

**Experiment title:**Orbital and magnetic order in strained LaMnO<sub>3</sub> thin films**Experiment number:**

HE-3123

**Beamline:**

ID08

**Date of experiment:**

from: 25/11/2009 to: 01/12/2009

**Date of report:**

25/02/2010

**Shifts:**

18

**Local contact(s):**

Dr Nicholas BROOKES (e-mail: brookes@esrf.fr)

*Received at ESRF:***Names and affiliations of applicants (\* indicates experimentalists):**

Carmela ARUTA\* CNR-INFM Coherentia &amp; University of Naples "FedericoII", Italy

Nicholas BROOKES\*, ESRF

Alice GALDI\* CNR-INFM Coherentia &amp; University of Salerno, Italy

Giacomo GHIRINGHELLI\* CNR-INFM &amp; Politecnico di Milano, Italy

Marco MORETTI SALA\* CNR-INFM &amp; Politecnico di Milano, Italy

Luigi MARITATO\* CNR-INFM Coherentia &amp; University of Salerno, Italy

Pasquale ORGIANI\* CNR-INFM Coherentia &amp; University of Salerno, Italy

**Report:**

The aim of the proposed experiment was the investigation of the magnetic and orbital phase diagram of La<sub>x</sub>MnO<sub>3-δ</sub> (LMO) thin films grown by Molecular Beam Epitaxy, as a function of several controlled structural parameters. Heavy-ion La/Mn stoichiometric ratio was varied from La-deficient, to 1:1-stoichiometric LaMnO<sub>3</sub>, and finally to La-rich. Different bi-axial strain was induced by growing LMO films on different substrates: namely SrTiO<sub>3</sub> (STO), where the films are expected to experience negligible strain, NdGaO<sub>3</sub> (NGO) and LaAlO<sub>3</sub> (LAO), where the films are expected to experience compressive strain of -1.2% and 3.0%, respectively. In order to investigate the electronic properties and the local structure of strained and unstrained LMO thin films grown on various substrates and having different thickness, we proposed to do polarization dependent X-ray Absorption Spectroscopy (XAS) and Resonant Inelastic X-ray Scattering (RIXS) characterization (both at the Mn L<sub>2,3</sub> and O K absorption edges). X-ray Magnetic Circular Dichroism (XMCD) and Linear Dichroism (LD) XAS allowed us the experimental determination of the orbital reconstruction and the magnetic order in manganite films. Complementary, RIXS is sensitive to the variations of the electronic excitation energies related to the tetragonal distortions, thus providing an extremely accurate information about the degree of hybridization of Mn<sub>3d</sub> and O<sub>2p</sub>, which depends on the Mn-O distances influenced by substrate-induced strain.

The following LaMnO<sub>3</sub> samples on different substrates and with different compositions, i.e. different values of the La/Mn ratio, have been measured:

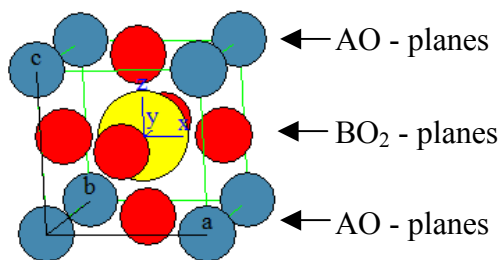
Substrate	La/Mn ratio				
	0.76	0.88	0.98	1.05	1.07
STO	ML080711F_STO (sample#06)	ML081013F_STO (sample#13)  ML081013L_STO (sample#03)*	ML081014F_STO (sample#05)	ML081216F_STO (sample#09)	ML081218F_STO (sample#04)
LAO	ML090217L_LAO (sample#08)		ML090216L_LAO (sample#02)		ML090218L_LAO (sample#07)
NGO	ML090217L_NGO (sample #12)		ML090216L_NGO (sample#01)*		ML090218L_NGO (sample#11)

