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Publication Date
2008-11-25
Magnetic structure near the Co/NiO(001) interface

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(Dated: August 29, 2008)

Abstract

We investigate the magnetic coupling at the Co/NiO interface using soft x-ray magnetic linear dichroism (XMLD) and circular dichroism taking explicitly into account the recently observed angular dependence of the XMLD with respect to the crystallographic axes. We find that the Co moments are aligned perpendicular to the NiO moments. We discuss the impact of the anisotropic XMLD on the intensity ratio of the two peaks at the Ni $L_2$ edge, which is commonly employed to determine the spin orientation in antiferromagnets using XMLD.

PACS numbers: 76.60.Cn, 75.70.Cn, 78.70.Dm
Controlling magnetic coupling at interfaces is one of the great challenges in tailoring magnetic heterostructures, e.g., for information storage and processing applications, today. Exchange interaction between two ferromagnetic (FM) or a FM and an antiferromagnetic (AFM) layer leads to collinear coupling of the magnetic moments. However, the FM moments align perpendicular to the AFM moments in case of frustrated interface coupling when the exchange field due to the FM spins acts equally on the AFM sublattices [1]. The relative orientation of FM and AFM moments near the interface is influenced by e.g. the interface plane orientation [2], magnetic layer thicknesses [3], intermixing at the interface [4, 5], and strain induced magnetoelastic effects [2]. The interplay of all these effects add to the complexity of adequately predicting magnetic coupling at interfaces.

Here we reinvestigate the magnetic structure near the Co/NiO interface using soft x-ray magnetic linear and circular dichroism (XMLD and XMCD). For the Co/NiO(001) interface plane it is well established that the projected Ni moments, \( H_p \), are along \( \langle 110 \rangle \) directions [4]. It is less clear whether the actual moments, \( H \), are in-plane or make an angle with the (001) plane, e.g., point along \( \langle 112 \rangle \) directions as in bulk NiO. Reorientation of the NiO moments near the Co/NiO interface was reported by Ohldag et al. [4]. However, the exact mechanism is largely unexplored. The element-, site-, and valence-specificity of soft x ray dichroism techniques [6] allow a thorough characterization of magnetic interfaces elucidating the basic principles of magnetic coupling. Taking into account the recently discovered strong angular dependence of XMLD [7] we find that the Co moments are oriented perpendicular to the NiO moments. We emphasize the importance of the XMLD anisotropy on the intensity ratio of the two peaks at the Ni \( L_2 \) edge, that are often used to determine the spin orientation in antiferromagnets using XMLD [8–10].

Measurements were performed on a 2.3 nm thick Co film deposited onto a freshly cleaved NiO(001) single crystal surface [4] and subsequently capped with 1 nm Pd. We employed photoemission electron microscopy (PEEM) to study the domain structure of AFM NiO using XMLD at the Ni \( L_{3,2} \) edges and determined the Co domain pattern employing XMCD at the Co \( L_{3,2} \) edges. Domain images were acquired with an Elmitec PEEM [11] on the SIM beamline [12] at the Swiss Light Source. The spatial variation of the x-ray absorption (XA) coefficient was measured with a spatial resolution of \( \sim 50 \) nm. The 100% linearly polarized x rays impinged on the sample at a grazing angle of 16° and the polarization axis could be rotated continuously through a 90° range.
We employed the Ni $L_{3,2}$ XMLD to determine the in-plane orientation of the Ni moments near the Co/NiO(001) interface. The inset in Fig. 1(a) shows the ratio of two PEEM images obtained at the two most pronounced features in the Ni $L_2$ edge shown in Fig. 1(a), i.e., at 870.3 eV and 871.5 eV. The linear polarization $\mathbf{E}$ was oriented in the (001) surface plane along the [110] direction. Stöhr et al. [13] have shown that the observed contrast is of magnetic origin since it disappears at the NiO Néel temperature. Local XMLD spectra were obtained by monitoring the photon energy dependence of selected domains on the NiO surface [marked areas in inset to Fig. 1(a)]. The XA spectra are shown in Fig. 1(a) and their difference, i.e., the XMLD spectrum, is shown in Fig. 1(b). To interpret the observed dichroism, we compare our experimental data with calculated XMLD spectra [7]. In near cubic symmetry the angular dependence of the XMLD can be described by a linear combination of two fundamental spectra, $I_0$ and $I_{45}$ [14, 15]. Figure 1(c) and 1(e) show the calculated in-plane XMLD spectra, $I_{XMLD} = I_{XA}(\mathbf{E} \parallel \mathbf{H}) - I_{XA}(\mathbf{E} \perp \mathbf{H})$ for $\mathbf{E} \parallel [110]$ and $\mathbf{E} \parallel [100]$, indicated by $I_{45}$ and $I_0$, respectively. The subscripts denote the angle $\phi$ of $\mathbf{E}$ with the [100] direction. $\mathbf{H}$ denotes the orientation of the Ni spin axis (in remanence). Comparing the calculated spectra it is seen that at the $L_2$ edge $I_{45} \approx -I_0$, whereas the $L_3$ structure is different for $I_0$ and $I_{45}$. In order to determine unambiguously the spin direction both the $L_2$ and $L_3$ structure need to be considered.

