

X-ray Resonant Diffraction Studies on the Phase Transition in $\text{Pr}_{0.6}\text{Ca}_{0.4}\text{MnO}_3$

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Introduction

Interatomic electronic correlations in transition metal oxides play a determining role in macroscopic electronic transport properties, including phenomena such as high-temperature superconductivity or colossal magnetoresistance [1]. The ground state of these materials is the result of the balance between the degrees of freedom of the electronic configuration, and it leads to phenomena such as charge localization and orbital and spin ordering, frequently accompanied by slight lattice deformations. Since the electronic configurations of the metal and oxygen atoms are still not well known, the existence of charge/orbital ordering and details about it are still being debated for many compounds. As part of a large amount of work that was done to characterize the electronic configuration in the valence bands, we studied the perovskite manganites $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$ that show colossal magnetoresistance below a phase transition temperature of about 230K.

Methods and Materials

By doping $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$, we can alter the balance between several degrees of freedom. With a ratio of $x = 0.3$ to 0.7 , a phase transition is observed and is assumed to be related to an ordering of the formal valences (charge ordering) of the Mn atoms together with an orbital ordering. Our group studied this compound previously by using the resonant x-ray scattering technique to show that the orbital and charge orders have different correlation lengths and that the charge ordering drives the orbital ordering [2]. Here we report on the continuation of this work performed at the CMC-CAT sector at the APS. In this study, we have focused on the incident energy dependence of the diffracted intensity as the incident energy is scanned through the Mn K edge. In this process, inner resonant electrons are virtually promoted to local unoccupied states, which, in turn, are sensitive to the electronic configuration of the resonant atom.

Results

We have measured diffraction peaks at the Mn K edge and uncovered a remarkable change in behavior through the phase transition. The peaks are characterized by measuring their energy and temperature dependencies as well as their dependence on the polarization direction of the incident and diffracted photons. In particular, to get more information, we have measured the peaks in the σ - σ and σ - π channels that lead to the different spectra, in part because of the different crystallographic rules that apply to each channel. For example, Fig. 1 shows the spectra of the (010) reflection in the σ - π channel taken at different temperatures above the phase transition. It shows three peaks, each of which has a Lorentzian shape with half widths that increase as a function of the energy. The peak at 6552 eV coincides with the main, strong resonance in the σ - σ channel, the behavior of which mirrors the charge

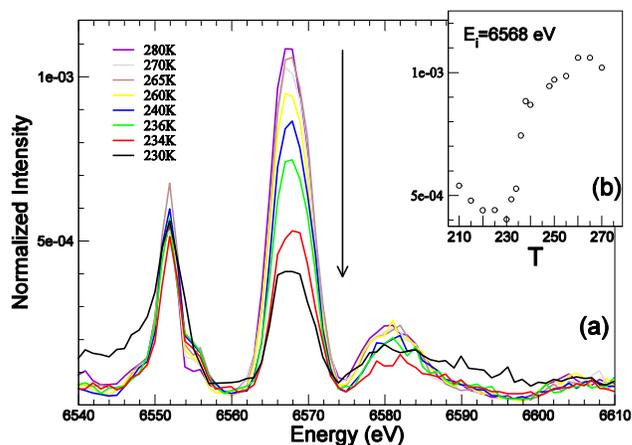


FIG. 1. (a) Intensity of the (010) reflection vs. energy and temperature at the Mn K edge in the s - p channel. (b) Intensity at 6568 eV vs. temperature.

order parameter and was reported on in Reference 3. Remarkably, the anomaly at 6568 eV increases as the temperature increases through the phase transition, characterizing a change of the electronic density around Mn atoms.

Discussion

The deduction of the precise electronic configuration around the Mn atom is not straightforward, since valence changes or structural changes (which may be due to an effect other than a valence change) give rise to similar effects on the intensity behavior but not the same effects. A further detailed study, together with theoretical calculations of the measured spectra, will be performed to provide a definitive conclusion about the existence of and the model for the charge/orbital ordering in this compound.

Acknowledgments

Use of the APS was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38. Brookhaven National Laboratory is supported under DOE Contract No. DE-AC02-98CH10886. Support from the National Science Foundation's Materials Research Science and Engineering Centers program under Grant No. DMR-0080008 is also acknowledged.

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