Correlation between Crystallographic and Magnetic Domains at Co/NiO(001) Interfaces

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Abstract

Using soft x-ray spectromicroscopy we show that NiO(001) exhibits a crystallographic and magnetic domain structure near the surface identical to that of the bulk. Upon Co deposition a perpendicular coupling between the Ni and Co moments is observed that persists even after formation of uncompensated Ni spins at the interface through annealing. The chemical composition at the interface alters its crystallographic structure and leads to a reorientation of the Ni moments from the $\langle 112 \rangle$ to the $\langle 110 \rangle$ direction. We show that this reorientation is driven by changes in the magnetocrystalline anisotropy rather than exchange coupling mediated by residual uncompensated spins.

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Elucidating the mechanisms behind the magnetic ordering and coupling at interfaces is crucial for the understanding of complex magnetic systems, such as nanomagnet assemblies and magnetic heterostructures. One particular area of interest concerns antiferromagnetic (AF) surfaces and their interfaces to ferromagnetic (FM) layers. Since AF systems are not sensitive to external fields they can induce an uniaxial anisotropy in an FM layer through AF/FM exchange coupling. By using such an interface it is possible to "pin" the direction of the FM layer magnetization via unidirectional exchange bias and to create a magnetic reference layer[1,2]. To achieve a deeper understanding of magnetic surfaces and interfaces it is important to reliably determine their spin structure and to correlate it to the chemical and structural properties. Today, magnetic dichroism in soft x-ray absorption (XA) has been established as an excellent tool to investigate magnetic interfaces with element-, valence-, and site specificity[3]. X-ray magnetic circular dichroism (XMCD) is sensitive to (unidirectional) FM order, while x-ray magnetic linear dichroism (XMLD) can also detect (uniaxial) AF order. A crystalline electric field with cubic symmetry induces only a weak angular dependence in XMCD spectra[4] but causes a very pronounced anisotropy in XMLD spectra. The spectral shape of the XMLD depends on the relative orientation of x-ray polarization vector, magnetic moments, and crystallographic axes[5]. Furthermore, non-magnetic sites with a distorted local cubic symmetry can give rise to an x-ray linear dichroism (XLD) as a consequence of to the search light effect[6].

Systems containing AF ordered NiO have been extensively studied in the past and are considered to be prototypical for AF/FM exchange coupling. NiO has a rather simple crystallographic structure and its magnetic order in the bulk is well established[7]. Nevertheless, AF surfaces and interfaces can show unexpected properties, e.g., the magnetically compensated NiO(001) surface experiences a spin reorientation upon deposition of an FM Co layer[8]. In this Letter we will present an x-ray dichroism spectromicroscopy study of the bare NiO(001) surface and the Co/NiO(001) interface to address the origin of the spin reorientation. Using Ni XMLD and O XLD, we find a crystallographic and magnetic configuration near the NiO(001) surface identical to that of the bulk, exhibiting an easy magnetic axis parallel to $\langle 112 \rangle$. Upon deposition of a thin Co layer and subsequent formation of a thin layer of uncompensated Ni moments, we find that the magnetic moments in NiO and Co are aligned perpendicular to each other along NiO $\langle 110 \rangle$ crystal axes. A careful analysis of the O XLD images indicates that the NiO surface in Co/NiO exhibits a tetragonal distortion

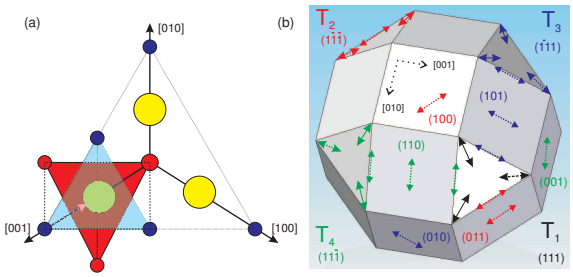

FIG. 1: (color online) (a) Atomic and spin structure in bulk NiO. Red and blue dots represent Ni atoms of opposite spin, while yellow dots represent O atoms. Antiferromagnetic order is caused by superexchange interaction along the Ni-O-Ni bond in the $\langle 100 \rangle$ direction, so that the net moment on each O site vanishes. Ni spins within a particular $\{111\}$ sheet are aligned parallel (red and blue planes). (b) Possible orientations of the *T(win)* domains, labeled T_1 to T_4, and walls in NiO. The possible spin axes in each T-domain are shown by double headed arrows with the same color. Also shown are the spin axes and domain wall orientations for domains walls between T_1 shown in black and the three other T-domains.

