Magnetic moment orientation and in depth distribution of dysprosium near the surface of DyCo$_{4.6}$ thin films determined by the analysis of their X ray circularly polarized absorption Dy $M_5$ spectra

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We have investigated the RE atomic distribution and its magnetic moment orientation at the region near the surface of DyCo$_{4.6}$  ferromagnetic amorphous films with perpendicular magnetic anisotropy (PMA), XMCD spectroscopy of the films at the Dy $M_5$ and Co $L_{2,3}$ edges using total electron yield (TEY) detection was performed at 2 K and 300 K temperatures, and at sample orientations ranged from 0° to 70° with respect to the magnetic easy axis. The measurements showed an apparent partial decoupling between the cobalt and dysprosium magnetic sublattices. At RT, the magnetic moment per atom of dysprosium was below the minimum value expected if all dysprosium moments were AF coupled to cobalt. At 2 K, the cobalt sublattice presented a stronger magnetic anisotropic behavior than the dysprosium sublattice. A detailed analysis of the circularly polarized spectra of the Dy $M_5$ edge, based on the deconvolution of the spectra in their related parallel, antiparallel and transverse to $J_z$ spectral components, demonstrates that the spectra are composed by dysprosium with different magnetic moment distributions. The fit of the Dy $M_5$ spectra using the $J_z$ spectral components evidenced a gradation of dysprosium concentration due to segregation at the region probed by TEY. The topmost layer was magnetically uncoupled from cobalt. At RT, 25% of the dysprosium magnetic moments in the under layer were found averaged oriented in the same direction as cobalt. The expected weak magnetic coupling of these dysprosium atoms to cobalt should explain the surprisingly lower magnetic anisotropy of the dysprosium sublattice compared to that of cobalt probed by TEY at 2 K.

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I. INTRODUCTION

RE-TM alloys are well known magnetic materials since decades ago [1, 2], constituting a key component in a variety of industrial and technological applications. A renewed interest in these materials has grown because their richness in magnetic behaviors and their relative flexibility for tailoring their properties to fit in specific magnetic applications and devices. RE-TM alloys are present in spring magnets [3–5], magnetic topological formations [6–8], spin wave functional devices [9] and all optical magnetic switching [10–12]. Their use is also favored by its simple thin film preparation process which can be done at RT.

The extraordinary magnetic properties of these alloys are based on the strength of the TM-TM exchange coupling, the high magnetic moment of the RE, and their RE-TM indirect exchange coupling interaction. The high orbital moment of its unquenched 4f orbital and the high energy of its spin-orbit coupling, makes the RE acts as a strong and localized magnetic moment whose orientation depends on the interatomic exchange and the crystal field at its particular local atomic environment. These two interactions are mainly provided by their TM neighbors whose RE-TM interaction energies overwhelms those of RE-RE. Since the RE-TM magnetic exchange coupling is antiferromagnetic in spin, RE-TM alloys are ferromagnetic for heavy REs like dysprosium. This interaction is usually treated as a molecular field whose intensity is calculated to be of the order of 200 T for crystalline DyCo$_5$ [13].

The energy of this interaction is, at least, one order of magnitude smaller than the TM-TM exchange. These range of values are comparable to $k_B T$ energies, giving rise to different magnetic configurations as a function of temperature and RE concentration. A characteristic parameter that define these ferrimagnetic alloys is their compensation temperature, $T_{Comp}$, the temperature where the magnetic moment of the RE and TM sublattices cancel to each other.

Giving the radical different magnetic behavior of the RE and the TM, the understanding of the magnetism of these alloys requires a precise characterization of the magnetic moment and anisotropy of the RE and TM sublattices separately. The perfect technique to do so is XMCD spectroscopy. The magnetic properties of the RE and TM sublattices can be studied separately by tuning the incident circularly polarized X rays to the corresponding absorption edges of the RE ($4d_{4,5}$, probing its 4f orbital) and the TM ($L_{2,3}$, probing its 3d orbital) [14–16]. This is actually the technique of choice for the study of RE-TM alloys in the form of thin films because it allows ex-situ sample preparation and it can be sensitive to regions at different depths of the sample by changing the way the X ray absorption spectra is detected. Surface sensitivity is attained using TEY...
these spin flop transitions.

The purpose of the experiment presented in this work is to improve the spectroscopical tools to understand the magnetic behavior of the TM and the RE sublattices, their mutual interaction and their magnetic anisotropy using the information extracted from the X ray absorption spectra of the RE atoms, and considering their possible inhomogeneous distribution in depth at the region probed by TEY.

For this experiment, we prepared DyCo thin films anisotropically uniaxial with high PMA energies. Their anisotropy fields at 2 K, $H_K$, were well above the range of available field intensity in the experiment. Measurements were done at 2 K and RT. These two temperatures were distant enough from the $T_{Comp}$ of the alloys to avoid possible spin flip (and flop) effects. Also, it permitted to measure in the regions where each of the sublattices were magnetically dominant (cobalt at RT, and dysprosium at 2 K). At 2 K, thermal disorder was reduced to a minimum.

The ferrimagnetic character of DyCo alloys permits to detect dysprosium atoms magnetically uncoupled from cobalt since they will be oriented in the opposite direction than their counterpart exchange coupled dysprosium atoms at temperatures above $T_{Comp}$ (cobalt sublattice magnetically dominant). Their detection requires a deconvolution of the RE $M_{4,5}$ spectra in their parallel, antiparallel and transverse components of $J_z$ which are specially well defined in dysprosium. This deconvolution has been presented before in DyCo films [24], but it was not linked to the moment orientation of the dysprosium magnetic moments as it is done and explained in detail in this work. Also, we use the XMCD spectra to deconvolve these components from the circularly polarized spectra, instead of relying completely on their theoretically calculated shapes, as the mentioned work tried. Actually, this is the first time that this kind of analysis is applied in a certainly complex system like the studied in this work.

We show the potential of this novel technique in the study of the interaction between RE and cobalt atoms at the region probed by TEY, by measuring at different orientation angles under a strong PMA. This technique is able to detect the presence of a RE segregated layer that affects in a significant way the measured moment of the RE. The presence of this layer seems intrinsic to the thin film growth in RE-TM alloys. Even removing the effect of this layer, the proportion of dysprosium atoms directly engaged in the PMA anisotropy of the alloy probed by TEY was not majority, possibly due to the extended thickness of the cobalt depleted layer caused by the dysprosium segregation.

The paper is organized as follows: First, sample preparation and experimental details for XMCD data acquisition are described. Next, after showing the VSM magnetometry characterization of the measured films, the XMCD experimental results for each elemental sublat-
tice are presented and discussed separately. The following section is dedicated to explain how the deconvolution of the Dy $M_5$ spectra is made, with the calculation details shown in an Appendix section. After this explanation, a model to fit the Dy $M_5$ of the measured samples using the deconvolved spectral components is proposed, which consisted in two layer with different dysprosium moment distributions. The results of the fits are presented and discussed. The final section are the conclusions of the work.

II. EXPERIMENT

The two studied DyCo thin films were prepared at RT by magnetron sputtering at a base pressure of $10^{-8}$ mbar and $10^{-3}$ mbar Ar pressure. They were grown onto silicon wafer substrates using two separate magnetron guns set at normal (cobalt gun) and at 30° angle incidence (dysprosium) with respect to the normal to the sample. Dysprosium and cobalt concentrations were calibrated using a quartz balance. The deposition method was different for each sample. Sample called DCC was prepared by codeposition of cobalt and dysprosium. Sample called DCM was grown by the alternate deposition of cobalt and dysprosium layers. The nominal thickness of the cobalt and dysprosium layers was 4.9 Å and 2.8 Å respectively. The topmost deposited layer was cobalt. This second preparation method was intended as a way to estimate the importance of the interdiffusion between the elements forming the alloy in their structure and magnetic properties. All the samples were protected with a 20 Å thick aluminum capping layer. The mean atomic concentration in the alloys was determined by electron induced fluorescence spectroscopy. The difference in concentration between both films was relatively small. Sample DCC contained more dysprosium (DyCo$_{4.4±0.05}$) than the DCM film (DyCo$_{4.6±0.05}$). Their nominal thickness was 35 nm.

