1-1-2011

Garnet Multilayer Thin Film Structure with Magnetostatically-Altered and Improved Magnetic Properties Prepared by RF Magnetron Sputtering

Mohammad Nur-E-Alam
Edith Cowan University

Mikhail Vasiliev
Edith Cowan University

Viacheslav Kotov

Kamal Alameh
Edith Cowan University

10.1109/HONET.2011.6149812

Garnet Multilayer Thin Film Structure with Magnetostatically-Altered and Improved Magnetic Properties prepared by RF Magnetron Sputtering

Mohammad Nur-E-Alam[1]*, Mikhail Vasiliev[1]*, Viacheslav Kotov[2] and Kamal Alameh[1, 3]

[1]Electron Science Research Institute, Edith Cowan University, 270 Joondalup Dr, Joondalup, WA 6027, Australia
[2]Institute of Radio Engineering and Electronics, Russian Academy of Sciences, 11 Mohovaya St, Moscow 125009, Russia
[3]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, m.vasiliev@ecu.edu.au

***Abstract*—We prepare an all-garnet multilayer film structure by sandwiching a magneto-soft garnet material in between two magneto-hard garnet materials with high bismuth substitution levels using RF magnetron sputtering technique and investigate the microstructure and the effects of magnetostatic inter-layer coupling on magnetic properties. Both types of the Bi-substituted magneto-optic garnet materials used possess excellent optical, magnetic and magneto-optical properties suitable for the application in different new and emerging technologies in optics and photonics. Garnet layers of composition type $Bi_2Dy_1Fe_4Ga_1O_{12}$ have strong perpendicular magnetic anisotropy and $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ magneto-soft layer features magnetization behavior similar to that of in-plane magnetized films. The all-garnet magnetostatically-coupled multilayer structure fabricated demonstrates an attractive combination of low coercive force and high uniaxial magnetic anisotropy, which is extremely useful for applications in nanophotonics, optical sensors and isolators.**

***Index Terms*—Magneto-optic materials, garnet multilayers, magnetic anisotropy, coercive force, nanophotonics.**

I. Introduction

Multilayer garnet thin film structures and nano-structured magnetic photonic crystals (MPCs) have attracted a world-wide research interest recently due to their application potential in information technology and all-optical reconfigurable signal processing devices. A number of research works have been conducted during the last decade to design and fabricate multilayer thin film structures and photonic crystals including ferrimagnetic, paramagnetic and dielectric materials of different composition types for use in various applications, for example in optical sensors, magnetic field visualizers and integrated optical isolators [1-10]. It is always challenging to fabricate high-quality thin film nano-structures (of either single- or multi-layer type) with a good degree of control over their magnetic behaviour, especially if the magnetic switching behaviour and magnetic anisotropy properties need to be adjustable. RF magnetron sputtering allows precise control over the thin films deposition process parameters, thus reducing the problem of synthesizing high-quality thin films and multilayer structures wherein each layer's chemical composition (stoichiometry) must be controlled accurately [10, 11]. To realise special magnetic behaviours, which are not attainable using single-layer thin films of a chosen garnet material type, we prepare an all-garnet multilayer structure using two record-performance highly-Bi-substituted iron garnet materials having very different magnetic behaviours (magnetic anisotropy, switching fields and saturation magnetizations). The two used Bi-substituted iron garnet (Bi:IG) thin film materials, which have previously been studied by our group in detail [12, 13], possess extra-ordinary optical and magneto-optical (MO) properties that are highly attractive for use in various existing and also new applications in optics, photonics and optoelectronics.

