Magnetic coupling across highly oriented oxidic interfaces: Fe_3O_4/NiO

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NiO thin films on Fe_3O_4 represent a fully oxidic model system for proximity effects at an antiferromagnet/ferromagnet interface. Our microspectroscopy studies reveal a strong influence of the crystallographic interface orientation on the magnetic coupling. Both the {110} and the {111} interfaces produce collinear coupling, while at the {001} interface a spin-flop coupling is observed. These differences are caused by the specific interfacial bonding and strain situation.

Two distinct magnetic materials in direct contact will experience a strong coupling across the interface mediated through short-ranged exchange interactions. This magnetic coupling causes a change in the magnetic properties of the individual constituents – a complex situation, which is sometimes termed “proximity effect”. The latter can have different facets reflected in a change of the magnetic anisotropy, the magnetic moments, the local spin configuration, or the magnetic ordering temperature. The interaction between a ferromagnet (FM) and an antiferromagnet (AF) gives rise to a particular proximity effect, which is known as *exchange bias* and exhibits all of the features mentioned above. Discovered in the 1950s by Meiklejohn and Bean [1], it has in the meantime become a key ingredient for the fabrication of high-density hard disks, magnetic-field sensors, and more sophisticated spintronic devices [2, 3].

A central aspect of the magnetic proximity effect is the crystallographic structure of the boundary region between the two materials, which may influence the interfacial coupling in two ways:

i) Breaking of translational symmetry causes electronic effects, for example, an anisotropic exchange interaction, altered crystal-field symmetry, and electronic hybridization across the interface. Those effects will be generally confined to the interface region.

ii) Epitaxial lattice mismatch causes magnetoelastic effects, which depend on the orientation of the planar epitaxial strain with respect to the crystal lattice. The strain-induced effects will extend farther into the sample.

In our work we addressed the question of how the crystallographic orientation influences the FM/AF magnetic proximity effect via exchange coupling and magnetoelastic effects in the specific system NiO on Fe_3O_4. We can distinguish two basic coupling geometries in FM/AF systems, namely, collinear and spin-flop coupling. In the latter configuration, the AF spin axis orients by 90° with respect to the FM.

The spectromicroscopy studies were carried out by means of a photoemission microscope, using x-ray magnetic circular (XMCD) and linear dichroism (XMLD) for the imaging of the domain structures in the FM and AF, respectively. The analysis of the spectroscopic data must take into account the delicate anisotropy of the XMLD signal in single-crystalline systems [4]. In spite of these complications it is possible to analytically calculate the absorption spectra for arbitrary orientations of the AF spin

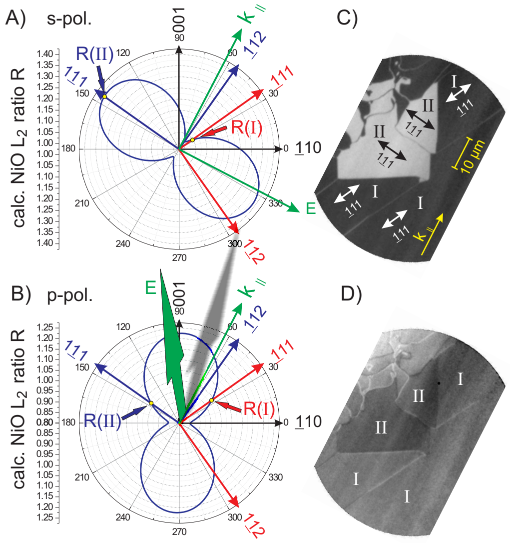

FIG. 1: 35 monolayers of NiO on Fe_3O_4(110). Polar plots in (A) and (B): calculated L_2 intensity ratio for every possible in-plane direction of the spin quantization axis, for the polarization in s- (A) and p-geometry (B). The plots include crystallographic directions for collinear and spin-flop coupling, as well as the surface projection of light incidence $\mathbf{k}_\parallel$ and light polarization $\mathbf{E}$. The calculated magnetic contrast for a given spin orientation in the sample plane is defined by its intersection with the L_2-ratio curve. (C) and (D): PEEM images. For collinear coupling we assign the axis $[\underline{1}11]$ to set I and $[1\underline{1}1]$ to set II. Conversely for spin-flop coupling, the assignment is $[1\underline{1}2]$ for set I and $[\underline{1}12]$ for set II. Only the collinear case matches the theoretical prediction, with set II being brighter in s and slightly darker in p geometry [points R(I) and R(II) in (A) and (B)].

quantization axis and for *s*- and *p*-polarized light, using only few fundamental spectra gained from atomic multiplet calculations (symmetry-adapted basis set). This allows a convenient comparison to the experiment, minimizing the computational effort.