X ray circularly polarized absorption spectra were obtained at the HECTOR endstation [25] of the BOREAS BL-29 beamline at the ALBA synchrotron using total electron yield detection. HECTOR has a cryomagnet that can apply up to ±6 T along the x-ray beam direction at different sample orientations, which in our case ranged from normal incidence (0°) to near grazing incidence (70°). The cryomagnet works at ultra-high vacuum conditions (pressure within the $10^{-10}$ mbar range). A liquid He cryostat permits to fix the sample temperature between 2 K and 350 K. Circularly polarized light was produced by an APPLE II elliptical undulator. Each XMCD spectra was the result of 4 spectra taken at opposite circular polarization helicities and magnetic field orientations.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Magnetometry

Both samples presented a PMA even at RT. Table I shows the magnetization in remanence of samples DCC and DCM measured using a Vibrating Sample Magnetometer (VSM). The values were obtained after magnetic saturation of the films with a field of 9 T at 5 K applied normal to their plane [26]. The same table gives the compensation temperatures, $T_{\text{Comp}}$, of the alloys and their coercive fields at 2 K and RT. $T_{\text{Comp}}$ temperature was smaller in sample DCM (90 K) than in sample DCC (120 K), in agreement with their different RE concentration, and with the expected by comparing with the reported by others in DyCo alloys of similar concentrations [5, 20], assuming a linear relationship between $T_{\text{Comp}}$ and the atomic concentration of the alloy [13]. The higher cobalt concentration in sample DCM causes a marked lower magnetic remanence at 10 K than in sample DCC. This explains the large difference between the coercive fields measured at 10 K, higher in sample DCM (3.5 T) than in sample DCC (1.9 T). However, sample DCM has a higher $H_C$ at RT, when its $M_S$ is higher. X ray reflectometry shows rougher surfaces in DCM than in DCC thin films, suggesting that the larger $H_C$ of the DCM thin film is probably caused by its higher density of domain wall pinning defects. The coercive fields of both samples were notoriously higher than the reported in DyCo$_4$ by others [5, 20], with $H_C$ values lower than 1 T at a similar decrement ($\Delta T=150$ K) in temperature from their $T_{\text{Comp}}$ (250 K). This might account for a higher PMA energy and/or domain wall pinning defects density in our samples. None of the samples presented triple hysteresis loops at fields below 6 T at temperatures close to their $T_{\text{Comp}}$ as the reported in samples of similar concentration and thickness [3, 20].

| Sample | $T_{\text{Comp}}$ (K) | Temperature (K) | $M_R$ (emu·cm$^{-3}$) | $H_C$ (T) |
|--------|----------------------|----------------|----------------------|----------|
| DCC    | 90                   | 2              | 125                  | 1.9      |
|        | 300                  |                | 250                  | 0.2      |
| DCM    | 125                  | 2              | 75                   | 3.5      |
|        | 300                  |                | 275                  | 0.4      |

B. XMCD hysteresis loops and magnetic moments

Figure 1 shows the hysteresis loops of sample DCC at RT and 2K for the cobalt and dysprosium sublattices obtained by measuring the intensity of the Co $L_3$ peak and the Dy $M_5$ peak respectively. Their change in shape
with field orientation at RT (figures 1(a) and (b)), from squared at 0° orientation to "S" shaped at 70° field orientation, shows that the samples had a PMA, as observed by VSM magnetometry. The loops of the cobalt and dysprosium sublattices are nearly identical. A small decoupling between both sublattices is only noticeable at high fields. The magnetization of cobalt remains constant at high fields up to 6 T whereas that of dysprosium seems to decreases steadily with the increasing field from zero field.

A similar effect, although much more pronounced than in the present case, has been observed in NdCo films [21]. The effect was demonstrated to be caused by a portion of paramagnetic Nd. A similar explanation would justify the observation in the present case since, at this temperature, any dysprosium magnetically uncoupled to cobalt would be oriented towards the field direction as cobalt, reducing its total magnetization.

At 2 K, only the hysteresis loop of the dysprosium sublattice in sample DCM was taken at near the plane field orientation (70°), shown in figure 1(c). The shape of the loop shows that at 6 T the films were far from being magnetically saturated, indicating that the PMA energy of the samples was strongly increased at this temperature. The lack of coercive field in the loop shows that the measured magnetic moment experience a progressive rotation with the applied field intensity. The loop shows a shape asymmetry between the positive and negative branches.

C. XMCD Cobalt

The magnetic moments of cobalt were deduced from their $L_{2,3}$ spectra. The method used to extract the cobalt absorption coefficient to correctly apply the XMCD sum rules [27, 28], which considers saturation effects, is fully described in [24]. The number of holes was calculated comparing their unpolarized absorption spectra with that of a pure cobalt reference sample deposited in similar conditions than the rest of the films. The number of holes for this reference sample was set to the tabulated for pure cobalt, 2.49, yielding a magnetic moment at 2 K (1.79±0.02$\mu_B$) similar to the measured by others (1.77 $\mu_B$) [29]. As it can be observed from figures 2(a) and 2(b), the shape of the spectra was almost identical to the cobalt reference except in their intensity which was lower and almost independent of the sample orientation angle or temperature. Their number of holes reduced to approximately 91% those of pure cobalt. The total magnetic moment of the samples together with the related orbital and spin components and orbital to spin ratios are shown in table 1. The total magnetic moment of the two alloys at 2 K and normal incidence (easy magnetic axis) was similar within the error, of the order of 1.33±0.03$\mu_B$. This value is in agreement with the expected from those observed in NdCo alloys as a function of the number of holes [21], and they would correspond to a RE concentration higher than the nominal, of the order of DyCo$_{3.5}$. The magnetic moment obtained at RT (only measured at normal incidence in DCC) was practically the same than at 2 K, 1.34±0.03$\mu_B$.

The effective spin and orbital magnetic moments of cobalt changed with the field orientation angle at RT. The effective spin moment $m_{eff}$ obtained from the sum rules is the sum of the spin moment $m_s$ and the dipolar moment $m_T$ which is angle dependent:

$$m_{eff}^{\theta} = m_s^{\theta} - 7m_T^{\theta}.$$ (1)

The dipolar moments at normal and in plane field orientation are -0.013 $\mu_B$ and 0.004 $\mu_B$, respectively; assuming a zero value for the dipolar moment when it is angle averaged [14, 30]. The orbital anisotropy is also oriented in the plane, $\Delta m_L = m_{0^\circ} - m_{70^\circ} = -0.02$. The anisotropy of this term is usually linked to the anisotropy in the orbital moment [14, 31, 62] as observed in other 5d-3d systems like Au/Co/Au [63]. This means that the cobalt sublattice of the alloys have an in plane magnetic anisotropy at RT, in coincidence with what we observed in NdCo alloys [21]. Actually, the in plane anisotropy of the cobalt sublattice is the expected for the oblate shape of the Nd and Dy 4f orbital, which is the one that generates the expected crystal field orientation for the RE out of plane anisotropy. Nevertheless, the anisotropy of the observed values is small, with values below the experimental error. This would discard cobalt as the origin of the PMA of the alloy at RT.
The total magnetic moment measured at 2 K is displayed in figure 2(a) as a function of the field orientation angle. It decreases in both samples with increasing field angle orientations because of the uncompleted magnetic saturation of the alloy at the 6 T applied field. Due to the higher strength of the exchange coupling between cobalt atoms compared to the indirect exchange interaction with the dysprosium moments, the orientation angle dispersion of their magnetic moments is expected to be much smaller than those of the dysprosium atoms. The dashed curve displayed in figure 2(a) is the fit of the averaged magnetic moments of the two samples using a cosine function plus a constant. The constant could be interpreted as the portion of the cobalt magnetic moment that it is oriented parallel to the field. This constant is only 10% of the total intensity, indicating that most of the cobalt magnetic moments were fixed at the easy axis direction.

The orbital magnetic moment at 0° field orientation was bigger than at RT, increased from 0.12 μB to 0.15 μB. Both samples showed similar values. For the rest of the field orientation angles, only the orbital to effective spin moments ratio can be compared because the uncompleted magnetic moment saturation. This ratio is the highest at 0° and it decreases significantly in both samples, from 0.14 at normal orientation, to 0.09 (0.10 at 70° in sample DCC). This indicates that the perpen-

| Sample | T | Field Orientation Angle | 0° | 20° | 45° | 70° |
|--------|---|-------------------------|----|-----|-----|-----|
| DCC    | RT | m_s         | 1.22 | -   | -   | 1.11 |
|        |    | m_o         | 0.12 | -   | -   | 0.16 |
|        |    | m_tot       | 1.34 | -   | -   | 1.25 |
|        |    | m_o/m_s     | 0.09 | -   | -   | 0.12 |
|        | 2 K | m_s         | 1.17 | 1.15 | 0.91 | 0.41 |
|        |    | m_o         | 0.16 | 0.10 | 0.08 | 0.06 |
|        |    | m_tot       | 1.33 | 1.25 | 0.99 | 0.50 |
|        |    | m_o/m_s     | 0.14 | 0.09 | 0.09 | 0.10 |
| DCM    | RT | m_s         | -    | -    | -   | 1.08 |
|        |    | m_o         | -    | -    | -   | 0.16 |
|        |    | m_tot       | -    | -    | -   | 1.24 |
|        |    | m_o/m_s     | -    | -    | -   | 0.15 |
|        | 2 K | m_s         | 1.15 | 1.10 | 0.79 | 0.54 |
|        |    | m_o         | 0.15 | 0.10 | 0.06 | 0.04 |
|        |    | m_tot       | 1.30 | 1.20 | 0.85 | 0.58 |
|        |    | m_o/m_s     | 0.13 | 0.09 | 0.08 | 0.09 |
| YCC    | 2 K | m_s         | 1.28 | -    | 1.24 | 1.28 |
|        |    | m_o         | 0.14 | -    | 0.19 | 0.16 |
|        |    | m_tot       | 1.42 | -    | 1.43 | 1.44 |
|        |    | m_o/m_s     | 0.11 | -    | 0.16 | 0.13 |
| YCM    | 2 K | m_s         | 1.26 | -    | 1.18 | 1.22 |
|        |    | m_o         | 0.07 | -    | 0.18 | 0.17 |
|        |    | m_tot       | 1.33 | -    | 1.36 | 1.39 |
|        |    | m_o/m_s     | 0.06 | -    | 0.15 | 0.14 |

