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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 sputtering technique, Co ion was doped in epitaxially growing Bi₃Fe₅O₁₂ garnet film. The intense enhancement of magnetization, coercivity, and optical absorption at 633 nm 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 Co₀.₅Fe₀.₅ spinel are embedded in epitaxially grown Bi₃Fe₅O₁₂.

1. INTRODUCTION

BiₓFeₓO₁₂ (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, Co²⁺ ion occupies octahedral (O) site contributing large magnetocrystalline anisotropy which results in high coercivity and that tetrahedral (T) site Co²⁺ strongly induces optical absorption and Faraday rotation due to crystal field transition (A_g → T_g) at around the wavelengths of 0.7 and 1.5 µm. These modification effects due to Co²⁺ 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, 633 nm and 1.5 µm[3]. The results of further investigation will be described.

2.EXPERIMENTAL

The films were deposited onto the (111) oriented Gdₓ(ScGa)₂O₁₂ garnet substrates kept at 500 °C in an oxidizing atmosphere by alternatingly sputtering the two ceramic targets T_A (=BIG=3Bi₂O₅·5Fe₂O₅) and T_B (=CoO) using Ar ion beam[3]. The sputtering time fraction R_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 9 min and t_B fixed at 1 min. 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_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_ST<1, main phase is epitaxially grown garnet phase (G). At R_ST=0.833, (111) oriented spinel phase (S) appears infrequently. In the range of 0≤R_ST≤0.75, G-phase, BiFeO₃, and Bi₂Fe₃O₇ do not appear, but S-phase and some unidentified phases are frequently observed. From these facts, we can deduce that, in the range 0≤R_ST≤0.8, S-phase is the main and G-phase does not grow if R_ST≤0.75.

In the range of 0.5≤R≤1, the specimens are ferromagnetic at room temperature. As shown in Fig.1, R_ST dependence of the...
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- Applied magnetic field I to Rim plane
- Applied magnetic field I to film plane

Filled circles and squares represent the films in which garnet phase was detected.

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<th>Fig.1</th>
<th>Room temperature saturation magnetization of Co-doped BIG films vs. $R_{ST}$.</th>
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saturation magnetization $M_s$ measured at room temperature has a prominent peak at $R_{ST}=0.75$ 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$^{3+}$ 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 $S_{ETF}$-phase, Co$_x$Fe$_{2-x}$O$_4$ whose $M_s$ increases monotonically as $x$ which may be proportional to $R_{ST}$ increases. The room temperature $M_s$ of CoFe$_2$O$_4$ is 320 emu/cc. The volume fraction of $S_{ETF}$-phase in a film and Co-content in $S_{ETF}$-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 $S_{ETF}$-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 $S_{ETF}$-phase may cause strong absorption because the transition is allowed.

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

4. CONCLUSION

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

References