Doping dependence of the photoemission spectra of La_{1-x}Sr_xFeO₃ epitaxial thin films

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Introduction

Since the discovery of high- T_c superconductivity in the cuprates, great interest has revived in perovskite-type transition-metal oxides because of their intriguing properties, such as metal-insulator transition (MIT), colossal magnetoresistance (CMR), and ordering of spin, charge, and orbitals [1]. La_{1-x}Sr_xFeO₃ (LSFO) has attracted much interest because it undergoes a charge disproportionation around x = 0.67 [2]. The central question about this system is how the electronic structure evolves from the charge-transfer-type insulator LaFeO₃ to the oxygen-hole metal SrFeO₃. To answer this question, we have performed *in-situ* photoemission spectroscopy (PES) and x-ray absorption spectroscopy (XAS) measurements of epitaxial thin films of LSFO (x = 0, 0.2, 0.4, 0.67). A systematic x-ray photoemission study of scraped bulk LSFO samples has been reported by Chainani et al. [3]. Structures in the valence band, however, were not clearly resolved partly because of the limited energy resolution (~ 0.8 eV). In this study, we have succeeded in obtaining bulk-sensitive high-quality spectra by using soft x-ray [4] and atomically flat surfaces of the thin films.

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 200 meV at the photon energy of 600 eV.

Results and Discussion

Figure 1 shows the doping dependence of the combined valence-band PES and the O 1s XAS spectra. In the PES spectra, one can observe three main structures A (e_g band), B (t_{2g} band), and C (Fe 3d - O 2p bonding states) and the satellite. A gap (absence of finite DOS at E_F) or a pseudogap (depression of DOS at E_F) was seen for all

values of x, which is considered to be a natural consequence of the wide insulating region of the LSFO phase diagram [7]. Structures A-C move toward E_F upon hole-doping. In addition, structure A becomes weaker with increasing x, indicating that holes are doped into the e_g band. In the XAS spectra, the empty Fe 3d states are split into two peaks, D and E, due to the e_g - t_{2g} crystalfield splitting. A new peak F grows within the band gap upon hole-doping. These results indicate that spectral weight is transferred from structure A below E_F to structure F above E_F , and the band gap is filled by the new spectral weight F as holes are doped. This spectral weight transfer with hole-doping is highly non-rigidband-like.



Fig. 1: Combined PES and XAS spectra of LSFO epitaxial thin films

References

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