FIG. 2. X ray absorption (XAS) spectra obtained at the Co L_{2,3} edge of a pure cobalt thin film compared with the spectra of cobalt in (a) DCC and (b) DCM thin films taken at 2 K and 0° (blue), 20° (green), 45° (orange), and 70° (red) beam incidence angles (magnetic field orientation angle) with respect to the normal to the sample. The most intense spectrum (black line) is from pure cobalt. In the inset, the XMCD spectra taken at different orientation field angles, using the same code color.

FIG. 3. Total magnetic moment of cobalt (solid dots) and dysprosium (empty dots) in samples DCC (red dots) and DCM (blue dots) measured at 2 K. The black dashed line in (a) is the fit to a cosine function plus a constant. Black dots an triangles are cobalt moments measured at RT. The estimated error of the measurement is the width of the dots.

| Sample | T | Field Orientation Angle | 0° | 20° | 45° | 70° |
|--------|---|-------------------------|----|-----|-----|-----|
| DCC    | RT | m_s         | 1.22 | -   | -   | 1.11 |
|        |    | m_o         | 0.12 | -   | -   | 0.16 |
|        |    | m_tot       | 1.34 | -   | -   | 1.25 |
|        |    | m_o/m_s     | 0.09 | -   | -   | 0.12 |
|        | 2 K | m_s         | 1.17 | 1.15 | 0.91 | 0.41 |
|        |    | m_o         | 0.16 | 0.10 | 0.08 | 0.06 |
|        |    | m_tot       | 1.33 | 1.25 | 0.99 | 0.50 |
|        |    | m_o/m_s     | 0.14 | 0.09 | 0.09 | 0.10 |
| DCM    | RT | m_s         | -    | -    | -   | 1.08 |
|        |    | m_o         | -    | -    | -   | 0.16 |
|        |    | m_tot       | -    | -    | -   | 1.24 |
|        |    | m_o/m_s     | -    | -    | -   | 0.15 |
|        | 2 K | m_s         | 1.15 | 1.10 | 0.79 | 0.54 |
|        |    | m_o         | 0.15 | 0.10 | 0.06 | 0.04 |
|        |    | m_tot       | 1.30 | 1.20 | 0.85 | 0.58 |
|        |    | m_o/m_s     | 0.13 | 0.09 | 0.08 | 0.09 |
| YCC    | 2 K | m_s         | 1.28 | -    | 1.24 | 1.28 |
|        |    | m_o         | 0.14 | -    | 0.19 | 0.16 |
|        |    | m_tot       | 1.42 | -    | 1.43 | 1.44 |
|        |    | m_o/m_s     | 0.11 | -    | 0.16 | 0.13 |
| YCM    | 2 K | m_s         | 1.26 | -    | 1.18 | 1.22 |
|        |    | m_o         | 0.07 | -    | 0.18 | 0.17 |
|        |    | m_tot       | 1.33 | -    | 1.36 | 1.39 |
|        |    | m_o/m_s     | 0.06 | -    | 0.15 | 0.14 |
dicular magnetic anisotropy of the alloy involves either an anisotropy in the orbital moment of cobalt in the same direction of the easy axis or a larger spin component at angles far from the easy axis.

In order to check that if this effect is caused by the magnetic interaction at cobalt with the RE, YCo films deposited in similar conditions of concentration, film thickness and deposition process were measured by XMCD. The chemical interaction of Yttrium is equivalent to that of RE, with a nearly empty 4f band, but without an occupied 4f orbital, i.e., with no 4f magnetic moment. Actually, their RE-Co phase diagram is very similar [34, 35]. Therefore, the atomic structure of the YCo alloy should be similar to that of DyCo. The magnetic moments of cobalt in YCo measured at RT (normal orientation only) and 2 K are displayed in table 1. The values are about 10 % smaller in the multilayer than in the continuous film and they increase in both films when temperature goes from RT to 2 K, in contrast to the observed in the DyCo alloys where there was almost no difference. The YCo alloys were magnetically soft and their magnetic anisotropy was in the plane. Their orbital to effective spin ratios were similar to the found at normal orientation in the DyCo films. But they did not show the strong decrease in their value with the increasing orientation angle as the observed in DyCo. This fact, together with the cosine like variation in the total magnetic moment of cobalt in the DyCo films, indicates that their probed cobalt atoms are mostly located at magnetically anisotropic environments. By comparison with the YCo alloy, the magnetic anisotropy found in the cobalt sublattice of the DyCo films must be induced by the dysprosium atoms. It would be expected, therefore, a similar angle variation in the magnetic moment of the dysprosium sublattice as the found in cobalt.

D. XMCD Dy

In dysprosium, the most relevant changes in the spectra occur in the Dy $M_5$ edge. Figure 4 shows the Dy $M_5$ taken in sample DCM at RT and 2 K, for the two circular polarizations, at different angles. Sample DCC showed similar changes. The Dy $M_5$ spectral shape is defined by three intense peaks, marked in the figure as peaks AP, TR and PP, which are located at photon energies 1294.5 eV, 1296.7 eV, and 1298.6 eV, respectively. Their relative intensity changes with the polarization sign and with the field orientation angle. At RT, the changes with the angle are small, but they are very evident at 2 K. At this temperature, peak PP is the most intense at $C^-$ polarization and normal incidence. Within the same polarization, peak PP decreases in intensity as the field orientation angle (incident X ray beam angle) increases. The opposite is observed using $C^+$ polarization. In this case, peak marked AP is the most intense at normal incidence. For both polarizations, peak TR increases in intensity when the incident angle increases. As it will be shown, peaks PP, AP and TR are associated to electronic transitions to states where the magnetic moment of the related 4f orbital is parallel, antiparallel or transverse to the circularly polarized x ray beam wave vector, respectively. It is important to notice that, at RT, peak PP is the most intense also in $C^-$ polarization. This is opposite to what happen at 2 K, specially at 70°, where the measured magnetic moment of dysprosium is the lowest and comparable to the measured at RT.

The XMCD spectra of Dy $M_{4.5}$ edge were analyzed following the process explained in [21]. Table III shows the magnetic moments of dysprosium in each of the samples at RT and 2 K. The relative weakness of the AF interaction with cobalt and the structural disorder of the alloy makes the magnetic moment of the dysprosium atoms to be dispersed in its orientation with respect to the cobalt magnetic moment forming what is called an aspermagnet [36]. The moment orientation of the RE atoms when the alloy is magnetized in the easy axis is usually taken as uniform and symmetrically distributed around this axis within a cone with a half opening angle $\theta_C$ ($\theta_C = \pi/2$ represents the distribution of moments in a semisphere). For field orientations away from the easy axis, a dispersion in the shape and opening angle of the cone is expected. In this case, we assume that the resulted distribution of the dysprosium can be approximated by a cone that has its symmetry axis deviated from the field direction an angle $\varphi_C$. The magnetic moment measured is the averaged sum within the cone of the magnetic moment component along the field orientation angle. At the easy axis, this sum is $M_{dy} \cos^2 \theta_C/2$, where $M_{dy}$ is 9.8 $\mu_B$, the dysprosium magnetic moment of its 4f orbital. The magnetic anisotropy would then change the value of the measured magnetic moment by opening the cone angle and changing its symmetry axis orientation.

