Element-specific study of the anomalous magnetic interlayer coupling across NiO spacer layer in Co/NiO/Fe/Ag(001) using XMCD and XMLD

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Co/NiO/Fe trilayers are grown on Ag(001) substrate using molecular-beam epitaxy and investigated by element-specific magnetic domain images using x-ray magnetic circular dichroism and x-ray magnetic linear dichroism techniques. By comparing the Co, Fe, and NiO magnetic domain images, we identify that the anomalous Co-Fe interlayer coupling from a 90° coupling to a collinear coupling with increasing the NiO film thickness is due to a transition from a collinear to 90° coupling at the NiO/Fe interface while retaining a 90° coupling at the Co/NiO interface. Uncompensated Ni spins are found at the Co/NiO interface but are absent at the NiO/Fe interface. No evidence of spiral NiO spin structure is found in this Co/NiO/Fe sandwich.

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The discovery of magnetic interlayer coupling between two ferromagnetic (FM) layers across a thin spacer layer[1] led to the discovery of the giant magnetoresistance (GMR) (Ref. 2) and has thereafter been stimulating a great activity in nanomagnetism research. Depending on the spacer-layer material, the interlayer coupling mechanism is quite different. For metallic spacer layers, whereas the GMR effect was discovered, the interlayer coupling has been extensively studied and the oscillatory interlayer coupling[3] are identified to be associated with the quantum-well states in the spacer layer at the Fermi level.[4,5] For insulating spacer layers, whereas the tunneling magnetoresistance[6,7] was discovered, the coupling mechanism has not been clearly understood.[8–10] In particular, the interlayer coupling across an antiferromagnetic (AF) insulating spacer layer is very ambiguous and confusing.[11,12] An anomalous 90° interlayer coupling across a thin AF NiO layer, which has become a model system for the study of this subject, was discovered in $Fe_3O_4/NiO/Fe_3O_4$,[13] $Ni_{80}Fe_{20}/NiO/Co$,[14] and Fe/NiO/Fe (Ref. 15) systems, and it is shown that this observed 90° coupling is different[15] from the conventional Slonczewski's 90° coupling in metallic systems.[16] Different and contradictious mechanisms have been proposed to account for this anomalous interlayer coupling across the NiO spacer layer. For example, the 90° coupling was proposed to be due to a spiral rotation of the NiO spins in the spacer layer by keeping a collinear magnetic coupling at both FM/AF microscopy interfaces,[13] whereas another proposal assumes a collinear NiO spin structure but leaving the interfacial coupling to be collinear and perpendicular at the two FM/AF interfaces, respectively.[14] Obviously, the key to resolve the coupling mechanism is a direct measurement of the NiO spin structure in the coupled magnetic sandwiches. Experimentally, this was not possible until the development of the x-ray magnetic linear dichroism (XMLD) technique which could probe the AF spin direction in certain AF materials.[17–20] In fact, XMLD technique has greatly advanced our knowledge in FM/AF bilayer systems in which the so-called exchange-bias effect was discovered.[21,22] In this Brief Report, we report a study of Co/NiO/Fe/Ag(001) trilayers using photoemission electron microscopy (PEEM) technique. By a direct comparison of the Co, Fe, and the NiO magnetic domains, we are able to resolve the confusing issue of the anomalous interlayer coupling across the NiO spacer layer. Specifically, we find that the anomalous transition of the Co-Fe interlayer coupling from a 90° coupling to a collinear coupling with increasing the NiO thickness is due to the fact that the NiO/Fe interfacial coupling undergoes a transition from a collinear to a 90° coupling while retaining a 90° coupling at the Co/NiO interface. We also identified uncompensated Ni spins at the Co/NiO interface but found no evidence of the NiO spiral spin structure.

