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Low-temperature magnetic transition of $LaCoO_3$ studied by Co *L* x-ray emission spectroscopy

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Introduction

$LaCoO_3$ exhibits a gradual non- to para-magnetic transition around 100 K. The ground state of Co is a low-spin (LS) state with S=0. It is now considered that an intermediate-spin (IS) state with S=1 is the first excited state and populated thermally through the transition [1]. By assuming these spin states, the magnetic susceptibility is explained qualitatively but not quantitatively [2]. The nature of the transition has yet to be understood fully.

We studied the Co 3*d* state of $LaCoO_3$ associated with the magnetic transition around 100 K by using Co 3*d*-2*p* x-ray emission spectroscopy (XES) in the Co 2*p* x-ray absorption (XA) region. Resonant XES (RXES) gives information on the change in the Co 3*d* electronic configuration through the polarization dependence of the spectra [3]. For example, if the initial state is totally symmetric, the elastic x-ray scattering is forbidden in a depolarized experimental configuration where the polarization vector of the incident photon is parallel to the scattering plane.

Results and discussion

The sample used was a $LaCoO_3$ single crystal whose surface was cleaned *in situ* by scraping. Figures 1(a) and 1(b) show the Co L_3, 3*d*-2$p_{3/2}$, XES spectra of $LaCoO_3$ measured in the depolarized configuration at 40 and 300 K, respectively, for the incident photon energy of 779.5 eV which corresponds to the Co 2$p_{3/2}$ XA peak position. Figure 1(c) shows the Co L_3 XES spectrum of $LaCoO_3$ for the incident photon energy of 820 eV well above the Co 2*p* XA threshold.

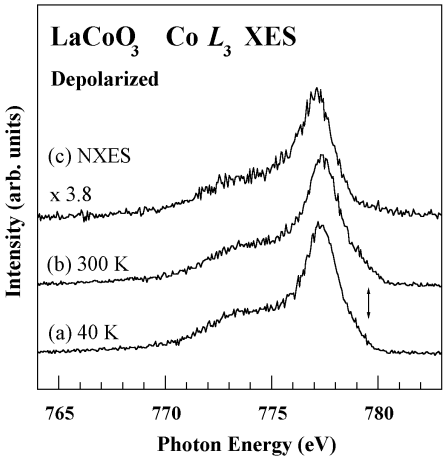

Fig.1. Co 3*d*-2$p_{3/2}$ (L_3) x-ray emission spectra of $LaCoO_3$ taken in the depolarized configuration. The incident photon energies were (a) and (b) 799.5 eV, where an arrow is shown, and (c) 820 eV. The sample temperatures were (a) and (c) 40 K and (b) 300 K.

The spectra in Figs. 1(a) and 1(b) resemble that in Fig. 1(c): the main peak is located around 777 eV and has a shoulder lower than the peak by about 4.5 eV in the three spectra. These structures have been also observed to be dominant in the other L_3 spectra for the measured excitation energies from 778 eV to 820 eV. This indicates that the Co 3*d* electron excited from the 2*p* core-level is delocalized in $LaCoO_3$, in accord with previous studies [1,4].

We notice, however, the change between the L_3 spectra in Figs. 1(a) and 1(b) around 779.5 eV just the same as the incident photon energy: the intensity increases with temperature. The intensity increase is mainly due to the elastic x-ray scattering. By assuming a non-distorted CoO_6 octahedron, the Co 3*d* electrons in a t_{2g}^6 configuration, that is, those in the LS state constitute a wave function being a basis for $^1A_1$. The elastic scattering for the system is forbidden in the depolarized configuration. On the other hand the IS state has a $t_{2g}^5 e_g^1$ configuration, and the initial state is no more represented by $^1A_1$. According to an analysis of the magnetic susceptibility of $LaCoO_3$ [1], almost all Co remains in the LS state at 40 K whereas over two thirds of Co is excited to the IS state at 300 K.

It is evident in the present study that the population of Co in the LS state decreases with temperature. We could not, however, confirm that the IS state is associated with the magnetic transition of $LaCoO_3$ around 100 K. A high-spin state in a $t_{2g}^4 e_g^2$ configuration, which was previously considered to be responsible for the transition, also allows the elastic x-ray scattering in the depolarized configuration. In order to identify the spin state involved in the transition of $LaCoO_3$ by using RXES, we have to investigate more detailed dependence of the spectra on incident photon energy and polarization along with numerical simulations of the spectra.

References

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