The individual performance characteristics of these two selected garnet materials of composition types $Bi_2Dy_1Fe_4Ga_1O_{12}$ $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ have also been reported to possess exceptional optical and MO qualities, as well as very good magnetic properties when mixed with extra co-sputtered bismuth oxide to obtain nanocomposite garnet-oxide derivative materials of type $(BiDyLu)_3(FeGaAl)_5O_{12}$:$Bi_2O_3$ [12, 13]. The present study is aimed at achieving improved magnetic properties in all-garnet multilayers by making triple-layer structures composed of different high-performance material layers that (due to being in close proximity geometrically) interact with each other magnetostatically, leading to improved magnetic properties. The material of composition type $Bi_2Dy_1Fe_4Ga_1O_{12}$ is a highly bismuth substituted iron garnet which possesses very high specific Faraday rotation (up to several thousand °/cm) in the visible range. This material possesses high uniaxial magnetic anisotropy (with its magnetization direction perpendicular to the film's plane) and also excellent magnetic memory properties. On the other hand, Bi- substituted lutetium iron garnet material of composition $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ features a magneto-soft behaviour with almost in-plane magnetization direction and high specific Faraday rotation [13].

In this paper, we report on the properties of an all garnet multilayer structure which is formed by sandwiching a garnet layer of composition type $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ in between two layers of $Bi_2Dy_1Fe_4Ga_1O_{12}$. The goal of this work is to study the engineering of magnetic properties in multilayers, and especially the ways of adjusting the coercive force and magnetic anisotropy properties. The experimental processes

for multilayer structure formation and characterization are detailed in section II, and the results achieved in our high-performance all-garnet multilayer structures are discussed in section III. Finally, we draw a conclusion in section IV.

II. Growth and Characterization of All-Garnet Multilayer Thin Film Structures

A garnet multilayer structure was prepared using RF sputtering deposition of highly Bi-substituted iron garnet materials of composition types $Bi_2Dy_1Fe_4Ga_1O_{12}$ (with perpendicular magnetization) and $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ (with its magnetization vector being almost in-plane). Fig. 1 shows the schematic diagram of our study and its approach as well as the type of materials and their measured Faraday rotation hysteresis loops (Fig. 1(a)). The multilayer structure contains a layer (thickness 500 nm) of $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ in between two layers of $Bi_2Dy_1Fe_4Ga_1O_{12}$ of the same thickness, as shown in Fig. 1(b). Fig.1(c) shows the expected (and desired) outcome in terms of the properties of this type of all-garnet multilayer structure, demonstrating a combination of low coercive force, low saturation field and high remnant magnetization.

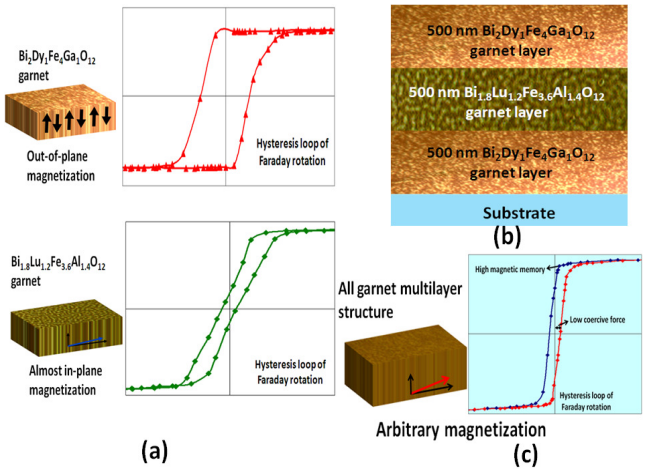

Fig.1. (a) Types of highly Bi-substituted iron garnet materials used and their Faraday rotation hysteresis loop behaviors; (b) a schematic diagram of a three-layer all-garnet structure[Substrate/(500 nm $Bi_2Dy_1Fe_4Ga_1O_{12}$)/(500 nm $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$)/(500 nm $Bi_2Dy_1Fe_4Ga_1O_{12}$)], and (c) the expected (hybrid-type) magnetic property outcomes of the all garnet magnetostatically-coupled multilayer structure with excellent magnetic switching properties.

The multilayer structure was deposited onto gadolinium gallium garnet (GGG) and also onto glass (Corning Eagle XG) substrates using RF magnetron sputtering. The structures were prepared on both substrate types in a single deposition run by sequential sputtering of layers using low-pressure argon plasma and a relatively low substrate temperature of 250 °C. Table 1 summarizes the conditions and process parameters that were used to prepare the three-layer garnet structures.