A Ag(001) single crystal is prepared by Ar ion sputtering and annealing in an ultrahigh vacuum system.[23] A 15 monolayer (ML) Fe film was deposited on top of the Ag(001) substrate followed by a wedged NiO film (0–4 nm) grown by a reactive deposition of Ni under an oxygen pressure of 1×10^{-6} Torr. Low-energy electron-diffraction (LEED) measurement after the NiO growth shows the formation of single crystalline NiO film, confirming the epitaxial growth nature of the NiO film on Fe(001).[24,25] Then a 2-nm-thick Co film was deposited onto half of the NiO wedge. This sample allows the study of both Co/NiO/Fe trilayers and NiO/Fe bilayers under the same growth condition. The absence of the LEED spots from the Co film shows the formation of polycrystalline Co film. The sample is covered by a 2 nm protection Ag film before bringing it to the PEEM-II station at the Advanced Light Source of the Lawrence Berkeley National Laboratory. As reported in the literature, Fe film grown on Ag(001) has a bcc structure with the Fe [100] axis parallel to the Ag [110] axis and the NiO film on Fe(001) has an fcc structure with the NiO [110] axis parallel to the Fe [100]

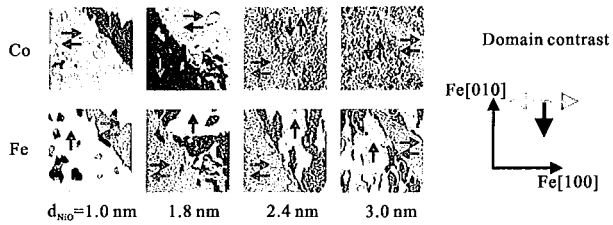

FIG. 1. (Color online) Co and Fe magnetic domains as a function of NiO thickness from Co(2 nm)/NiO/Fe(15 ML)/Ag(001). The correspondence between domain contrast and spin orientation is shown in the schematic drawing as a reference. Arrows represent the spin orientations. The result shows that the Co and Fe have a 90° coupling for $d_{NiO}<2.0$ nm and a collinear coupling for $d_{NiO}>2.0$ nm.

axis.[26] To avoid confusion, all the crystalline axes in this Brief Report are specified according to the bcc Fe unless otherwise specified.

We first present the result of magnetic interlayer coupling between Co and Fe films. The ferromagnetic Co and Fe domains (Fig. 1) are obtained by taking the ratio of the corresponding Co and Fe L_3 and L_2 absorption edges with circular polarized incident x rays.[27] In our measurement, the x ray is in the (100) plane and makes an incident angle of 60° with respect to the surface normal direction ([001] direction). Since the XMCD signal is determined by the projection of the ferromagnetic spin to the incident x-ray direction, the ferromagnetic spin orientation (arrow symbols in Fig. 1) can be determined from the domain contrast with the white, dark, and gray domains corresponding to spins parallel to [010], [0–10], and [±100] axis, respectively (see the schematic drawing in Fig. 1). After assigning the spin direction to the Fe and Co domains, we immediately identified that the Fe and Co spins are coupled perpendicularly to each other (90° coupling) for NiO thickness thinner than ~2 nm ($d_{NiO}<2$ nm) and collinearly for $d_{NiO}>2$ nm. It should be mentioned that below 1 ML NiO, we observed a ferromagnetic interlayer coupling between the Fe and Co films which is not surprising because the Fe and Co films should be partially connected under this condition. Note that Slonczewski's 90° coupling comes from interfacial roughness, thus cannot explain our observation because it would lead to a 90° coupling at thicker NiO film where a rougher NiO film is expected. Another observation is that there coexist both "FM coupling" and "AF coupling" in the collinear coupling regime ($d_{NiO}>2.0$ nm). As it will be explained later, this coexistence is not due to inhomogeneity of the sample but is energetically degenerate due to 90° coupling at both Co/NiO and NiO/Fe interfaces.

