1-1-2012

New Class of Garnet Nanocomposites for Use in Magnetic Photonic Crystals Prepared by RF Magnetron Co-sputtering

Mohammad Nur E Alam

Mikhail Vasiliev
Edith Cowan University

Kamal Alameh
Edith Cowan University

10.1109/HONET.2012.6421460

This is an Author's Accepted Manuscript of: Nur E Alam, M. , Vasiliev, M. , & Alameh, K. (2012). New Class of Garnet Nanocomposites for Use in Magnetic Photonic Crystals Prepared by RF Magnetron Co-sputtering. Proceedings of IEEE - International Symposium on High Capacity Optical Networks & Enabling Technologies (HONET). (pp. 184-187). Istanbul, Turkey. IEEE. Available here

New Class of Garnet Nanocomposites for Use in Magnetic Photonic Crystals Prepared by RF Magnetron Co-sputtering

Mohammad Nur-E-Alam[1], Mikhail Vasiliev[1], and Kamal Alameh[1,2]

[1]Electron Science Research Institute, Edith Cowan University, 270 Joondalup Dr, Joondalup, WA 6027, Australia
[2]Department of Nanobio Materials and Electronics, Gwangju Institute of Science and Technology, World Class University (WCU), Gwangju Institute of Science & Technology (GIST), Gwangju 500-712, South Korea
Email: m.nur-e-alam@ecu.edu.au

***Abstract*—A new class of magneto-optic garnet nanocomposite materials is prepared using two Bi-substituted iron garnet materials of composition types $(Bi,Dy)_3(Fe,Ga)_5O_{12}$ and $Bi_3Fe_5O_{12}$. A composition adjustment approach is applied by varying the radio frequency (RF) powers driving each sputtering target during the deposition process. This new class of nanocomposite materials exhibits simultaneously high specific Faraday rotation, MO figure of merit, and effective uniaxial magnetic anisotropy after being crystallized through optimized annealing processes. We demonstrate experimentally that the excellent combination of materials' properties obtained in this garnet nanocomposite is particularly advantageous for developing magneto-photonic crystals as well as optical sensors and isolators.**

***Index Terms*—Magneto-optic materials, garnet, specific Faraday rotation, MO figure of merit, red-shift, magnetic anisotropy, magneto-photonic crystals.**

I. Introduction

Nowadays, nanocomposite-type materials are becoming more attractive for use in nanophotonics, integrated optics and broadband communication systems. The main challenge in the field of magneto-optic (MO) materials is the development of next-generation nanocomposite-type material systems which can possess simultaneously a high MO figure of merit and high effective uniaxial ($K_u - 2\pi M_s^2 > 0$) magnetic anisotropy (sufficient for orienting the magnetization of thin films along the normal to film's plane). Bi-substituted iron garnets of different compositions are still the main class of materials used in applied magneto-optics, since these possess record optical and MO properties in the infrared and visible spectral regions. Bi-substituted ferrimagnetic iron garnets in thin film form (obtained using RF magnetron sputtering or Pulsed Laser Deposition) containing a high volumetric fraction of the garnet phase with good surface quality and microstructure can possess attractive magnetic properties and high specific Faraday rotation. The MO properties (Faraday rotation) of these classes of garnets are strongly dependent on the level of bismuth substitution in thin film materials found in the garnet films, which are often prepared by liquid-phase epitaxy (LPE) [1-2].

LPE films having a record-high Bi-content demonstrated a linear growth of specific Faraday rotation with increasing bismuth content which has been obtained by applying compositional changes from $Lu_3Fe_5O_{12}$ to $Bi_{2.4}Lu_{0.6}Fe_5O_{12}$ [1]. We have also synthesized co-sputtered nano-composites of type ($Bi_3Fe_5O_{12}$-$Bi_2Dy_1Fe_4Ga_1O_{12}$) to investigate the sources of any possible disagreements between the measured properties of highly Bi-substituted garnets and the relevant theory predictions. This co-sputtering approach was mainly an attempt to increase the number of bismuth atoms per unit formula in sputtered garnet films. Another indirect reason to co-sputter these two compositions from two different targets has been related to the fact that so far we have not been able to 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 has been one of our selected ways 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 an out-of-plane magnetization due to having some Dy substitution level. The flexibility of the co-sputtering approach has also enabled many final stoichiometry variations within nanocomposites. Another typical approach to crystallize the sputtered garnets containing five iron atoms is to prepare a garnet multilayer structure (sandwiching a $Bi_3Fe_5O_{12}$ layer in between two $Bi_2Dy_1Fe_4Ga_1O_{12}$ layers, where the layer thicknesses need to be optimized), which is being implemented by our research group and be reported elsewhere, as a future extension of this research.