After the deposition of multilayer structures, a conventional oven annealing system (CF 1200X, MTI Corporation) was used to crystallize the structures in order to synthesize the garnet phase in each layer. To find the most suitable annealing regime for simultaneously crystallizing both material types within these garnet multilayer structures, a significant amount of experimental work was performed. In this study, we used a range of annealing temperatures between 610-650 °C and crystallization process durations ranging between 1-10 hours. All annealing experiments were conducted using slow ramp-up and ramp-down process rates of 3 °C/min to avoid any significant micro-crack formation.

Table.1. Typical sputtering conditions and process parameters used to produce all-garnet multilayers (triple-layer thin films).

Sputtering parameters	Comments & values
Garnet targets (oxide-mix stoichiometries)	$Bi_2Dy_1Fe_4Ga_1O_{12}$ & $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$
Sputter gas and pressure	Argon (Ar), P(total) = 1 mTorr
Base pressure	P(base) < 1-2E-06 Torr (high vacuum)
RF power densities	6.09 W/cm^2 (280 W) for $Bi_2Dy_1Fe_4Ga_1O_{12}$ target & 3.81 W/cm^2 (175 W) for $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ target
Substrate surface temperature during deposition	250 °C
Substrate stage rotation rate	50-53 rpm
Oven annealing regimes used (temperature and time)	610-650 °C and between 1-10 hours with 3 °C/min ramp-up and ramp-down rates

The transmission and reflection spectra of annealed multilayer structures have been measured using a Beckman Coulter D 640B UV/Visible spectrophotometer. The measurements of total Faraday rotation angle of the structures and the Faraday hysteresis loops were performed using a Thorlabs PAX polarimeter system and a custom-made calibrated electromagnet. A Leitz Orthoplan polarization microscope was used to observe (in transmission mode) the magnetic domain structures within our multilayers. Finally, we made a film cross-section using focused ion beam (FIB) milling of an annealed all-garnet multilayer on a GGG substrate and observed its microstructural properties.

III. Optical, Magnetic and Microstructure Properties of All Garnet Multilayer Structure

The physical thicknesses of each layer of the multilayer thin film 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 layer thicknesses of the structure using specialized thickness-fitting software (MPC optimization software made at ECU) and the optical transmission spectra of both the as-deposited and post-annealed structures. Fig. 2 shows the transmission spectra of a three-layer garnet structure obtained from an amorphous and a post-annealed sample, compared to the modelled

transmission spectra of the same structure. The measured transmission spectra are very similar to the modelled transmission spectrum but they show less transparency by several percent in the visible spectral range and are also more transparent in the near IR region as compared to the modelled one. This is due to slight variations in the refractive index and (especially) the variations in absorption spectra of both garnet types with the thermal treatment regime.

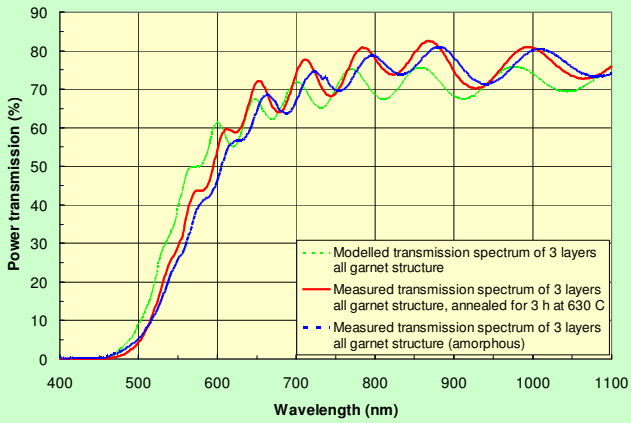

Fig.2. Transmission spectra of [GGG / (500 nm $Bi_2Dy_1Fe_4Ga_1O_{12}$) / (500 nm $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$) / (500 nm $Bi_2Dy_1Fe_4Ga_1O_{12}$)] multilayer garnet structure, (green dotted line) is modeled transmission spectrum of this structure, (red line) is the measured transmission spectrum of a structure which was annealed with an optimized annealing regime (3h at 630 C) and (blue dotted line) is the measured transmission spectrum of this structure obtained immediately after the deposition.

