value of the anisotropy constant K_6^6 [9]. The last term is the Zeeman Energy, and the external field is along the a-axis ($\varphi = 0$) direction in the basal plane.

We use $S = 2.5$, $g = 4/3$, corresponding to a total angular momentum $J = 15/2$, and the nearest neighbor exchange is $J_1 = 44k_B$. The temperature dependence of the second neighbor exchange, $J_2 = -J_1/4cos\ \phi(T)$, is represented by using the experimental value [10] of the temperature dependence of the bulk turn angle $\phi(T)$. We use a

self-consistent local field algorithm [11], which allows calculating both the thermal average value of the total angular momenta ($\{<J_n>\}$: $n = 1,\ldots N$), and the orientation of the spins in each layer ($\{\varphi_n\}$: $n = 1,\ldots N$), taking into account the impact of the modifications of the exchange energy due to the reduced coordination near the surfaces.

Two features are worth noticing. At low temperatures, the surface spins align more easily with the easy axis of the hexagonal anisotropy than spins from inner atomic layers, due to the reduced exchange field in the surface region. For a given temperature, the formation of a surface lockin phase requires a modified helical state, in the inner layers, that fits the restrictions imposed by the surface spins along two easy directions of the basal plane anisotropy. Thus the number of lockin phases, and their temperature intervals, depend on the thickness of the dysprosium film.

Results and discussion

In Fig. 1, we show typical surface lockin patterns for a 16 atomic layers films (panels a and b) for low values of the temperature (100 K) and external field. The surfaces have spins along the easy axis, and by increasing the external field strength from 0.6 to 0.8 kOe, the lockin pattern change with a reduction in the modified helix number of spins, and a larger number of spins aligned parallel to the external field.

Most interestingly, for the 28 atomic layers film, the anisotropy lockin of surface spins leads to alternate helicity. Assuming a turn angle around $\pi/3$, the film would fit more than two helical periods. As shown in the panels (c) and (d), there are strong surface effects, at a temperature of 96 K, and external field values of 0.24 and 0.4 kOe, and the helical state in the inner atomic layers is forced to change helicity so as to fit the alignment of surface spins with the anisotropy easy axis directions.

In thinner films, with thickness smaller than the helix period, the number of surface lockin phases is small (see Fig. 2). For a 10 atomic layers films (thickness of 28 Å), with a small external field strength of 0.24 kOe, there is a transition around 90 K, from the ferromagnetic state to a surface lockin phase, which is stable in the temperature interval between 90 and 107 K. The magnetization displays a flat plateau, indicating small changes in the magnetic structure (shown in panel a). At 107 K, there is a transition to another lockin phase, symmetrically arranged with respect to the external field. This phase (shown in panel b), formed at a temperature interval of weaker anisotropy, displays larger thermal variation of the magnetization, and evolves continuously to the spin-flop-like phase of high temperature (panels c and d). In the inset, we show the bulk magnetization curve for the same value of the external field, for comparison purposes.

In Fig. 3, we show the angular profiles at each temperature (in steps of 1 K) and we focus on the nature of the surface lockin phases shown in Fig. 2. The profiles corresponding to the asymmetrical lockin phase (shown from 90 to 107 K), and the symmetrical lockin phase (shown from 110 to 120 K), have smaller temperature variation than the profiles in the spin-flop like phase, confirming the surface lockin nature of the low temperature phases ($T < 120$ K).

In Fig. 4 we show the sequences of phases for a 16 atomic layer film (44.8 Å), for an external field strength of 0.15 kOe. There are three surface lockin phases, associated to plateaus in the magnetization curve. Starting at $T = 85$ K, until 90 K, there is a surface lockin phase with a magnetic pattern similar to the low temperature phase of the 10 atomic layer film shown in Fig. 2, covering a $2\pi/3$ total angle inner helical region. The other two surface lockin phases cover wider angles ($4\pi/3$ and $5\pi/3$) in the helical region of the inner layers. At $T = 120$ K, the anisotropy effects no longer exist and a spin-flop like phase

