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Influence of the crystal structure of thin Co films on X-ray magnetic linear dichroism—Comparison of *ab initio* theory and reflectometry experiments

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We report an investigation of the influence of the crystal structure of Co thin films on the X-ray magnetic linear dichroism (XMLD) spectrum. We compare XMLD spectra measured in reflection at the 3*p*-edges for two distinct orientations of the magnetization in the crystalline Co film with *ab initio* calculated spectra. The latter was computed for the face-centered cubic as well as the hexagonal-close packed crystal structures of Co. We find that the XMLD signal is strongly dependent on the magnetization direction with respect to the crystal axes as well as strongly influenced by the crystal structure. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4869292]

I. INTRODUCTION

Magneto-optical methods like the X-ray magnetic circular dichroism and the transversal magneto-optical Kerr effect are powerful tools to investigate ferromagnetic materials.[1] However, they are not very sensitive to the crystal structure. The X-ray natural linear dichroism (XNLD) and the near edge X-ray absorption (XANES), in contrast, are very sensitive to the orientation of the crystallographic structure. X-ray magnetic linear dichroism (XMLD) is both sensitive to the crystallographic structure as well as sensitive to the magnetization direction. Although XNLD in non-cubic crystals can be larger than XMLD, XNLD vanishes in cubic structures. Here, we concentrate on investigating the influence of the crystal structure on the XMLD signal. Compared to the commonly used total electron yield (TEY) technique, photon-in–photon-out measurements in reflection have the advantage that buried structures can be investigated[2] and, in addition, these can be used much easier with applied high magnetic fields. XMLD spectra measured using TEY at the 2*p* edges of transition metals and their compounds were successfully compared with *ab initio* calculations.[3–6] Recently, also the usefulness of XMLD measured in reflection was proven at the 3*p* edges of 3 *d* transition metals.[7] Nevertheless, investigations of the influence of the crystallographic structure itself on the XMLD are still missing. To this end, we have calculated the XMLD signal in reflection at the 3*p* (*M*) edge of Co for three different crystal structures as well as performed measurements on a thin crystalline Co film and have compared the computed and measured data.

II. METHODOLOGY

A. Calculations

The electronic structures (ES) of fcc and hcp Co were calculated with the WIEN2k code.[8] For the exchange-correlation term, the local spin density approximation[9] was used. The spin-orbit interaction (SOI) was included in a second-variation scheme.[10] The following parameters were employed: the energy cutoff, given as the product of the muffin-tin radius and the maximum reciprocal space vector was $R_{\mathrm{MT}}\ K_{\max} = 8.5$, the largest reciprocal vector in the charge Fourier expansion, $G_{\max}$, was set to 14 $\mathrm{Ry}^{1/2}$, and the maximum value of partial waves inside the muffin-tin spheres, $l_{\max} = 10$. The Brillouin zone sampling was done on grid of $27 \times 27 \times 27$ ($22 \times 22 \times 11$) *k*-points for the fcc (hcp) structure, whereas for the squared momentum matrix elements among all band and *k*-wave vectors combinations corresponding to the optical transitions,[11] a finer mesh of $81 \times 81 \times 81$ ($44 \times 44 \times 22$) was used, respectively. Careful Kramers-Kronig transformations (in-house code) were performed and finally the complex values of the permittivity tensor elements ϵ_{ij} were obtained. Subsequently, the optical response for the multi-layered model, here simply air/Co/air, with a 3 nm metal thickness, was described by our in-house developed optical program for solving the light propagation based on Yeh's formalism as extended in Ref. 12. The experimental lattice constants of $a_{hcp} = 2.51$ Å, $c_{hcp} = 4.07$ Å, and $a_{fcc} = 3.425$ Å were used.[13]

B. Experiment

XMLD-in-reflection (ΔR-XMLD) measurements were performed at BESSY II. The investigated sample was a 3 nm Co layer deposited on a crystalline GaAs substrate with a

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150 nm crystalline cubic Ag buffer layer on it. To prevent oxidation, the sample was capped with 3 nm Au. LEED measurements revealed a fourfold symmetry that can be related either to a fcc structure or to a hcp one with a $(11\bar{2}0)$ surface. The XMLD measurements were performed with the BESSY-Polarimeter[14] using linear polarized light of the undulator beamline U125-2-SGM2[15] with a spectral resolution across the Co $3p$ edge near 60 eV of $\Delta E/E = 3000$. The angle of incidence was set to near-normal incidence, i.e., $\theta = 85^{\circ}$ (see Fig. 1). The depicted crystallographic axes are crystal direction along [100], ([110]) and all equivalent directions and they belong to a cubic fcc structure with its orientation parallel to the sample edges. Two measurements were performed. The alignment of the electric field (**E**) was set parallel to the [110] direction in one case and normal to the [110] in a second one. The applied magnetic field with a magnetic flux density of about $B = 250\,\mathrm{mT}$ could be aligned perpendicular and parallel to plane of incidence, i.e., also perpendicular and almost parallel to **E**.