instead of the rhombohedral contraction that is typically observed in the bulk of the material. We therefore conclude that the reorientation is driven by strain-induced changes in the magnetocrystalline anisotropy at the interface, rather than the AF/FM exchange coupling mediated by residual uncompensated spins, as initially suggested[8].

To determine the Co and Ni spin structures as well as to image the crystallographic domains we used photoemission electron microscopy (PEEM). PEEM is a full-field imaging technique where the lateral distribution of the secondary electrons is magnified using electrostatic or magnetic lenses onto a two-dimensional electron detector[9]. Since the secondary electron yield is proportional to the XA cross section, a direct image of the lateral variation is obtained. The PEEM2 endstation installed on bending magnet beamline 7.3.1 at the Advanced Light Source allowed us to explore the dichroic contrast mechanisms to determine the NiO(001) surface and the Co/NiO(001) interface structure. Images and spectra shown in this manuscript were acquired with linear or circular polarized x-rays, whereas the degree of circular polarization was 75%. For our studies we prepared a NiO(001) surface by ex situ cleaving of a bulk single crystal. After insertion into the UHV system the surface was cleaned in situ by annealing at 450 K for 30 min.[10]. A 3 nm Co film was deposited onto the

clean NiO(001) surface at ambient temperature using a deposition rate of ∼0.2 nm/min. in a background pressure better than 10^{-9} mbar.

We will first introduce the possible crystallographic and magnetic domains in AF ordered bulk NiO. The AF superexchange interaction of the Ni-O-Ni bonds along ⟨001⟩ directions leads to the formation of FM ordered {111} planes, where spins in adjacent {111} planes are aligned antiparallel, c.f., Fig. 1(a). Below the Néel temperature ($T_N \approx 525$ K) the cubic NiO crystal experiences a contraction along the ⟨111⟩ direction due to exchange striction. As a result the coordination around each O and Ni site becomes slightly rhombohedral with the symmetry axis parallel to ⟨111⟩[11,12]. Due to this lattice contraction the initially cubic crystal splits up into four crystallographic *T(win)* domains, T_1 to T_4, as shown in Fig. 1(b). The spin within each T-domain can be aligned either along ⟨112⟩ (parallel to a bisector of the triangle) or ⟨110⟩ (parallel to an edge of the triangle). It was shown by Yamada *et al.*[11,12] that within the domains the easy axis is aligned along ⟨112⟩ and that in the domain walls the spin aligns along the ⟨110⟩. Possible domain walls are limited since two adjacent T-domains need to share a common ferromagnetic plane which has to be a mirror plane.

To determine the structural domain pattern of the bare surface, we acquired local O K edge XA spectra from two different domain areas on the NiO(001) surface. The spectra together with the difference signal (i.e., the XLD) are shown in Fig. 2(a). The domain images in Fig. 2(b) and 2(c) represent the difference normalized to the sum of two images taken at the photon energies labeled A and B in the spectrum[13]. Since each O atom is surrounded by six Ni atoms whose moments are antiferromagnetically aligned, the O XLD shown in Fig. 2(a) can not be due to a magnetic effect. Kinoshita *et al.*[14] showed that the XLD is due to the rhombohedral crystal distortion and can therefore be employed to map structural domains in the surface near region.

