Materials Science

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Effect of oxygen partial pressure on the magnetic properties of $Ba(Fe_{0.5}Mn_{0.5})O_{3-\delta}$ epitaxial thin films

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Introduction

Magneto-dielectric materials have attracted considerable attention due to a possible application for novel magneto-electric devices. In the course of our intensive studies on Ba-based perovskite magneto-dielectric single-crystalline films, we view $Ba(Fe_{0.5}Mn_{0.5})O_{3-\delta}$ (BFMO) as one of the potential candidates of the room temperature ferromagnetic insulator. As is well known, however, the presence of oxygen deficiencies in the films has significant influences on the magnetic and dielectric properties. In the present studies, we investigated the effect of oxygen deficiencies on the structural and magnetic properties of the films. The correlation between the magnetic properties and the valence state of the Fe and Mn ions will be communicated.

Experimental Procedure

BFMO epitaxial films were synthesized in vacuum by pulsed laser deposition on (001) $SrTiO_3$ substrates at 873 K. To control the amount of the oxygen deficiencies, O_2 gas was introduced in the deposition chamber during a post-process of substrate cooling in the pressure of 0, 5 and 10 mTorr. Each sample was labeled samples-0, -5 and -10, respectively. Structural characterizations were carried out by XRD and TEM. Magnetic properties were measured by a SQUID magnetometer. The valence state of the Fe and Mn ions was evaluated by XPS analysis by using the soft X-ray radiation at KEK-PF.

Results and Discussions

The (001) epitaxial grown BFMO thin films on STO(001) substrate had a pseudo tetragonal perovskite crystal structure. It was found that the lattice constants significantly decreased with increasing oxygen pressure during the post process. This may suggest that the radius of the Fe and Mn ions that is strongly correlated with the valence state of the ions, systematically changed.

The magnetization loops measured at 300 K are shown in Fig. 1. As is obvious in the figure, all the samples exhibited a ferromagnetic nature even at 300 K and the saturation magnetization increased with increase in the oxygen partial pressure. The maximum saturation magnetization of 43 emu/cc could be obtained for the sample-10, which is attributable to 0.315 μB/f.u.

Figures 2 show the $2p_{1/2}$ and $2p_{3/2}$ core level XPS spectra for the Mn and Fe ions of the samples-0 and -10. As is clearly indicated in Fig. 2(a), the peak of the Mn spectra with a higher binding energy significantly stands out with

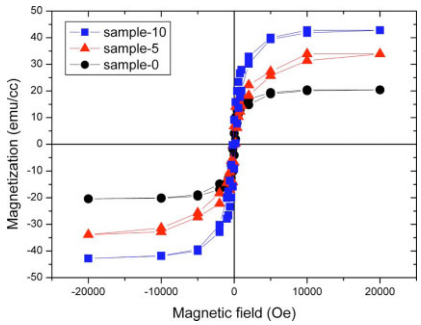

Fig. 1 Magnetization curves measured at 300K for the sample-0, sample-5 and sample-10.

increasing oxygen partial pressure (see the results of a peak separation). This simply means that the valence state of the Mn ions in the majority part of the films changed from Mn^{3+} to Mn^{4+} with increase in the oxygen partial pressure. On the other hand, the peak of the Fe spectra (Fig. 2(b)) shows an opposite tendency to the Mn case. That is, the peak shifted toward the lower binding energy side with decrease in the amount of the oxygen deficiency, which suggested the value of the Fe ions tends to change from Fe^{4+} to Fe^{3+} with decreasing amount of the oxygen deficiency. [1]

These results suggested that the amount of the $Fe^{3+}(3d_5)$ and $Mn^{4+}(3d_3)$ portion increased with increasing oxygen partial pressure. Hence, the increased amount of the $Fe^{3+}(3d_5)$–O^{2-}–$Mn^{4+}(3d_3)$ superexchange coupling with 180° bonding angle may be the main reason for the enhancement in the ferromagnetic spin alignment in the BFMO sample-10.

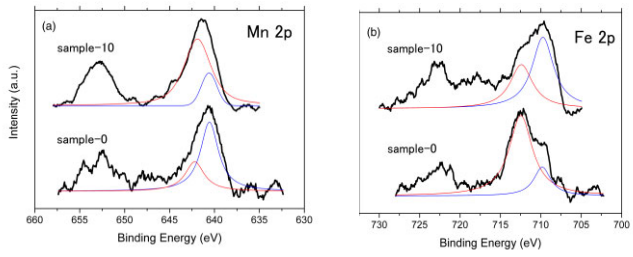

Fig. 2 The $2p_{3/2}$ *and* $2p_{1/2}$ core level XPS spectra for the Mn (a) and Fe (b) ions, measure by using soft x-ray radiation with $h\nu$=3100 eV for the sample-0 and -10.

References

[1] H. Falcón et al., Appl. Catal. B, 53 37 (2004).

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