Absence of induced moment in magnetic tunnel junction barriers

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(Received 22 July 2005; revised manuscript received 26 September 2005)

To confirm the theoretical underpinnings of spin-polarized transport, we have performed x-ray magnetic circular dichroism experiments on samples exhibiting a ferromagnet-insulator interface grown using processes which have yielded junctions with appreciable tunneling magnetoresistance. Within experimental sensitivity, no magnetic moment induced due to the proximity of the ferromagnet was detected on any atomic barrier site using this element-specific technique.

DOI: XXXX PACS number(s): 75.70.Cn, 72.25.Mk, 87.64.Ni

Many magnetic tunnel junction studies have been performed using ferromagnetic electrodes sandwiched between the ultrathin insulator Al_2O_3,[1] and more recently MgO.[2–4] Alongside technological applications, much fundamental research has focused on understanding the electronic nature of the spin-polarized tunneling current through such barriers. Experimentally, the spin-dependent tunneling technique,[5] which uses a superconducting electrode to probe tunneling from a ferromagnet across an ultrathin insulator, has been favored since it can yield both the amplitude and the sign of spin polarization at the ferromagnet-insulator interface. However, all such experiments using Al_2O_3 tunnel barriers yielded a positive sign of the ferromagnet's spin polarization,[6] sometimes in contradiction with that inferred from the ferromagnet's spin-polarized density of states at the Fermi level.

This issue was partly resolved through similar experiments using a transition metal oxide barrier such as $SrTiO_3$.[7,8] A combination of interfacial bonding and/or metal-induced gap states with *d*-type electronic character can account for the transmission of the correct sign of spin polarization in this case. To account for such experimental results, a group of theoreticians has examined the Co/Al_2O_3,[9] Co/O vacuum,[10] $Co/SrTiO_3$,[11] and Fe/O vacuum[12] interfaces. Their preeminent work predicts the appearance of a magnetic moment induced on the barrier's interfacial layer due to contact with a ferromagnetic electrode, through a direct or indirect exchange mechanism. Using the element-specific technique of x-ray magnetic circular dichroism (XMCD), such moments may be calculated from x-ray absorption spectra (XAS) taken at either the *L* edge corresponding to $p \rightarrow d$ transitions for transition metals, or at the *K* edge corresponding to $s \rightarrow p$ transitions for lighter species such as O and Al. We have performed XMCD experiments at the European Synchrotron Radiation Facility (ESRF) on samples with Co/Al_2O_3 and Fe/MgO(001) interfaces, grown using processes which have yielded working magnetic tunnel junctions.[2,13] We measure no dichroism at the O *K*, Mg *K*, or Al *K* edges, suggesting in the case of O sites an induced moment no greater than our $0.05\mu_B$ experimental sensitivity, which would be lower than the $0.07-0.2\mu_B$ calculated.[9,12]

The $Co/Al_2O_3/Co$ tunnel junctions were grown by sputtering as described in Ref. 13. The Al_2O_3 layers were either sputtered directly from a stoichiometric target or obtained from Al layers oxidized in an Ar/O_2 plasma. The Fe/MgO/Fe tunnel junctions were epitaxially grown on MgO-buffered GaAs(001) substrates as described in Ref. 2.

XMCD experiments were performed at the ESRF's ID08 beamline, which exhibits a helicity of the incoming photon beam with $\approx 100\%$ polarization. In the discussion below, parallel and antiparallel alignments of photon helicity and magnetization define the positive and negative photon helicities, respectively. The energy resolution at 850 eV is $\Delta E/E \sim 2\times 10^{-4}$, so that our experimental setup is well adapted to this study. Measurements were performed in total electron yield mode on samples with the interface of interest lying within the $\sim 30-50$ Å electron escape depth of the surface. All spectra were recorded at $T=10$ K and at normal incidence in a magnetic field ($H=\pm 3$T) large enough to saturate the magnetization of all the samples.

