In-situ photoemission study of Nd$_{1-x}$Sr$_x$MnO$_3$ epitaxial thin films

Hiroki WADATI*, Riichiro TAKAISHI†, Akira CHIKAMATSU‡, Masaru TAKIZAWA†, Hiroshi KUMIGASHIRA‡, Atsushi FUJIMORI†, Masaharu OSHIMA‡, Mikk LIPPMAA*, Masashi KAWASAKI†, Masashi KAWASAKI‡, Hideomi KOINUMA§

†Department of Physics and Department of Complexity Science and Engineering, University of Tokyo, Kashiwa 277-8561, Japan
‡Department of Applied Chemistry, University of Tokyo, Tokyo 113-8656, Japan
§Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan
‡Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
§Materials and Structures Laboratory, Tokyo Institute of Technology, Yokohama 226-8503, Japan

Introduction

Hole-doped perovskite manganese oxides $R_x$Sr$_x$MnO$_3$ ($R$ = rare earth) have attracted much attention because of their interesting magnetic and transport properties such as colossal magnetoresistance and metal-insulator transition [1]. Among them, Nd$_{1-x}$Sr$_x$MnO$_3$ (NSMO) is especially interesting because for $x = 0.5$ it undergoes a transition from the ferromagnetic metal (FM) to a charge-ordered insulator (COI) upon cooling [2]. In this study, we have performed in-situ photoemission spectroscopy (PES) and x-ray absorption spectroscopy (XAS) measurements of epitaxial thin films of NSMO ($x = 0, 0.2, 0.4, 0.55$) grown on SrTiO$_3$ (001) substrates. We have succeeded in obtaining bulk-sensitive high-quality spectra by using soft x-rays and atomically flat surfaces of the thin films.

Experimental

The NSMO thin films were fabricated in a laser MBE chamber connected to a synchrotron radiation photoemission system at BL-2C of Photon Factory [3]. NSMO thin films were deposited on Nb-doped TiO$_2$-terminated SrTiO$_3$ (001) substrates [4] at 1050 °C at an oxygen pressure of $1 \times 10^{-4}$ Torr. The fabricated NSMO 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

In Fig. 1 shows the doping dependence of the valence-band PES spectra. One can observe four main structures, labeled as A, B, C, and D. From Mn 2p → 3d (643 eV) and Nd 3d → 4f (979 eV) resonant photoemission spectra, structures A and B are ascribed to the Mn $e_g$ and $t_{2g}$ states, respectively, and structures C and D are ascribed to the Mn 3d - Nd 4f - O 2p mixed states. A gap (absence of finite density of states at $E_F$) is seen for all values of $x$, which is considered to be a natural consequence of the insulating behavior of this system at room temperature. The satellite structures are not observed. Structures A-D 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.

From the binding-energy shifts of core levels, it was found that the chemical potential is shifted monotonically without any sign of suppression. The values of the shifts are almost the same as those of La$_{1-x}$Sr$_x$MnO$_3$ [5, 6]. Since the electronic phase separation results in the pinning of the chemical potential, the monotonic chemical-potential shift of NSMO suggests the absence of phase separation on a microscopic scale as in the case of LSMO. This result is considered to reflect the narrower composition range of charge-ordered states than that of, for example, Pr$_{1-x}$Ca$_x$MnO$_3$, which shows charge ordering over a wide composition range around $x = 0.5$.

Fig. 1: PES spectra of NSMO epitaxial thin films.

References

* wadati@wyvern.phys.s.u-tokyo.ac.jp