At RT, the dysprosium magnetic moment has almost

![FIG. 4. Dy $M_5$ spectra of sample DCM obtained at RT (spectra at (a) and (b)) and 2 K (spectra (c) and (d)). The spectra taken with right handed circular polarization are (a) and (c). (b) and (d) are the spectra taken using left handed circular polarization. RT spectra were taken at 0° (dark blue line) and 70° (red line) incident angles. 2 K spectra were taken at 0° (dark blue line), 20° (light blue line), 45° (orange line) and 70° (red line) incident angles.](image-url)
some of them, the exchange coupling to cobalt should be dysprosium atoms with their moment orientation opposite to that of those AF coupled to cobalt. Theoretically, calculations estimated that the averaged magnetic moment of Dy decays to half its value at about 300 K in crystalline DyCo5 [37]. The lower values observed in the present case should have to do with the intrinsic disorder of the measured alloys, with dysprosium atoms distributed in different atomic environments where, in some of them, the exchange coupling to cobalt should be weaker than in crystalline DyCo5 or possibly nonexistent. The analysis of the Dy M5 spectra exposed in a later section searches to understand how these dysprosium atoms are distributed through the depth proved by TEY and how this can affect the TEY measurements including the possible presence of segregated dysprosium at the interface with the aluminum capping layer.

Figure 3 (right side scale) displays the variation of the dysprosium magnetic moment as a function of the field orientation angle at 2 K compared to that of cobalt. Both samples have similar values which decay with the angle at a slower pace than in the cobalt sublattice. This magnetically less anisotropic behavior of dysprosium is somehow contrary to what expected: the magnetic moment of the RE should be, compared to cobalt, the one fixed with the highest energy to the easy axis since the PMA in these alloys must stems from it, as deduced from the analysis done in the previous section (III C). The total magnetic anisotropy of the alloy should depend on the distribution of the crystal field orientation at the RE sites [2], and its influence on the magnetic moment of the bonding TMs at each RE site. Following the single ion anisotropy model [2], both crystal field and RE-TM indirect exchange are expected to be tightly related. Therefore, the RE environments that most contribute to the magnetic anisotropy of the alloys should be strongly exchange coupled to the TM as well.

The magnetically less anisotropic character of the measured dysprosium sublattice indicates that only a portion of the dysprosium atoms probed by TEY should be active magnetic anisotropy generators. The magnetic field felt by dysprosium is the sum of the external applied field and the molecular field, which is mainly provided by the interatomic exchange interaction with cobalt atoms, whose strength can be estimated in, at least, more than 150 T [21]. As it has been observed, the rotation of the cobalt moment at grazing orientations is small. Therefore, the effective molecular field felt by the dysprosium should be closer to that of the applied field at those sample orientation angles, meaning that a portion of the probing dysprosium should be somehow decoupled or poorly coupled to cobalt, as stemmed from the analysis done at RT. One of the challenges in the analysis of the Dy M5 spectra, shown in the next sections, is to determine the location of the dysprosium which are apparently weakly interacting to cobalt.

A way to compare the magnetization measured in the bulk with the magnetization measure near the surface (XMCD using TEY) is to evaluate how these magnetic moments found by XMCD fit with the deduced from VSM, shown in table 7. When the magnetic moment of the cobalt used to calculate the total magnetization of the alloy is the deduced by XMCD, 1.33 µB, i.e., the half value of its total magnetic moment. This means that there should be dysprosium atoms with their moment orientation opposite to that of those AF coupled to cobalt. Theoretical calculations estimated that the averaged magnetic moment of Dy decays to half its value at about 300 K in crystalline DyCo5 [37].

| Sample | T  | Field Orientation Angle | 0° | 20° | 45° | 70° |
|--------|----|-------------------------|----|-----|-----|-----|
| DCC    | RT | m^s_1 | -1.92 | - | - | -1.92 |
|        |    | m_o  | -1.42 | - | - | -1.41 |
|        |    | m_{Tz} | -0.09 | - | - | -0.10 |
|        |    | m_{tot} | -2.78 | - | - | -2.74 |
|        | 2 K | m^s_1 | 2.78 | 2.51 | 2.19 | 1.46 |
|        |    | m_o  | 4.19 | 3.81 | 3.42 | 2.23 |
|        |    | m_{Tz} | 0.26 | 0.23 | 0.19 | 0.13 |
|        |    | m_{tot} | 8.18 | 7.43 | 6.67 | 4.35 |
| DCM    | RT | m^s_1 | - | - | - | -0.91 |
|        |    | m_o   | - | - | - | -1.44 |
|        |    | m_{Tz} | - | - | - | -0.07 |
|        |    | m_{tot} | - | - | - | -2.81 |
|        | 2 K | m^s_1 | 2.84 | 2.64 | 2.14 | 1.61 |
|        |    | m_o   | 4.21 | 3.93 | 3.17 | 1.61 |
|        |    | m_{Tz} | 0.28 | 0.26 | 0.21 | 0.15 |
|        |    | m_{tot} | 8.2 | 7.66 | 6.19 | 4.73 |

no difference at both orientations, as expected from the hysteresis loops. The PMA of dysprosium has not enough energy to distort its magnetic moment distribution cone at RT. There is not a significant variation between the values obtained in both samples, which are about -2.8 µB (sample DCM only measured at 70° field orientation). This quantity is small. The lowest magnetic moment that can be measured assuming that all dysprosium atoms are AF coupled to Co, i.e., uniformly distributed within a cone of half opening angle $\theta_C = \pi/2$ (semisphere), is $M_{Dy}/2 = -4.9 \mu_B$, i.e., half the value of its total magnetic moment. This means that there should be dysprosium atoms with their moment orientation opposite to that of those AF coupled to cobalt.  

TABLE III. Effective spin moment, $m^s_1$, orbital moment, $m_o$, dipole moment of spin, $m_{Tz}$, and total magnetic moment, $m_{tot}$, of dysprosium obtained by XMCD in samples DCC and DCM. All moments are given in $\mu_B$ units. The estimated error bars is of 2% in $m^s_1$, $m_o$ and $m_{Tz}$. $m_{Tz} = m_o \frac{m^s_1}{m_o} \left[ \frac{m^s_1}{m_o} - m_o \right]$ [15], using the theoretical value of $\frac{m^s_1}{m_o} = 0.475$ [16], $m_{tot} = m_o + (m^s_1 - 6m_{Tz})$.
segregation. If that was happening in our samples, their cobalt and dysprosium moments in the bulk should be
higher than the measured by XMCD. For instance, if the cobalt moment in the bulk raised to 1.50 $\mu_B$, the related
dysprosium moment should be 4.7 $\mu_B$, which is almost 2 $\mu_B$ higher than the measured by XMCD. This would
set the surface at a higher $T_{\text{Comp}}$ than in the bulk. But none of our samples showed wing side hysteresis loops at
7 T at temperatures near the $T_{\text{Comp}}$, as the reported in [5, 20] in alloys of similar thickness and concentration.

The $T_{\text{Comp}}$ of a ferrimagnetic RE-TM alloy essentially depends on the magnetic exchange strength between the
RE and the TM atoms. If the segregated RE atoms at the region near the surface do not bond tight to cobalt,
their exchange interaction will be weaken. The effect, in terms of magnetic moments, will be similar to an increase of
the $T_{\text{Comp}}$ because, above this temperature, there will be dysprosium moments that will not be AF coupled to
cobalt, reducing the total moment of the dysprosium sub-
lattice. Below $T_{\text{Comp}}$, the effect will be the opposite, i.e.,
they will add up in moment to the moment of the AF
coupled-to-cobalt dysprosium. This is not an unlikely situation regarding the magnetic moments of dysprosium
measured by XMCD, which are well below the expected if all RE atoms were AF coupled to cobalt. Moreover, if
RE segregation occurs because it favors RE-RE metal-
lic bonding. To demonstrate the presence of these two kinds of RE atoms requires a technique able to detect
them. The next section shows a way to do it in a quan-
titative way by analyzing the circularly polarized Dy $M_5$
spectra. Actually, dysprosium is specially well adapted for this kind of analysis, although it should be applicable
to most of the REs, with the exception of those with $J = 0$ like Gd$^{3+}$.

IV. SPECTRAL ANALYSIS METHOD FOR DY $M_{4,5}$ SPECTRA

Like in most of the RE, the electrons in the 4f orbital
dysprosium are well screened by the valence band or-
bitals and behave as in an isolated atom. Actually, the
Dy $M_{4,5}$ edge spectrum, which involves electronic transi-
tions form the 3d to the 4f orbital, is practically insen-
sitive to the dysprosium chemical environment and it is well fitted by calculating it using only intratomic interac-
tions[35, 39]. The relative intensity of these transitions can be notably influenced by the bonding with other magnetic atoms through its indirect exchange mag-
netic interaction and the resulting crystal field by breaking
its degeneracy in the $J_z$ momentum. Their relative
weakness permits to treat them as perturbations whose
most important effect is to orient the magnetic moment of
4f orbital along a specific direction, i.e., to break the
degeneracy of the quantum number $M$ of the 4f angular
moment component $J_z$.