We first discuss results obtained on a Si//Ta (50 Å)/Co (150 Å)/Al_2O_3 wedge/Co (30 Å)/Au (20 Å) sample with an Al_2O_3 wedge 30 mm long and with a final thickness of 30 Å which was sputtered from a stoichiometric target. Given a 1000×50 μm^2 photon beam incident on the sample with the length along the wedge direction, our measurements offer a ± 1 Å resolution on Al_2O_3 thickness. Figure 1 presents x-ray absorption spectra (XAS) at (a) the Co $L_{2,3}$, (b) the Al *K*, and (c) the O *K* edges for positive and negative helicities taken for $d_{Al_2O_3} \approx 3$ Å. Using the sum rules, spectra taken at the Co $L_{3,2}$ edges reveal spin and orbital moments $1.64\mu_B$/atom and $0.15\mu_B$/atom, respectively, in good agreement with the calculated values $1.63\mu_B$/atom and $0.12\mu_B$/atom.[14] Within the 0.2 eV experimental energy resolution, no multiplet structure indicative of oxidation was observed.[15] No dichroism was observed at either the Al *K* or O *K* edges. Similarly, negative results were obtained on a thicker 7 ± 1 Å part of the Al_2O_3 wedge, and on a sample with an Al_2O_3 layer synthesized from the plasma oxidation of Al (data not shown).

We now turn to MgO results. Given the pulsed-laser deposition growth of MgO, no wedge capability could be utilized to probe a barrier thickness dependence of the induced moment. Therefore, the experiment was conducted on

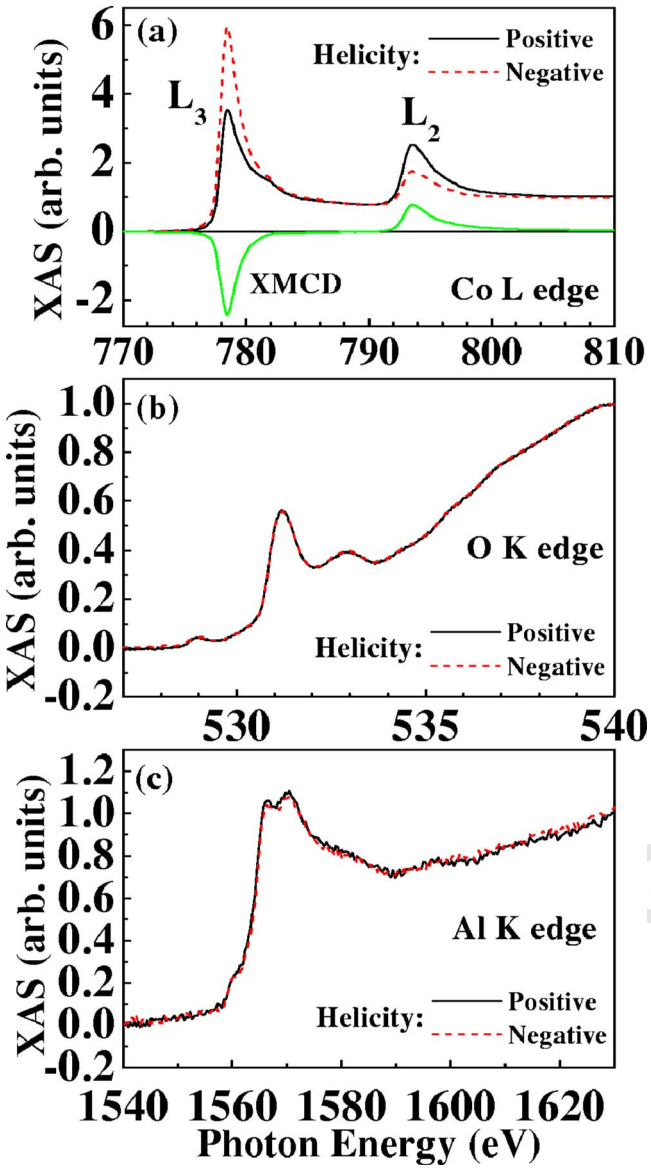

FIG. 1. (Color online) Si//Ta (50 Å)/Co (150 Å)/Al_2O_3 (3±1 Å)/Co (30 Å)/Au (20 Å) at T=10 K: XAS for the positive and negative helicities at the (a) Co $L_{2,3}$, (b) Al K, and (c) O K edges. Panel (a) also shows the resulting XMCD spectrum. No corrections have been made.