In both the measurements and *ab initio* calculations, ΔR-XMLD was determined in an analogous way. Specifically, for incident p-polarized light, the XMLD was calculated as $\Delta R\text{-XMLD}_p = [R_{pp}(M_{\perp}) + R_{pp}(-M_{\perp}) - R_{pp}(M_{\parallel}) - R_{pp}(-M_{\parallel})]/2$, where $M_{\perp}$, $M_{\parallel}$ denote the in-plane magnetization direction being perpendicular and parallel to the incident plane, respectively, and R_{pp} is the reflectance of p-incident light. Note that frequently the normalized XMLD signal is used. This is calculated as $\Delta R\text{-XMLD}/2\bar{R}$, with the average reflectivity $\bar{R}$ for all magnetization directions. However, to suppress artificial structures in spectral regions with a low reflectivity, we used ΔR-XMLD instead.

III. RESULTS AND DISCUSSION

First-principles calculations (not shown here) indicated that the XMLD anisotropy at the $3p$ edges is a consequence of the common interactions of the magnetic exchange of the valence (semicore) states of the $3p$ edges (M_2, M_3) and the cubic crystal-field splitting of the $3d$ density of states, i.e., to the largest extent of the spin-polarized parts of the e_g and t_{2g} states. The complexity stems from two major facts, first, the exchange splitting of the $3p$ states and their splitting due to the spin-orbit interaction are of the same magnitude[16] and second, such semicore states are not localized, but k-wave vector dependent. This is in contrast to the XMLD at the $2p$ (L_2, L_3) edges, where the core states are well localized, have no k-wave vector dependence, and the core level spin-orbit interaction is more than an order of magnitude larger than the core level exchange splitting.[3] A detailed analysis of the XMLD at the $3p$ edges of ferromagnetic $3d$ transition metals is given in Refs. 7 and 16.

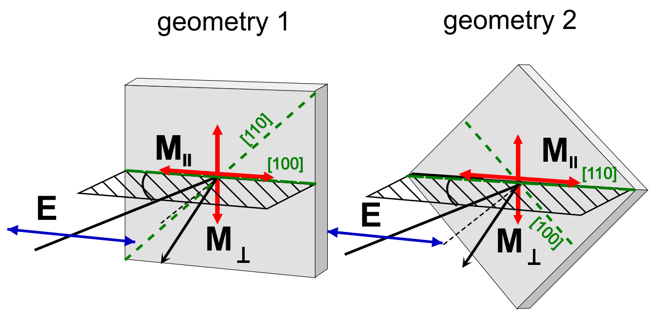

FIG. 1. Experimental geometries. The light is reflected off the sample at near-normal incidence ($\theta = 85^{\circ}$). The E-field vector of the light is aligned nearly parallel to either the fcc-crystal directions [100] or [110]. The sample is magnetized parallel and subsequently, perpendicular to the electric field vector to measure the XMLD.

In Fig. 2, we show our main result, ΔR-XMLD, for the $3p$ edges of fcc and hcp Co. The measured spectra (symbols) for **E** ∥ [100] (upper panel) and E ∥ [110] (lower panel) are given for fcc Co (see also Fig. 1). We find the signal to be strongly dependent on the magnetization direction (magneto-crystalline anisotropy) and to exhibit an inversion of the ΔR-XMLD spectra at the $3p$ edges when comparing **E** ∥ [100] and [110]. In the same way, our computed ΔR-XMLD spectra for fcc and for hcp Co with (0001) surface and $(11\bar{2}0)$ surface were processed. Note that within the hcp (0001) plane the primitive vectors ($\mathbf{a}_1$, $\mathbf{a}_2$) are 120° apart and therefore the [110] direction is a 60° away from $\mathbf{a}_1$ ($\mathbf{a}_2$). However, for the $(11\bar{2}0)$ surface, we have for the XMLD first that **E** is parallel to the $[1\bar{1}0]$ magnetization and then parallel to the magnetization along the $[14\,\overline{14}\,15]$ direction both containing an angle of 45°, see Fig. 3.

The calculated ΔR-XMLD spectrum of fcc Co (blue line in Fig. 2) shows an inversion of its spectral shape for **E** ∥ [100] and **E** ∥ [110], similar to what is measured, despite the double peak structure for the first lower lobe in the