The domain pattern in Fig. 2(b) was obtained with purely linear polarization parallel to the surface plane, which allows us to distinguish between T-domains that have different in-plane components of the domain axis. The image reveals two different areas separated by wall planes parallel to the [001] and [010] directions. Because the angle between polarization vector and ⟨111⟩ axis is 39° for $T_{1,2}$ domains and 75° for $T_{3,4}$ domains, we conclude that the two different areas contain these two sets of T-domains. To further investigate the surface near crystal structure, we determined the domain pattern using elliptically polarized radiation providing both out-of-plane and in-plane components of the polarization vector

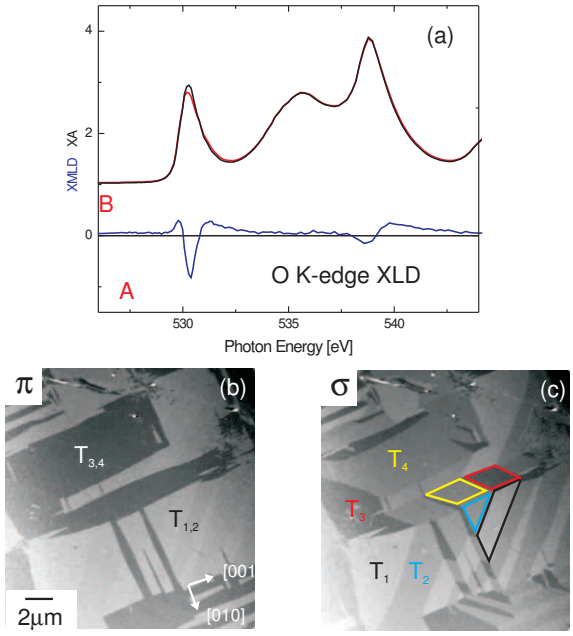

FIG. 2: (color online) O K edge XA spectra and PEEM images from a clean NiO(001) surface. (a) O K spectra (top) obtained using linearly polarized x-rays (π) from T_1 and T_3 domain areas of the NiO(001) surface as indicated in panel (c) together with the difference spectrum in blue (grey) below. The absorption intensity and the difference are plotted in arbitrary units. (b) PEEM images obtained using π polarized x-rays. (c) Domain image measured with elliptically polarized x-rays (σ). Images obtained with π-polarized x-rays are only sensitive to the in-plane orientation of the domain axis, while images obtained with σ-polarized radiation also reveal the out-of-plane orientation.

(see Ref. 10). The results are shown in Fig. 2(c), where initially dark areas are now split up into dark and grey domains, while initially bright areas are split up into white and lighter grey domains. These out-of-plane domains are separated by walls parallel to (110) and $(1\bar{1}0)$. Taking into account the orientation of the observed domain walls we can now assign each area in the image to a particular T-domain in accordance with Fig. 1(b). The observed contrast follows nicely the angle of the effective polarization vector and the postulated T-domain pattern[15]. We conclude that the structural T-domain pattern observed at the surface of NiO(001) is compatible with the bulk crystallographic structure.