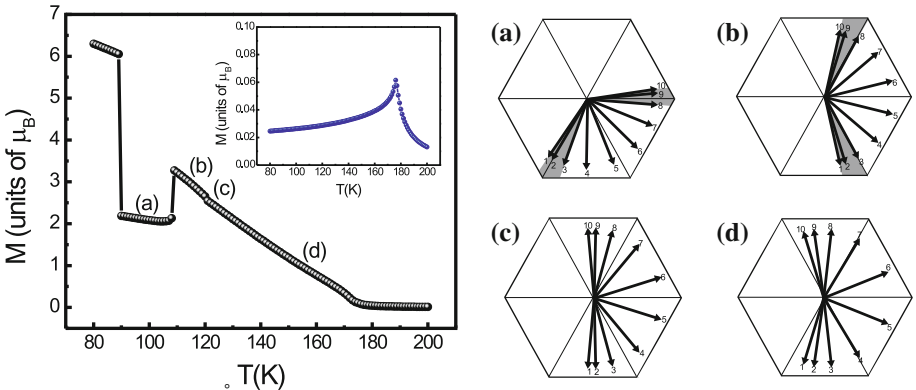

Fig. 2 Magnetic moment per atom for a 28 Å dysprosium thin film for an external field of 0.24 kOe along the *a*-axis. In the inset, we show the magnetic moment per atom for bulk dysprosium for the same value of the external field. The spin panels are schematic representations of the magnetic phases at selected temperatures, and the numbers indicate the atomic layers. See text for details

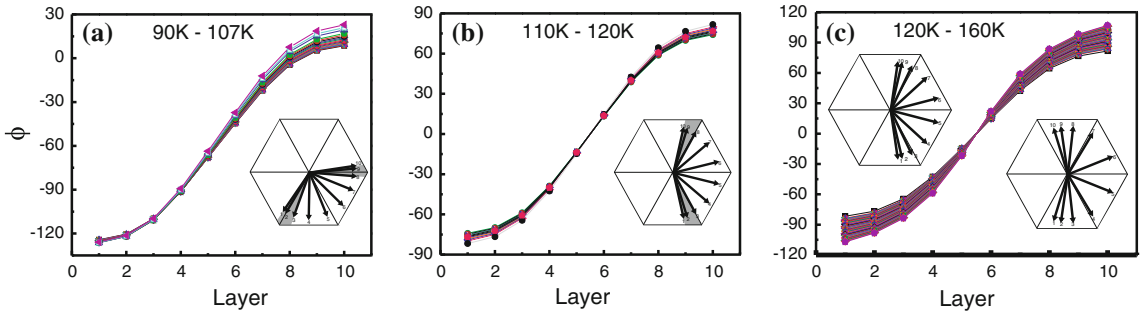

Fig. 3 Angular profiles at selected temperature intervals, indicating the locking of surface spins to the hexagonal anisotropy easy axis. The spin panels show the magnetic structure in each temperature interval

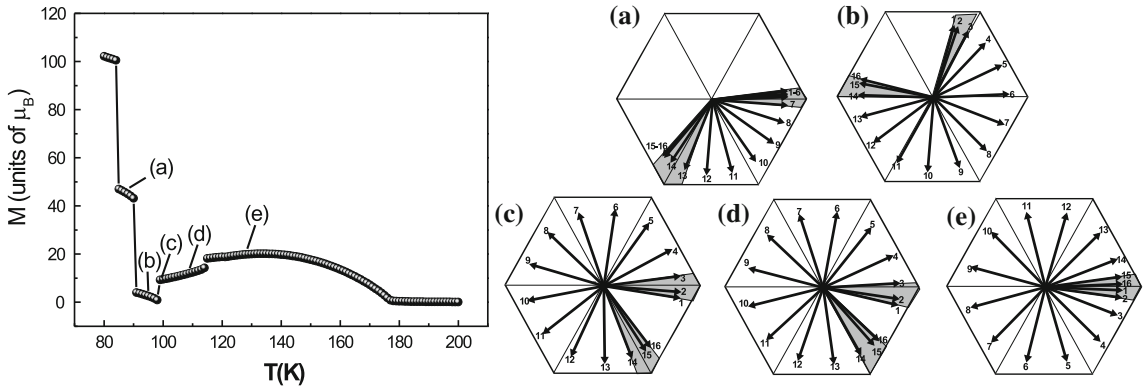

Fig. 4 Magnetic moment per atom for a 44.8 Å (16 atomic layers) dysprosium thin film for an external field of 0.15 kOe along the *a*-axis. The spin panels are schematic representations of the magnetic phases at selected temperatures, and the numbers indicate the atomic layers. See text for details

is formed, with the spins arranged symmetrically with respect to the external field direction.

In summary, we have shown that the reduced exchange energy of surface atoms allows the nucleation of large magnetic moment surface modified helical states, which are strongly affected by anisotropy and external fields. These surface lockin phases, have values of the magnetic moment per surface area much larger than the corresponding bulk values (Fig. 2), even though the external field strength (below 1 kOe) is much smaller than the constitutive dysprosium (exchange) fields.

Acknowledgements The authors acknowledge financial support from CAPES and FAPERN. The work of A.S.C. was supported by CNPq Grant No. 350773, the work of A.L.D. was supported by CNPq Grant No. 309676, and the work of F.H.S. Sales was supported by FAPEMA grant No. BD-00461/09.

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