We then studied the NiO AF spin structure to identify its role in the Co-Fe interlayer coupling. We took the Ni L2 energy spectra from NiO(3.0 nm)/Fe/Ag(001) to identify the existence of the XMLD effect. An external magnetic field is applied to align the Fe spin parallel to the [010] axis and two energy spectra were taken with the x-ray linear polarization vector $\vec{E}$ parallel (ϕ=0°) and perpendicular (ϕ=90°) to the Fe spin direction (Fig. 2). The spectra are normalized by the peak intensity at the lower peak energy of 870.3 eV for a

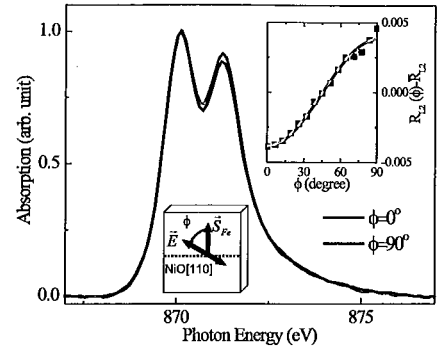

FIG. 2. (Color online) Normalized energy spectra from the Ni L2 edge of NiO for two orthogonal linear polarizations. The inset shows the sinusoidal dependence of the L2 ratio as a function of the polarization angle ϕ.

clear view of the difference in the two spectra. The L2 ratio [defined as $R_{L2}=I(870.3\text{ eV})/I(871.3\text{ eV})$, where I is the normalized intensity], which is a direct measure of the XMLD effect, is plotted in the inset as a function of the x-ray polarization angle. The L2 ratio has a sinusoidal dependence on the polarization angle with the minimum and maximum values being at ϕ=0° and ϕ=90°, respectively, showing that the NiO spin direction is in the NiO [±110] direction. As shown by previous works,[28,29] for easy axis in the NiO [±110] direction the L2 ratio of NiO should reach its maximum value as the NiO spin is parallel to the x-ray polarization direction. Therefore the result of Fig. 2 shows that the NiO spins in the d_{NiO}=3.0 nm sample are parallel to the Fe[100] axis, i.e., orthogonal to the Fe spin direction.

We then imaged the NiO AF domains in the Co/NiO/Fe trilayers using PEEM by dividing two images taken at 870.3 and 871.3 eV using linear polarized x rays and compared the image with the Co and Fe ferromagnetic domains. Figure 3(a) shows the element-specific magnetic domain images of the Co(2.0 nm)/NiO(1.1 nm)/Fe(15 ML) trialyers in which the Co and Fe layers have a 90° coupling. XMCD image of the NiO is also taken to single out the induced uncompensated ferromagnetic Ni spins at interfaces. From the domain images, we find that the NiO AF spins are collinearly coupled to the Fe spins but 90° coupled to the Co spins and

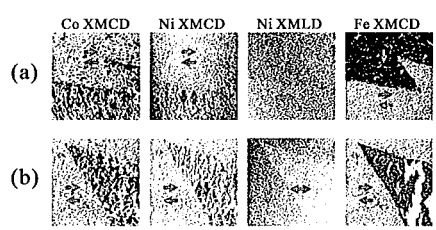

FIG. 3. (Color online) Co and Fe XMCD images together with Ni XMCD and XMLD images taken at (a) d_{NiO}=1.1 nm and (b) d_{NiO}=3.0 nm from Co(2 nm)/NiO/Fe(15 ML)/Ag(001) sample. The arrows represent the spin direction. The NiO/Fe interfacial coupling changes from a collinear coupling at d_{NiO}=1.1 nm to a 90° coupling at d_{NiO}=3.0 nm. The Co/NiO interfacial coupling remains a 90° coupling at all NiO thicknesses with an induced Ni XMCD signal at the Co/NiO interface.