We now employ a combination of O XLD, Ni XMLD, and Co XMCD images to follow

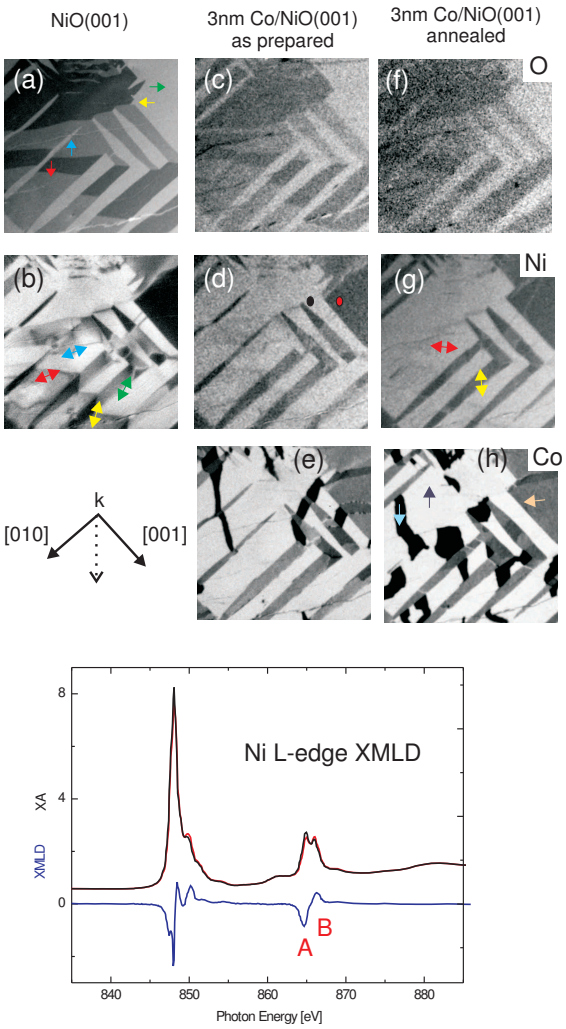

FIG. 3: (color online) Crystallographic, antiferromagnetic, and ferromagnetic domain structures near the NiO(001) surface and Co/NiO(001) interface. Images obtained by O K XLD (first row), Ni L_2 XMLD (second row), and Co L_2 XMCD (third row) for the clean NiO surface (first column), the exchange coupled Co/NiO bilayer (second column), and after annealing at 550 K for 20 min. (third column). Bottom panel shows the Ni $L_{3,2}$ XA spectra obtained from different AF domains in panel(b) and the difference spectrum in blue (grey) below. XA intensities and their difference are plotted in arbitrary units.

the evolution of the NiO and Co magnetic domain structures upon deposition of the 3 nm Co film onto the NiO(001) surface and to evaluate the impact of subsequent annealing. The as prepared NiO(001) surface is represented in the first column of Fig. 3 with the structural

domains obtained at the O K edge [Fig. 3(a)] and the antiferromagnetic domain structure obtained at the Ni L_2 edge [Fig. 3(b)] .

Three levels of contrast are observed in the O XLD image in the geometry with the x-ray angle of incidence of 30° and the x-ray polarization at 45° to the [100] axis. We find that dark (grey) areas are indicative of T_2 (T_1) domains, while T_3 and T_4 domains can not be distinguished and appear in bright grey. A map of the local orientation of the Ni moments [Fig. 3(b)] was obtained by determining the ratio A/B of the signals measured at photon energies labeled A and B in the Ni XA spectrum displayed at the bottom of Fig. 3. With the polarization vector oriented at 45° relative to the [100] axis, bright (dark) domains correspond to Ni moments oriented parallel (perpendicular) to the incoming x-ray beam direction[5]. The in-plane orientation of the AF spin axis obtained from the XMLD contrast is indicated by colored arrows in the figure. Overall, the magnetic domain pattern matches nicely the crystallographic domain pattern derived from the O XLD. In addition, we can distinguish the two domains T_3 and T_4, since the AF spin axis is parallel to {211} and hence the contrast does not disappear in this geometry. Our interpretation of the NiO L_2 XMLD allowing for its angular dependence indicates that the domain pattern near the sample surface is compatible with the spin arrangement expected for a bulk-truncated NiO(001) surface in contrast to earlier results that did not take the anisotropic XMLD into account[8].

Deposition of 3 nm Co onto the NiO(001) surface does not qualitatively change the crystallographic domain pattern derived from the O XLD. The overall contrast shown in Fig. 3(c) is reduced as expected due to attenuation of the O K signal by the Co layer: the contrast between the brightest (white) and darkest (black) domains is reduced from 16% to 5%, while the contrast between domains with intermediate brightness (gray) and white domains is reduced from 7.5% to 2.5%. The Ni L_2 XMLD contrast [Fig. 3(d)] is also reduced by a factor of three upon Co deposition but the four domains of different brightness can still be observed. Altogether we do not observe any changes in the NiO domain structure upon Co deposition.

