

Magnetic origin of magneto-dielectric Ba(Co_{0.85}Mn_{0.15})O_{3-δ}

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1 Introduction

We have ever investigated the structural, magnetic, and dielectric properties of Mn doped BaCoO_{3-δ}. In particular, Ba(Co_{0.85}Mn_{0.15})O_{3-δ} (BCMO) sintered ceramics was found to exhibit unique physical properties. The BCMO ceramics are ferromagnetic below about 35 K and paramagnetic above that temperature. Moreover, the BCMO ceramics show abrupt increase in resistivity below 35 K. Thus, the BCMO ceramics behave as a ferromagnetic insulator at lower temperature below 35 K, and as a paramagnetic semiconductor above that temperature. However, the details of the magnetic transition occurred at that temperature range has been still uncertain. In the present studies, we have tried to evaluate the magnetic origin of the BCMO ceramic samples using the magnetic Compton scattering (MCP) as well as the X-ray photoelectron spectroscopy (XPS).

2 Experiment

BCMO was prepared through a conventional ceramic process from the desired amount of BaCO₃, Co₃O₄ and Mn₃O₄ powders. We have carried out the magnetization measurements using a SQUID magnetometer. The valence state of 3d transition metal ion was measured by soft X-ray photoelectron spectroscopy (XPS) at KEK-BL27A. MCP measurements were also performed at various temperatures.

3 Results and Discussion

The BCMO samples are found to be mainly consisted of the mixture of the phases with the 10H and 12H hexagonal crystal structures, which is consistent with our previous works. [1] The temperature dependence of the magnetic moment of the samples applying magnetic field of 50 Oe is also shown in Fig. 1. As can be seen in the figure, some kind of magnetic transition can be observed around 30 K. According to the inverse susceptibility ($1/\chi$) versus temperature plot that is shown in Fig. 1(b), the plots form a straight line above 50K suggesting that the Curie-Weiss law dominates. The values of the remanent magnetization at 8K are 0.42 μ_B /f.u.. Consequently, some kind of ferromagnetic-paramagnetic transition occurred at around 30–50K. We also accurately determine the magnetic transition temperature to be 32K by Arrott plot of the magnetization data below 50K.

To investigate the ferromagnetic origin of the BCMO samples, we performed the XPS analysis using synchrotron soft-X-ray radiation of $h\nu=3100$ eV to investigate the valence state of the B site transition metal ions. Figure 2 shows the Mn2p core level spectra of the

samples. Comparing the observed value of the peak shift with the reliable database, the Mn ions are speculated to exist as tetravalent state in the present samples. Since MCP measurements revealed that the phase transition at simply caused by thermal spin fluctuation, the origin of the magnetic ordering of BCMO is expected to be super-exchange coupling of Co⁴⁺(d⁵)-O²⁻-Mn⁴⁺(d³) with bonding angle of 180° and/or Mn⁴⁺(d³)-O²⁻-Mn⁴⁺(d³) with bonding angle of 90° [2]. This is because the Kanamori-Goodenough rule for super-exchange coupling, implies that the exchange interaction for these combinations can be ferromagnetic.

References

- [1] T. Inoue *et al.*, *IEEE Trans. Magn.* **41**, 3496 (2005).
[2] R. Shinoda *et al.*, *J. Appl. Phys.* **113**, 17E307 (2013).

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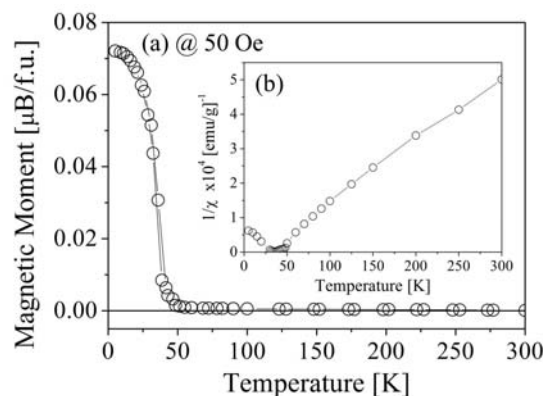


Fig. 1 (a) Temperature dependence of the magnetization at 50 Oe. (b) $1/\chi$ versus T plots

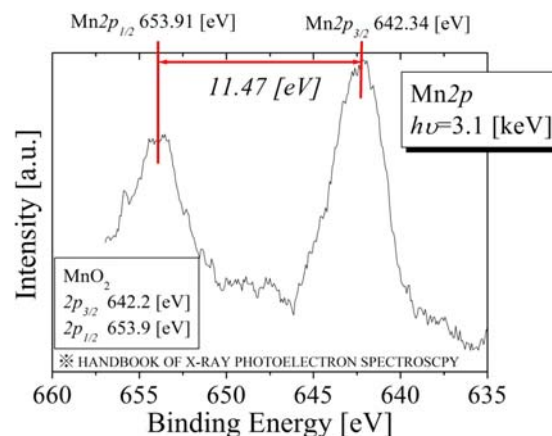


Fig. 2 Mn2p core level XPS spectra using synchrotron soft-X-ray radiation of $h\nu=3100$ eV.