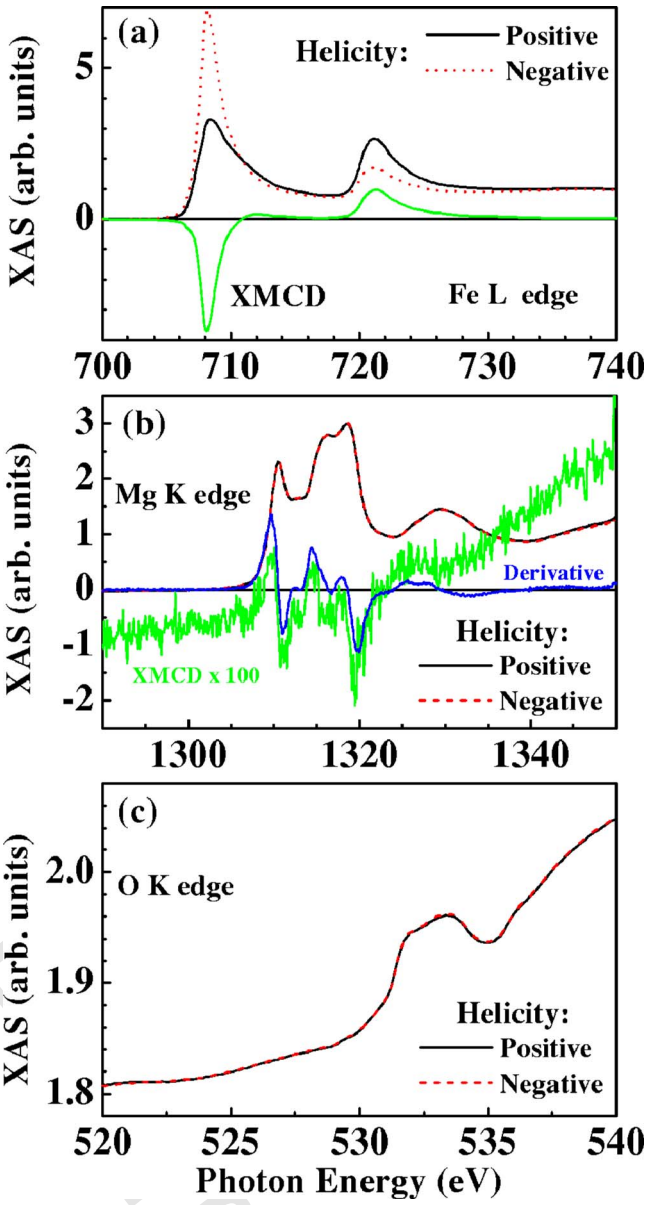

FIG. 2. (Color online) MgO(001)//MgO (100 Å)/Fe (250 Å)/MgO (10 Å)/Fe (20 Å)/Pt (20 Å) at T=10 K: XAS for the positive and negative helicities at (a) the Fe $L_{2,3}$, (b) Al K, and (c) O K edges. Panels (a) and (b) also show the resulting XMCD spectrum. No corrections have been made.

several samples individually grown with varying MgO thickness. Figure 2 presents typical results obtained for this series with XAS spectra at (a) the Fe $L_{2,3}$, (b) the Mg K, and (c) the O K edges on the sample MgO(001)//MgO (100 Å)/Fe (250 Å)/MgO (10 Å)/Fe (20 Å)/Pt (20 Å). Using the sum rules, the spin and orbital moments of Fe were found to be $2.19\mu_B$ and $0.03\mu_B$ per atom, within the error bar of the respective bulk values of $2.25\mu_B$ and $0.08\mu_B$ per atom.[14] Again, no multiplet structure indicative of oxidation is present. Since growth studies of MgO on Fe point to an interfacial FeO layer,[16] this implies that our XAS spectra only probe the Pt capping layer, top Fe layer, and a portion of the MgO layer. Our barrier atomic site XAS measurements are therefore sensitive to the top, unoxidized Fe/MgO interface. Nevertheless, the XMCD spectrum taken at the Mg K edge is quite similar to the derivative of the spectrum taken with positive helicity, so that it cannot be attributed to dichroism. No dichroism is present at the O K edge either. Spectra taken at the O edge differ somewhat from that found for Al_2O_3, as expected from the different electronic environments of O in the two samples.

