1-1-2012

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ABSTRACT

RF co-sputtered nanocomposite ($Bi_3Fe_5O_{12}$-$Bi_2Dy_1Fe_4Ga_1O_{12}$) thin films are prepared using composition adjustment approach, implemented by varying the RF powers at each sputtering target. A new MO nanocomposite material is obtained by means of applying optimized annealing crystallization process to these as-deposited garnet layers. The optimally annealed garnet nanocomposites exhibit a significant red shift of the main MO transitions and show strong Faraday rotation of about 8.8°/μm at 532 nm simultaneously with having a high MO quality and magnetization direction perpendicular to the film plane.

INTRODUCTION

Bi-substituted iron garnets in thin film form are the best semi-transparent magneto-optic materials for photonics applications in the visible and near-infrared spectral regions. These materials can possess attractive magnetic properties and high specific Faraday rotation, if the deposited layers contain a high volumetric fraction of the garnet phase with good surface quality and

V. Rajendran, R. Yuvakkumar, K. Thyagarajah and K.E. Geckeler (eds.)
Applications of Nano Materials: Electronics, Energy and Environment, pp. 143–148 (2012).

microstructure. Significant numbers of studies have been conducted to understand the relationship between the MO properties (Faraday rotation) and the Bi-content in each material system. A strong dependence of Faraday rotation on the level of bismuth substitution in thin film materials was seen in the garnet films prepared by liquid-phase epitaxy (LPE). The extrapolated value of the specific FR of the fully bismuth-substituted $Bi_3Fe_5O_{12}$ was evaluated (based on these investigations) to be about 6 deg/micron at the wavelength of 633 nm. Another comparative study has been reported on the epitaxial $Bi_3Fe_5O_{12}$ films prepared by pulsed laser deposition (PLD) and reactive ion beam sputtering (RIBS) techniques where the Faraday rotation measured in PLD-BIG films at 635 nm was about 7.8°/μm, and 6.9°/μm in RIBS-BIG films.[1–3] Recently, electron cyclotron resonance (ECR) sputtered $Bi_3Fe_5O_{12}$ films have been shown to demonstrate the specific Faraday rotation of about 8.4°/μm at 633 nm.[3, 4] LPE films having a record-high Bi-content demonstrated a linear growth of specific Faraday rotation with increasing bismuth content which was obtained by the approach of applying compositional changes from $Lu_3Fe_5O_{12}$ to $Bi_{2.4}Lu_{0.6}Fe_5O_{12}$.[2] In order to investigate the source of such disagreements between the measured properties of highly Bi-substituted garnets and theory predictions and also to try and increase the number of bismuth atoms per unit formula in our sputtered films, we synthesized co-sputtered nano-composites of type ($Bi_3Fe_5O_{12}$-$Bi_2Dy_1Fe_4Ga_1O_{12}$). Another reason we decided to co-sputter these two compositions from two different targets was the fact that so far we could not successfully crystallize any RF-sputtered garnet-type materials from targets containing five iron atoms per stoichiometric formula unit (without any Ga or Al dilution), despite multiple trials. Therefore this co-sputtering approach was our selected way of synthesizing a garnet composition as close as possible to $Bi_3Fe_5O_{12}$ which still contained some Ga dilution and was expected to have out-of-plane magnetization due to some Dy substitution. The flexibility of co-sputtering approach also allows many final stoichiometry variations within nanocomposites.

In this paper, we report on the successful synthesis of a new type of high performance nanocomposite MO material of potential usefulness in magnetic photonics crystals (MPC) which exhibits a significant red-shift of the main MO transitions (thus improving its Faraday rotation), the existence of which we found conclusively after a series of sensitive magnetic circular dichroism (MCD) measurements.

NANOCOMPOSITE THIN FILMS GROWTH AND CHARACTERIZATION

Garnet nanocomposite films (about 1 μm thick) were sputtered onto the paramagnetic $Gd_3Ga_5O_{12}$ (GGG) substrates from two separate oxide-base-mixed targets. The sputtering conditions and process parameters used to deposit the nano-composite layers are detailed in Table 1.

