Structural analysis and spin state transition in cobalt oxide $Sr_{0.75}Y_{0.25}CoO_{3-\delta}$

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Cobalt oxides with perovskite structure exhibit variety of physical phenomena caused by the various orderings of the charge, spin, and orbital. It is well known that Co3+ ions have a spin degree of freedom such as low-spin (LS) (t2g6, S = 0), high-spin (HS) (t2g4eg2, S = 2) and intermediate-spin (IS) (t2g5eg1, S = 1) state.

The physical properties of perovskite cobalt oxides Sr1-xRexCoO3- δ (Re : Y, Er, Ho, etc.) has been investigated, and it has been found that Sr0.75Y0.25CoO3- δ (SYCO) has the highest ferromagnetic transition temperature (Tc ~ 340 K) among cobalt perovskite oxides [1]. The magnetic susceptibility of SYCO follows a Curie-Weiss law with the negative Curie-Weiss temperature above Tc. The crystal structure in the highest temperature phase is tetragonal I4/mmm with a cell of 2ap×2ap×4ap, where ap is the cell length of the primitive perovskite unit cell. The CoO6 octahedral layers and the CoO4.25 layers are alternately stacked along the c-axis. Recently, resonant X-ray scattering study revealed that the origin of ferromagnetic phase transition at Tc is caused by the eg orbital ordering of IS sate of Co3+ ions [2]. Furthermore, the sudden decrease of the magnetic susceptibility with the hysteresis was observed below 250 K in the temperature dependences of magnetization (M-T curve) [3]. To clarify the origin of the unique magnetic properties, it is important to reveal the details of crystal structure below Tc. In this study, we have investigated the crystal structure and magnetic property of SYCO in a wide temperature range, by synchrotron powder diffraction (SPD) and magnetization measurements. SPD experiments were carried out using one-dimensional Si microstrip detector system in the temperature range of 100K – 700K at the BL02B2 beamline of SPring-8, Japan.

In the SPD patterns, the multistep structural phase transitions were observed in wide temperature range. Especially, at Tc, we observed the peak splittings of fundamental Bragg reflections and that the superlattice reflection intensities changed from those above Tc. Furthermore, we observed the difference of SPD patterns in between cooling and warming processes at 220 \sim 280 K. This temperature range corresponds to the sudden decrease of the magnetic susceptibility with the hysteresis in the M-T curve. From these results, it was suggested that the magnetic property of SYCO below Tc is due to the spin state transition of Co3+ accompanied by the structural phase transition. In this presentation, the details of structural analysis and spin state of Co3+ are also discussed.

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