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Microscopic origin of perpendicular magnetic anisotropy in amorphous Nd-Co homogeneous and compositionally modulated thin films studied by XMCD

R. Cid*[1], J. Díaz[1], L. M. Álvarez-Prado[1], J. M. Alameda[1], S. M. Valvidares[2], J. C. Cezar[2], N. B. Brookes[2]

[1] Dpto. Física, Facultad de Ciencias, Universidad de Oviedo - CINN, C/ Calvo Sotelo s/n, 33007 Oviedo, Spain

[2] European Synchrotron Radiation Facility, 6 Rue Jules Horowitz, BP220, 38043

*Corresponding author: cidrosalia.uo@uniovi.es

Abstract. Amorphous Nd-Co films deposited by DC-magnetron sputtering presented perpendicular magnetic anisotropy (PMA) with energies, K_N, of the order of 10^6 erg/cc at RT. To understand the origin of their PMA, we measured the orbital and spin magnetic moments in Co and Nd by XMCD at the Co $L_{3,2}$ and Nd $M_{5,4}$ edges in two kinds of samples of similar thickness (30 nm) and composition: one compositionally modulated Nd/Co film (CM) with strong PMA ($K_N \sim 10^7$ erg/cc at 10 K) and a homogenous alloy (A) with not strong enough PMA to see stripe domains for such thickness. The XMCD analysis evidenced the significant role of Nd in the PMA of these films.

1. Introduction

Rare-earth-Transition-metal (RE-TM) amorphous thin films have been of great interest and their properties have been intensively studied since the discovery of perpendicular magnetic anisotropy in many of them [1]. The origin of the perpendicular magnetic anisotropy (PMA) is still controversial and has been attributed to many different causes, e. g., pair ordering, columnar microstructures, strain, exchange, bond-orientational anisotropy, single-ion anisotropy [2] ….

This PMA is strongly dependent on growing conditions. We have observed anisotropy energies at room temperature (RT) for $NdCo_5$ as high as $2x10^6$ erg/cm^3 [3], not far from the crystalline value, which increased in an order of magnitude when temperature is lowered to 10 K.

When the RE has $L \neq 0$, the single-ion anisotropy model is the one with better experimental confirmation from magnetization measurements [4, 5]. A way to increase the effect of the single-ion anisotropy in an amorphous alloy is to modulate it in composition. In this paper, we compare the individual magnetism of RE and TM sublattices measured by XMCD in a homogeneous amorphous alloy (A) and a compositionally modulated film (CM) of similar composition and thickness.

2. Experimental Details

Both A and CM films were grown at RT by DC-magnetron co-sputtering (50 mmϕ Co and Nd targets) onto Si100 substrates placed at 18 cm away from the targets. Their thicknesses, measured by X-ray small angle reflectivity, were close to 30 nm. Ar pressure during evaporation was $P_{Ar} = 3x10^{-3}$ mbar. The base pressure of the vacuum chamber was below $5x10^{-9}$ mbar. The angle of incidence of sputtered atoms with respect to the normal to the substrate was 0º for Co and 36º for Nd. The sputtering powers were fixed at 0.05 kW for Nd and 0.12 kW for Co that, according to the quartz monitor, correspond to individual growth rates of 1.0 and 1.5 Å/s respectively. For the CM sample, the Nd and Co magnetrons were used alternatively for short periods of time, resulting in a modulation in atom concentration that consisted of less than one monoatomic layer of Nd and near 2 atomic layers of Co.

A 2 nm thick capping layer of Al was deposited to avoid oxidation. X-ray diffraction verified that both A and CM films were amorphous. Film composition was determined by electron microprobe. Their stochiometry were $NdCo_{3.8}$ for sample A and $NdCo_{3.5}$ for sample CM.

The XMCD (X-ray magnetic circular dichroism) measurements at the $L_{2,3}$ edges of Co and $M_{4,5}$ edges of Nd were done on the high field magnet end station, at ID08, ESRF. The spectra were obtained in total electron yield (TEY) mode measuring the drain current from sample to ground. All the spectra were normalized to the incident beam intensity using the electron yield from a gold grid. The XMCD signal was determined by reversing both the ellipticity of the beam between circular left and right, and the applied magnetic field between +5 T and -5 T, which was always collinear with the photon flux. For each sample, different measurements were taken at several incidence angles between normal (0º) and grazing (75º) incidence and temperatures between 10 K and 300 K. Fluorescence yield was used to obtain a qualitative insight about the magnetization processes (i.e. hysteresis loops).