Figure 3(e) shows the contrast derived as the ratio of the Co L_3 and L_2 intensities obtained using circular polarization. Domains with the Co moment parallel to the x-ray beam appear in black and white, while grey areas indicate domains with the Co moment perpendicular to the x-ray beam. Careful, domain by domain, comparison of the Co and

NiO images in the entire field of view reveals that the in-plane components of the Co and Ni moments are aligned perpendicular. The Ni L_2 XMLD and hence the AF NiO domain structure remains unchanged. We did not detect any XMCD at the Ni $L_{3,2}$ edge on the as prepared samples suggesting that the AF surface is indeed magnetically compensated and the observed perpendicular coupling is expected in this case.

To further investigate the AF/FM coupling at the Co/NiO(001) interface, the sample was annealed at 550 K for 20 min. leading to the formation of a 1-2 ML thick $CoNiO_x$ interface alloy[16]. A small Ni XMCD signal (not shown) indicative of the presence of uncompensated Ni moments and the formation of a partially magnetic uncompensated interface is now detected but the relative orientation between moments in the two layers — the AF NiO in Fig. 3(g) and the FM Co in Fig. 3(h) — remains perpendicular. This shows that the presence of uncompensated moments alone does not lead to parallel alignment of AF and FM spins. Our findings are supported by theoretical models analyzing the relative alignment between the AF and FM moments at interfaces[17]. They predict for large AF grains - like single crystals in our case - perpendicular spin coupling.

We find that upon annealing the Ni spins undergo a reorientation from the $\langle 121 \rangle$ direction towards $\langle 011 \rangle$ in plane direction. We propose that this spin reorientation is due to strain induced by the formation of the $CoNiO_x$ interface alloy. While the lattice constant of NiO is 4.176 Å, the lattice constant of CoO is 4.26 Å. Consequently, formation of $NiCoO_x$ at the interface will induce compressive strain in the NiO lattice near the surface. Our conclusion is supported by calculations by Finazzi *et al.*[18] evaluating the impact of uniform strain on the magnetic order in NiO thin films. They found that compressive tetragonal strain preferentially stabilizes domains with out-of-plane spin orientation while tensile strain leads to in-plane spin orientation. This is consistent with our observation of an in-plane orientation of the Ni moments upon annealing, i.e., in the case of compressive strain in the surface near region of NiO(001).

To experimentally confirm the origin of the reorientation, we closely evaluate the contrast observed at the O K edge after annealing [Fig. 3(f)]. Compared to the as prepared interface we find that the contrast in the O XLD images across a (001) wall remains approximately the same at 4.5%, while contrast across a (011) wall is reduced by a factor of 5 to 0.5%. This disappearance of the $\{011\}$ walls is strong evidence that the crystallographic structure at the interface changes from a rhombohedral contraction along the $\langle 111 \rangle$ directions, found

in pure NiO, to tetragonal contractions along [100], similar to CoO. Consequently, only T-domains separated by {001} walls remain as they exist in CoO. Since the easy axis in CoO is close to $\langle 011 \rangle$[19] we conclude that the spin reorientation at the Co/NiO interface is driven by strain-induced changes in the magnetocrystalline anisotropy at the Co/NiO interface.

In summary, using soft x-ray spectromicroscopy we showed that NiO(001) exhibits a crystallographic and magnetic domain structure near the surface that is identical to that of the bulk. Upon Co deposition perpendicular coupling of Co and Ni moments is observed. The perpendicular coupling persists even after the formation of uncompensated Ni moments at the interface induced through annealing. The $CoNiO_x$ at the interface has a different crystallographic structure than the sharp interface and causes a reorientation of the Ni moments from $\langle 112 \rangle$ to $\langle 110 \rangle$. Our results provide evidence that the reorientation is driven by strain-induced changes in the magnetocrystalline anisotropy rather than the exchange coupling mediated by residual uncompensated spins as has been previously suggested[8].

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