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XMCD Study on Electronic and Spin States of Co in $La_{1-x}Sr_xCoO_3$ ($0 \leq x \leq 0.6$)

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Introduction

The end-member compound $LaCoO_3$ is considered to be a nonmagnetic semiconductor in the low-spin state of Co^{3+} at low temperature. Magnetic susceptibility slowly increases with temperature and reaches a maximum broad peak at $T \cong 100$ K [1,2]. Nonmagnetic $LaCoO_3$ is considered to transform into paramagnetic $LaCoO_3$ with the spin-crossover phenomenon around 100 K.

When holes can be doped into $LaCoO_3$ by substituting Sr for La, the transport mechanism of $La_{1-x}Sr_xCoO_3$ compounds changes from that of a nonmagnetic semiconductor to that of a ferromagnetic metal for $x \geq 0.2$ up to the other end-member of $SrCoO_3$. As well as $LaCoO_3$, the complicated spin-state of $La_{1-x}Sr_xCoO_3$ affects the magnetic and electronic properties, which vary as functions of doped hole concentration x. The origin of itinerant electrons was considered to be the freezing of super-paramagnetic clusters ($x < 0.3$) and long-range ferromagnetic order ($x \geq 0.3$) [3,4]. A site-selective and valence-selective XMCD study is required for the Co *K* absorption edge to elucidate the relationship between magnetic property and crystal structure in terms of the hybridization between the 3*d* and 4*p* states through the ligand 2*p* state.

Experimental

Powder crystals of $La_{1-x}Sr_xCoO_3$ were synthesized from appropriate molar mixtures, having the Sr contents of x = 0, 0.2, 0.3, 0.4, 0.5 and 0.6.

XANES and XMCD experiments were carried out on the BL-3A. The horizontally polarized white X-rays were monochromatized by the Si(111) double-crystal monochromator. The beam size limited by a slit was $3^{(H)}$ x $2^{(V)}$ mm^2 at the sample position. The intensity after transmitting through the sample was measured with 300 mm ionization chamber which was filled with 75% N_2 + 25% Ar gas.

The difference in the absorption coefficients for right- and left-circularly polarized X-rays was measured with spins parallel and antiparallel to the direction of light travel. The incident beam was guided into a synthetic single crystal of (001) diamond with a thickness of 0.492 mm in order to produce circularly polarized X-rays. A standard transmission setup was used with the Faraday arrangement, where X-rays irradiates the sample through a pair of pinholes in rare-earth magnets in a magnetic field of 0.4 T.

Results and discussion

In Fig. 1, a negative XMCD peak was clearly observed for pure $LaCoO_3$ at E = 7.719 keV within the threshold region of the main edge, suggesting the existence of the intermediate-spin state of Co^{3+}. A positive XMCD peak appeared at E = 7.723 keV by Sr substitution of La in $LaCoO_3$ ($x \geq 0.2$) in accordance with the low-spin state of Co^{4+} and X-ray absorption near-edge-structure (XANES) spectra. A negative XMCD peak was also observed at the pre-edge, which can be explained as the hybridization with the neighboring Co^{4+} in the dipole transition. The dispersion-type XMCD signals at the main edge may be rationalized with the hypothetical double-exchange interaction between Co^{3+} and Co^{4+}.

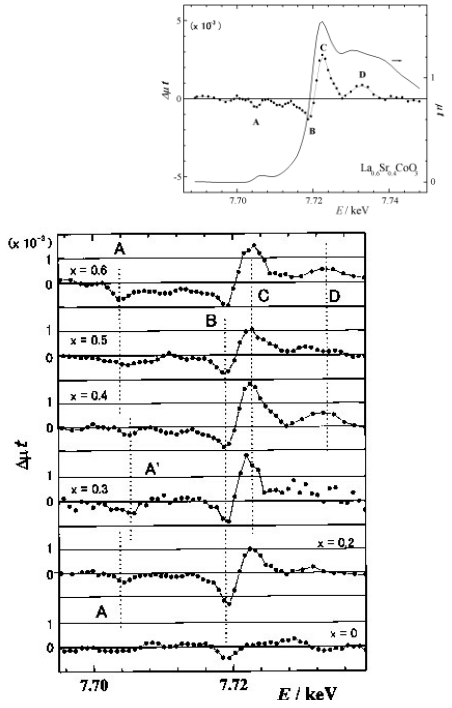

Fig. 1: Compositional change of the XMCD spectra of $La_{1-x}Sr_xCoO_3$ ($0 \leq x \leq 0.6$) at the Co *K* absorption edge.

References

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