Temperature-dependent study of charge disproportionation in La_{1-x}Sr_xFeO₃

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Introduction

Carrier doping into a Mott insulator causes various intriguing properties. Recently, charge ordering (CO) and its associated metal-insulator transition (MIT) have attracted great interest in relation to charge stripes in high- T_c cuprates [1] and giant magnetoresistance in manganites. $La_{1-x}Sr_xFeO_3$ (LSFO) with $x \sim 0.67$ exhibits charge disproportionation (CD), a unique type of CO, below 190 K [2]. Matsuno et al. [3] studied detailed temperature dependent changes near the Fermi level (E_F) in the photoemission spectra of LSFO with x = 0.67. The intensity at E_F was found to change dramatically across the transition temperature. In their measurements, however, the O 2p contribution overwhelmed the Fe 3dcontribution due to the low photon energies hv = 21.2 -100 eV [4] and information directly related to Fe 3d states has been lacking. In this work, we address the question of how the electronic structure of LSFO changes as the charge disproportionation occurs by measuring soft x-ray photoemission and absorption spectra of epitaxiallygrown high-quality thin films prepared in situ.

Experimental

The LSFO thin films were fabricated in a laser MBE chamber connected to a synchrotron radiation photoemission system at BL-2C of Photon Factory [5]. LSFO thin films were deposited on Nb-doped TiO₂-terminated SrTiO₃ (001) substrates [6] at 950 °C at an oxygen pressure of 1×10^{-4} Torr. The fabricated LSFO thin films were transferred into the photoemission chamber under an ultrahigh vacuum of 10^{-10} Torr. The PES spectra were taken at room temperature with the total energy resolution of about 250 meV at the photon energy of 710 eV (Fe $2p \rightarrow 3d$ resonance). After the measurements, we confirmed that for x = 0.67, there is a jump of resistivity at 190 K (T_{CD}).

Results and Discussion

Figure 1 shows the temperature dependence of the valence-band photoemission spectra of LSFO (x = 0.67) taken at $h\nu = 710$ eV (Fe $2p \rightarrow 3d$ resonance). The spectra change gradually with temperature. The spectra clearly indicate the transfer of spectral weight from lower

to higher binding energies with decreasing temperature within 2 eV of E_F , i.e., within the e_g band. The energy range at which spectral weight transfer occurs is about 2 eV, which is large compared with the transition temperature $T_{CD} = 190$ K (~ 22 meV). Spectral weight transfer over such a wide energy range was also reported in other transition-metal oxides such as La_{1-x}Sr_xMnO₃ [7]. We also observed the temperature dependence of the photoemission spectra taken at 600 eV and the O 1*s* x-ray absorption spectra. Similar temperature dependence was observed in the x = 0.4 sample, suggesting a local charge disproportionation even at x = 0.4. This is consistent with the previous report [3].



Fig. 1: Temperature dependence of the valence-band photoemission spectra of LSFO (x = 0.67) taken at hv = 710 eV (Fe $2p \rightarrow 3d$ resonance).

References

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