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 very significant "red-shift" of the main MO transitions (thus improving its Faraday rotation in the visible range), the existence of which is conclusively confirmed, after a series of sensitive magnetic circular dichroism (MCD) measurements.

The growth and characterization processes used to synthesize high-Bi-content garnets and to investigate the properties of this new class of nanocomposite materials are detailed in section II, and the results achieved using this material system are discussed in section III. Finally, a conclusion is drawn in section IV.

II. Growth and Characterization of ($Bi_3Fe_5O_{12}$-$Bi_2Dy_1Fe_4Ga_1O_{12}$) Garnet Nanocomposites

978-1-4673-2890-6/12/$31.00

Garnet nanocomposite films (about 1 µm thick) were sputtered from two separate oxide-base-mixed targets. Table 1 summarizes the sputtering conditions and process parameters used to prepare the nano-composite layers on polished glass (Corning 1737) and Gadolinium Gallium Garnet (GGG (111)) substrates. After the sputtering process, the as-deposited garnet-type layers (amorphous in their phase) were crystallized by using an oven annealing process, which was optimized for this material type by running many annealing trials followed by MO characterization. Some of the already-annealed films were subsequently re-annealed at a slightly higher temperature to check if this reannealing process changes any of their MO properties. In particular, some films were initially annealed at 520°C for 1h and then re-annealed for an additional 1 hour at 530°C. After that, the samples, which were annealed previously at 530°C for 1hr, were annealed at two different temperatures namely, 540°C and 550°C. The crystallized (optimally annealed as judged by the best-obtained specific Faraday rotations at 532 and 635 nm) nanocomposite films were characterized using a Beckman Coulter DU 640B UV/Visible spectrophotometer, magnetic circular dichroism (MCD) spectroscopy, and a Thorlabs PAX polarimeter system used in conjunction with a custom-made calibrated electromagnet.

Table.1. Sputtering conditions and process parameters used to synthesize the magneto-optic nanocomposite layers deposited onto GGG substrates.

Sample process parameters	*Comments & values*
Garnet targets (oxide-mix-type stoichiometries) and target size	$Bi_2Dy_1Fe_4Ga_1O_{12}$ & $Bi_3Fe_5O_{12}$, 3´´ (diameter) with the material layer thickness of 0.125´´, bonded to a 0.125″ Cu backing plate
Sputter gas and pressure	Argon (Ar), P(total) = 1 mTorr
Base pressure	P(base) < 1-2E-06 Torr (high vacuum)
RF power densities 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 surface temperature during deposition	250 °C
Substrate stage rotation rate	32-40 rpm
Substrate-target distance	18-20 cm
Oven annealing regimes used (temperature and time)	510-550 °C and between 60-90 minutes with 3-5 °C/min ramp-up and ramp-down rates

III. PROPERTIES OF GARNET NANOCOMPOSITES

The physical thicknesses of all thin films were measured during the deposition processes using an in-situ laser reflectometer system which had an estimated measurement accuracy of ± 2%. Also, after the deposition, we re-confirmed the actual thicknesses of the garnet layers using specialized thickness-fitting software (in-house-developed MPC optimization software package, algorithm written by Dr. Mikhail Vasiliev) and the optical transmission spectra of both the as-deposited and post-annealed garnet layers. The absorption spectra (shown in Fig. 1) of the composite films prepared on glass substrates were derived using iterative fitting of the spectral features observed in the measured and modeled transmission spectra of the films. The effects of substrates on the optical absorption were also observed very carefully and similar trends of absorption spectra were found in all composite garnet materials deposited onto GGG substrates.

