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Spin orientation in an ultrathin CoO/PtFe double-layer with perpendicular exchange coupling

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ABSTRACT

We studied by soft X-ray absorption spectroscopy the magnetization axis in a 4 nm thin CoO (111) layer exchange-coupled to an ultra thin $L1_0$ PtFe layer with perpendicular magnetic anisotropy. The angular dependence of the linear magnetic dichroism at 10 K and the relative variations of the spectral features provide a full description of the spin orientation in this antiferromagnetic layer. The spins are found in the film plane, pointing along the 110 direction. This results is discussed in relation to the film strain and the preferential occupation of t_{2g} orbitals. The strong orthogonal coupling between Co and Fe spins should be at the origin of the robustness of the exchange bias effect found in this bilayer system.

1. Introduction

Antiferromagnetic/ferromagnetic (AFM/FM) bilayers with controllable perpendicular magnetization are of a great interest for potential logic devices with high density, thermal stability and low critical current for current-induced magnetization [1,2]. To have an adequate description of AFM/FM couplings, crystallographic order, epitaxial strain, spin orientation and competing anisotropies are essential parameters.

The PtFe alloy in the chemically ordered $L1_0$ phase has one of the strongest magnetocrystalline anisotropy energies. The $L1_0$ phase, formed by alternating Fe and Pt atomic planes along the *c*-axis of the tetragonal structure, leads to an enhanced uniaxial anisotropy along the ordering axis [3] even in the thinnest limit of a few monolayers (MLs) [4,5]. $L1_0$ PtFe(001) layer with tetragonal axis perpendicular to the surface provides then an ideal out-of-plane spin network with enhanced perpendicular magnetic anisotropy to study the exchange-coupling properties with thin AFM layers. Among them CoO films are specially relevant for spintronic devices. Bulk CoO has a Néel temperature (T_N) of 293 K, close to room temperature (RT), and a magnetic moment of 3.98 μ_B [6,7], with a large orbital contribution. In the paramagnetic phase, bulk CoO crystallizes in the rocksalt structure with alternate Co and O (111)-planes (Fig. 1). Below T_N, AFM ordering goes along with a monoclinic distortion [6,7]. The bulk CoO AFM structure is described as a stacking of uncompensated FM hexagonal planes of Co^{2+} $3d^7$ ions coupled antiferromagnetically along the trigonal elongation (Fig. 1). In thin CoO layers grown on different substrates, polarization dependent soft X-ray absorption spectroscopy shows that the strain induced by epitaxy leads to significant modifications in the magnitude and the orientation of the magnetic moments [8], confirming the important sensitivity to magnetostriction. These results motivate our combined investigation of structural distortion and spin orientation in an ultrathin CoO/PtFe double-layer with perpendicular exchange coupling.

2. Experiments

The films have been grown by molecular beam epitaxy and studied *in situ* by grazing incidence X-ray diffraction (GI-XRD) at the French CRG BM32 beamline at the European Synchrotron Radiation Facility (ESRF, France). The Pt-terminated $L1_0$ PtFe(001) thin layer grown by thermal deposition of about 3 MLs of Fe on an ultrahigh vacuum cleaned Pt(001) substrate [9] hold at 600 K is in coherent epitaxy on Pt(001). A CoO layer with 4 nm thickness was grown by reactive molecular beam epitaxy on the ultrathin PtFe (001) layer hold at 523 K [10]. GI-XRD shows that the growth by reactive molecular beam epitaxy of the cobalt oxide on a Pt-terminated PtFe/Pt(001) surface gives rise to a hexagonal (111)-like surface. The strain imposed by the substrate on the

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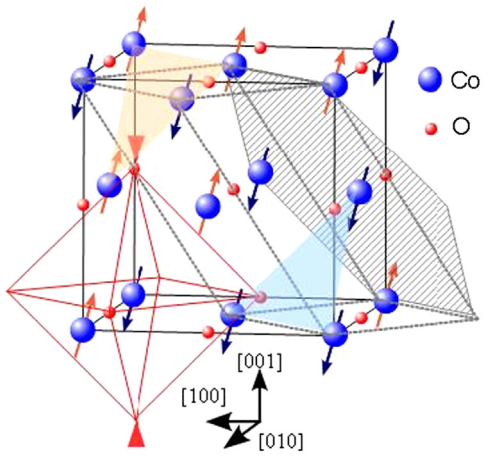

Fig. 1. Bulk CoO structure showing the Co(111) FM sheets coupled antiferromagnetically along the [111] direction. The hatched surface corresponds to the hexagon lying on the FePt layer in the CoO/PtFe bilayer. A CoO_6 octahedron is shown.

CoO layer leads to a slight monoclinic distortion at room temperature. The monoclinic distortion can be described as a tetragonal distortion with $c/a = 1.008$, with an additional slight trigonal distortion along the rocksalt [111] direction. The [001] axis of the tetragonal distortion is not perpendicular to the surface, but tilted by $\theta_0 = 54.74^\circ$ in relation to the normal surface. The growth proceeds in fourfold equivalent well-crystallized domains, so that the [001] axis is fourfold degenerated. The detailed growth procedure and X-ray diffraction study will be described elsewhere [10]. The double layer shows perpendicular magnetic anisotropy and an exchange bias shift of 800 Oe at 10 K [11].