that the uncompensated ferromagnetic Ni domains follow the Co domains. Here we find no evidence of the spiral spin structure in the NiO film because a spiral NiO spin structure would result in an averaged NiO spin direction in the Fe $[\pm 1 \pm 10]$ axis which should produce a maximum/minimum L2 ratio at 45° polarization angle. In addition, the identical Co and Ni XMCD domains show that the uncompensated Ni ferromagnetic spins are induced at the Co/Ni interface, which is not surprising because a rougher interface is expected at the Co/NiO interface than at the NiO/Fe interface. Using the same method, we took and assigned the spin directions for Co(2.0 nm)/NiO(3.0 nm)/Fe(15 ML) trialyers where the Co and Fe layers have a collinear coupling [Fig. 3(b)]. We find that NiO AF spins in this case are 90° coupled to both Co and Fe spins and that the uncompensated ferromagnetic Ni spins again are from the Co/NiO interface. The results of Figs. 3(a) and 3(b) clearly explain the Co-Fe interlayer coupling in Fig. 1: the observed transition from 90° to collinear Co-Fe interlayer coupling with increasing NiO thickness is due to a transition of collinear to 90° interfacial coupling at the NiO/Fe interface while retaining a 90° coupling at the Co/NiO interface. This also explains why there coexist parallel and antiparrallel alignments between the Co and Fe spins in the collinear coupling regime ($d_{NiO} > 2.0$ nm) because the coupling mechanism here comes from 90° coupling at both Co/NiO and NiO/Fe interfaces so that the Co-Fe parallel and antiparrallel alignments are energetically degenerate.

Theoretically, a 90° coupling is expected at the FM/AF interface for a perfect compensated AF interface.[30] It is suggested that uncompensated spins of the AF layer due to roughness and defects could induce different types of interfacial coupling.[27,31,32] Since an increase in the NiO thickness in Co/NiO/Fe/Ag(001) is likely to change the roughness at the Co/NiO interface rather than at the NiO/Fe interface, it is quite interesting that we observe a 90° coupling and uncompensated Ni spins at the Co/NiO interface for all NiO thicknesses, but both a collinear coupling ($d_{NiO} < 2.0$ nm) and a 90° coupling ($d_{NiO} > 2.0$ nm) at the NiO/Fe interface. It should be pointed out that the measured Ni XMCD signal could, in principle, come from both the Co/NiO and NiO/Fe interfaces. To further separate the contributions from the NiO/Fe interface, we performed XMCD and XMLD measurement on NiO/Fe(15 ML)/Ag(001) bilayers. We observe

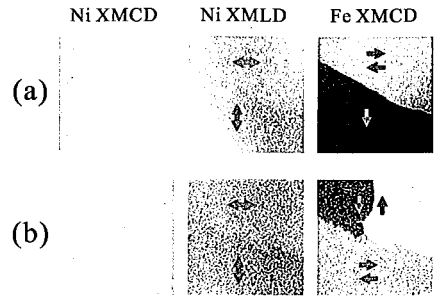

FIG. 4. (Color online) Fe XMCD image together with Ni XMCD and XMLD images taken at (a) $d_{NiO}=1.1$ nm and (b) $d_{NiO}=3.0$ nm from of NiO/Fe(15 ML)/Ag(001). The arrows represent the spin directions. The NiO/Fe interfacial coupling changes from a collinear coupling at $d_{NiO}=1.1$ nm to a 90° coupling at $d_{NiO}=3.0$ nm. No induced Ni XMCD signal is detected.

the collinear to 90° coupling switching with increasing NiO thickness in the bilayer system (Fig. 4) consistent with the result reported before.[26] More importantly, no Ni XMCD signal is detected for all NiO thicknesses, suggesting that the all the uncompensated Ni spins in Co/NiO/Fe sandwich come from the Co/NiO interface. We are unclear, however, on whether the observed uncompensated Ni signal at the Co/NiO signal is responsible for the different Co/NiO and NiO/Fe interfacial magnetic couplings. Further investigation is needed to clarify this issue.

In summary, the Co-Fe magnetic interlayer coupling in Co/NiO/Fe/Ag(001) trilayers switches from a 90° coupling to a collinear coupling with increasing the NiO thickness. This anomalous coupling is due to a switching of the NiO/Fe interfacial coupling from a collinear to a 90° coupling with increasing the NiO thickness while retaining a 90° coupling at the Co/NiO interface. Induced uncompensated Ni spins are observed at the Co/NiO interface but not at the NiO/Fe interface. No evidence of spiral NiO spin structure is found in this system.

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