

## Magnetic Properties of MBE Grown $La_{1/3}Y_{1/3}Sr_{1/3}MnO_3$ Thin Films and Superlattices W.A. Ruiz, C. Kengle, J. Payne, D. Brown, M.P. Warusawithana, T.M. Pekarek

## Abstract

We have investigated the magnetic properties of thin films related to the standard CMR system  $La_{2/3}Sr_{1/3}MnO_3$  where Y substituted for 50% of the La atoms. These  $La_{1/3}Y_{1/3}Sr_{1/3}MnO_3$  films were grown as a random alloy where La, Y, and Sr atoms randomly occupied the A-site or as a superlattice where each unit-cell-thick layer stacked along the crystallographic (001) direction contained only one of the atoms La, Y, and Sr occupying the A-site. One of the key magnetic features of  $La_{2/3}Sr_{1/3}MnO_3$  is a prominent ferromagnetic transition near 350 K. We find the substitution of La with Y suppresses this ferromagnetic transition in both the random alloy and the superlattice samples. In the superlattice sample we find a magnetic transition that is coincident with a metal-to-insulator transition we observe in electronic transport. In the random alloy sample, we see a similar magnetic transition but at lower temperatures where we find the sample is too insulating to measure electronic transport. We will compare our measurements on these  $La_{1/3}Y_{1/3}Sr_{1/3}MnO_3$  samples with CMR thin films of  $La_{2/3}Sr_{1/3}MnO_3$ .



(Top) Bulk  $La_{1-x}Sr_xMnO_3$  Phase diagram. [Hemberger *et al*, PRB 66, 094410 (2002).] (Bottom) Mechanism for double exchange interaction.

- $La_{1-x}Sr_{x}MnO_{3}$  has been extensively documented in bulk crystals with respect to temperature and strontium concentration.
- Undoped LaMnO<sub>3</sub> is a Mott insulator that is paramagnetic at high T and antiferromagnetic at low T.
- Around x = 0.4,  $La_{1-x}Sr_xMnO_3$  is a paramagnetic insulator at higher temperatures but at lower temperatures it transitions to a ferromagnetic conductor.
- This ferromagnetic-conducting state is kinetically favorable around x = 0.4 at lower temperatures because the double exchange interaction allows for the delocalization of electrons across spin aligned manganese atoms, reducing free energy.

Department of Physics, University of North Florida, Jacksonville, Florida 32224, USA



- Samples grown using Molecular Beam Epitaxy (MBE).
- Carefully calibrated molecular beams of constituent elements produce thin films one atomic layer at a time.
- Constituent elements sources are heated in effusion cells to evaporate material into the chamber, forming collimated molecular beams.
- Ozone was introduced into the chamber to form oxides, allowing us to keep the chamber pressure low (5  $\times$  10<sup>-7</sup> Torr) due to ozone's high oxidizing potential.



XRD shows that both the  $La_{1/3}Y_{1/3}Sr_{1/3}MnO_3$  random alloy sample and the  $LaMnO_3$ -YMnO<sub>3</sub>-SrMnO<sub>3</sub> superlattice sample are clamped to the NdGaO<sub>3</sub>(110) substrate. Furthermore, the superlattice peaks observed in the XRD data indicates that the ordered stacking sequence of the superlattice sample persists throughout the thickness of the film.



YMnO<sub>3</sub> SrMnO<sub>3</sub> LaMnO<sub>3</sub> random alloy Reflection High Energy Electron Diffraction (RHEED) images taken *in situ* during the growth of the films indicate epitaxial layer-by-layer growth. RHEED images during the growth of a supercell: (left) end of LaMnO<sub>3</sub> monolayer, (left center) end of YMnO<sub>3</sub> monolayer, (right center) end of SrMnO<sub>3</sub> monolayer. Surface reconstructions observed in RHEED patterns indicate the degree of octahedral distortions in the strained superlattice. RHEED image at the (right) end of the  $La_{1/3}Y_{1/3}Sr_{1/3}MnO_3$  random alloy sample.

Front view picture of the chamber used in our growths.



Magnetization versus temperature for the  $La_{1/3}Y_{1/3}Sr_{1/3}MnO_3$  superlattice sample is shown in the bottom trace. The key feature is a ferromagnetic transition near 85 K. The top traces show the raw data for the combined supperlattice and NGO substrate as well as the NGO substrate alone.





Temperature (K) (Bottom) As expected  $La_{2/3}Sr_{1/3}MnO_3$  when grown as a random alloy where La and Sr atoms randomly occupy the A-site of the perovskite structure, has a conducting (and ferromagnetic) ground state. (Top) Replacing half of the La atoms with Y atoms,  $La_{1/3}Y_{1/3}Sr_{1/3}MnO_3$ , leads to an insulating ground state. While both La and Y are in a 3+ configuration, the ionic size effects that leads to octahedral distortions play a key role in modifying the electronic properties. (Middle) Growing this in the form of a superlattice, LaMnO<sub>3</sub>-YMnO<sub>3</sub>-SrMnO<sub>3</sub>, the ordered interfaces influence the electronic transport leading to conducting tendencies and associated magnetic order to appear around 100 K.

DMR-This research was supported by the UNF Terry Presidential Professorship, a UNF OUR Research grant, the Florida Space Grant Consortium, a Purdue University Academic Reinvestment Program and by the National Science Foundation (NSF) Grant Nos. DMR-14-29428 and 16-26332. 

dM/dT is shown versus T for the superlattice sample. The minimum is taken to be the ferromagnetic transition temperature.

## Acknowledgements