Therefore, to a good approximation, only the intensity,
but not the shape, of the Dy $M_{4,5}$ lines are modulated by
the orientation of the total angular moment of the 4f or-
bital with respect to the direction of the polarized X ray.
The allowed transitions in the dipole approximation are
those in which $\Delta J = 0, \pm 1$. If light is circularly polar-
ized, only $\Delta m = \pm 1$ transitions are allowed. Therefore,
if the beam is perfectly oriented parallel (antiparallel) to
the magnetic moment of the 4f orbital, the resulting
$M_{4,5}$ spectrum will be built with only those excitations
where $\Delta J = 0, \pm 1$ and $\Delta m = 1(-1)$. We will call to
this spectral components PP if $\Delta m = 1$ and AP when
$\Delta m = -1$. The circular dichroism spectrum, XMCD,
is the difference between these two spectrum. The sum
rules related to these transitions gives rise to the XMCD
sum rules that permits to determine both the magnetic
spin and orbital moment of the 4f orbital. $\Delta m = 0 tran-
sitions occurs when the light is linearly polarized along
the magnetic moment direction. This spectral compo-
nent is not present in the XMCD spectrum because de-

deps on $\langle M^2 \rangle$ [39]. We will call to this component TR.

The excitations of all the three spectral components
PP, AP and TR occurs when the 4f magnetic moment
and the incident circularly polarized light are not par-
allel. This is because the electric field felt by the 4f
electrons is equivalent to the sum of two circularly polar-
ized fields with opposite helicities, and a linear polarized
field aligned with the direction of the magnetic moment.
The detailed calculation of the amplitude of these fields is
explained in appendix $A$. The dependency of the ampli-
ditude of these polarized field with the magnetic moment
orientation angle $\theta$ is:

$$P = \frac{e^{\pm i\phi}}{\sqrt{2}} \left[ \frac{(\cos \theta \pm 1)}{\sqrt{2}} P^{-1}_{2m} + \frac{(\cos \theta \mp 1)}{\sqrt{2}} P^{1}_{2m} + P^{0}_{2m} \sin \theta \right]$$

$$P^{-1}_{2m}, P^{1}_{2m} \text{ and } P^{0}_{2m} \text{ are the dipole operators for right }$$

and left circular polarizations, and linear polarization
along the $z$ axis, respectively. Taking into account the
previous expressions, the modulation of the intensity of
each of the spectral components (PP, AP and TR) with
the orientation angle $\theta$ is given by the following expres-
sion (see appendix $A$):

$$PP + AP + TR = \left[ \frac{(\cos \theta \pm 1)^2}{4} A^{J}_{J'} + \frac{(\cos \theta \mp 1)^2}{4} A^{J}_{J'} + \frac{\sin^2 \theta}{2} A^{J}_{J'} \right]$$

where $A^{J}_{J'}$ are the reduced angular integers. They

are directly related to the transitions $\Delta J = 0, \pm 1$ and
FIG. 5. Angle coefficients for PP, AP, and TR as a function of (a) magnetic moment orientation angle with respect to the circularly polarized incident beam, \( \theta \), and (b) cone half opening angle \( \theta_C \).

\[
PP + AP + TR = \frac{1}{3} \left( 1 - \cos^6 \left( \frac{\theta_C}{2} \right) \right) A_{J'J'}^{-1} + \frac{1}{3} \sin^3 \left( \frac{\theta_C}{2} \right) A_{J'J'}^{-1} + \frac{1}{3} \left( \cos^3 \theta_C - 3 \cos \theta_C + 2 \right) A_{0}^{0}.
\]

Figure 5(b) shows the coefficients of each excitation component as a function of the cone half opening angle \( \theta_C \). The coefficients of the TR and AP components increment its value at large angles, but not as fast as in the case of a defined angle orientation. This is better observed when the ratio between the TR and AP components, shown in both figures, are compared. These ratios are substantially smaller in the cone angle coefficients (figure 5(b)).

When the cone has an inclination angle \( \varphi_C \) with respect to the beam direction, the components PP’ AP’ and TR’ of the magnetic moment in the cone are projected in the beam axis following expression 3 substituting the angle \( \theta \) by \( \varphi_C \). The PP’ component is calculated using equation 4 the AP’ component is the same as the equation 4, but interchanging the factors \( A_{J'J'}^{-1} \) by \( A_{1J'J'} \), and the TR’ component is calculated by multiplying TR by \( \sin^2 \theta_C / 2 \).

Since the cross section values of the angular integers are tabulated \( \text{[38, 39]} \) and the angle dependent modulation of the PP, AP and TR components are known (equations 3 and 4), it is possible to determine the orientation of the magnetic moment of any RE in any chemical environment just by identifying their PP, AP and TR components and checking their relative intensities. This is something that can be inferred also by comparing the magnetic moment deduced from the application of the XMCD sum rules and compare it with the total magnetic moment of the RE. But this is only valid if there is a single magnetic form of dysprosium. Things become more complicated if the analyzed material contains RE in different magnetic states, i.e., RE with different magnetic moment orientation distributions. The deconvolution of the spectra in their PP, AP and TR components is then required. This could be done by their direct calculation using numerical methods as the employed by Thole et al. \( \text{[38]} \). This is the approach used in \( \text{[24]} \). But it is important to notice that the build of the RE \( M_{4,5} \) spectrum only requires, to a good approximation, the shape of the spectral component, which can be isolated by spectral methods. The Dy \( M_{5} \) spectrum is specially suited for this kind of analysis.

Due to the large value of the orbital moment, \( L \), of the 4f orbital in dysprosium, the splitting between the PP and AP components in the \( M_{5} \) edge is the largest among the RE. Dy is the RE that has the lowest overlap between these two sets of excitations, which is estimated in less than 5% \( \text{[39]} \). This eases the extraction of both spectral components from any XMCD spectra at the Dy \( M_{5} \) edge. Figure 4 shows the XMCD spectrum for Dy \( M_{5} \). Only the PP and AP components are in the spectra, which are well distinguished because they have opposite sign. These two spectral shapes can be taken, as a first approximation, to the PP and AP true components of the spectra. The TR spectral component is extracted by subtraction of these two approximated components to the related circularly polarized spectra. For the subtraction, each of the components must be multiplied by a coefficient which is related to its relative spectral intensity, which depends on the sum of all the RE magnetic moment orientation dis-
tributions probed. These coefficients must be the same in both circularly polarized spectra, bearing in mind that the PP and AP components are interchanged in the two circular polarization spectra taken at the same magnetic field direction. The determination of the value of these coefficients is not precise, since it depends on how well the shape of the TR component is known. In our approach, we used the spectral lineshape theoretically calculated by Thole et al. [38]. This so extracted TR component contains the overlap region between the PP and AP components. This produces some shape differences between the TR obtained at different beam orientations angles, because the proportion of the TR component respect to the PP and AP components changes. In our case, the spectra taken at 70° has, clearly, the lowest overlap proportion in its TR component. Then, the PP-AP overlapping region can be extracted by subtracting this TR component to the TR component withdrawn at 0°, being the former conveniently normalized to the intensity of the 0° TR component. This overlapping component is added to the PP and AP components directly extracted from the XMCD spectra obtained at any angle and temperature. The process is then repeated for the extraction of the related TR components, reducing in this way its overlapping portion, and approaching to the true TR component.

The result of applying this process in our samples is shown in figure 6. Although the overall shape of each of the components is very similar for any spectra, they were not exactly the same. For instance, we observed a reduction in the energy distance between the AP and PP peaks position with increasing field orientation angle, which is visible in their XAS spectrum shown in figure 7(a) for sample DCM. The same observation was made in sample DCC and at RT. The reason of this is beyond the scope of this work. More specific experiments are required to determine if these differences, which are small in any case, are caused by experimental factors or because the spectra are actually sensitive to the chemical and/or crystal field environments [21, 41, 42]. However, such differences imposed that the deconvolution of each spectrum and, later on, its model fitting, had to be done using its own PP, AP and TR components. We noticed that the obtained coefficients for each of the components used in the deconvolution of the spectra have little variations when using the corrected or the uncorrected spectral components.

Figure 8 shows the normalized coefficients of the three spectral components obtained from the deconvolution of the spectra of the two samples DCC and DCM at RT and 2 K, as a function of the beam and magnetic field orientation angle. The plot was done to compare it with the angle dependent coefficients plotted in figures 5(a) and 5(b), which are cross section independent. The normalization of the coefficients was done by dividing all the components by the intensity of the PP component measured at normal incidence, which is the one with the highest intensity, and corrected by their relative cross sections. Cross sections were obtained from [39]. The used cross section ratio between the PP and the AP components were 3.4, and 1.85 between PP and TR components. The TR component is divided by 2, since there are two possible traversal directions to $J_z$. The coefficients obtained at 2 K had the TR component smaller than the AP component at any angle. There is not any dysprosium moment orientation with such a coefficient configuration in figure 5. The same occurs at RT. In this case, the AP experimental coefficient is overwhelm higher than the PP coefficient. Moreover, the PP coefficient reaches such a value only when the dysprosium magnetic moment orientation is in the plane. All these means that the dysprosium spectra of the DCC and DCM samples can not be understood using a single distribution of dysprosium magnetic moments.