\* also measured by RIXS

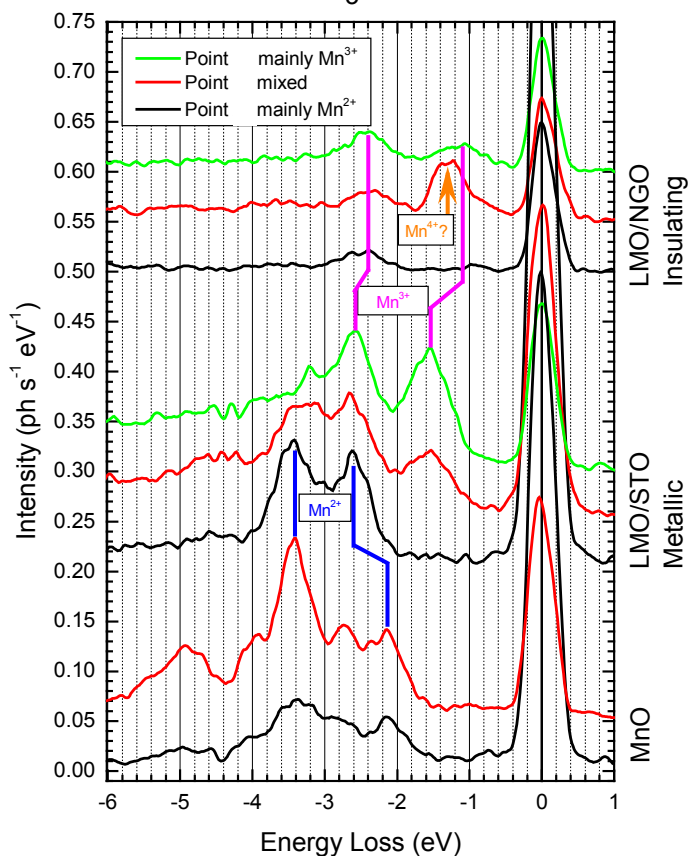
Linear and circular dichroism (LD and XMCD) have been performed on all samples. The LD on a single crystal of MnO has been also measured as a reference sample. Mn L- and O K- edges LD have been measured in zero-field-cooled (ZFC) at 300K and 10K, both with and without 1T applied magnetic field. Mn L-edge XMCD has been measured at 1T, 300K and 10K, both in Grazing and in Normal Incidence (GI and NI) to check the magnetic anisotropy. In few cases, we have also measured at 5T and 3 T and sample#13 has been measured also in field-cooled (FC) at 100K and 10K. Samples with different La/Mn content on SrTiO<sub>3</sub> substrates (negligible-strain case) were investigated. Data are presently under investigation.

The three following LMO samples were selected to be investigated by RIXS:

Sample	Physical properties	Characteristic XAS feature
ML081013L_STO (sample#03)*	Metallic and Ferromagnetic ( $T_{MI} = 380K$ ; $T_{Curie} = 360K$ )	presence of Mn <sup>2+</sup> features
ML090216L_NGO (sample#01)*	Insulating	No Mn <sup>2+</sup> features
MnO single crystal	Insulating	reference sample



Mn L<sub>3</sub> RIXS



As previously said, RIXS spectra provide an extremely accurate information about the degree of hybridization of Mn<sub>3d</sub> and O<sub>2p</sub>, which substantially depends on the Mn-O structural arrangement. In the B-site (see figure aside), the atom is tetrahedrally coordinated with 6 oxygen atoms, while in the A-site, the atom has 12 first-neighbors in an almost-spherical coordination. As a consequence, sizeable differences in RIXS spectra are expected to occur (for instance) in these two limit-cases. As a preliminary step, XAS experiments were also performed in RIXS chambers on a metallic and ferromagnetic LMO sample grown on SrTiO<sub>3</sub> substrate, and on an insulating LMO sample grown on NdGaO<sub>3</sub> substrate (see aside). Three significant energies were selected in the XAS spectra to run RIXS experiments. In particular, the selected energies were at about 640.0eV (E<sub>1</sub>), 641.5eV (E<sub>2</sub>) and 642.2eV (E<sub>3</sub>); the E<sub>1</sub> and E<sub>3</sub> energies correspond to Mn<sup>2+</sup> and Mn<sup>3+</sup> transition energies, respectively. E<sub>2</sub> can't be triavially associated to a single Mn characteristic transition energies, but most likely to some a Mn<sup>2+</sup>/Mn<sup>3+</sup> mixed characteristics. As a reference, we also decided to perform RIXS experiments on a MnO single crystal. The 3 samples give very different results. The spectra at point A in MnO and LMO/STO are very different, indicating a very different crystal field in the two situations for the Mn<sup>2+</sup>. Calculation of the RIXS spectra for Mn<sup>2+</sup> for different crystal field are mandatory to understand experimental data.

By considering the original proposal, all the aimed goals were achieved.