Thus, both sets of samples show strongly magnetic, nonoxidized transition metals at the upper interface with the barrier to be probed. However, our XAS data show no dichroism at the absorption edges of the barrier materials. Therefore, the results presented above, which reflect the accumulation of many spectra, do not validate the picture of

induced moments on elemental sites within tunneling barriers due to the contact with a ferromagnet.

Our experiments focused mainly[23] on elements such as Mg, Al, and O with only *K* and *L* shells. Beamline ID08 at the ESRF implements a monochromator which provides the highest flux in the 850 eV range so as to probe $L_{2,3}$ edges in 3*d* transition metals. While the photon flux decreases by factors of 1.6, 6, and 20 in the energy ranges of the O, Mg, and Al *K* edges, respectively, and the $2p \rightarrow 4s$ transitions are not as probable as the usual $2p \rightarrow 3d$, the good signal-to-noise ratio on individual scans presented above, even for Al, validate our experimental conditions.

Finally, we discuss an upper limit to a possible experimental error resulting in a nonzero amplitude of such induced moments. At the Co/Al_2O_3 interface, covalent bonding between the O 2*p* and the Co 3*d* orbitals was calculated[9] to lead to an induced magnetic moment of $\approx 0.07\mu_B$ on the O site. In the case of Co-Al bonding, these calculations predict a smaller induced moment. Regarding the Fe/MgO(001) interface, Fe was calculated to induce a $\approx 0.2\mu_B$ moment on the O site.[12] In this latter case, the epitaxial nature of our sample placed the experiment in quite ideal conditions to verify the calculation. In a separate experiment on the similar beamline ID12B, Pellegrin *et al.* measured a 1.6% dichroic signal at the O *K* edge for $La_{0.7}Sr_{0.3}MnO_3$,[17] associated with a calculated $0.08\mu_B$ moment on the O site induced by hybridization.[18] This indicates an experimental sensitivity in our case to an induced moment of $\sim 0.05\mu_B$ in a *bulk* film with a magnetic environment. Compounding this issue, our experiment endeavored to measure an induced moment at the *K* edge along a two-dimensional (2D) plane of sites outside a magnetic environment. Other experiments have demonstrated that it is possible to measure a moment on oxygen sites adsorbed onto Co(001) and Ni(001),[19,20] though not quantitatively due to difficulties in interpreting the spectra within the *K*-edge sum rules.[21,22]

In conclusion, we have performed XMCD experiments on samples with Co/Al_2O_3 and Fe(001)/MgO(001) interfaces to test preeminent theory on the electronic character of the tunneling current through insulating layers as manifested by the appearance of induced moments on the barrier's atomic sites. Despite appropriate samples and experimental conditions, we observed no dichroism at the O *K*, Mg *K*, or Al *K* edges. In particular regarding oxygen, our experimental sensitivity was $0.05\mu_B$ of induced moment per O site, to be compared with calculations ranging from 0.07 to $0.2\mu_B$. Thus our results suggest that no moment greater than $0.05\mu_B$ per site is induced on O. We look forward to future experimental endeavors to observe magnetic moments induced on tunnel barrier atomic sites using the more refined x-ray resonant magnetic scattering or x-ray photoemission electron microscopy techniques.

The authors wish to thank C. Martínez-Boubeta and A. Cebollada, as well as A. Vaurès, for sample growth, and A. Fert for useful discussions. M.B. gratefully acknowledges support through an Eiffel scholarship from the French Ministry of Foreign Affairs.

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