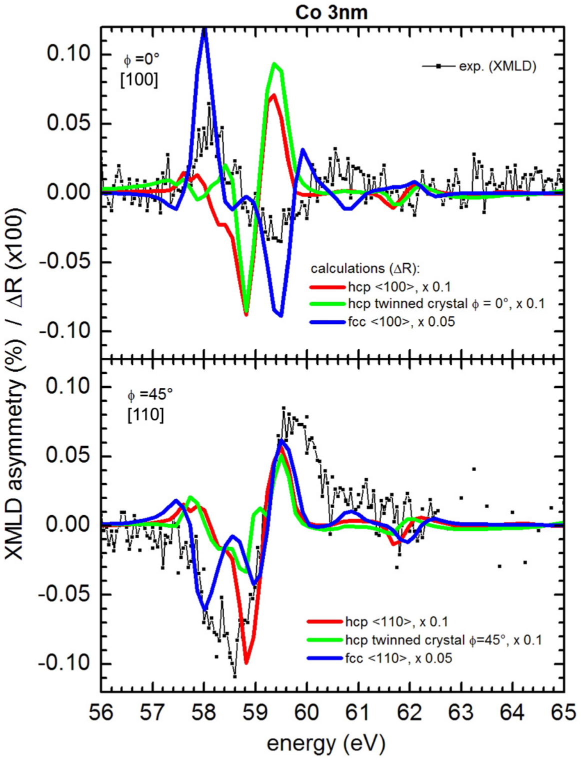

FIG. 2. The XMLD-in-reflection, measured at near-normal incidence ($\theta = 85^{\circ}$), for the magnetization directions **E** ∥ [100] (upper panel) **E** ∥ [110] (lower panel) for fcc Co and calculations for hcp Co within (0001) plane and within the $(11\bar{2}0)$ plane for the equivalent directions (see text). The calculations were shifted by 1.2 eV to align with the experimental edge and smeared with 0.27 eV.

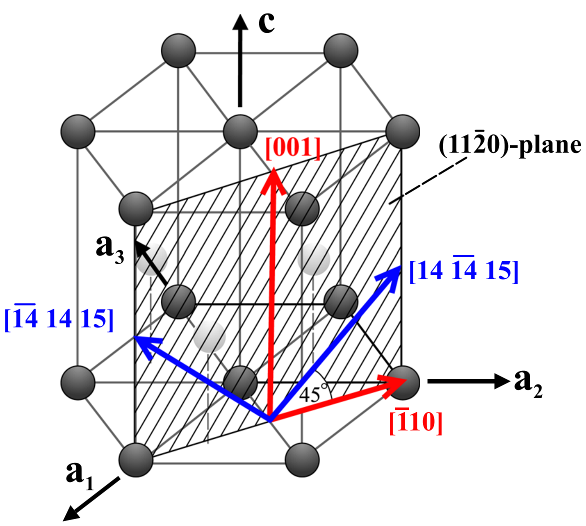

FIG. 3. The magnetization directions for the XMLD calculations within the $(11\bar{2}0)$ (surface) plane of hcp Co. For details, see text.

ΔR-XMLD ($\mathbf{E} \parallel [110]$) spectrum. For $\mathbf{E} \parallel [110]$, the calculated ΔR-XMLD well reproduces the second maximum on the high energy side.

The ΔR-XMLD spectrum of hcp Co with a (0001) surface (red curve in Fig. 2) does not show such a spectral change for $\mathbf{E} \parallel [100]$ and [110], as seen for the fcc structure. The absolute values of the calculated signals were reduced by a factor of 5×10^{-4} for hcp and 2×10^{-4} for fcc Co to fit the experimental data. This is done because the calculations were performed for perfect conditions, i.e., without considering roughness or additional broadening, which would decrease the reflectivity. Also interference effects may play a role here.

For a hcp Co phase with a $(11\bar{2}0)$ surface (see Fig. 3) and the electric field parallel to the sample edges, $\mathbf{E} \parallel [001]$, the four directions of the magnetization would be $[\bar{1}10]$, [001], $[1\bar{1}0]$, and $[00\bar{1}]$. For the $\mathbf{E}$-field orientated 45° to the sample edges, see Fig. 3, the magnetic field within the $(11\bar{2}0)$ plane is tilted by 45°, 135°, 225°, and 315° with respect to the c-axis. This corresponds approximately to the $[\bar{14}\ 14\ 15]$, $[14\ \bar{14}\ 15]$, $[14\ \bar{14}\ \bar{15}]$, and $[\bar{14}\ 14\ \bar{15}]$ directions. The calculated ΔR-XMLD of the parallel (45°) orientation is given in the upper (lower) panel of Fig. 2 (green curves). The ΔR-XMLD asymmetry here is about twice larger for the parallel than for the 45° orientation, but again we do not obtain a sign change in the ΔR-XMLD spectrum between the two orientations.

IV. CONCLUSIONS

On the basis of our first-principles calculations we can demonstrate the strong influence of the crystallographic structure on the angular dependence of the XMLD. Comparing the fcc phase and the hcp phase with a $(11\bar{2}0)$ surface, we could identify that only the fcc phase causes a change in the sign of the calculated XMLD spectra when rotating the orientation of the light's electric polarization by 45°, e.g., from $\mathbf{E} \parallel [100]$ to $\mathbf{E} \parallel [110]$. Our calculations for the fcc structure reproduce the spectral shape of the experimental data, which leads to the assumption that the prevailing phase of the sample is fcc Co. XMLD-in-reflection might therefore be an appropriate tool to distinguish crystal structures of buried layers with fourfold symmetry.

The spectral shape of the calculated ΔR-XMLD spectra is found to agree reasonably well with the measured ones despite the minimal smearing width or much larger intensity. A decrease of the calculated intensity and possibly also an improvement of the spectral shape towards better agreement with experimental data might be accomplished if more sophisticated multi-layered optical models, i.e., including capping and substrate layers are taken into account as well as inclusion of sample roughness and optical background.

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