3. Results and discussion

The element-specific hysteresis loops measured by XMCD (in fluorescence mode) at 10 K and grazing incidence (θ = 75º) for samples A and CM are shown in Fig. 1. From the XMCD magnetization curves, measured at maximum L_3 and M_4 peak intensities for Co and Nd respectively, we conclude that CM exhibits a much greater PMA than A (in which case, if it exists, it is not enough to produce out-of-the-plane magnetization -stripe domains- and so, transcritical loops [3] , not even at 10 K).

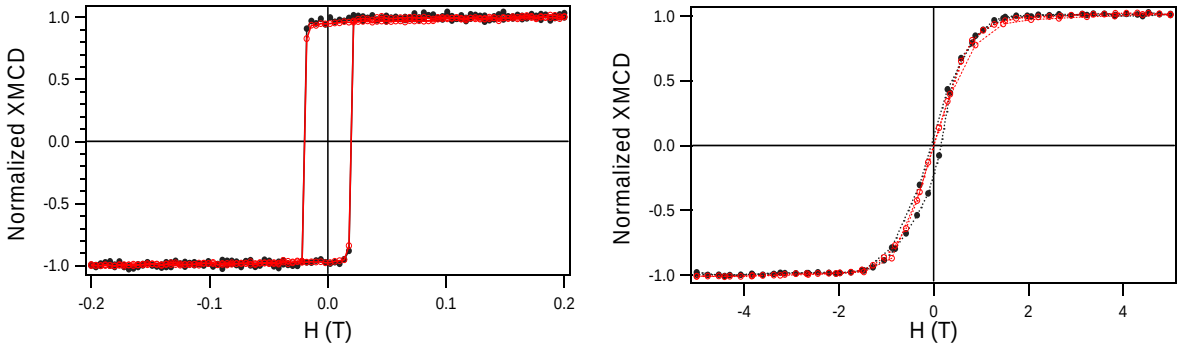

Fig. 1. XMCD magnetization curves at 10 K and grazing incidence for samples A (left) and CM (right). Co XMCD-closed circles, Nd XMCD- open circles. A presents a tipical easy-plane hysteresis curve, while in CM the transcritical hysteresis curve evidences the existence of PMA.

Saturation effects can appreciably distort the measured signal at $M_{4,5}$ edges of RE [6] and the $L_{2,3}$ edges of TM [7] when TEY detection is used. Saturation occurs when the measured signal is no longer

proportional to the photoabsorption cross section and intensities of prominent absorption peaks are reduced or ‘‘saturated”. We calculated the correction factor f (θ, λ_x, λ_e) at each energy point of the spectra as proposed by Nakajima et al. [8], assuming an electron sampling depth λ_e of 25 Å in all the studied samples.

Fig. 2 shows normalized absorption spectra for Co and Nd when the photon spin was parallel (XAS^+) and antiparallel (XAS^-) to the magnetization direction and their difference (dichroic signal). Fig. 2(a) refers to Co in A and (b) to Nd in CM, both measured at 10 K and normal incidence. The whiteline intensities were obtained after substracting a two steplike function from the original data. In the case of Co, we used the usual steplike function described by Chen et al. [9]. For Nd, the value of the steplike function in between edges is determined phenomenologically from the minimum value of the experimental data in this region.

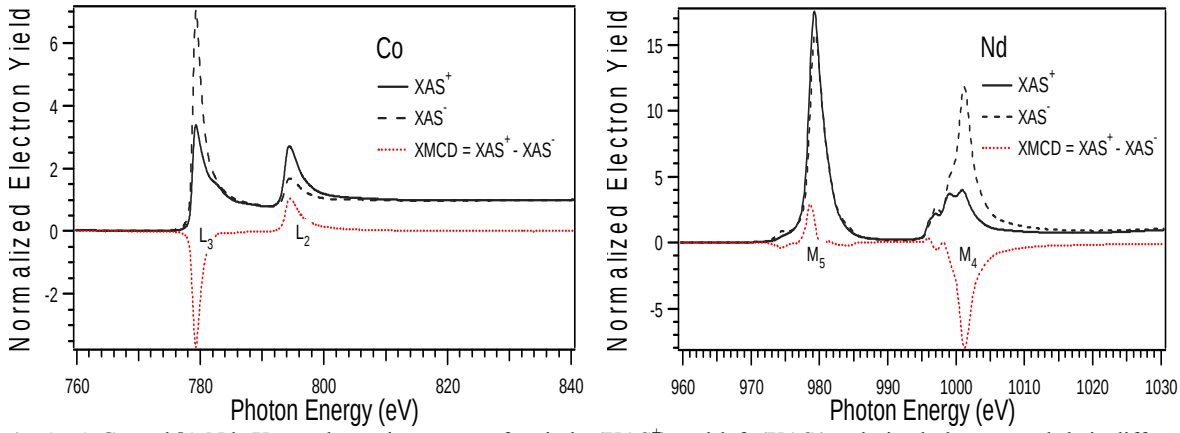

Fig. 2. a) Co and **b)** Nd X-ray absorption spectra for ritght (XAS^+) and left (XAS^-) polarized photons and their difference (XMCD effect).

