



Experiment title: Determination of the magnetic phase diagram of SMO/LMO superlattices

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Report:

During our experiment we measured $(\text{SrMnO}_3)_n/(\text{LaMnO}_3)_{2n}$ superlattices (SLs) with different periodicity, i.e. $n=1, 5, 8, 16$ to study electronic and magnetic interfacial properties in samples with several hole dopings related to the $\text{Mn}^{3+}/\text{Mn}^{4+}$ mixed valence at the interfaces. The SrMnO_3 (SMO) and LaMnO_3 (LMO) constituent blocks in form of thin films were also measured as reference samples. All samples were 20nm thick and grown by molecular-beam epitaxy on (100) SrTiO_3 (STO) substrate. X-ray circular dichroism (XMCD) and linear dichroism (XLD) measurements in x-ray absorption spectroscopy were performed at the Mn L-, La M- and O K-edges, in fluorescence and total electron yield mode. The measurements were carried out from room to 10 Kelvin to cross the metal-insulator and magnetic transitions and with applied magnetic fields up to 5T. In particular, field cooling and zero field cooling measurement cycles were performed.

The ferromagnetism (FM) was investigated by XMCD both in grazing (GI) and normal incidence (NI) configurations. We observed that all samples have a FM phase content, also the pure LMO film. The hysteresis loops in fig.1 are obtained by the maximum peak intensity of the XMCD (about 642 eV) as a function of the applied magnetic field. It can be observed that the FM easy-axis results oriented in the *ab*-plane and the anisotropy ratio is higher in the SL with $n=1$, decreases with n and is the lowest in the LMO film. In addition, apart from the SL with $n=1$ which hysteresis loop is very narrow, the coercivity enhances with decreasing the temperature in the other SLs and in LMO.

The FM phase observed in the LMO in form of thin film is not unusual and can have different explanations, as the lanthanum deficiency, the oxygen excess¹ or the structural distortions related to the in-plane lattice mismatch with STO substrate². Therefore, the origin of the FM phase is still not clear and has to be further investigated.

XLD measurements were employed to study the orbital occupation and the antiferromagnetic (AF) properties. Assuming that the orbital contribution to XLD is negligibly sensitive to the temperature, we obtained the magnetic part of the linear dichroism signal (XMLD) by subtraction of the orbital contribution at room temperature from the measurements taken below the magnetic ordering temperature: $I(\text{XMLD}) = \text{XLD}(10\text{K}) - \text{XLD}(300\text{K})$. XMLD at $B=0\text{T}$ is affected by both FM and AF phases, while with $B=1\text{T}$ we single out only the AF phase (fig.2). In the case $n=1$, XMLD spectra at $B=0\text{T}$ and $B=1\text{T}$ have similar shapes because the AF and FM easy axes are both in plane, but the features at $B=1\text{T}$ are weaker probably because the AF phase content is lower. Differently, for $n=5, 8$, and LMO, XMLD spectra at $B=0\text{T}$ and $B=1\text{T}$ are completely reversed each other indicating that the AF phase is out-of-plane. While in SLs with $n=5$ and $n=8$, FM and AF phases contents are comparable, in pure LMO AF phase is predominant. Moreover, all the AF spin directions are in agreement with the preferential orbital occupation obtained by the XLD spectra (fig.3). Pure LMO has XLD spectra typical of the preferential occupation of out-of-plane orbitals, i.e. $e_g(3z^2-r^2)$ or $(y^2-z^2)/(z^2-x^2)$. $\text{SMO}_n/\text{LMO}_{2n}$ SLs with $n=1$ show a weaker XLD signal with an in-plane preferential orbital occupation, in agreement with a mixing of orbital disordered FM phase and in-plane orbital ordered AF phase. The preferential orbital occupation becomes more and more out-of-plane with increasing n in the $n=5$ and $n=8$ superlattices, thus with increasing the LMO bulk contribution and decreasing the interfaces number. Therefore, redistribution of the charge carriers in the $e_g(x^2-y^2)$ orbitals of the interfacial MnO_2 doped planes has been observed, unlike the $e_g(3z^2-r^2)$ preferential occupation in the thicker layers, giving rise to different ferromagnetic (FM) and antiferromagnetic (AF) spin-orbital coupled states. As a consequence, the interfacial FM phase does not homogeneously extend in the whole superlattice, but it is overwhelmed by the AF bulk phase stabilized by the $e_g(3z^2-r^2)$ orbitals in the superlattices with $n \geq 5^3$.

