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Probing Local Magnetic Order in Transition Metal Oxides

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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 (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 ab-

sorption 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_β 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 X21A of the National Synchrotron Light Source at Brookhaven National Laboratory. SPXANES spectra were collected by monitoring the K_β 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 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 E between 6.535 and 6.55 kiloelectronvolts (keV)) and the main edges (energy E 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 La_{1-x}

Ca_xMnO_3 ($x = 0, 0.3, 0.5,$ 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 inves-

tigate 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.

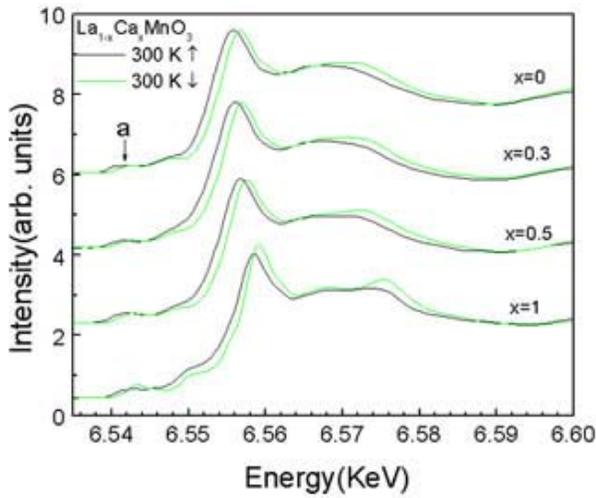


Figure 1. SPXANES spectra of $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ for $x = 0, 0.3, 0.5,$ and 1 . The solid and green lines correspond to the spin up and down channel, respectively, measured at 300 Kelvin.

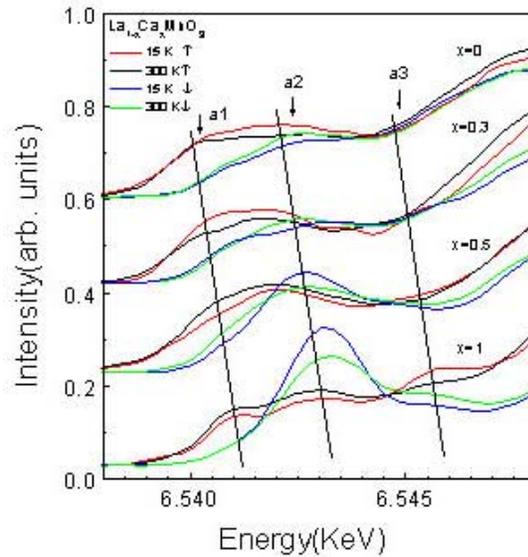


Figure 2. Temperature-dependent pre-edge SPXANES spectra of the region near feature a in figure 1. The three straight lines indicate the a1, a2, and a3 features.

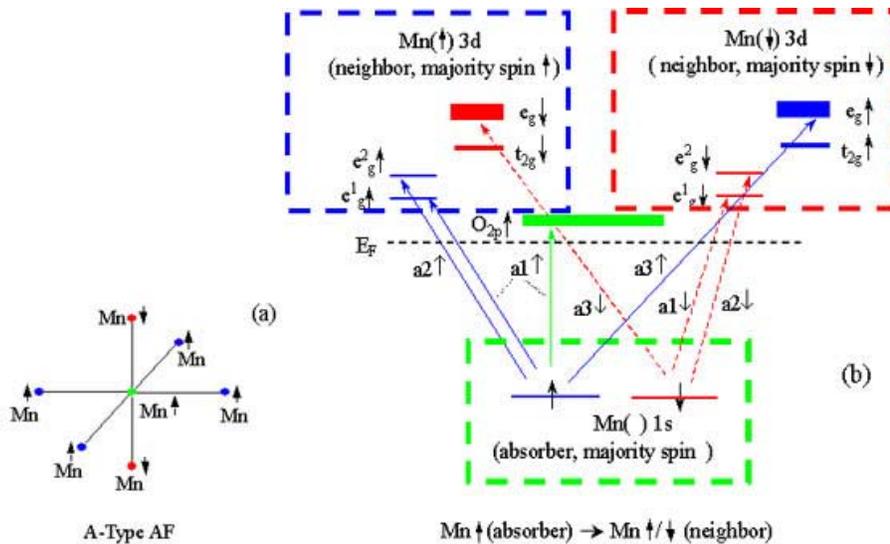


Figure 3. Schematic diagram showing the transitions in the pre-edge for spin-polarized absorption of LaMnO_3 in the low-temperature A-type antiferromagnetically ordered state. (a) Local magnetic ordering of magnetic ions. (The oxygen atoms are not shown for clarity.) (b) Allowed transitions for excitation 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).