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Shifts:	Local contact(s):	Received at ESRF:
18	Dr Nicholas BROOKES (e-mail: brookes@esrf.fr)	
Names and affiliations of applicants (* indicates experimentalists): Carmela ARUTA* CNR-INFM & University of Rome "Tor Vergata", Italy Giuseppe BALESTRINO CNR-INFM & University of Rome "Tor Vergata", Italy Norberto G. BOGGIO* CNR-INFM & University of Rome "Tor Vergata", Italy Nicholas BROOKES*, ESRF Giacomo GHIRINGHELLI* CNR-INFM & Politecnico di Milano, Italy Maurits HAVERKORT* II. Physikalisches Institut, Universität zu Köln, Germany Pier Gianni MEDAGLIA CNR-INFM & University of Rome "Tor Vergata", Italy Andrea PIAZZALUNGA CNR-INFM & Politecnico di Milano, Italy		

## **Report:**

The aim of our experiment was to study the effect of the strain induced by the substrate on the electronic properties on  $La_{0.7}Sr_{0.3}MnO_3$  (LSMO) thin films. We used X-ray Absorption Spectroscopy (XAS) and Resonant Inelastic X-ray Scattering (RIXS) measurements. The change in the MnO<sub>6</sub> octahedra distortion (out-of-plane compression in case of SrTiO<sub>3</sub> substrate and elongation in case of LaAlO<sub>3</sub> substrate) influences the *3d* states population, which can be monitored by linear dichroism (LD) in XAS. Moreover the variations in the Mn-O distances induced by the strain can affect the *dd* excitation energy and the degree of hybridization of Mn *3d* and O *2p* states, thus modifying the RIXS spectra.

First of all, we recorded LD-XAS spectra at the Mn L-edge and O K-edge, by using two orthogonal linear polarization, vertical (V) and horizontal (H), of the synchrotron radiation with an incident angle of 20° from the sample surface. We measured 4 LSMO films grown on STO with the same thickness but different degree of oxygenation, 7 fully oxygenated LSMO films grown on STO and 4 on LAO with different thickness, 2 LSMO films grown on STO with Nb and SrRuO<sub>3</sub> buffer layers. In addition we measured as a reference the pure STO and LAO substrates and a MnO bulk single crystal. In figure 1 (left panel) we show the results at Mn L-edge of LSMO films with a thickness of 100 unit cells (u.c.) and different oxygenation, as deduced by the different metal-insulator transition temperatures  $(T_{MI})$ . Moreover, on the right panel of figure 1 we show the results obtained on LSMO films on LAO, optimally oxygenated and with different thickness. The different LD behaviour in case of STO and of LAO is a consequence of the different preferential occupation of the  $x^2-y^2$  and the  $3z^2-r^2$  orbitals, respectively. Those results are confirmed by the spectra recorded at the O K-edge and they are in agreement with our theoretical calculations. Details of this study are reported in the paper we have recently published [C. Aruta, G. Ghiringhelli, A. Tebano, N.G. Boggio, N.B. Brookes, P.G. Medaglia and G. Balestrino, Phys. Rev. B 73, 235121 (2006)]. In addition, we have submitted for publication a manuscript where we explain the intensity behaviour of the LD spectra as a function of thickness in the framework of strain driven competition and nano-scale coexistence of the orbital ordered  $3z^2 r^2$ antiferromagnetic-insulating and the orbital disordered ferromagnetic metallic phases.

Furthermore, we have studied the dd and charge transfer excitations of LaMnO<sub>3</sub> (LMO, bulk single crystal cleaved in air) and on two selected LSMO thin films (100 u.c. thick) grown on LAO (LSMO324) and on STO (LSMO286) substrates. We have measured RIXS spectra excited at 3 different energies below the main

 $L_3$  peak of Mn, in the region where the emission spectra have a predominant Raman character. The F excitation is on the main  $L_3$  peak, excitations E and D are 1 eV and 2 eV below excitation F, respectively. The combined energy resolution measured from the elastic peak was 450 meV: we had to reduce the resolving power of the instrumentation for intensity reasons, but for LMO we could check that with 320 meV resolution no clear sharpening of the spectra features could be observed. All the measurements were made at room temperature.



**FIGURE 1** Average between experimental XAS signals taken in V- and H-polarizations (V+H)/2 and LD=V-H spectra in percent of the absorption data of LSMO strained films.



**FIGURE 2**: The RIXS spectra of LMO as excited on the main  $L_3$  peak of Mn. The *dd* excitations dominate the energy interval from 0 to 6 eV loss, whereas charge transfer excitations and fluorescence characterize the 6 eV to 12 eV range. The black and red curves correspond to linear polarization of the incident photons perpendicular and parallel to the scattering plane respectively.



**FIGURE 3:** The RIXS spectra of LSMO films measured with three different excitation energies and with two linear polarizations. We can notice that the spectra are very similar in shape, indicating that only minor differences in the average crystal field of  $Mn^{3+}$  ions correspond to the different strain conditions. On the other hand we can notice that in the film grown on LAO the *dd* features are sharper and stronger than in in that grown on STO, indicating a lower degree of hybridization and a higher localization of the *3d* states. On the other hand both LSMO samples give RIXS spectra rather similar to those of LMO (insulating antiferromagnetic material), but characterized by broader features and higher intensity of the charge transfer/fluorescence peak.