A Resonant X-ray Scattering Study of Octahedral Tilt Ordering in LaMnO₃ and P_{1-x}Ca_xMnO₃

M. v. Zimmermann,¹ C. S. Nelson,¹ Y.-J. Kim,¹ J. P. Hill,¹ D. Gibbs,¹ H. Nakao,² Y. Wakabayashi,² Y. Murakami,² Y. Tokura,³ Y. Tomioka,³ C.-C. Kao,⁴ D. Casa,⁵ C. Venkataraman,⁵ T. Gog⁵

¹ Dept. of Physics, Brookhaven National Laboratory, Upton, NY, U.S.A.

² Photon Factory, Institute of Materials Structure Science, Tsukuba, Japan

³ Joint Research Center for Atom Technology (JRCAT), Tsukuba, Japan

⁴ National Synchrotron Light Source, Brookhaven National Laboratory, Upton, NY, U.S.A.

⁵ CMC-CAT, Advanced Photon Source, Argonne National Laboratory, Argonne, IL, U.S.A.

Introduction

During the last several years, there has been enormous interest in x-ray resonant scattering studies of orbital ordering in transition metal oxides. ¹⁻⁹ Here, we report an x-ray scattering study of octahedral tilt ordering in the manganite series Pr_{1-x}Ca_xMnO₃ with x=0.4 and 0.25 and in LaMnO₃.

Methods and Materials

The sensitivity to tilt ordering is achieved by tuning the incident x-ray energy to the $L_{\rm I},\,L_{\rm II},$ and $L_{\rm III}$ absorption edges of Pr and La, respectively. The polarization dependence is predominantly σ to $\pi,$ and the azimuthal dependence follows a sin-squared behavior. These results lead to a description of the cross section in terms of Templeton scattering in which the tilt ordering breaks the symmetry at the rare earth site.

Results

Figure 1 shows the energy dependence of the scattering at the (0,1,0) orbital wavevector of Pr_{0.75}Ca_{0.25}MnO₃. A large resonant signal is visible at ħω=6.547 keV near the Mn K edge. Remarkably, there is also a resonant feature at the Pr LII edge with ħω=6.44 keV. The latter resonance arises from octahedral tilt ordering. The most interesting result of the present work is our observation that octahedral tilt ordering persists above the orbital ordering transition temperatures in all three samples. Indeed, we identify separate structural transitions, associated with the onset of orbital and tilt ordering, respectively,

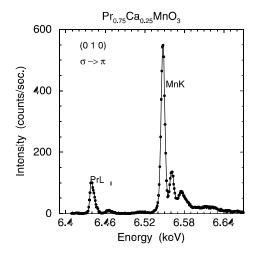


FIG. 1. The energy dependence of the scattering at the (0,1,0) orbital wavevector of $Pr_{0.75}Ca_{0.25}MnO_3$.

Discussion

The main features of the resonant profiles for Pr_{0.75}Ca_{0.25}MnO₃, are reproduced above the orbital ordering temperature. Importantly, from the perspective of the theory of Benedetti et al.,¹⁰ no changes in the L-edge lineshapes are observed with increasing temperature through the orbital transition. From this we conclude that the main features of the L-edge resonant profiles reported here originate in octahedral tilt ordering and that any contributions to the L-edge resonances arising from the Mn orbital order are too small to be detected. A full discussion of the present results has been accepted by *Physical Review B*.⁸

Acknowledgments

These experiments were carried out both at the NSLS and at the APS. The work at Brookhaven, both in the Physics Department and at the NSLS, was supported by the U.S. Department of Energy, Division of Materials Science, under Contract No. DE-AC02-98CH10886. Support from the Ministry of Education, Science and Culture, Japan by the New Energy and Industrial Technology Development Organization (NEDO), and by the Core Research for Evolution Science and Technology (CREST) is also acknowledged. Work at the CMC beamlines is supported, in part, by the Office of Basic Energy Sciences of the U.S. Department of Energy and by the National Science Foundation, Division of Materials Research. Use of the Advanced Photon Source was supported by the Office of Basic Energy Sciences of the U.S. Department of Energy under Contract No. W-31-109-Eng-38.

References

- ¹ Y. Murakami et al., Phys. Rev. Lett. **80**, 1932 (1998).
- ² S. Ishihara et al., Phys. Rev. Lett. **80**, 3799 (1998).
- ³ Y. Murakami et al., Phys. Rev. Lett. **81**, 582 (1998).
- ⁴ M. Fabrizio et al., Phys. Rev. Lett. **80**, 3400 (1998).
- ⁵ I.S. Elfimov et al., Phys. Rev. Lett. **82**, 4264 (1999).
- ⁶ M. Benfatto et al., Phys. Rev. Lett. **83**, 636 (1999).
- ⁷ M. v. Zimmermann et al., Phys. Rev. Lett. **83**, 4872 (1999).
- ⁸ M. v. Zimmermann et al., Phys. Rev. B (in press).
- ⁹ C.S. Nelson et al., Phys. Rev. B (submitted).
- ¹⁰ P. Benedetti et al., Phys. Rev. B (in press).