The orbital and spin moments for Co were calculated using the sum rules introduced by Thole et al. [10,11], which have been extensively proved to work for TM. We neglected the contribution of the magnetic dipole operator $\langle T_z \rangle$ assuming that an amorphous system is the limit case of a randomly oriented polycrystal, as expressed by Stöhr and König [12].

Unfortunately, the spin sum rule cannot be applied in RE so straightforward as in TM because this rule relies on the difference in integrated intensity between the spin-orbit split $M_{4,5}$ edges. But the 4*f*-3*d* exchange and electrostatic interactions mix intensities between the two edges (jj mixing). Therefore, a correction factor must be included to take into account the deviation from this sum rule. For that purpose, we assumed an atomic-like behaviour of Nd and use the deviation factors calculated by Teramura et al. [13]. This assumption is based on the fact that in RE and actinides the crystal field is smaller than electrostatic and spin-orbit interactions, and so, the ratio of $\langle L_z \rangle$, $\langle S_z \rangle$ and $\langle T_z \rangle$ is approximately conserved. We also took the value of the ratio $\langle T_z \rangle / \langle S_z \rangle$ from ref. [13].

The orbital to spin moment ratio ($r=\mu_L/\mu_S$) in Co at 10 K and 0º obtained by the sum rules was 0.13 for sample CM and 0.11 for sample A. For comparison, r is 0.12 for Co when form clusters of about 1200 atoms in Au [14]. For hcp-Co, the value of r is found in between 0.099 and 0.12 [9,15]. It is interesting to note that r is as high as 0.26 in epitaxial $Nd_{0.19}Co_{0.81}$ with c axis in plane [15]. From polarized neutrons in crystalline $NdCo_5$ [16], r_{Co}= 0.23 for the Co_{2c} and 0.13 for the Co_{3g} site, at similar temperature and magnetic field direction conditions to those of [15], were found.

This ratio was r_{Nd} ~ -2.2 in Nd, approximately constant for all temperatures and samples under study, in agreement with the atomic value calculated by Teramura et al. [13] (r_{Nd} = 2.375). This result reinforces the validity of our assumptions and shows that J is a good quantum number for Nd, i.e., it behaves atom-like. Although r_{Nd} was similar in both samples, the value of the Nd orbital moment, $\langle L_z \rangle^{Nd}$, was significantly higher in sample CM than in sample A. At 10 K and 0º, $\langle L_z \rangle^{Nd}$ was 0.16 μ_B

per hole in A and 0.30 μ_B per hole in CM. Moreover, the CM film showed a reduction of $<L_z>^{Nd}$ with temperature slower than in the A film ($<L_z>^{Nd}$ (300 K)/ $<L_z>^{Nd}$ (10 K) ~ 0.4 in CM, but 0.25 in A).

The structure of Nd_xCo_{1-x} amorphous alloys is considered sperimagnetic, where the Co sublattice is basically collinear, while Nd moments are distributed in a cone with the axis pointing in the Co magnetization direction, whose angle is slightly affected by a magnetic field [17]. This is due to the different strengths of 3*d*-3*d* and 3*d*-4*f* exchange interactions and the effects of a random crystalline field acting on Nd and Co ions, whose strength is comparable to the 3*d*-4*f* exchange interactions, but much smaller than 3*d*-3*d* interactions. The validity of the atomic picture for Nd extracted from the constant value of r_{Nd} implies a correlation between its orbital and total magnetic moments, making $<L_z>^{Nd}$ suitable to give us information about the angle of the cone in the sperimagnetic structure. From the values of $<L_z>^{Nd}$ obtained by XMCD, it is evidenced that this angle is substantially smaller in sample CM than in A. This fact could be explained by a slightly anisotropic crystal field axis distribution for Nd in sample CM, induced by the fabrication process, in contrast to the random crystal field axis distribution present in sample A.

In addition to this, $<L_z>^{Nd}$ showed a variation the order of 4% between 0º and 75º at 10 K only in sample CM, indicating a preference of the Nd orbital moments cone to be oriented perpendicular to the film plane. This anisotropy decreased when increasing temperature, which evidences the leading role of Nd in the PMA found in sample CM.

4. Conclusions

XMCD measurements evidenced the fundamental role of Nd in the origin of the PMA present in Nd-Co alloys. The significant increase in the PMA for the CM sample indicated that this PMA is highly correlated with the local order at Nd atoms.

5. Acknowledgements

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