For the projected moment $\mathbf{H}_p \parallel \langle 110 \rangle$ in the (001) plane the angular dependence of the XMLD is [14]

$$I_{XMLD} = I_{XA}(\mathbf{H}_p \parallel [110]) - I_{XA}(\mathbf{H}_p \parallel [1\overline{1}0]) = I_{45} \cos^2 \tau \sin 2\phi,$$

where $\tau$ is the (out-of-plane) angle between $\mathbf{H}_p$ and $\mathbf{H}$, while $\phi$ is the (in-plane) angle of $\mathbf{E}$ with respect to [100]. Interestingly, Eq. (1) shows that for $\mathbf{E}$ in the (001) plane and $\mathbf{H} \parallel \langle 110 \rangle$ always the pure $I_{45}$ spectrum is obtained. The measured $I_{45}$ (dark minus bright domains) is reproduced in Fig. 1(b) and the very good agreement with the calculated spectrum in Fig. 1(c) is evident. The XMLD signal given in Eq. (1) reverses for a rotation of either $\mathbf{H}$ or $\mathbf{E}$ by 90°, hence the sign of the XMLD determines which domains are parallel and which ones are perpendicular to $\mathbf{E}$. We use this to assign the in-plane Ni spin orientation of the domains. As indicated in Fig. 2(a) we find that the dark and bright domains correspond to Ni moments oriented parallel and perpendicular, respectively, to $\mathbf{E}$ (yellow double headed
To verify the assignment of the domains and to determine the angle $\tau$, i.e., the out-of-plane orientation of $\mathbf{H}$, we monitored the ratio $R_2$ — that is the XA intensity at 871.5 eV normalized to the value observed at 850.3 eV — while rotating the linear polarization $\mathbf{E}$ from out-of-plane ($\varepsilon = 0^\circ$) to in-plane ($\varepsilon = 90^\circ$), i.e., from the [001] to [110] direction. For the dark domains with spin oriented along the [110] direction, $R_2$ stays constant as shown in Fig. 3 by red (solid) dots although the relative orientation of $\mathbf{E}$ and $\mathbf{H}_p$ changes from perpendicular to parallel. For the bright domains with spin projection parallel to the x-ray beam, i.e., $\mathbf{H}_p \parallel [110]$, the rotation of $\mathbf{E}$ from $\varepsilon = 0^\circ$ to $90^\circ$ does not change the relative orientation of moments and polarization, it is always perpendicular. However, the $R_2$ ratio is not constant. As the experimental data shown by blue (open) dots in Fig. 3 indicates, a $\cos 2\varepsilon$ (i.e., $\sin^2 \varepsilon$) dependence [blue (thick) line] is observed.

The $\varepsilon$ dependence of the XA spectrum for the two different projected spin directions can be derived as [14]

$$ I_{\mathbf{H}_p \parallel [110]}^{\mathbf{E}}(\varepsilon, \tau) = B + \frac{1}{2} I_{45} (\cos^2 \tau \sin^2 \varepsilon - \sin 2\tau \sin 2\varepsilon), $$

$$ I_{\mathbf{H}_p \parallel [T0]}^{\mathbf{E}}(\varepsilon, \tau) = B - \frac{1}{2} I_{45} \cos^2 \tau \sin^2 \varepsilon, $$

$$ B = I_{\mathbf{E} \parallel [001]}^{\mathbf{E}}(\tau) + \frac{1}{4} I_0 (3 \cos 2\tau - 1) \sin^2 \varepsilon. $$

(2)

This demonstrates the strong angular dependence and varying $I_0/I_{45}$ contributions as a function of moment direction. Obviously, $I_{\mathbf{E} \parallel [001]}^{\mathbf{E}}(\tau)$ is the same for both domains. Substituting $I_{45} = -I_0$ (valid for the Ni$^{2+}$ L$_2$ edge [7, 16]) into Eq. (2) it can be verified that the $\varepsilon$ dependence of the dark domains only vanishes if $\tau = 0$, in which case

$$ I_{\mathbf{H}_p \parallel [110]}^{\mathbf{H}}(\varepsilon) = I_{\mathbf{H}_p \parallel [T0]}^{\mathbf{H}}(\varepsilon) = I_{\mathbf{E} \parallel [001]}^{\mathbf{H}} = \text{constant}, $$

$$ I_{\mathbf{H}_p \parallel [T0]}^{\mathbf{H}}(\varepsilon) = I_{\mathbf{E} \parallel [001]}^{\mathbf{H}} + I_0 \sin^2 \varepsilon. $$

(3)

Since the $R_2$ value of $I_0$ is positive [c.f., Fig. 1(e)] also the experimental $\varepsilon$ dependence of the bright domains is confirmed.

