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Trevor Tyson, Assistant Professor,  
Department of Physics, New Jersey  
Institute of Technology, Newark, NJ  
Tyson@admin.njit.edu

## Probing Local Magnetic Order in Transition Metal Oxides

Q. Qian<sup>1</sup>, T.A. Tyson<sup>1</sup>, C.-C. Kao<sup>2</sup>, M. Croft<sup>3</sup> and A. Yu. Ignatov<sup>1</sup>

<sup>1</sup>New Jersey Institute of Technology; <sup>2</sup>Brookhaven National Laboratory, NSLS; <sup>3</sup>Rutgers University

*In the transition metal oxide  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  (with  $0 < x < 1$ ), electrical conductivity is strongly influenced by external magnetic fields, making this material an exceptionally good candidate for a new generation of magnetic sensors that may significantly improve magnetic data storage. To probe the local spin arrangement in transition metal oxides, we have developed a method to directly assess the local magnetic ordering about specific metal atoms. This method promises to lead to a better understanding of magnetism in magnetic oxides and to assist in improving theoretical models of correlated electron systems.*

Intriguing phenomena such as high-temperature superconductivity (electrical conductivity without resistance) and colossal magnetoresistance (change in electrical resistance due to the presence of a magnetic field) have stimulated renewed interest in the physics of transition metal oxides based on perovskite ( $\text{CaTiO}_3$ ), by using techniques such as x-ray absorption spectroscopy (XAS) and x-ray emission spectroscopy (XES).

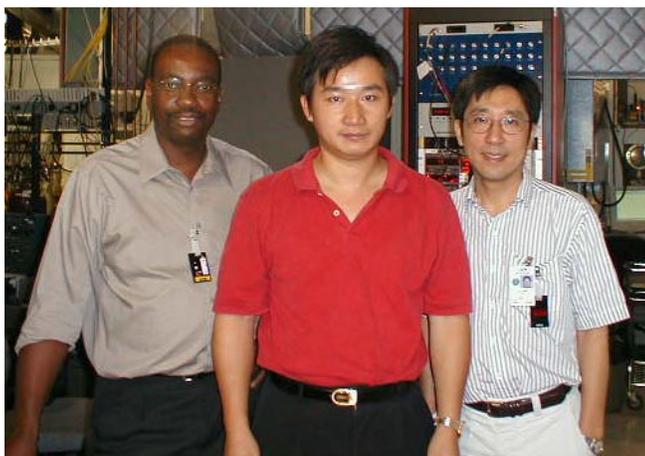
High resolution XAS work has provided evidence of intriguing temperature dependencies of the manganese K-XAS pre-edges of  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  materials which accompany changes upon crossing phase boundaries involving combinations of insulating, paramagnetic, antiferromagnetic, ferromagnetic, and charge-orbital ordered phases. But these interphase changes are not well understood. Consequently, we used a powerful combination of XAS and XES, called spin-polarized x-ray absorption near edge

spectroscopy (SPXANES), to extract spin-polarized x-ray absorption spectra and integrate the results into a model which is sensitive to local magnetic order.

SPXANES is based on energy resolving the 3p to 1s transition ( $K_\beta$  emission) and measuring the emission from the main or satellite lines of the spectra as a function of the incident x-ray energy. Previous studies focused on the nature of the splitting between spin up and spin down channels in the main line, but no emphasis was made on the pre-edge region and on the

temperature-dependent changes in SPXANES.

Manganese SPXANES measurements were performed at beamline X21 of the National Synchrotron Light Source. SPXANES spectra were collected by monitoring the  $K_\beta$  fluorescence yield at two energies, specific to spin up and down final states, while the incident energy across the near-edge region was scanned. Measurements were performed for  $x = 0, 0.3, 0.5,$  and  $1$  (corresponding to  $\text{LaMnO}_3$ ,  $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ ,  $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$  and  $\text{CaMnO}_3$ , respectively) at two temperatures: 15 and 300 Kelvin (K).



*Members of the team of scientists who performed the study (from left to right): Trevor Tyson (lead author), Qing Qian, and Chi-Chang Kao.*

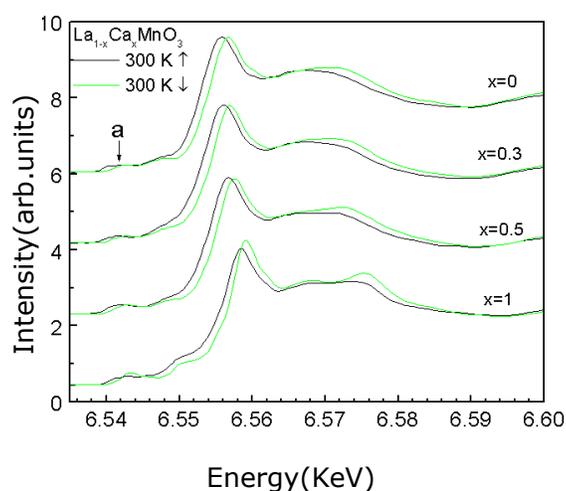
All the K-edge SPXANES spectra of  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  are shown in **Figure 1**. They reveal a marked splitting between the spin up and spin down channels of both the pre-edges (energy between 6.535 and 6.55 kiloelectronvolts (keV)) and the main edges (energy between 6.55 and 6.58 keV).

