Chapter 5

Conclusion and Future Work

5.1 Conclusion

5.1.1 Experimental Setup

A. XAS system

Soft x-ray absorption experimental setup has been established. The specially designed rotary sample holder employed in polarization-dependent XAS, eliminates the difference in optical path and probing area and provides reliable XAS measurements. The polarization dependence and linear dichroism in XAS are useful techniques in determining the orbital polarization of transition metal oxide compounds.

B. PLD system

Pulsed laser deposition is an extremely useful technique for fabricating thin films with smooth surfaces and interfaces. With this PLD setup, we realized the epitaxy of strained perovskite films caused by substrates demonstrated that our PLD system is suitable for making thin films of manganite compounds. Furthermore, strain effect has been discussed to elucidate the

degrees of freedom which control the physical properties. A PLD thin film growth system could provide us an opportunity to study strongly correlated materials.

5.1.2 Orbital Ordering in $La_{0.5}Sr_{1.5}MnO_4$

We found that the conventional model of orbital ordering of $3x^2-r^2/3y^2-r^2$ type in the e_g states of $La_{0.5}Sr_{1.5}MnO_4$ is incompatible with measurements of linear dichroism in the Mn $2p$-edge x-ray absorption, whereas these e_g states exhibit predominantly cross-type orbital ordering of x^2-z^2/y^2-z^2. LDA+U band-structure calculations reveal that such a cross-type orbital ordering results from a combined effect of antiferromagnetic structure, Jahn-Teller distortion, and on-site Coulomb interactions.

5.1.3 Orbital Polarization in Strained $La_{0.5}Sr_{0.5}MnO_3$

We report spectroscopic evidence for the existence of orbital-mediated phases in strained $La_{0.5}Sr_{0.5}MnO_3$ thin films. Measurements of polarization-dependent soft x-ray absorption reveal that electronic states responsible for lowest-energy excitations in C-type anti-ferromagnetic (AFM) $La_{0.5}Sr_{0.5}MnO_3$ films have an orbital symmetry of $3z^2-r^2$, while those in A-type AFM films have an orbital symmetry of x^2-y^2. Compared with band-structure calculations in the LDA+U approximations, we results indicate that such orbital polarizations in strained films result from a combined effect of tetragonal Jahn-Teller distortion and Coulomb interactions of Mn $3d$ electrons.

5.1.4 Orbital Symmetry in Na_xCoO_2

Measurements of polarization-dependent soft x-ray absorption reveal that the electronic states determining the low-energy excitations of Na_xCoO_2 have predominantly a_{1g} symmetry with significant O $2p$ character. In contrast to the prediction of band theory, doping-dependent O $1s$ x-ray absorption shows a large transfer of spectral weight, providing spectral evidence for strong electron correlations of the layered cobaltates. We also found that Na_xCoO_2 exhibits a charge-transfer electronic character rather than a Mott-Hubbard character.

5.2 Future Work

The recent discovery of superconductivity in cobalt oxides with two-dimensional triangular-lattice CoO_2 layers has attracted much interest. Therefore, to study Co oxides with different two-dimensional structures is essential. Matsuno [1] *et al.* realized that possibility of growing a novel compound, Sr_2CoO_4, which has a crystal structure identical to superconducting Cu oxides, $La_{2-x}Sr_xCuO_4$, i.e., the K_2NiF_4-type structure. Single crystal of Sr_2CoO_4 compounds has never been synthesized before, even in the form of bulk polycrystal. They have first succeeded in fabricating a single-crystalline thin film of Sr_2CoO_4 by employing the pulsed laser deposition method. The Sr_2CoO_4 has a square-lattice CoO_2 layers in contrast to the triangular-lattice CoO_2 layers in Na_xCoO_2. Matsuno *et al.* also indicated that Sr_2CoO_4 has

both ferromagnetic and metallic behaviors, with a Curie temperature at Tc $\sim$ 250 K, rather than superconductivity. Sr_2CoO_4 may be regarded as a quasi-two-dimensional metallic ferromagnet, which has never been observed in the K_2NiF_4-type family. Therefore, the electronic structure of Sr_2CoO_4 is one of our focused subjects.

Reference

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Appendix A

List of Publications

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