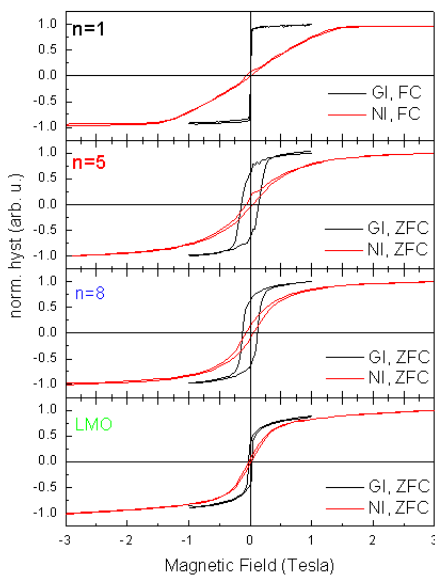


Fig.1 Hysteresis cycles GI vs NI at 10K.

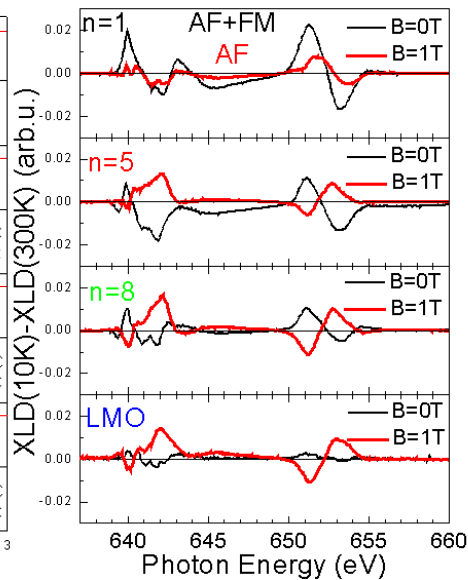


Fig.2 Field cooling XMLD

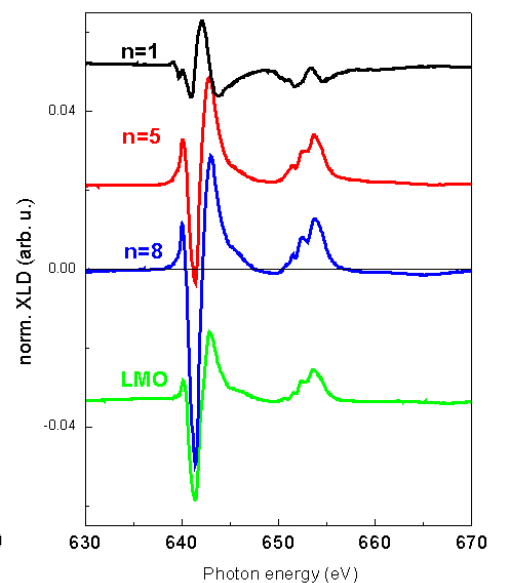


Fig.3 XLD at 300K with no magnetic field, normalized to the sum of the XAS Mn L_3 peak height signals

¹ A. Gupta et al. Appl. Phys. Lett. **67**, 3494 (1995); C. Aruta et al. J. of Appl. Phys. **100**, 023910 (2006)

² Shuai Dong, Rong Yu, Seiji Yunoki, Gonzalo Alvarez, J.-M. Liu and Elbio Dagotto, Phys. Rev. B **78**, 201102(R) (2008)

³ C.Aruta et al. to be published