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Magnetic and Optical Studies of Structure of Co-Doped Bismuth-Iron-Garnet

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Abstract Using an alternating reactive ion beam sputtering technique, Co ion was doped in epitaxially growing $\text{Bi}_3\text{Fe}_5\text{O}_{12}$ garnet film. The intense enhancement of magnetization, coercivity, and optical absorption at 633nm was observed. The Faraday hysteresis loop for slightly Co-doped film showed anomalous behavior. Those phenomena can be understood considering the composite film structure in which fine particles of $\text{Co}_{3-x}\text{Fe}_x\text{O}_4$ spinel are embedded in epitaxially grown $\text{Bi}_3\text{Fe}_5\text{O}_{12}$.

1. INTRODUCTION

$\text{Bi}_3\text{Fe}_5\text{O}_{12}$ (BIG) is a magnetically soft ferrimagnet with garnet structure. For near infrared light, BIG is transparent and shows giant Faraday rotation (θ_F) [1,2]. It is well known that, in magnetic oxides, Co^{2+} ion occupies octahedral (O) site contributing large magnetocrystalline anisotropy which results in high coercivity and that tetrahedral (T) site Co^{2+} strongly induces optical absorption and Faraday rotation due to crystal field transition (${}^4A_2 \rightarrow {}^4T_1$) at around the wavelengths of 0.7 and 1.5 μm . These modification effects due to Co^{2+} ion are also expected in BIG, which are worth investigating from both viewpoints of fundamental research and application of BIG. On the Co-doped BIG films prepared by reactive alternating ion beam sputtering, we have reported the enhancement of magnetization and coercivity and the different values of coercivity derived from the θ_F hysteresis loops measured at different wavelengths, 633nm and 1.5 μm [3]. The results of further investigation will be described.

2. EXPERIMENTAL

The films were deposited onto the (111) oriented $\text{Gd}_3(\text{ScGa})_5\text{O}_{12}$ garnet substrates kept at 500°C in an oxidizing atmosphere by alternately sputtering the two ceramic targets $T_A (= \text{BIG} = 3\text{Bi}_2\text{O}_3 \cdot 5\text{Fe}_2\text{O}_3)$ and $T_B (= \text{CoO})$ using Ar ion beam [3]. The sputtering time fraction $R_{\text{ST}} = t_A / (t_A + t_B)$ was varied from 0 to 1. t_A and t_B are sputtering time in one period for respective targets. t_A was varied from 0 to 9min and t_B fixed at 1min. The total sputtering time of each run was set at 600 min. The films were characterized at room temperature using X-ray diffractometer (XRD), AFM (atomic force microscope), vibrating sample magnetometer (VSM), infrared and visible spectrometer, and Faraday rotation spectrometer.

3. RESULTS AND DISCUSSION

In the range of $0 < R_{\text{ST}} < 1$, no indication of periodic layered structure was found in XRD peaks measured at low and high diffraction angle. BIG/CoO layered structure will be hardly formed because of high substrate temperature. In the range of $0.833 < R_{\text{ST}} < 1$, main phase is epitaxially grown garnet phase (G). At $R_{\text{ST}} = 0.833$, (111) oriented spinel phase (S) appears infrequently. In the range of $0 \leq R_{\text{ST}} \leq 0.75$, G-phase, BiFeO_3 , and $\text{Bi}_2\text{Fe}_4\text{O}_9$ do not appear, but S-phase and some unidentified phases are frequently observed. From these facts, we can deduce that, in the range $0 \leq R_{\text{ST}} \leq 0.8$, S-phase is the main and G-phase does not grow if $R_{\text{ST}} \leq 0.75$.

In the range of $0.5 \leq R_{\text{ST}} \leq 1$, the specimens are ferromagnetic at room temperature. As shown in Fig. 1, R_{ST} dependence of the

Fig.1 Room temperature saturation magnetization of Co-doped BIG films vs. R_{ST} .

Fig.2 Optical absorption coefficient of Co-doped BIG films at 633nm vs. R_{ST}

Fig.3 Faraday and magnetization hysteresis loops of the sample of $R_{ST}=0.9$. θ_F was measured at 633nm.

saturation magnetization M_s measured at room temperature has a prominent peak at $R_{ST} \approx 0.75$ and the peak value of 300 emu/cc is about twice of that of BIG. Existence of G - or S -phases was not confirmed by means of XRD. The coercivity H_C increases monotonically with decrease of R_{ST} . At $R_{ST}=0.9$ and 0.5 , H_C are 1 and 1.8 kOe, respectively. R_{ST} dependence of M_s can not be understood by the substitution of Co^{2+} for Fe^{3+} in G -phase. It will be natural to consider the formation of ferromagnetic phase with high M_s other than G -phase. The most plausible phase is S_{CF} -phase, $Co_xFe_xO_4$ whose M_s increases monotonically as x which may be proportional to R_{ST} increases. The room temperature M_s of $CoFe_2O_4$ is 320 emu/cc. The volume fraction of S_{CF} -phase in a film and Co-content in S_{CF} -phase decrease as R_{ST} increases, which may explain R_{ST} dependence of M_s .

The crystal structure of either G - and S -phases contains T - and O -sites. The crystal field transition of Co^{2+} at the T -site, which is allowed one, results in optical absorptions at wavelengths of 0.6, 1.4 and 2.5 μm . The absorption measured at 633nm shown in Fig.2 decreases monotonically and intensely as R_{ST} increases, which suggests that this absorption is caused by Co^{2+} on the T -site in S_{CF} -phase, since Co^{2+} is hardly introduced into G -structure. In S -structure, Co^{2+} substitutes preferentially at O -sites, however, small amount of Co^{2+} at T -sites in S_{CF} -phase may cause strong absorption because the transition is allowed.

In a film containing small amount of Co^{2+} , a peculiar behavior of θ_F hysteresis loop measured at 633nm was observed. As shown in Fig.3, the sign of θ_F changes before the applied field decreased from saturation to 0 Oe. The hysteresis loop measured in the same field strength range using VSM shows step-like change. These can be understood by exchange coupling effect between magnetically soft BIG of giant θ_F and magnetically hard S_{CF} -phase.

4. CONCLUSION

We can deduce that the films of $R_{ST} \approx 0.9$ are composites in which fine particles of S_{CF} -phase which are hardly detected by XRD because of the small size and volume fraction are embedded in epitaxially grown BIG films.

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