In **Figure 2**, we expand

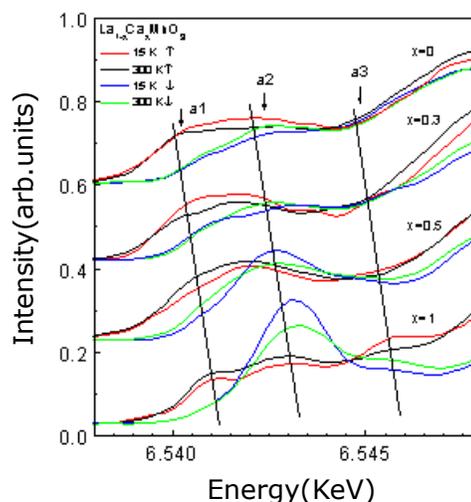
the pre-edge region of the SPXANES spectra. Three peaks (labeled a1, a2, and a3) in  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  ( $x = 0, 0.3, 0.5, \text{ and } 1$ ) are observed. By examining the changes in the pre-edge spectra for temperatures above and below magnetic ordering temperature, we have developed an electron excitation model to understand the origin of the features a1, a2 and a3.

In **Figure 3**, we display our transition model, which shows that the changes in the pre-edge region with temperature can be directly linked to changes in magnetic ordering of the manganese ions around absorbing manganese sites. The model also shows a transition from a random local magnetic arrangement to an ordered arrangement.

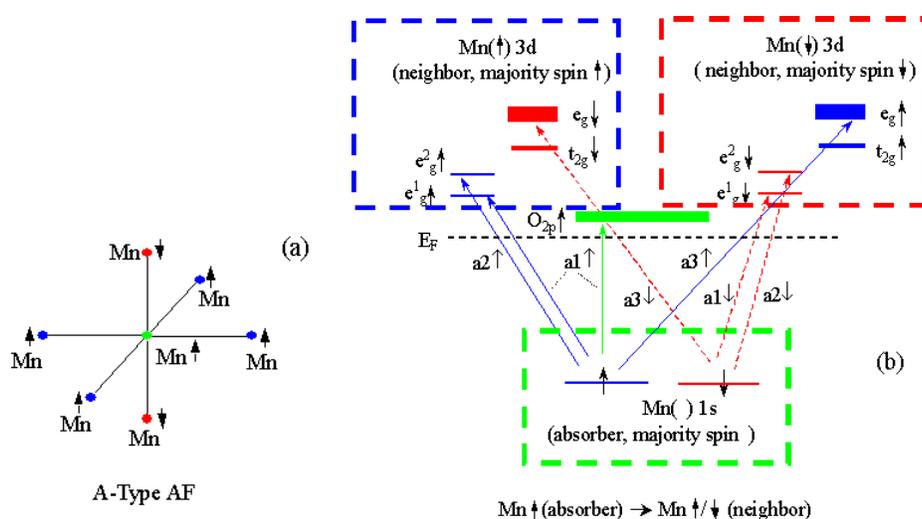
Our model could be used to investigate a great range of perovskite transition metal materials, and, more generally, octahedrally-coordinated transition metal materials. Our model and SPXANES measurements could also be used to predict magnetic ordering changes in materials such as thin films, single crystal or powders.



**Figure 1.** SPXANES spectra of  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  for  $x = 0, 0.3, 0.5, \text{ and } 1$ . The solid and green lines correspond to the spin up and down channel, respectively, measured at 300 Kelvin.



**Figure 2.** Temperature-dependent pre-edge SPXANES spectra of the region near feature a in Figure 1. The three straight lines indicate the a1, a2, and a3 features.



**Figure 3.** Schematic diagram showing the transitions in the pre-edge for spin-polarized absorption of  $\text{LaMnO}_3$  in the low-temperature A-type antiferromagnetically ordered state. (a) Local magnetic ordering of magnetic ions. (The oxygen atoms are not shown for clarity.) (b) Allowed transitions for excitation of manganese, in which the d final state of a neighbor is of the same spin polarization as the absorber (left panel) or the spin polarization is reversed (right panel).