Soft x-ray linear dichroism study of La_{1-x}Sr_xMnO₃ epitaxial thin films

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Introduction

Hole-doped perovskite manganese oxides $R_{1,x}A_{x}$ MnO3, where R is a rare-earth (R = La, Nd, Pr) and A is an alkaline-earth atom (A = Sr, Ba, Ca), have attracted much attention because of their remarkable physical properties, such as colossal magnetoresistance, and ordering of spin, charge, and orbitals [1]. It was reported that the magnetic and electronic phases can be controlled for thin films of La_{1,x}Sr_xMnO₃ grown on perovskite substrates with various lattice parameters [2]. Recently, Huang et al. [3] succeeded in observing the orbital states of the bulk samples of $La_{1,x}Sr_{1+x}MnO_4$ by measuring the linear dichroism (LD) in the Mn 2p x-ray absorption spectra. In this study, in order to observe the orbital states, we have performed in-situ x-ray absorption spectroscopy (XAS) measurements of epitaxial thin films of La_{0.6}Sr_{0.4}MnO₃ grown on LaAlO₃ (001) (under compressive strain) and SrTiO, (001) (under tensile strain) substrates by laser molecular beam epitaxy (laser MBE).

Experimental

The La_{0.6}Sr_{0.4}MnO₃ thin films were fabricated in a laser MBE chamber connected to a synchrotron radiation photoemission system at BL-2C of the Photon Factory [4]. La_{0.6}Sr_{0.4}MnO₃ thin films were deposited on LaAlO₃ (001) substrates at 500 °C and on SrTiO₃ (001) substrates at 1050 °C at an oxygen pressure of 1×10^4 Torr. The fabricated La_{0.6}Sr_{0.4}MnO₃ thin films were transferred into the photoemission chamber under vacuum of 10^{-10} Torr. The XAS spectra were taken at room temperature in the total-electron-mode.

Results and Discussion

Figure 1 (a) shows the polarization-dependent XAS and LD spectra of a $La_{1,x}Sr_xMnO_3$ epitaxial thin film grown on LaAlO₃ (001) substrates. The line shape of LD was very similar to that of LaSrMnO₄ reported by Huang *et al.* [3]. LaSrMnO₄ is expected to exhibit $3z^2 - r^2$ "ferroorbital" ordering, and Huang *et al.* succeeded in reproducing the LD spectrum of this material by multiplet calculations for Mn³⁺ ions with occupied d_{3z_2,r_2} orbitals. Since the LD signal from Mn⁴⁺ sites is considered to be negligible, the present result indicated that the e_g electrons in a $La_{0.6}Sr_{0.4}MnO_3$ thin film grown on LaAlO₃ (001) were

mainly in $d_{3:2-r^2}$ (out-of-plane) orbitals. This can be easily explained by the compressive strain from LaAlO₃ (001) substrates, which is expected to split the energy levels of e_g orbitals and stabilize the out-of-plane orbitals. These results are consistent with the phase diagram proposed by Konishi *et al.* [2]. Figure 1 (b) shows the result of a La₁. _xSr_xMnO₃ epitaxial thin film grown on SrTiO₃ (001) substrates. The LD signal was expected to be negligible due to the isotropy of orbitals in the ferromagnetic state. However, the experimental LD was very similar to the case of LaAlO₃ (001). This LD signal was unexpected, and further experimental and theoretical studies are needed to resolve this issue.

We are now constructing a new experimental setup for LD measurements which can eliminate experimental artifacts related to the difference in the optical path and to the probing area [5].



Fig. 1: Polarization-dependent XAS and LD spectra of $La_{0.6}Sr_{0.4}MnO_3$ epitaxial thin films grown on (a) $LaAlO_3$ (001) and (b) SrTiO_3 (001) substrates.

References

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