

Strain Modified Properties of LCMO Thin Films

J. Dvorak, Y. Idzerda, J. Holroyd (Montana State U.), S. Ogale, S. Shinde and V. Venkatesan (U. of Maryland)
Beamline(s):U4B

Introduction: We have carried out a systematic study of the effects of lattice strain on the electronic and magnetic properties of $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ (LCMO) thin films using soft X-ray XAS and MCD. LCMO has two very interesting physical properties: it exhibits colossal magnetoresistance (CMR) near room temperature¹ and is a half metallic ferromagnet^{2,3}. These two properties make it a key candidate for advanced electronic applications based on spin polarized electron transport. The CMR phenomenon occurs near the Curie temperature of this material, at which LCMO undergoes a ferromagnetic-paramagnetic phase transition, a small structural transition, and a metal-insulator transition. This fascinating coupling between the structural and magnetic degrees of freedom suggests that it may be possible to influence the magnetic and transport properties of LCMO by controlling the structure. Structural control is possible by growing epitaxial layers of LCMO on single crystal substrates of differing lattice constants. This confines the in-plane lattice constants of the LCMO film, which in turn introduces lattice strain. The strain may vary as a function of the film thickness due to various stress relieving mechanisms. We have characterized the electronic and magnetic properties of a series of LCMO films ranging in thickness from 50 to 2500 angstroms grown on three different substrates: strontium titanate (STO), neodymium gallium oxide (NGO), and lanthanum aluminum oxide (LAO). The STO introduces a +0.9% in-plane strain (tensile) and the LAO introduces a -2.1% in-plane strain (compressive), while the NGO is lattice matched. We have developed a simple model that allows us to understand the observed behavior of these films in terms of the substrate induced strain.

Methods and Materials: The films were grown by pulsed laser deposition from polycrystalline targets on single crystal substrates of STO, NGO, and LAO(100). The Ca $L_{2,3}$ edge, O K edge, Mn $L_{2,3}$ edge, and La $M_{4,5}$ edge XAS spectra were taken with both s and p polarized radiation. In addition, the magnetic properties of the films were characterized as a function of temperature by MXCD spectra taken on the Mn $L_{2,3}$ edge utilizing circularly polarized radiation. All spectra were taken at the U4B beamline at the NSLS.

Results: We will concentrate on the results of the O K edge spectra, shown in Figure 1. The O spectra are of interest because they track the unoccupied density of states of the solid, and hence give information about the electronic structure of stressed overlayer. Furthermore, due to the dipole transition rule, p polarized light will show enhanced spectral weight with electronic states that are oriented out of plane. Referring to Fig.1, it can be seen that the films grown on NGO show the least polarization dependence, and therefore have the least amount of out of plane anisotropy. Applying in-plane stress, however, alters the observed spectra. Both in-plane tension and compression affect the in-plane absorption (s pol.) similarly near 530 eV. We attribute this to changes in the O $2p - \text{Mn } t_{2g}$ hybridization due to strain in the films. In addition, close inspection of the spectra reveal an interesting difference along the leading edge between 528-529 eV. For the thinnest films under tensile stress, the s polarized absorption is greater in this region, and we see an opposite effect for films under compressive stress. This opposite effect correlates well with the structural changes that occur in these films; the unit cell elongates perpendicular to the surface for films under compressive stress, while it shortens for films under tensile stress. We have developed a simple model of how the Mn-O octahedra of LCMO reorient under strain, and how this affects the O $2p - \text{Mn } 3d$ hybridization. This allows us to assign the 528-529 eV transitions to changes in the O $2p - \text{Mn } e_g$ hybridization. The effect is observed to be opposite for tensile versus compressive stress. Finally, it is observed that for the 2500 Å films, the spectra are similar regardless of the substrate, implying a similar electronic structure for the thickest films. We attribute this to inelastic strain relief mechanisms, such as defect formation, that allow the film to relax towards its "bulk like" electronic structure as the thickness increases beyond a critical thickness.

Conclusions: Polarization dependent XAS spectra at the O 1s edge show changes in the O $2p - \text{Mn } 3d$ hybridization. The thinner films are elastically strained, and this strain affects the electronic structure of the films. It is observed that both in-plane compression and tension increase the in-plane O $2p - \text{Mn } t_{2g}$ hybridization. However, in-plane compression increases the out-of-plane O $2p - \text{Mn } e_g$ hybridization, with in-plane tension having the opposite effect. Since the Mn e_g orbitals are the important orbitals for conductivity and electron transport in LCMO, we suggest that the CMR behavior of these films will have a strong anisotropy as well.

References:

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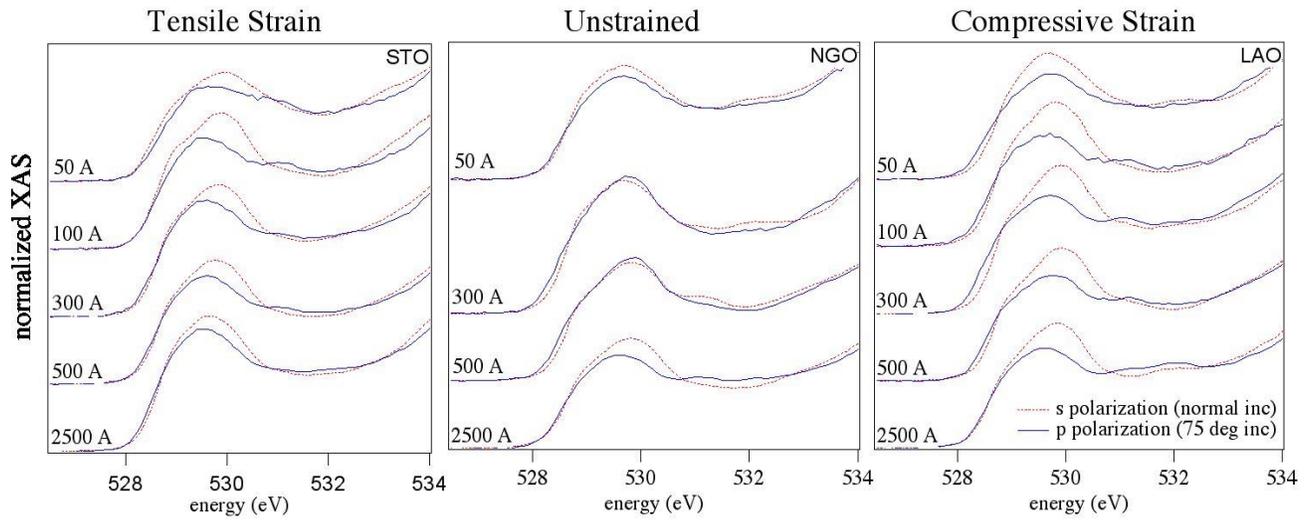


Figure 1: Polarization dependence of O 1s XAS spectra as a function of LCMO film thickness for thin films under tensile strain (STO substrate), unstrained (NGO substrate), and compressive strain (LAO substrate).