This finding supports the conclusion derived from the low magnetic moment value of dysprosium at RT in section III D the observed spectra must be the result of
FIG. 7. (a) XAS spectrum \((C^+ + C^-)\) of sample DCC taken at 2 K and at different orientation angles: 0° (dark blue line), 20° (light blue line), 45° (orange line) and 70° (red line). (b) PP and AP components, and (c) TR components extracted from the spectra of the DCC sample at 2 K. The colors code is the same as in (a).

the combination of the spectra from, at least, two different distribution of dysprosium magnetic moments. One of them must have the dysprosium poorly magnetically coupled or even uncoupled from cobalt. For this to happen, a possibility is to have this dysprosium physically separated from the DyCo alloy by segregation at the interface with the aluminum cap layer. RE segregation is a common effect in RE-TM alloys, with the RE always located at the free surface region [17, 18, 21, 22]. Then, the total spectra can not be built by the simple summation of two spectra. That would be the case if the two types of dysprosium atoms were uniformly distributed all through the depth of the film. When TEY detection is used, the measured spectral intensity is build from the secondary electrons current which depends on the depth from where they are generated by two factors: their scape depth in the excited layers \((\lambda_e)\), and the X ray intensity which is absorbed as it propagates through them. This causes distortions in the measured spectra with respect to the true absorption coefficient of the analyzed sample. These saturation effects becomes more important as the angle of incidence is increased, because the X ray penetration depth becomes similar to the secondary electrons scape depth [43]. Such effects are taken into account when the spectra are analyzed to recover the absorption coefficients that were originated using a method which is fully described in [21]. Note that this analysis process assumes the presence of a single layer.

Then, two steps are required to untangle the experimental spectra in their contributions from the different regions at different depths from the films surface. The first step is to model the experimental spectra assuming the presence of, at least, two layers with different thickness, dysprosium concentrations, magnetic moment distributions and secondary electron scape lengths. The modeling of the spectra is done using the absorption cross section of the PP, AP and TR components (i.e., the related peak shapes \(A_{J^{-}J'}\), \(A_{J^{+}J'}\) and \(A_{J^{0}J'}\)), multiplied by their corresponding angle dependent coefficients shown before. The sum of the contribution of each layer is weighted depending on their specific thickness and depth. The resulting spectral intensity follows the expression:

FIG. 8. Normalized PP (blue dots), AP (red dots) and TR (yellow dots) components in Dy \(M_5\) spectra of samples (a) DCC and (b) DCM taken at 2 K as a function of the circularly polarized incident beam. Empty squares are related to the components of the Dy \(M_5\) spectra taken at RT.

larger than expected intensity of component AP found in the spectra is by the presence of dysprosium with low magnetic moment (large cone opening angle) at the topmost surface of the alloy.

The intensity of the secondary electrons collected by the detector depends on the depth from where they are generated by two factors: their scape depth in the excited layers \((\lambda_e)\), and the X ray intensity which is absorbed as it propagates through them. This causes distortions in the measured spectra with respect to the true absorption coefficient of the analyzed sample. These saturation effects becomes more important as the angle of incidence is increased, because the X ray penetration depth becomes similar to the secondary electrons scape depth [43]. Such effects are taken into account when the spectra are analyzed to recover the absorption coefficients that were originated using a method which is fully described in [21]. Note that this analysis process assumes the presence of a single layer.

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\[ I(E) \approx \frac{I_{0n}}{\cos \theta_i} \left( \mu_1(E) \frac{1 - e^{-\frac{z_1}{\lambda_{ei1}}}}{\lambda_{ei1} \mu_1(E)} + 1 + \mu_2(E) \frac{e^{-\frac{z_2}{\lambda_{ei2}}}}{\lambda_{ei2} \mu_2(E)} + 1 \right) \] (5)

This equation has been calculated assuming that the first layer is thinner than the absorption coefficient \( z << \mu_{1,2}(E) \), which is applicable in our case.

In the second step, the modeled spectrum passes through the same process as the actual experimental spectrum, described in [21] to extract what it could be called the effective absorption coefficient. This is a necessary step to compare the saturation corrected experimental effective absorption coefficient, where only a single layer is assumed in the spectrum saturation correction process, with the modeled one.

The parameters to enter in the modeling of the spectra are the thickness of the first layer, \( \tau_1 \), the dysprosium concentration \( \rho_1 \) of the top layer and the underlayer, the secondary electron escape length \( \lambda_{ei1} \) for each of the layers, the overall escape length used to correct saturation effects \( \lambda_{ei1} \), and the opening and inclination angles of the cone of each layer, \( \theta_{C1} \) and \( \varphi_{C1} \). The model that significantly best fitted the spectra required an additional parameter for each layer which represented the portion of dysprosium atoms with moments in the opposite orientation.

Some assumptions were done to chose the value of some of the parameters of the model. The cobalt concentration of the first layer was set to zero in the first layer in base of the expected RE segregation. The secondary electron escape length was set to 25 Å for both layers. This is the length used in the saturation correction of the experimental spectrum, which was the one that worked the best in these alloys [21]. The validity of this parameter to correct the saturation effects was tested by measuring the experimental ratio between the AP and PP component extracted from the XMCD spectra. This ratio has to be constant for any field orientation and probed sample. Its value was identical to the theoretical one [39] within the experimental error. The goodness of the fits were evaluated by minimizing a \( \chi^2 \) function. Different values of the above mentioned fixed parameters were tried to check the incidence in the results of the fits. Their variation over a relatively wide range of values did not change in a substantial way the conclusions of the spectral fits here presented.

The accuracy of the fitted parameters and the discrimination accuracy of the possible configurations was increased by fitting at the same time the spectra taken at opposite circular polarizations. Both spectra are symmetrical in their spectral components and, therefore, they must use the same fitting parameters: the TR component does not change with circular polarization light and the PP and AP components becomes AP and PP components in the opposite helicity. Obviously, the condition for the fits of yielding the same magnetic moment value as the experimental one was imposed. The inadequacy of the spectra to be fitted with a single component was tested. The quality of the fits was very low in this case, with \( \chi^2 \) values two orders of magnitude higher than the result from the proposed models, confirming the previous conclusion extracted from the deconvolution of the spectra in their PP, AP and TR components.

The spectral fittings were done using the PP and AP components with (corrected) and without (uncorrected) the overlapping component, using the related TR component in each case. We will only show the results corresponding to the corrected components. The results were essentially the same using the uncorrected components.

The described analysis of the spectra used two layers for the spectra modeling. Include a larger number of layers might compromise the accuracy of the analysis due to the consequent increase in the number of variables. Also, the thickness of the effective probed layer, which is estimated to be of the order of 2 to 3 nm, leaves little room for a better in depth accuracy for the method, because the intensity is exponentially reduced with depth. The use of two layers is also justified by posterior measurements on the same samples by X ray photoemissional which showed a significantly larger dysprosium concentration at the surface than the nominally expected. The next section discusses the results of the fits.

### A. Results and discussion of the fits

Table IV shows the result of the fits with the lowest \( \chi^2 \) of the spectra taken at RT and 2 K. Figure 9 shows the fitting of the circularly polarized spectra of sample DCC at RT an 0° incident angle as an example of the quality of the fit. In the model used for these fits, the top layer had its average moment direction opposite to that of the dysprosium in the under-layer. The underlayer had two dysprosium moment components that shared the same direction, but having their average moment oriented opposite to the top layer. The thickness of the effective probed layer, which is estimated to be of the order of 2 to 3 nm, leaves little room for a better in depth accuracy for the method, because the intensity is exponentially reduced with depth. The use of two layers is also justified by posterior measurements on the same samples by X ray photoemission which showed a significantly larger dysprosium concentration at the surface than the nominally expected. The next section discusses the results of the fits.
This effect was even stronger in the fits of the spectra taken at 2 K. The possible explanation to this will be discussed later on.

