



	Experiment title: Weak ferromagnetism in the CuO <sub>2</sub> planes of La <sub>2-x</sub> Sr <sub>x</sub> CuO <sub>4</sub> thin films: doping dependence and effects from a ferromagnetic coupled layer	Experiment number: HE 3111
Beamline:	Date of experiment: from: July 5 <sup>th</sup> to: July 12 <sup>th</sup> 2010	Date of report: 9/08/2011
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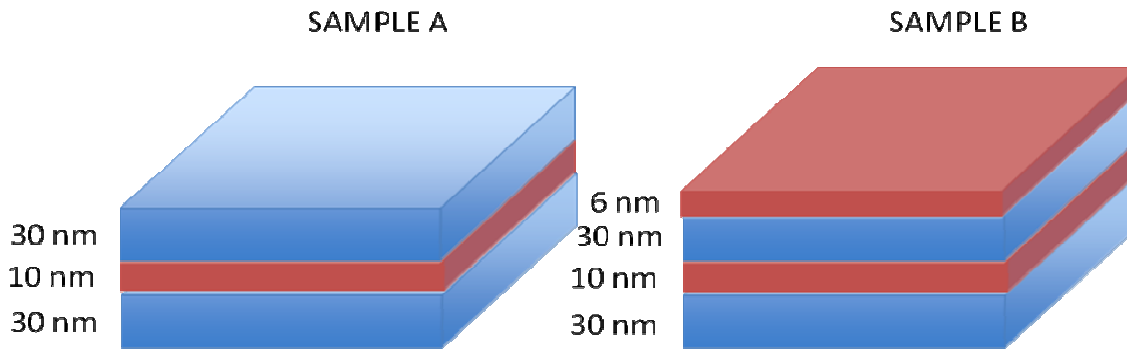
## Report.

The latest advancements in the epitaxial growth have demonstrated that transition metal oxide (TMO) heterostructures with atomic-scale perfection can now be realized. These achievements opened the road to realize novel functionalities based on novel physical phenomena occurring at the interfaces. Studies on manganites/cuprates multilayers have shown that the physics arising at their interfaces is of extreme importance in determining the overall characteristics of the system. For this reason the characterization of the peculiar electronic and structural properties of the interfaces, compared with the bulk material and even with the surfaces itself, is deserving great attention [1].

In this context, one of the most interesting issue of condensed matter physics, which remains concealed, is the interplay between magnetism and superconductivity in single phase cuprates and in superconducting/ferromagnetic (S/F) heterostructures. One of the most interesting result, to understand the interplay between superconductivity and magnetism in cuprates was reported by Chackalian et al. [2].

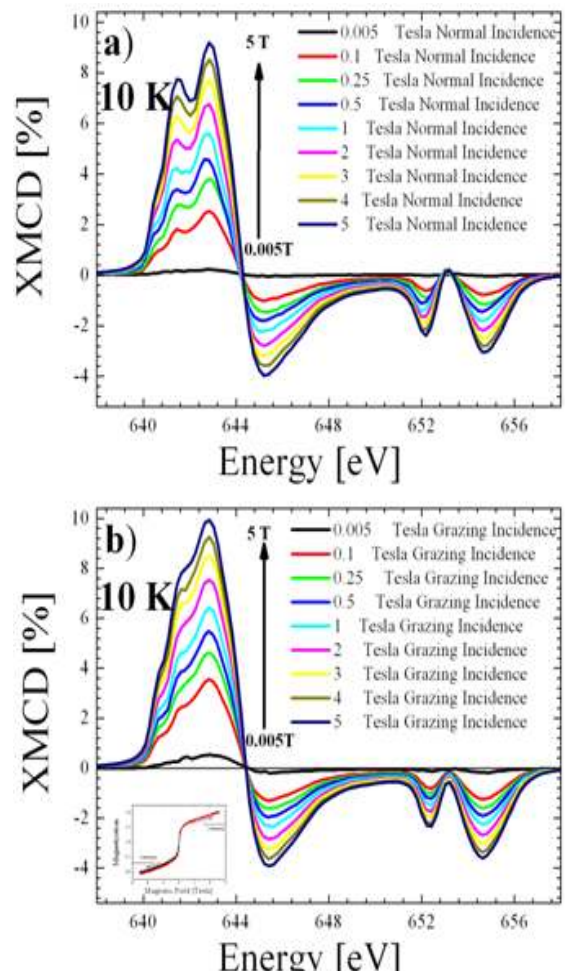
Using the x-ray circular and linear dichroism techniques on YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (YBCO)/La<sub>0.66</sub>Ca<sub>0.33</sub>MnO<sub>3</sub> (LCMO) superlattice (SL), it was shown that a peculiar electronic state is realized at the interface. This electronic state was composed by a bonding Mn3d-O2p-Cu3d molecular orbital [2], also responsible for an unusual antiparallel alignment of the Cu and the Mn spins at the interface [3].