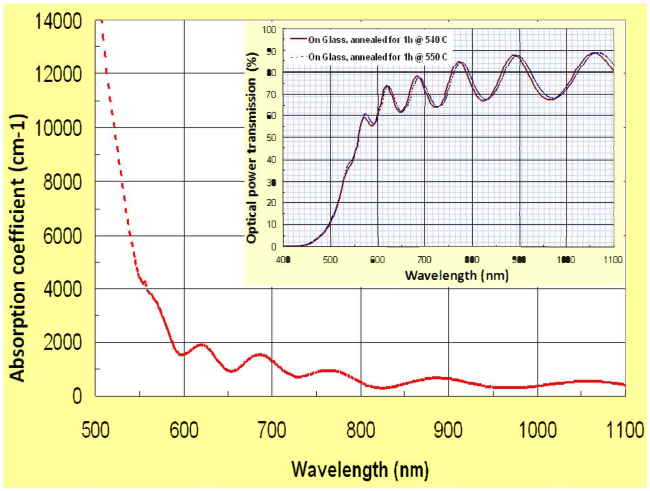

Fig.1. Measured absorption spectrum of a ($Bi_3Fe_5O_{12}$-$Bi_2Dy_1Fe_4Ga_1O_{12}$) garnet nanocomposite film prepared on a glass substrate, and the inset shows the transmission spectra of two co-sputtered films from the same deposition batch but processed using different annealing regimes.

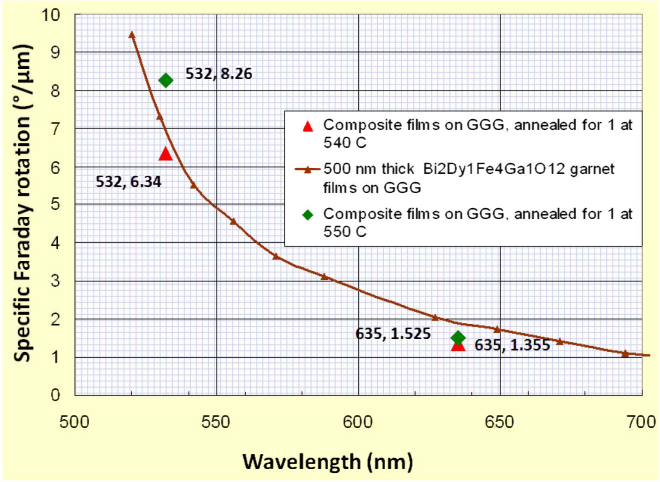

Fig.2. Specific Faraday rotation data points of garnet nanocomposite films sputtered onto a GGG substrate (measured at 532 nm and 635 nm); the obtained data are presented in comparison to these obtained in optimally annealed $Bi_2Dy_1Fe_4Ga_1O_{12}$ garnet films.

The co-deposited optimally annealed films exhibited a rather high specific Faraday rotation (more than 8.0°/µm at 532 nm, compared to around 6.9 °/µm measured in typical $Bi_2Dy_1Fe_4Ga_1O_{12}$ films, which, typically, required annealing at temperatures greater than 650°C) simultaneously with good surface quality. Fig. 2 shows the specific Faraday rotation data points obtained in optimally-annealed composite films prepared on GGG substrates. Almost 100% remnant magnetization behavior was observed in the composite films during the specific Faraday rotation

measurements. The presence of strong remnant magnetization behavior in the composite films resulted from high uniaxial magnetic anisotropy (which ensures that the magnetization direction of films is perpendicular to the film plane). We believe that the hysteresis loops of Faraday rotation in these nanocomposites will be nearly-square in shape, and the hysteresis loop will be very similar to that observed in a 1 µm thick garnet layer of composition type $Bi_2Dy_1Fe_4Ga_1O_{12}$ [3]. Measurements of the hysteresis loops of Faraday rotation as well as other characterization experiments (such as microstructure and crystal structure analyses) will be conducted for this nanocomposite material type, and the results will be published elsewhere.

