Magnetic properties of (Bi,*L*)(Fe$_{1-x}$Co$_x$)O$_3$ (*L* = La, Nd, Sm, Gd, Dy) multiferroic thin films and consideration on the origin of their magnetic properties

Akita Univ., ◦Soumyaranjan Ratha, Kotaro Takeda, Daichi Yamamoto, Munusamy Kuppan, Genta Egawa, Satoru Yoshimura

E-mail: syoshi@gipc.akita-u.ac.jp

Introduction : The applications of multiferroic thin films are effective in magnetic devices to decrease the power consumption because it is possible to realise the magnetization switching by electric field. Bismuth Ferrite (BFO) is a better candidate for this application as it shows multiferroic properties at room temperature. The antiferromagnetic property of pure BFO needs to be changed to ferrimagnetic property for application point of view by substituting some atoms with it. In (Bi,Ba)FeO$_3$, which we substituted Ba in A-site of BFO with an appropriate percentage, shows relatively high saturation magnetization (M_s) (90 emu/cm^3) [1], but the ratio of perpendicular coercivity against in-plane coercivity ($H_{c\perp}/H_{c//}$) (0.8) & magnetic Kerr rotation angle (Θ_k) (0.03º) were not enough for application. In our previous study, we found remarkable improvement in magnetic properties like M_s of 80 emu/cm^3, relatively high $H_{c\perp}/H_{c//}$ of 1.6 [2] and very high Θ_k of 0.67º in (Bi,La)(Fe,Co)O$_3$ by replacing La as A-site substituent. In this study, we are going to investigate the improvement of magnetic properties and their origin of BFO based thin films by substituting other Lanthanoids in A-site and Co in B-site in (Bi$_{0.6\text{-}0.4}L_{0.4\text{-}0.6}$)(Fe$_{0.7}Co_{0.3}$)O$_3$ (*L*=La, Nd, Sm, Gd, Dy).

Experimental : Multilayers of Ta (5 nm) / Pt (100 nm) / (Bi$_{0.6\text{-}0.4}$La$_{0.4\text{-}0.6}$)(Fe$_{1-x}$Co$_x$)O$_3$ (x = 0-0.3) or (Bi$_{0.6\text{-}0.4}L_{0.4\text{-}0.6}$)(Fe$_{0.7}Co_{0.3}$)O$_3$ (*L*=La, Nd, Sm, Gd, Dy) (200 nm) were deposited onto a thermally oxidized Si wafer using a UHV sputtering system. After dry-etching of substrate surface, the Ta seedlayer, Pt underlayer and (Bi,*L*)(Fe,Co)O$_3$ layer were deposited at room temperature, 400 °C, and 695 °C respectively. The film thickness and deposition temperature of the Ta seedlayer and Pt underlayer were optimized to obtain a strong (111) orientation of the Pt underlayer. The VHF (40.68 MHz) plasma irradiation during the RF sputtering deposition of Pt film and reactive pulsed DC sputtering deposition of (Bi,*L*)(Fe,Co)O$_3$ films were performed with an electric power of 5 W to obtain the crystal grain growth of Pt film and (Bi,*L*)(Fe,Co)O$_3$ films. The frequency pulsed DC was 100 kHz with the duty ratios between sputtering ON and OFF is 2–3 (the time of sputtering ON is 6 μs and that of OFF is 4 μs). The concentrations of substituent were confirmed by dispersive X-ray spectroscopy. The hysteresis curve (*M-H*) and Θ_k were measured by vibrating sample magnetometer and MOKE measurement equipment respectively.

Results : Table 1 shows the magnetic properties of (Bi$_{0.6\text{-}0.4}$La$_{0.4\text{-}0.6}$)(Fe$_{1-x}$Co$_x$)O$_3$ (x = 0-0.3) or (Bi$_{0.6\text{-}0.4}L_{0.4\text{-}0.6}$)(Fe$_{0.7}Co_{0.3}$)O$_3$ (*L*=La, Nd, Sm, Gd, Dy) thin films. The magnetic properties of (Bi,Ba)FeO$_3$ are also shown as a reference. The Co substitution in B-site has a great role in improving every magnetic property by increasing the concentration up to 30%. Therefore, Co substitution in B-site is key factor to obtain good magnetic properties. And also enhanced M_s and $H_{c\perp}/H_{c//}$ are observed by substituting Neodymium instead of Lanthanum in A-site at a percentage around 40-60%. Therefore, suitable Lanthanoid substitution in A-site is also key factor to obtain good magnetic properties. The magnetic properties like M_s, $H_{c\perp}/H_{c//}$ and Θ_k are increased drastically compared with the previously observed value in (Bi,Ba)FeO$_3$. The highest M_s of 140 emu/cm^3 and large perpendicular magnetic anisotropy are found in (Bi,Nd)(Fe,Co)O$_3$, which is suitable for magnetic recording devices, and maximum Θ_k of 0.67º is found in (Bi,La)(Fe,Co)O$_3$, which is suitable for optical devices.

Table 1 Magnetic properties (M_s, $H_{c\perp}$, $H_{c\perp}/H_{c//}$, Θ_k) of all substituted thin films.

Substitution element	Ba	La					Nd	Sm	Gd	Dy
$Co^{2+}/(Fe^{3+}+Co^{2+})$	0	0	0.05	0.12	0.16	0.28	0.25	0.25	0.24	0.27
M_s (emu/cm^3)	90	15	23	48	69	80	140	110	65	85
$H_{c\perp}$ (kOe)	2.0	0	0.4	3.1	3.1	2.6	2.1	1.9	2.2	1.9
$H_{c\perp}/H_{c//}$	0.8	0	0.2	0.9	1.2	1.6	2.6	2.1	28	1.7
Θ_k (°@750nm)	0.03	0		0.08	0.12	0.67	0.34	0.21	0.19	0.23

Reference

1) S. Yoshimura et al., *JJAP-STAP*, **57**, 0902B7 (2018).
2) M. Kuppan et al., *SCIENTIFIC REPORTS*, **11**, 11118 (2021).