The total moment of dysprosium in the underlayer was $3.8 \mu B$. It matched the expected from the VSM data (see table I) using the cobalt moment deduced from XMCD. It is important to notice that this value was obtained considering the presence of a significant portion of dysprosium atoms whose average magnetic moment orientation was the same as cobalt. There is no other way to obtain such a low magnetic moment in dysprosium. This is the reason why the AP component is so intense at this temperature. The presence of this phase in the underlayer indicates that it should also exists in the bulk, although in a lower proportion in base of the estimated value of its dysprosium moment in that region. The magnetic interaction of this kind of dysprosium with cobalt must be either too weak or null and it might be caused by a segregation process and/or by disorder. The relatively narrow moment distribution of this dysprosium compared to the fitted on the top layer could be explained by being the former in closer interaction with other magnetic atoms increasing the mean field felt by them, whereas the latter might be mixed with those of the capping layer at the interface. At low temperature, this type of dysprosium in the under-layer should contribute to increase the total magnetic moment of the dysprosium sublattice. The presence of these weakly coupled dysprosium atoms at the region probed by TEY would explain the apparent increase in the $T_{Comp}$ at the surface with respect to the bulk measured by XMCD using TEY detection, since its average magnetic moment is oriented in the same direction as the applied field. It should be also the cause of the observed small decoupling between the cobalt and dysprosium hysteresis loops (see section III A).

At 2 K, the underlayer has a single component, but the top layer required two types of dysprosium with opposite averaged moment directions. The moment distribution for this layer caused a reduction of its total moment to almost zero, becoming close to a magnetically dead layer. This change in the magnetic behavior of the top layer could be attributed to the presence of some oxidized dysprosium which is AF below 4 K [44]. The oxidation state does not cause any change in the Dy $M_{4,5}$ spectra but it does in the Co $L_{2,3}$ spectra. Cobalt spectra did not show any feature related to oxidized cobalt, what it could be explained by the expected location of the possibly oxidized segregated dysprosium at the interface with the aluminum cap layer. Figure 11 shows the fitted components for both layers at the two circular polarizations for 0° and 70° incidence angles. For C+ polarization, blight blue line for the top layer and dark blue line for the under layer. For C− polarization, blight orange line for the top layer and red line for the under layer.

![Graphs and Diagrams](image-url)
samples (see table IV). A higher insight of the magnetic distribution were similar in both different local anisotropy energies. The variation in the uniaxial PMA where dysprosium atoms are subject to is the qualitatively expected behavior in the presence of field orientation angle $\theta_{C2}$: cone inclination angle with respect to the beam (field) orientation in the underlayer; $m_{C2}$ estimated magnetic moment of the underlayer; $\rho_2$: effective atomic concentration of dysprosium at the under layer; AP$_1$: proportion of antiparallel dysprosium in the first layer (only at 2 K); $\chi^2$: statistical goodness of the fit.

| Sample T | Field Orientation Angle | 0°   | 20°  | 45°  | 70°  |
|----------|-------------------------|------|------|------|------|
| DCC      | $\tau_1$                | 1.6(1) Å | -    | -    | 1.5(1) Å |
|          | $\theta_{C1}$           | 90°  |      |      | 90°  |
|          | $\theta_{C2}$           | 53.5(1)° | -    | -    | 54.1(1)° |
|          | $AFC_2$                 | 0.26(1) | -    | -    | 0.26(1) |
|          | $m_{C2}$                | -3.8(1) | -    | -    | -3.8(1) |
|          | $\rho_2$                | DyCo$_{3}$ | -    | -    | DyCo$_{4.9(3)}$ |
|          | $\chi^2$                | 0.6  | -    | -    | 0.8  |
| 2 K      | $\tau_1$                | 2.1(1) Å | 2.0(1) Å | 2.8(1) Å | 5.6(1) Å |
|          | $\theta_{C1}$           | 90°  |      |      | 90°  |
|          | $\theta_{C2}$           | 35.9(1)° | 45.7(2)° | 47.5(2)° | 40.2(1)° |
|          | $\varphi_{C2}$          | 0    | 10°  | 9°   | 47°   |
|          | $m_{C2}$                | 8.9  | 8.2  | 8.1  | 5.8   |
|          | $\rho_2$                | DyCo$_{0.1}$ | DyCo$_{0.0}$ | DyCo$_{0.2}$ | DyCo$_{0.8}$ |
|          | $\chi^2$                | 1.74 | 3.23 | 2.0  | 0.9   |
| DCM      | $\tau_1$                | -    | -    | -    | 1.5 (1) Å |
|          | $\theta_{C1}$           | -    | -    | -    | 90°   |
|          | $\theta_{C2}$           | -    | -    | -    | -56.7(1)° |
|          | $AFC_2$                 | -    | -    | -    | 0.26(1) |
|          | $m_{C2}$                | -    | -    | -    | -3.7(1) |
|          | $\rho_2$                | -    | -    | -    | DyCo$_{5.3}$ |
|          | $\chi^2$                | -    | -    | -    | 0.4   |
| 2 K      | $\tau_1$                | 2.6(2) Å | 2.4(2) Å | 3.3(2) Å | 3.7(2) Å |
|          | $\theta_{C1}$           | 90°  |      |      | 90°  |
|          | $\theta_{C2}$           | 27.0(1)° | 32(1)° | 44(1)° | 48(1)° |
|          | $\varphi_{C2}$          | 0(1)° | 6(1)° | 25(1)° | 46(1)° |
|          | $m_{C2}$                | 9.3(2) | 9.0(2) | 7.8(2) | 6.1(2) |
|          | $\rho_2$                | DyCo$_{4.9(1)}$ | DyCo$_{5.3(1)}$ | DyCo$_{5.4(1)}$ | DyCo$_{5.4(1)}$ |
|          | $\chi^2$                | 3.3  | 3.3  | 3    | 2.8   |

FIG. 11. Espectral components of the circularly polarized spectra Dy $M_5$ taken at 2 K and at (a) 0° and (b) 70° orientation field angles for sample DCC, and at (c) 0° and (d) 70° for sample DCM. For C$^+$ polarization, bleight blue line for the top layer and dark blue line for the under layer. For C$^-$ polarization, bleight orange line for the top layer and red line for the under layer.

respect to the expected in the bulk. Attending to the results of the fits obtained at RT, the increment in the dysprosium total magnetic moment can be explained by the portion of the weakly magnetically exchange-coupled-to-cobalt dysprosium atoms which, at this temperature, are ferromagnetic. The magnetic anisotropy of these dysprosium atoms is expected to be lower than those exchange coupled to cobalt, what would explain the apparent lower anisotropy of the dysprosium sublattice. Also, the anisotropy field $H_k$ is expected to decrease due to the increase in the magnetization. In reference to this, the deviations of the dysprosium cone distribution with respect to the beam (angle $\varphi_{C2}$) and the increasing opening of the cone angle (angle $\theta_{C2}$) with the increasing field orientation angle $\theta_i$ with respect to the easy axis is the qualitatively expected behavior in the presence of an uniaxial PMA where dysprosium atoms are subject to different local anisotropy energies. The variation in the dysprosium magnetic distribution were similar in both samples (see table IV). A higher insight of the magnetic interactions and anisotropy of the dysprosium and cobalt atoms could be gain by calculating them in base to the observed magnetic moment distributions resulted from the fits [46].

The required use of dysprosium with opposite average moment directions in the fits of the spectra at RT and at 2 K is consequent with the observations done in the previous section, where it was shown that the intensity of the deconvolved AP component was too high...
are not contemplated in the fit, where the concentration decreasing its density and concentration. These details should extend the thickness of the top layer dysprosium, of dysprosium within the aluminum capping layer that film surfaces. Moreover, it is expected some diffusion instance, they are below the estimated roughness of the ers. These values might not be the actual ones. For and its highest value is not higher than two monolay-

tions, which might change with the temperature, and not the spectral weight given to the top layer in equation 5 could be attributed, at least partially, to the simplicity of this section. These consisted in an increased value of both parameters with the incident angle, indicating that the spectral weight given to the top layer in equation [47] was below the required in the fits. The second inconsist-

ty was an increase in the values of these parameters when the temperature decreased to 2 K. These two effects could be attributed, at least partially, to the simplicity of the model, which does not consider any gradation in the film driven by the increasing energy of the anisotropy field that reduced the magnetization in the direction of the applied field [17]. More measurements done at dif-

ferent temperatures are needed to fully test this expla-

nation, what will definitively prove the high sensitivity of the proposed analysis method to the variations in the magnetic moment of the REs at the region probed by TEY.

FIG. 12. (a) Comparison between the magnetic moment of dysprosium in the underlayer resulted from the “two layers model” fits (red dots, sample DCC; blue dots, sample DCM), and the magnetic moments measured considering a single dysprosium magnetic state, as a function of the field orientation angle. (b) Comparison between the spectra of the Dy $M_5$ circularly polarized spectra of samples DCC and DCM taken at 2 K and at 70°. Dark ($C^+$) and light ($C^-$) blue lines for sample DCM and red ($C^+$) and orange ($C^-$) lines for sample DCC.