In this experiment we used X-ray absorption spectroscopy experiments have been performed at the ID08 beamline of the ESRF, employing the high resolution Dragon monochromator and the XAS end-station equipped with a 6T superconducting magnet. By using simultaneously Total Electron Yield (TEY) and Fluorescence Yield (FY) detection methods - x-ray magnetic dichroism (XMCD) and x-ray linear dichroism (XLD), at the Cu and Mn  $L_{2,3}$  edges on (LSMO)/LSCO SL's, were performed to investigate the relationship between the spin moments of Cu and Mn. In particular, using two different stacking configurations (Fig.1a, LSCO on top of LSMO, sample A, Fig1b LSMO on top of LSCO, sample B) we were able to discriminate interface from bulk effects in the system.



**Fig.1** sketch of the LSCO/LSMO SL's studied, which are characterized by two different layer terminations, i.e. in sample A they are terminated by a 30nm thick LSCO film, and in sample B by a 6nm LSMO layer.

Up to 3,2 Cu L-edge XAS spectra, acquired with plus and minus elicitities, have been averaged to eliminate any source of systematic error and to reduce the noise level. Here we show data acquired at low temperatures (5K) with the SL in the superconducting state. We have performed measurements in grazing incidence (GI) and normal incidence (NI) conditions. Before each measurement, the magnetization of the LSMO layer was saturated by applying a field above the coercive value in both out of plane and in plane directions (we used 0.5 Tesla). In fig. 2 we show the Mn-L edge XMCD spectra taken on sample B and relative to the 6 nm LSMO layer, the spectra are similar for both normal incidence and grazing incidence conditions at field above 0.1. In particular the XMCD signal does not saturate at high field, but increases until 5 Tesla, anyway reaching a value 1.5 lower of that one measured on isolated LSMO thin film. The corresponding XMCD measured on Cu for sample B show a dichroism opposites to the one measured on the LSMO up to 1 Tesla, and opposite to the applied magnetic field. This result shows that the aligned Cu magnetic moments are antiparallel to the Mn spins (Fig. 3). However, at high magnetic fields, 4 Tesla, the dichroism changes sign becoming parallel to the Mn moment in LSMO and parallel to the external field, but its absolute value remains approximately



**Fig.2** Mn edge XMCD spectra on sample B acquired in normal a) and grazing incidence b) as function of the magnetic field, inset magnetization of the LSMO measured during acquisition of XMCD spectra.

constant. The magnitude of the XMCD is similar in grazing and normal incidence.

This result is noteworthy, since at grazing incidence the LSMO spin have mainly an in plane component, while the Cu spin in LSCO are expected to be mainly in the  $\text{CuO}_2$  plane for any orientation of the magnetic field. Its behavior at low fields is opposite but qualitatively proportional to the XMCD measured on Mn. The TEY XMCD at high field (4 Tesla) on sample A and the one measured on sample B are very similar. As report in ref. [4], the  $\text{Cu}^{2+}$  spins are mainly in the  $\text{CuO}_2$  planes. Thus the measured XMCD is associated to the canting of the Cu spins out of the  $\text{CuO}_2$  planes[4]. The absolute value of out of plane component is similar at low and high field and matches with the values measured on several LSCO superconducting samples. The results obtained on our LSCO/LSMO SL's and to the comparison with YBCO/LCMO SL's, we can see that the XMCD and correspondent spin moment measured is at least a factor two smaller. This is exactly the same ratio between the bulk Cu-spin moment measured in LSCO and YBCO films and single crystals. Thus, again it appears that the spin moment measured is related to the canting of the Cu spins, which creates an out of plane component aligned by an external magnetic field, or by the coupling with a ferromagnetic layer. However, the antiparallel orientation of the Cu and Mn spin at low fields has not a straightforward explanation. By using fully linear polarized light and grazing incidence (GI) conditions ( $\vartheta = 70^\circ$  from the surface normal), we have studied the orbital anisotropy of Cu and Mn 3d states, with particular attention towards the differences between interface and bulk electronic properties. In Fig. 4, we show XAS spectra at the Cu L<sub>23</sub> edges of samples A and sample B measured in TEY (circles) and FY (lines) modes, with linear polarization parallel ( $I_{ab}$ ) and perpendicular ( $I_c$ ) to the interface. The bulk-like FY spectra of the two samples are identical, while we can notice relevant differences in the TEY mode, in particular for the  $I_c$  XAS spectra.

