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## **Report:**

The latest advancements in the epitaxial growth of oxides have demonstrated that oxide interfaces and heterostructures with atomic-scale perfection can now be realized. These achievements opened the road to realize novel functionalities based on novel physical phenomena. Studies on manganites/cuprates heterostructures and multilayers have shown that the physics of the states arising at these interfaces is of extreme importance in determining the overall characteristics of the system. For this reason the characterization of the peculiar features of the interfaces if compared with the bulk material and even with the surface itself is deserving great attention. The close relationship between superconductivity and magnetism was considered as strong indication of a magnetic origin of the effective electron-electron interaction leading to the superconductivity in an hole doped antiferromagnetic Mott insulator. One way to study this phenomenon consists in growing high quality Manganites/Cuprates superlattices and bilayers, and to analyze the reciprocal influence in a variety of experimental conditions. In the experiment HE2525 we already performed X-ray Magnetic Circular Dichroism (XMCD) at the copper L<sub>2,3</sub> edge of undoped (La<sub>2</sub>CuO<sub>4</sub> (LCO)), underdoped, and optimally doped La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> (LSCO), NdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> (NdBCO) and YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> (YBCO) compounds[see report of HE2525]. To elucidate the origin of the spin moment, the observed XMCD signal was studied as function of the temperature and of the magnetic field. The data showed that the undoped compounds, characterized by a long range antiferromagnetic order, develop at the Cu<sup>2+</sup> sites a Weak Ferromagnetic (WF) component induced by the external magnetic field. In doped compounds (even in the optimally doped samples)WF survives in the superconducting state.

In this experiment, using the high resolution Dragon monochromator of ID08 and the 6T superconducting magnet, we have studied the origin and coupling of the magnetism and superconductivity at the interface

between  $Pr_{0.7}Ca_{0.3}MnO_3$  and  $La_{1.85}Sr_{0.15}CuO_4$  epitaxial films deposited by Reflection High Electron Diffraction (RHEED) assisted pulsed laser deposition. By using simultaneously Total Electron Yield (TEY) and Fluorescence Yield (FY) detection methods at the Mn ( $L_{2,3}$  edge), we have measured the XMCD and Linear Dichroism (LD) of Manganites  $Pr_{0.7}Ca_{0.3}MnO_3$  (PCMO 25u.c) /Cuprates  $La_{1.85}Sr_{0.15}CuO_4$  (LSCO 8 u.c) superlattices (ten times repeated), bilayers PCMO(250 u.c )/LSCO (76 u.c.) and single PCMO films with 5 u.c. and 250 u.c. respectively.

The main idea was to obtain information about the effect on the magnetic properties at the  $L_{2,3}$  edge of the Mn induced by the strain and proximity effect, changing the thickness of the PCMO film, without and with cuprates at the interface. In the latest case, the effect of the cuprates was studied for PCMO film completely relaxed (bilayer) and completely strained (multilayer), respectively.

The quality of our samples is demonstrated in figure 1, where we have measured the classical ferromagnetic hysteretic XMCD vs magnetic field signal of a PCMO film characteriyed by only 5 u.c. deposited on the top of a TiO<sub>2</sub> terminated SrTiO<sub>3</sub> (STO) single crystal .



Figure 1. Magnetic field as function of the integrated value of the XMCD (obtained by the difference of XAS spectra measured with the left and right circularly polarized light respectively), peak at the L<sub>3</sub> edge of Mn is shown for PCMO 5 u.c. film.

A comparison of the XAS and LD spectra among the samples is presented in fig.2. According to the literature(see ref.[1] and ref.[2]) in manganite thin films the LD at the  $L_{2,3}$  edge of the Mn is strongly dependent on the strain, and therefore is able to give information about the strain effect. We observe a clear difference among different samples. A PCMO thick film deposited on STO, exhibit a linear dichorism characteristic of a relaxed sample, which is characterized by out of plane lattice parameter longer than the in plane ones (in pseudocubic units). A similar behaviour is observed in the case of the PCMO/LSCO bilayer. In the case of a thin film, however, the strain is tensile and it is dominating the dichroism, which is actually opposite, i.e. the in plane lattice parameters is expanded respect the out of plane lattice ones. Finally no dichroism is observed in the PCMO/LSCO multilayer. In the latest case we consider that Pr/La interdiffusion at the interface and strain effect, could explain this result. It must be noted that superconductivity is completely suppressed in the superlattice. This is a first evidence that the interface play an important role on the properties of these samples.