A result of this procedure is shown in Fig. 1 for the example of the {011} interface. For clarity, we anticipate that the XMCD in the Fe_3O_4 substrat yielded two different $\langle 111 \rangle$-type in-plane easy-axes for the domains, and that the exchange coupling limits the NiO spin axis alignment to the sample plane as well. Furthermore we classify the coupling angle in term of collinear (near 0°) or spin-flop (near 90°). The blue dumbell-shaped curves in panels (A) and (B) depict the angular dependence of the XMLD signal with respect to the crystalline axes. For the comparison with the experiment we define two sets of domains (I and II) with different contrast levels in the panels (C) and (D), each type correlated with an easy-axis in the substrate. As apparent in the PEEM images, this domain contrast inverts, when going from s- to p-polarized light, but the relative contrast between type I and type II domains is smaller than in the s-polarized case. The expected contrast level for an arbitrary spin quantization axis in the film plane is given by its intersection with the blue contrast curve. From inspection of (A) it becomes clear that a spin-flop coupling should yield a bright contrast for set I and a dark contrast for set II, quite opposite to the experimental observation. A similar contradiction is encountered for p-polarized light. The experimental data can be explained consistently only if a *collinear* coupling between FM and AF is assumed [4, 5].

At first glance, this result contradicts Koon's theory [6] which predicts spin-flop or collinear coupling, if the AF surface termination is compensated or uncompensated, respectively. In our case, we thus would expect spin-flop coupling for a ferromagnet in contact with the compensated {110} and {001} NiO surfaces. In our experiments we find spin-flop coupling only for the {001} interface and collinear coupling for {110} and {111}. In order to understand this discrepancy, we have to consider that the interface has two sides, i.e., a NiO and an Fe_3O_4 side. We must take into account the full crystalline and magnetic structure of the transition Fe_3O_4 (ferrimagnetic, spinel) $\rightarrow$ NiO (two sublattices, rocksalt). In all three crystalline orientations investigated, one can find a configuration where the magnetic unit cells of NiO and Fe_3O_4 match. This implies that the two magnetic sublattices (spin up and spin down) of NiO will experience different magnetic environments at the interface. Consequently, in a more realistic picture the interface cannot be compensated anymore and a tendency for collinear coupling should result. In fact, the microspectroscopy studies of the Ni L_3 XMCD signal reveal different amounts of uncompensated magnetic moments in NiO at each interface orientation (Fig.2). We note that these uncompensated moments may also result from a reconstruction of the interface, leading to the formation of an interfacial $NiFe_2O_4$ (NFO) phase.

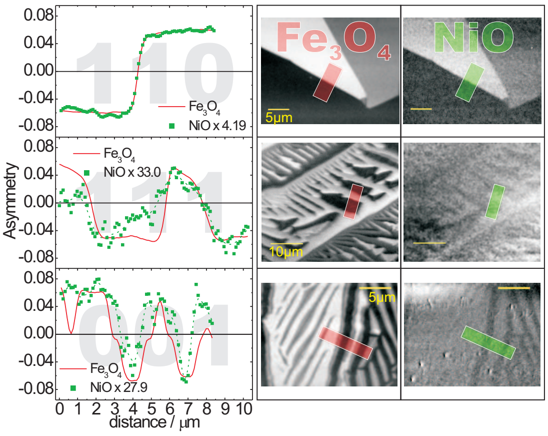

FIG. 2: Comparison of the amount of uncompensated magnetization at the interface for the three orientations {110}, {111}, and {001}. For comparison, the NiO XMCD has always been scaled to match the size of the Fe_3O_4 XMCD.

The extreme case is the {111} interface, where locally only one type of AF sublattice meets the interface (locally uncompensated). In principle, partial compensation is realized for all interface orientations investigated in this study, rendering spin-flop coupling unlikely. Experimentally, however, we find perpendicular coupling for the {001} interface, contradicting the structural argument given above.

This indicates that a further mechanism must be at play, favoring a perpendicular over a collinear spin arrangement, thereby overcoming the influence of exchange interactions and the particular interface structure. Our analysis shows that magnetoelastic effects can indeed lead to spin-flop coupling at the {001} interface if the magnetoelastic energy gain dominates over interfacial exchange coupling. It is interesting to note that for $NiFe_2O_4$ coupled to magnetite, the situation would be the same as for NiO – so spin-flop coupling could result even for a NFO-type reconstructed zone at the interface.

Further details may be found in Refs. [4, 5].

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