The results of sputtering provided the garnet-type layers in amorphous phase onto the substrates and these amorphous layers were later crystallized by using an oven annealing process which we optimized for this material type by many annealing trials followed by MO characterization. Some of the already-annealed films were subsequently re-annealed at a slightly higher temperature to see if that changed any of MO properties. In particular, we initially annealed some films for 1h at 520°C and then re-annealed them for an additional 1 hour at 530°C. Later multiple crystallized nanocomposite films were characterized using magnetic circular dichroism (MCD) spectroscopy. MCD measurements are remarkably sensitive and allow detection of the very small spectral changes in the locations of the MO transitions fundamentally responsible for the observed features in the FR and ellipticity spectra of garnet materials. We show that it was possible to confirm that two slightly different annealing temperatures led to forming two different garnet compositions (the one with larger Bi-substitution crystallized at a colder temperature) showing an apparent red-shift in their MCD spectra with respect to each other as well as to other known garnets.

Table 1: Sputtering Conditions and Process Parameters Used to Synthesize the Magneto-Optic ($Bi_3Fe_5O_{12}$+ $Bi_2Dy_1Fe_4Ga_1O_{12}$) Nanocomposite Layers onto GGG Substrates

Sputtering target stoichiometries	$Bi_3Fe_5O_{12}$ (materials' purity 99.99%) and $Bi_2Dy_1Fe_4Ga_1O_{12}$ (99.99%)
Target size	3´´ (diameter) with the material layer thickness of 0.125´´, bonded to a 0.125″ Cu backing plate
Background Pressure	P(base) ~ 1–2x10^{-6} Torr
Process gas and pressure	Argon, P(Ar) = 1mTorr
RF power density at targets	4.35 W/cm^2 (200 W) for $Bi_3Fe_5O_{12}$ and 1.63 W/cm^2(75 W) for $Bi_2Dy_1Fe_4Ga_1O_{12}$
Substrate-target distance	18–20 cm
Substrate temperature during deposition	250°C

RESULTS AND DISCUSSION

The co-deposited annealed films exhibited rather high specific Faraday rotation of about 8.8°/µm at 532 nm (compared to around 6.9°/µm measured in typical $Bi_2Dy_1Fe_4Ga_1O_{12}$ films which usually required annealing at temperatures greater than 650°C) simultaneously with good surface quality. The MCD measurements performed over the spectral region 300–600 nm revealed a significant red-shift of the main MO transitions in all of our co-sputtered samples, in comparison with the spectral locations of these transitions measured in LPE garnet films with much smaller Bi-substitutions. The spectral dependencies of MCD signals were obtained using a sensitive dichrometer which measured the absorption difference between the left- and right-circularly polarized (LCP and RCP) light waves propagating through samples in the presence of strong magnetic field parallel to the light beam. The MCD signals from all samples were recorded in arbitrary units and plotted as a function of wavelength as shown in Figure 1. Our samples annealed at 520°C exhibited the first MCD peak position at 515 nm, and the samples annealed at 530°C displayed a two times broader MCD peak at 495 nm with a small shoulder at 515 nm, as distinct from the first MCD peak position at 442 nm observed in LPE films of composition $Bi_{0.45}Tm_{2.55}Fe_{3.8}Ga_{1.2}O_{12}$.[2] The long-wavelength negative peak of the FR moved from its position at 475 nm observed in $Bi_{0.45}Tm_{2.55}Fe_{3.8}Ga_{1.2}O_{12}$ up to 535 nm and even to 545 nm for nano-composite $Bi_3Fe_5O_{12}$- $Bi_2Dy_1Fe_4Ga_1O_{12}$ films annealed at 530°C and 520°C, respectively.