The XAS spectra in LSCO are in general characterized by a very strong anisotropy in the electronic states, due to the almost complete occupation of the  $3d_{z^2}$  orbitals. The unique feature in the FY  $I_c$  spectra of LSCO is a small peak close to 931 eV, at the same position of the main absorption that occurs for in plane 3d states. In sample A, LSCO terminated, we can notice that the TEY spectra is quite similar to the FY spectra. Small differences between TEY and FY are associated to a non-negligible contribution of the surface layer in TEY (see below) and to self absorption effects in the FY mode. On the other hand, in sample B, the TEY and FY XAS acquired with polarization

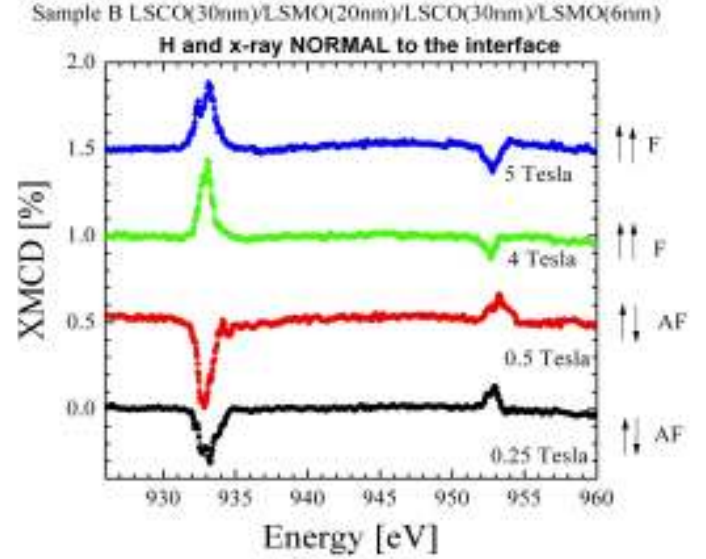


Fig. 11: Cu edge XMCD spectra measured at 5 K on sample B in normal incidence conditions as function of the magnetic field.

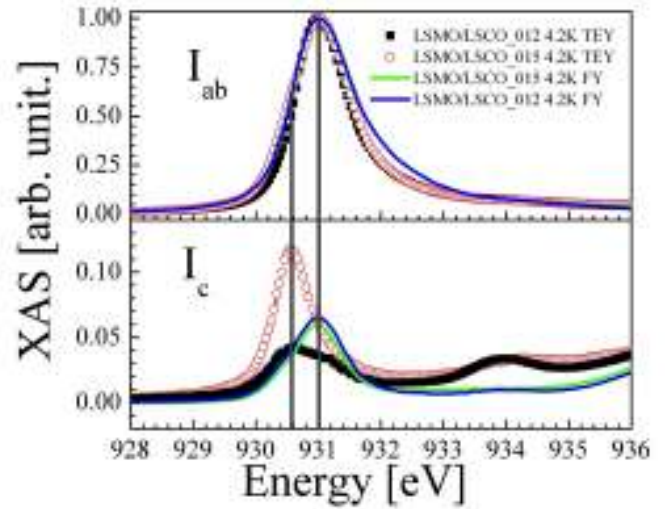


Fig. 4: X-ray absorption spectra on sample A and sample B measured in TEY (sample A-filled squares, sample B-open red circles) and FY (sample A-blue line, sample B-green line) modes. The upper panel is for linear polarization of light parallel to the interface and the lower pane is for perpendicular polarization. In the inset of the lower panel a sketch of the experimental configuration.

perpendicular to the interface differ quantitatively and qualitatively. Moreover, the TEY spectra of sample B, acquired in perpendicular polarization, is also quite different from the equivalent TEY spectra of sample A. Since TEY XAS on sample B has a main contribution from the LSCO/LSMO interface, this result suggests that some electronic changes are effectively occurring. In particular we can clearly notice that the Cu  $3d_z^2$  peak is shifted to lower energy, which suggests the realization of new molecular orbital at the interface. Actually, a close look to the TEY data acquired on sample A, suggests that a similar peak, much less intense, is also present in this sample, however it cannot be due to the LSCO/LSMO interface since the interface is 30 nm below (LSCO is 30 nm thick), indicating that a similar molecular orbital can be created at the LSCO-Vacuum interface. The presence of the new state can be interpreted as a localization of holes into the Cu  $3d_z^2$  orbitals, due to a strong hybridization between Cu and O along the Cu-O-Mn bonds suggested by Chakhalian et al. [2]. However, in our samples we have not clarified yet, if this consideration should be validated. Although a more accurate analysis of these findings is needed in order to get a clear picture. These results at the interface, show that the out of plane Cu-spin component is aligned AF with the Mn one realizing a magnetic dichroism opposite to the Mn magnetization. This happens at low magnetic field, until the external field is too low to overcome the out of plane magnetic interaction  $J_z$ . On the other hand the XLD experimental results obtained on LSCO/LSMO interface effectively suggest that not only a magnetic, but also a small electronic reconstruction occurs at the cuprate manganite interfaces.

## Reference

- [1] M. Huijben et al., Nat. Mater. 5, 556 (2006).
- [2] J. Chakhalian, et al. Nat. Phys. 2, 244 (2006)
- [3] J. Chakhalian, et al. Science 318,1114 (2007).
- [4] G. M. De Luca, et al. Phys. Rev. B 82, 214504 (2010).