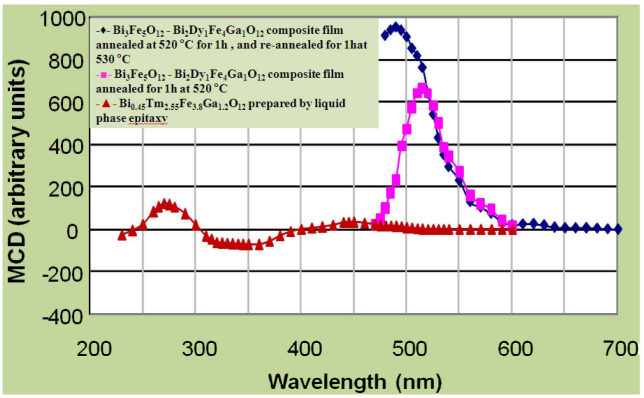

Fig.3. Measured spectral dependencies of MCD signal in (-♦-) RF co-sputtered ($Bi_3Fe_5O_{12}$-$Bi_2Dy_1Fe_4Ga_1O_{12}$) composite film of thickness about 1 µm, annealed for 1 hour at 520 ^oC at first stage and later re-annealed for 1 hour at 530 ^oC; (-■-) co-sputtered composite film of the same stoichiometry when still mainly in its amorphous phase and of the same thickness, but only annealed for 1 hour at 520 ^oC; 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 the same during all measurements.

The MCD measurements (which 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) were performed over the spectral region 300-600 nm in our composite garnet films. A significant spectral "red-shift" effect affecting the main MO transitions in all of our co-sputtered samples was observed in the MCD spectra, in comparison with the spectral locations of these transitions measured in LPE garnet films having much smaller Bi-substitutions. The spectral dependencies of MCD signals were obtained using a sensitive dichrometer that measured the absorption difference between the left- and right-hand circularly polarized light waves propagating through samples in the presence of strong magnetic field directed in parallel to the light beam. Fig. 3 shows the obtained MCD signals (recorded in arbitrary units) from all the samples, plotted as a function of wavelength. The samples annealed at 520^{o}C exhibited the first MCD peak position at 515 nm, while the samples annealed at 530^{o}C displayed a twice 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^{o}C and 520^{o}C, respectively. A slight difference in the annealing temperature led to a significant difference in the material phase content, which can be explained by the fact 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 effective crystallization of each garnet type, with less Bi-rich compositions requiring higher temperatures. It was also expected that less Bi-rich (but still highly Bi-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 above-discussed experimental results demonstrate the feasibility of synthesising new garnet nanocomposite materials and engineering their magnetic and MO properties, making them attractive for various applications in nanophotonics, optical sensors and isolators.

IV. Conclusion

RF magnetron co-sputtered ($Bi_3Fe_5O_{12}$-$Bi_2Dy_1Fe_4Ga_1O_{12}$) thin films have been developed, demonstrating new and interesting MO nanocomposite materials with rather high Bi-substitution well in excess of two formula units. Experimental results have shown that this new class of garnet nanocomposites exhibits simultaneously a high Faraday rotation, good MO quality, magnetization direction perpendicular to the film plane, and notable spectral red-shift features in their MCD spectra.

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.

Acknowledgment

This research is supported by the Faculty of Computing, Health and Science, 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).

References

[1] A. K. Zvezdin and V. A. Kotov, *Modern Magnetooptics and Magnetooptical Materials* (Institute of Physics Publishing, Bristol and Philadelphia), ISBN 075030362X, 1997.

[2] N. Adachi, V. P. Denysenkov, S. I. Khartsev, A. M. Grishin and T. Okuda, "Epitaxial $Bi_3Fe_5O_{12}$ (001) films grown by pulsed laser deposition and reactive ion beam sputtering techniques", J. Appl. Phys., 88, 1, 2734-2739, 2000.

[3] Mohammad Nur-E-Alam, Mikhail Vasiliev, Kamal Alameh and Viatcheslav Kotov, "Garnet multilayer thin film structure with magnetostatically-altered and improved magnetic properties prepared by RF magnetron sputtering," In Proc. Int. Conf. on High-capacity

Optical Networks and Enabling Technologies (HONET 2011), pp-177-181, December 19-21, 2011, Saudi Arabia.