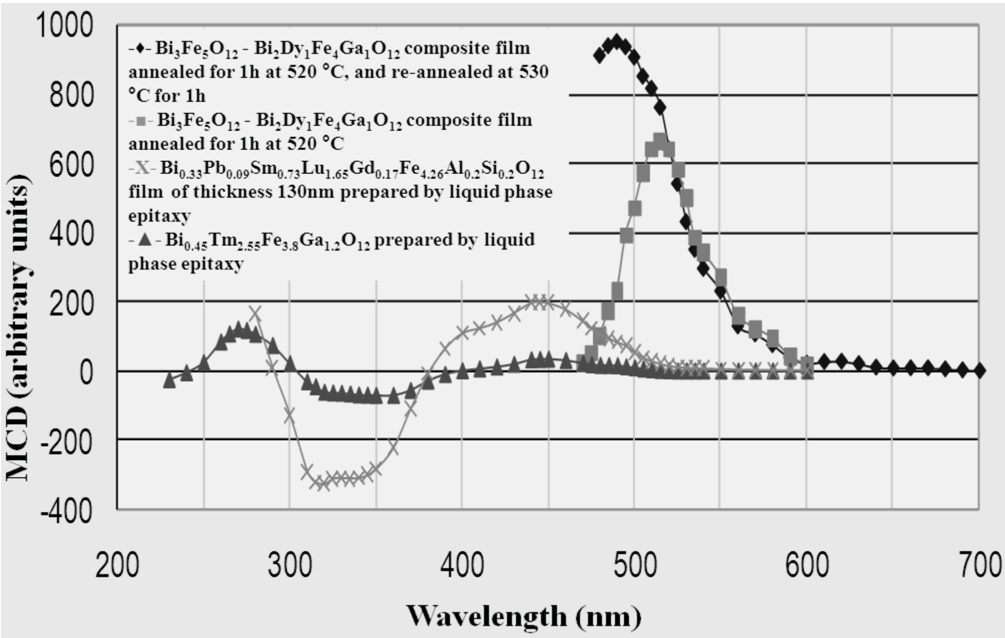

Fig. 1: Spectral dependencies of MCD signal (in arbitrary units) measured in different garnet samples. (-♦-) RF co-sputtered ($Bi_3Fe_5O_{12}$-$Bi_2Dy_1Fe_4Ga_1O_{12}$) composite film of thickness about 1 μm, annealed for 1 hour at 520°C at first stage and re-annealed for 1 hour at 530°C; (-■-) co-sputtered composite film of the same stoichiometry when still in its amorphous phase and the same thickness, but only annealed for 1 hour at 520°C; (-X-) LPE film - $Bi_{0.33}Pb_{0.09}Sm_{0.73}Lu_{1.65}Gd_{0.17}Fe_{4.26}Al_{0.2}Si_{0.2}O_{12}$ of thickness 130 nm, and (-▲-) $Bi_{0.45}Tm_{2.55}Fe_{3.8}Ga_{1.2}O_{12}$ film prepared by LPE. Sensitivity scale of the dichrometer was same during all measurements.

These results indicate that a slight difference in the annealing temperature led to a measureable difference in the material phase content and that two different garnet materials with non-identical Bi-substitutions may have formed at each of these temperatures since their MCD signatures were red-shifted with respect to one another. As is relatively well known (and confirmed by our experience), there exists a minimum annealing temperature suitable for effectively crystallizing each garnet type, with less Bi-rich compositions requiring higher temperatures. It was also expected that less Bi-rich (but still highly substituted) garnets would exhibit smaller red-shifts in their MCD signatures with respect to the reference MCD peaks measured in LPE garnets having very limited Bi-substitutions. The ways of synthesizing garnets with very high Bi-substitutions using RF-sputtering and co-sputtering are still a subject of ongoing research within our group.

CONCLUSION

RF magnetron co-sputtered ($Bi_3Fe_5O_{12}$-$Bi_2Dy_1Fe_4Ga_1O_{12}$) thin films resulted in obtaining new and interesting MO nanocomposite materials after annealing near 520°C, which exhibited simultaneously a high Faraday rotation, good MO quality, magnetization direction perpendicular to the film plane, and notable red-shift features in their MCD spectra. The results indicate that we synthesized a new garnet material type with rather high Bi-substitution well in excess of two formula units, which will be useful for applications in magnetic photonic crystals (MPC).

ACKNOWLEDGEMENT

We acknowledge the support of Edith Cowan University and the World-Class University Program funded by the Ministry of Education, Science, and Technology through the National Research Foundation of Korea (R31–10026).

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