Additional evidence of the domain orientation is obtained by studying the $I_0/I_{45}$ composition of the XMLD. Using Eq. (2) we can extract the $I_0$ spectrum, since we precisely know the $I_{45}$ spectrum, which is measured pure in the (001) plane [c.f., Eq. (1)]. The extracted $I_0$
spectrum is displayed in Fig. 1(d) and shows a good agreement with the calculated spectrum in Fig. 1(e).

Finally, we discuss the magnetic coupling between the Co and NiO moments. Indeed, a main reason to determine the correct orientation of each NiO domain is to be able to determine unambiguously the relative orientation of the Co and NiO moments. We determined the Co spin direction of the domain structure using XMCD. Figure 2(b) displays the ratio of two PEEM images obtained with left and right circularly polarized x rays at the Co $L_3$ edge. The Co moments are collinear with the x-ray beam in areas that appear in white and black, while grey areas indicate perpendicular alignment of the Co moments to the x-ray beam. Comparing the NiO domains in Fig. 2(a) with the Co domains in Fig. 2(b) indicates spin-flop coupling, i.e., the Co moments are aligned perpendicular to the Ni moments. This is expected for a magnetically compensated surface like the NiO(001) [17]. While a fraction of an atomic layer of uncompensated Ni moments is present at the interface, Stiles et al. [17] have shown that its presence does not necessarily lead to parallel FM/AFM exchange coupling. The impact of oxidation/reduction reactions and magnetoelastic effects at the NiO interface will be discussed elsewhere [18].

To summarize, we revisited the magnetic coupling at the Co/NiO interface using spectroscopy taking into account the angular dependence of the XMLD. The two orientations $H \parallel \langle 100 \rangle$ and $\langle 110 \rangle$ in the (001) plane are each associated with a unique $L_{3,2}$ XMLD spectrum, i.e., $I_0$ and $I_{45}$, respectively. For these two spectra the $L_2$ structure has opposite sign, whereas the $L_3$ structure is distinct. From the angular dependence we establish that with $E$ in-plane we measure the pure $I_{45}$, where the sign of the XMLD unambiguously determines the orientations of the different NiO domains. The orientations are further confirmed by rotating $E$ out-of-plane. The angular dependence and relative $I_0/I_{45}$ contributions of the XMLD reveal that, in our samples near the Co/NiO interface, the Ni moments are oriented in the surface plane, without significant out-of-plane contribution. Finally, the measured Co $L_{2,3}$ XMCD shows that the Co moments of each domain are perpendicular to the NiO moments, hence demonstrating spin-flop coupling.

The Advanced Light Source is supported by the Director, Office of Science, Office of Basic Energy Sciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231. The authors thank Andreas Scholl for the sample preparation.
This relation is a peculiarity of the Ni$^{2+}$ L$_2$ edge and not generally true for other transition metal L$_2$ edges [14, 15].
FIG. 1: (Color online) (a) Local Ni$^{2+} L_{3,2}$ XA spectra and NiO domain pattern (inset). Spectra obtained in the areas indicated by the blue (dashed) circle and red (drawn) circle are shown by curves with the blue (open) dots and red (solid) dots, respectively. The ⟨100⟩ directions of the NiO(001) plane and the orientation of the linear x-ray polarization, $E \parallel [110]$, are indicated in the inset. (b) Experimental XMLD $I_{45}$ spectrum obtained as the difference between the two local XA spectra shown in (a). Also shown (d) the experimentally obtained XMLD $I_0$ spectrum and (c) and (e) results of multiplet calculations for the Ni$^{2+} L_{3,2}$ XMLD $I_{45}$ and $I_0$ spectra.


FIG. 2: (Color online) Comparison of PEEM images of (a) antiferromagnetic NiO domains and (b) ferromagnetic Co domains in 2.3 nm Co/NiO(001). The arrows indicate the magnetic orientations and directions. A perpendicular coupling of the ferromagnetic and antiferromagnetic layers is observed.

FIG. 3: (Color online) Ni\textsuperscript{2+} L\textsubscript{2} peak intensity ratio, R\textsubscript{2}, as a function of the orientation of the x-ray linear polarization relative to the sample normal, i.e., E is along the [001] surface normal for $\varepsilon = 0^\circ$, while E is in the sample surface plane for $\varepsilon = 90^\circ$. The dependence obtained in the area marked by the blue (dashed) and red (drawn) circle is shown by the blue (open) and red (solid) dots, respectively. The blue (thick) line is a cos 2$\varepsilon$ fit and the red (dashed) line has a constant value.