**Figure 2**. LD and XAS spectra for PCMO thin film (black solid line), PCMO thick film (red solid line), PCMO\_LSCO bilayer ((blue solid line) and PCMO\_LSCO multilayer (magenta solid line), are shown. The spectra were taken with the electric E vector perpendicular (V polarization) and E parallel (H polarization) to the film c-axis

To estimate the magnitude of the magnetic moment from the XMCD, we applied the well known sum rules, taking into account the correction factor necessary in the case of manganites from ref.[3]. In table 1 the value of the spin moment calculated for the  $Mn^{3+}$  is reported at 10 K and 5T. Also the estimated ferromagnetic T<sub>c</sub>, obtained from the temperature dependence of the XMCD is reported.

SAMPLE	T <sub>c</sub>	$\begin{array}{c} \mu_B/Mn \text{ atom} \\ (Mn^{3+}) \end{array}$
PCMOthin	180	1.52
PCMOthick	190	2.32
PCMO_LSCObilayer	190	3.29
PCMO_LSCOmultilayer	190	2.99

**TABLE 1:** estimated T<sub>c</sub> and low temperature magnetic moments on the samples studied. Note that the data have been obtained after zero field cooling and in an applied field of 5 Tesla

The  $T_c$  of the thick ferromagnetic insulating film is of the order of 120 K at zero field, and does not decrease much in the ultrathin film. Applying a magnetic field of 5 tesla we can see (figure 3) that there is a shift of the  $T_c$  toward higher temperature on all of the samples analized. In particular, the magnetic moment is strongly reduced in the thin film, suggesting that in thin films, strongly strained, there could be a strong tendency toward phase separation of the sample into antiferromagnetic and ferromagnetic regions, which could explain the reduced magnetization. Measurement of the LD as function of the magnetic field, indeed show a pronounce effect in the case of interface the thin films, which could be correlated to the appearance of large antiferromagnetic regions in place of ferromagnetic ones.

Very interstingly, the PCMO/LSCO multilayer show an higher  $T_c$  and an higher magnetization. In our opinion this is related to the interdiffusion La/Pr, which could realize a PrLaMnO fase, with enhanced ferromagnetic properties.

However, very surprisingly the PCMO/LSCO bilayer show a similar enhanced magnetization but a lower ferromagnetic  $T_c$ . Since in this sample there is only one interface, and since the PCMO layer is buried below a thick superconducting LSCO film, we postulate that what we are looking at here is an interface effect. Indeed, we are very sensitive to the interface (the intensity of the photons arriving to the sample is at least

reduced by a factor Exp(2)) and the TEY contribution is dominated by the interface layer. At the moment we do not have hints about the mechanism leading to the reduced ferromagnetic T<sub>c</sub>, but we suppose that this should be related to the presence of the interface with the superconducting LSCO sample.



Figure 3. Temperature dependence from 9 K to 300 K of the spin magnetic moment obtained from the XAS and XMCD data at 5 T using the sum rules: PCMO thin ( black solid line), PCMO thick (red solid line), PCMO\_LSCO bilayer (blue solid line) and PCMO\_LSCO multilayer (magenta solid line) film respectively

## References

[1] C. Aruta, G. Ghiringhelli, A. Tebano, N. G. Boggio, N. B. Brookes, P. G. Medaglia, and G. Balestrino, *Phys. Rev. B* 73, 235121 (2006)

[2] A. Tebano et al., Phys. Rev. B 74, 245116 (2006).

[3] M.C. Richter et al., Journal of Alloys and Compounds 362, 41-47 (2004)