The orientation of the spin moments in the CoO layer at low temperature ($T \approx 10$ K) was investigated *ex situ* by soft X-ray absorption spectroscopy (XAS) using linear dichroism at Co $L_{2,3}$ edges at the PGM beamline of the Laboratorio Nacional de Luz Sincrotron (LNLS, Brazil). The spectral resolution was $E/\Delta E = 6000$ and the degree of linear polarization was close to 100%. The sample was cooled down with an applied magnetic field of 5 kOe perpendicular to the surface plane. The magnetic field was then set to zero and the spectra were collected using the total electron yield. The sample was allowed to rotate about a vertical axis, with the polar angle (θ) defined as the angle between the surface normal and the X-ray propagation (Fig. 2a, inset). In this experimental geometry the variation of the escape length of the electrons as a function of θ should be corrected in order to recover the real X-ray absorption signal. This has been systematically corrected using the standard procedure for electron yield saturation effects [12]. To check this correction, for each θ, the X-ray absorption spectrum collected with vertical polarization was used as reference. At each angle, the X-ray linear dichroism (XLD) is obtained by two ways: (1) from the difference between the absorption with horizontal and vertical X-ray polarizations and (2) with horizontal X-ray polarization from the difference between the absorption at θ and at $\theta = 0^\circ$. The two methods give the same signals within the accuracy of the measurements.

3. Results

Fig. 2a shows the Co L_3 edge XAS spectra at 10 K for four θ values from $\theta = 0^\circ$ (polarization vector in the surface) to $\theta = 70^\circ$ (polarization vector towards the surface normal). The spectra, normalized far from $L_{2,3}$ edges, show a clear linear dichroism. Four main features (labeled A–D) are observed in the XAS and XLD signals. They correspond to transitions towards orbitals of different symmetries and then show distinct variations as a function

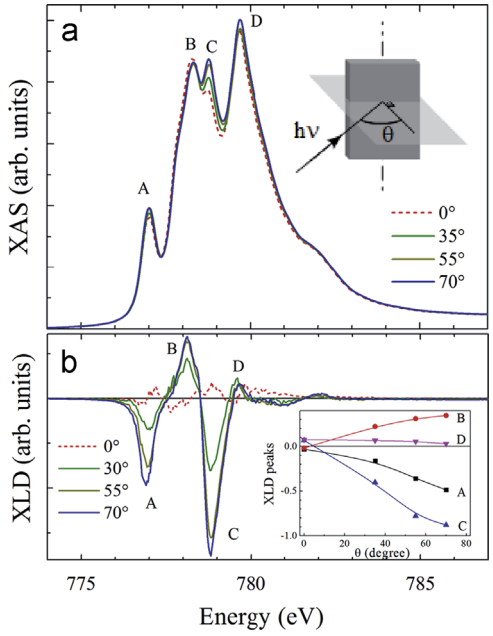

Fig. 2. (a) Co L_3 edge XAS as a function of θ. Inset: experimental geometry; (b) XLD as a function of θ. Inset : angular variation of the intensity of the main XMLD features.

of θ (Fig. 2b, inset). Taking the $\theta = 0^\circ$ spectrum as reference, the feature D at higher energy is almost constant, feature B increases with θ, while A and C decrease.

Magnetic and non-magnetic effects mixed by the local crystal field symmetry contribute to the polarization contrast. However, we have observed that the angular dependence dramatically decreases when the temperature increases and almost vanishes above 300 K, when the magnetic contribution to the anisotropy has vanished [11]. The XLD signal at 10 K, then, measures essentially the magnetic dichroism, i.e. the charge anisotropy associated with the magnetic moment anisotropy through the spin–orbit coupling [13,14,8]. Relative to the magnetic axis, this anisotropy due to local exchange fields and spin–orbit coupling can be written as $I_{XMLD} \propto |\vec{m} \cdot \vec{E}|^2$, with $\vec{E}$ the electric field polarization and $\vec{m}$ along the magnetic axis. A $\cos^2\theta$ dependence is then expected for the magnetism-sensitive transitions. The feature C shows the largest dichroism and fits well with a $\cos^2\theta$ function, with a minimum at $\theta = 90^\circ$. From multiplet calculations it has been demonstrated that the situation when the polarization vector is perpendicular to the magnetic axis, $\vec{m} \perp \vec{E}$, corresponds to C minimum and B maximum [13,14]. This is obtained for $\theta = 90^\circ$ within an accuracy of a few degrees. We can then conclude that the Co spins are essentially parallel to the surface plane. As the FePt spin axis is perpendicular to the layer, the coupling between Co and Fe spins at the interface is orthogonal (Fig. 3).

Additional information on the orientation of the spins within the plane can be drawn from the relative variation of the different features. For the same relative orientations of $\vec{E}$ and $\vec{m}$, multiplet calculations show quite different XAS and XLD features when the spin axis $\vec{m}$ are along ⟨110⟩ or along ⟨100⟩ direction [13]. In the first case A and C features have the same variation with θ, in the second their variations are opposite. The experimental data show that A and C vary in the same way (Fig. 2b, inset). We deduce that the Co spins are pointing along the ⟨110⟩ direction.

4. Discussion

The relationship between the orientation of the magnetic moments and the sign of the crystal field effect has been demonstrated