Driving Mechanism Behind Diverse Electronic States in Manganese Perovskites $Nd_{1-x}Sr_xMnO_3$

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Introduction

Manganese perovskites have much attracted interest due to many intriguing phenomena such as colossal magnetoresistance (CMR), and the charge / orbital-ordering. $Nd_{1-x}Sr_xMnO_3$ is a suitable system to understand the electronic structure for these phenomena, because it shows various electronic phases, such as paramagnetic insulator (PI), ferromagnetic metal (FM), antiferromagnetic metal (AFM) and antiferromagnetic insulator (AFI) depending on temperature and x [1]. In order to clarify the driving mechanisms leading to various aspects of the *3d* states, we have performed high-resolution Mn *2p-3d* resonance photoemission (RPES), X-ray absorption spectroscopy (XAS) of $Nd_{1-x}Sr_xMnO_3$

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Methods and Materials

Single crystalline $Nd_{1-x}Sr_xMnO_3$ (x = 0.40, 0.47, 0.50, 0.55, 0.63) samples were prepared by the floating-zone method. The compounds with x = 0.40 and 0.47 undergo a PI-to-FM transition at T_C ~ 290 K and 275 K, respectively, where the FM state is stable down to the lowest temperatures below T_C. The PI-FM transition is also seen for x = 0.50 (T_C ~ 255 K), while this compound is a charge-ordered insulator (COI) below T_{COI} ~ 160 K. The x = 0.55 sample shows a PI-to-AFM transition at T_N ~ 220 K, in which AFM phase consists of antiferromagnetically stacking 2 dimensional ferromagnetic sheets due to the $d(x^2-y^2)$ orbital ordering. The x = 0.63 sample undergoes a PI-to-AFI transition at T_N ~ 220 K. We have performed high-resolution soft X-ray bulk sensitive Mn *2p-3d* RPES and XAS with an energy resolution of 100 meV at BL25SU in SPring-8. The base pressure was 4×10^{-8} Pa. The sample surface was obtained by fracturing *in situ*.

Results

Figure 1 shows the Mn *2p-3d* RPES spectra of $Nd_{1-x}Sr_xMnO_3$ measured at the Mn *2p-3d* resonant maximum ($h\nu$ = 643 eV). It has been reported that a possible Auger contribution is negligible within the energy region from E_F to 3 eV at hν = 643 eV [2]. For each x, the Mn *3d* spectral function in the ground-state (GS) at a low-temperature is noticeably different from that in the PI phase near room temperature [3]. It is found that the spectral line shape in the GS is closely related to the shape of MnO_6 octahedra depending on x due to a *static* Jahn-Teller (JT) effect, while the line shape in the PI phase is qualitatively similar to each other irrespective of x. In the PI phase, however, Mn *3d* spectra show the finite intensity at E_F, indicating that the compounds in the PI phase are neither simple band insulator nor Mott insulator.

Discussion

The small but finite spectral weight at E_F in the PI phase suggests that there are "incoherent" hopping electrons whose energy is very a close to the Fermi level. We note that the relatively low resistivity [4] in the PI phase also supports this scenario. One of the acceptable scenarios to explain finite spectral weight at the Fermi level is a *dynamic* JT polaron model suggested by Millis *et al* [5]. Therefore, we conclude that the competition of both the *static* and *dynamic* Jahn-Teller effects at high and low temperatures, respectively, are responsible for the actual electronic states.

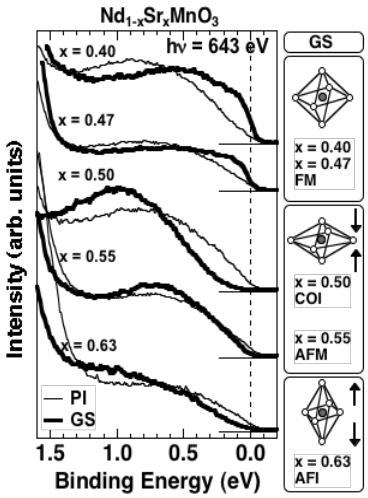

Fig. 1. Mn 3d spectral functions in PI (fine line) and GS(thick line). It is shown beside the plot that the prospective shape of the MnO_6 octahedron in GS reported from neutron diffraction [1].

Acknowledgements

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[1] R. Kajimoto et al., Phys. Rev. B **60**, 9506 (1999)
[2] H. Fujiwara et al. J. Electron Spectrosc. Relat. Phenom. **144-147**, 807 (2005)
[3] A. Sekiyama et al., cond-mat/0401601,
[4] H. Kuwahara et al., J. Appl. Phys. **93**, 7367 (2003)
[5] A. J. Millis et al., Phys. Rev. Lett. **77**, 175 (1996)