A plausible explanation for this effect might have to do with the sensitivity of the dysprosium spectrum to the moment distribution across the thickness of the film and not to its chemistry. The magnetization of the probed dysprosium increases from RT to 2 K. Pure dysprosium becomes ferromagnetic at temperatures below 80 K and oxidized dysprosium is AF at temperatures below 4 K. This means that the changes in the orientation of the dysprosium moment should be more gradual from the top layer to the under-layer at 2 K than at RT, as the fits suggests. As the orientation of the applied magnetic field deviates from the easy axis, the top layer with its nearly zero average magnetic momentum would penetrate into the film driven by the increasing energy of the anisotropy field that reduced the magnetization in the direction of the applied field [17]. More measurements done at dif-

ferent temperatures are needed to fully test this expla-

nation, what will definitively prove the high sensitivity of the proposed analysis method to the variations in the magnetic moment of the REs at the region probed by TEY.
V. CONCLUSIONS

To summarize, the magnetism of the dysprosium and the cobalt sublattices in ferromagnetic DyCo thin films with PMA has been investigated by XMCD spectroscopy using TEY detection. Some unexpected results were observed in the magnetic behavior of the dysprosium sublattice. The measurements done at the Co L_{2,3} showed that the PMA of the films must stem from the RE sublattice. However, at 2 K, when the PMA energy of the films was the highest, the probed cobalt sublattice resulted to be magnetically more anisotropic than the dysprosium sublattice. Additionally, the magnetic moment of the dysprosium sublattice measured at RT was lower than expected if all moments were AF coupled to cobalt.

To understand the cause of these apparent anomalies, a method to analyze the circularly polarized Dy M_{5} spectra obtained by TEY has been presented, which is based in the deconvolution of their parallel, antiparallel and transverse to J_{z} spectral components. This is the first time that this kind of analysis is applied in this kind of systems. This spectral analysis reveals the presence of a relatively large proportion of dysprosium with its average moment oriented in the same direction than cobalt, and a thin layer of segregated dysprosium at the top of the alloy which should be not exchanged coupled to cobalt. Some of the dysprosium in this layer is AF coupled at 2 K, suggesting that either it is partially oxidized or there exist a proportion of dysprosium which is magnetically dead at this temperature. The apparently lower magnetic anisotropy of dysprosium with the field orientation angle is explained by the thickness of the segregated dysprosium layer at the region near the surface. A similar kind of measurements should be conducted using bulk detection sensitivity to determine the precise distribution of RE sites that contribute to the PMA of the alloy. This seems mandatory to be able to predict with accuracy the possible magnetic configurations resulting from the interaction between RE and TM sublattices.

The presented results show that, if the RE is segregated at the surface, care must be taken to remove the effect that it causes in the value of the RE magnetic moment measured using TEY detection. This affects, for instance, to the T_{comp} values of RE-TM alloys measured at the surface in this way, which could be overestimated.

The proposed analysis method can serve to detect the presence of RE in different magnetic states, either in the surface or in the bulk. The method can be extended to the circularly polarized spectra of other RE at the M_{4,5} edge, whenever the angular moment J of its 4f orbital is different of zero (i.e., it can not be applied to Gd^{3+}). In those other RE, the identification of the PP, AP and TR spectral components requires a different methodology than the presented here for dysprosium since their overlap is much important. This might not be a problem because these components are well approximated by theory. They can be also isolated by spectroscopic methods using circular but also linear polarized spectroscopy at the related RE M_{4,5} edges. Nevertheless, more additional work is needed to improve and test the accuracy of the method by better characterizing the region probed with other surface sensitive techniques.

The experiment confirms the presence of segregated RE reported in other RE-TM systems. The preparation method of the alloy did not produced substantial differences in the magnetism of the cobalt and dysprosium sublattices at the surface of the alloy, indicating that the origin of their different macroscopic behaviors has to be found in the bulk or they are caused by their different microstructure. The no-dependence of the RE segregation with the deposition method (co-sputtering and alternate deposition) remarks that the segregated RE is an immediate post deposition process, which can proceed during time as it has been observed in GdCo [17]. This is also demonstrated in the DCM film whose last deposited layer, before depositing the aluminum capping layer, was cobalt. This means that RE atoms could diffuse also in the bulk at defects like voids, cracks and grain borders within the bulk. This might affect magnetic properties like, for instance, the coercive field of the thin film since it would contribute to the formation of domain wall pinning defects.

Appendix A: Orientation dependence of light absorption

Here, it is shown the details of the calculation of the intensity absorbed by circularly polarized light when the orientation of the angular moment of the 4f orbital forms an angle \( \theta \) with the direction of the wavevector \( \vec{k} \) which is chosen parallel to the \( z \) axis. The components of the electric field of the circularly polarized light in the frame of the 4f angular moment, with its \( z_{m} \) axis along its quantization axis, are calculated by making a first rotation of an angle \( \varphi \) in the \( xy \) polarization plane (\( z \) axis as the rotation axis) followed by a rotation of an angle \( \theta \) around the resulted \( y \) axis after the first rotation.

\[
\begin{pmatrix}
\epsilon_{x_{m}} \\
\epsilon_{y_{m}} \\
\epsilon_{z_{m}}
\end{pmatrix} = \begin{pmatrix}
\cos \varphi & -\sin \varphi & 0 \\
\sin \varphi & \cos \varphi & 0 \\
0 & 0 & 1
\end{pmatrix} \begin{pmatrix}
\cos \theta & 0 & -\sin \theta \\
0 & 1 & 0 \\
\sin \theta & 0 & \cos \theta
\end{pmatrix} \begin{pmatrix}
\epsilon_{x} \\
\epsilon_{y} \\
\epsilon_{z}
\end{pmatrix}
\]

Therefore, for right (\( - \)) and left (\( + \)) circular polarization:

\[
\begin{pmatrix}
\epsilon_{x_{m}} \\
\epsilon_{y_{m}} \\
\epsilon_{z_{m}}
\end{pmatrix} = \frac{\epsilon_{\pm \varphi}}{\sqrt{2}} \begin{pmatrix}
\cos \theta \\
\mp i \\
\sin \theta
\end{pmatrix}
\]

By using this result, the electric dipole operator \( P \) written in the frame of the 4f orbital reads:

\[
P = \frac{\epsilon_{\pm \varphi}}{\sqrt{2}} \left[ P_{m}^{0} \cos \theta \mp i P_{m}^{0} + P_{m}^{0} \sin \theta \right]
\]

\[\text{(A3)}\]
The superscript in the dipole operator indicates the polarization state of the electric field in the frame of the 4f orbital: 0 is linear, 1 circular left handed and +1 circular right handed. The electric dipole operator can be expressed in the components parallel to the quantization axis $z_m$ of the 4f orbital using the relations:

$$P^0_{zm} = \frac{1}{\sqrt{2}} [P^1_{zm} + P^1_{zm}] \quad (A4)$$

and

$$P^0_{ym} = \frac{i}{\sqrt{2}} [P^1_{zm} - P^1_{zm}] \quad (A5)$$

Then

$$P = e^{\pm i\varphi} \left[ \frac{(\cos \theta \pm 1)}{\sqrt{2}} P^1_{zm} + \frac{(\cos \theta \mp 1)}{\sqrt{2}} P^1_{zm} + P^0_{zm} \sin \theta \right]$$

(A6)

The absorption cross section for the excitation from a state $|\alpha mJ\rangle$ to a state $|\alpha' m' J'\rangle$ is, in the dipole approximation:

$$\sigma_{\alpha mJ \rightarrow \alpha' m' J'} = 4\pi^2 \alpha_0 \hbar \omega |\langle \alpha mJ | P | \alpha' m' J' \rangle|^2$$

(A7)

The cross section for circular polarized light is obtained substituting equation 2 in equation 3.

Then

$$\sigma_{\alpha mJ \rightarrow \alpha' m' J'} = 4\pi^2 \alpha_0 \hbar \omega S_{\alpha J \alpha' J'} \left[ \frac{(\cos \theta \pm 1)^2}{4} A_{J'J'}^1 + \frac{(\cos \theta \mp 1)^2}{4} A_{J'J'}^1 + \frac{\sin^2 \theta}{2} A_{J'J'}^0 \right]$$

(A8)

absorption cross sections for the corresponding situation in which the polarized beam and the magnetization are parallel ($q = 0, \pm 1$). Then, the $\theta$ dependent functions that multiply to each of the related $q = 0, \pm 1$ cross sections are the coefficients for the TR ($q = 0$), PP ($q = -1$) and AP ($q = +1$) components:

$$PP + AP + TR = \left[ \frac{(\cos \theta \pm 1)}{4} A_{J'J'}^1 + \frac{(\cos \theta \mp 1)}{4} A_{J'J'}^1 + \frac{\sin^2 \theta}{2} A_{J'J'}^0 \right]$$

(A9)

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