The reflection spectrum measurement of samples was also performed using the UV/Visible spectrophotometer. A non-polarizing beam-splitter cube (the reflectance of the cube at glass-air interfaces was about 4%) was used to make the reflectivity measurements by re-directing part of the light source beam to be reflected off samples, with a silver mirror (100 nm silver layer deposited onto a glass substrate) being used as a reference mirror which also deflected the light toward the sample placed next to a cube side opposite to the side next to which the mirror was placed. The reflection coefficient of the mirror is wavelength dependent but the variations were small enough to be neglected, and we used a spectrally-flat 97% reflectance approximation for our silver mirror. The wavelength dependency of the cube's internal diagonal reflectance was also measured and used to calculate the sample's own reflectance at each wavelength from the spectrophotometer data obtained in (the only available) transmission measurement mode with and without the sample placed next to the cube's side surface. The sample was placed with the garnet layer surface firmly against the glass cube side during the measurement. From the measurements of the transmission and reflection spectra, we derived the absorption spectrum of our three-layer garnet structure using the simple formula $A=1-T-R$ (%), where A is the absorbed power fraction, T is the power transmission coefficient, and R is power reflectivity. There was a strong agreement observed between the derived absorption spectrum and the modelled absorption spectrum of the annealed three-layer garnet structure, as is shown in Fig. 3.

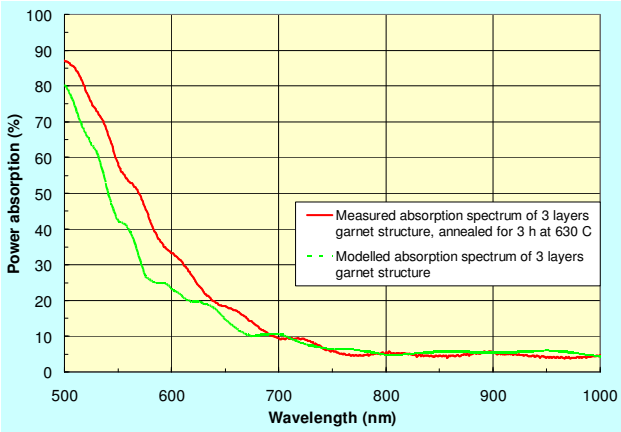

Fig.3. Absorption spectra of [GGG / (500 nm $Bi_2Dy_1Fe_4Ga_1O_{12}$) / (500 nm $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$) / (500 nm $Bi_2Dy_1Fe_4Ga_1O_{12}$)] multilayer garnet structure, (green dotted line) is modeled absorption spectrum of this structure, (red line) is the measured absorption spectrum of a structure which was annealed using an optimized annealing regime (3h at 630 C)

The measurements of Faraday rotation (total polarization plane azimuth rotation angle) for the all-garnet multilayer structure and the specific Faraday rotation for single garnet layers as well as the hysteresis loop of Faraday rotation have been performed using a 532 nm plane-polarized solid-state laser. There were two distinctly different shapes of hysteresis loop observed in two different types of single-layer garnet thin films each having 1 μm thickness. A nearly-square hysteresis loop with almost 100% remanence was observed in a 1 μm-thick single layer of $Bi_2Dy_1Fe_4Ga_1O_{12}$ garnet material. The measured coercive force for thin (1000 nm) $Bi_2Dy_1Fe_4Ga_1O_{12}$ garnet films sputtered at 250 °C onto GGG substrates was in excess of 200 Oe (red curve), and their saturation field was about 500 Oe. Note that the values of coercive force for this type of material are also dependent on the films' thickness, and coercive force values of about 1000 Oe were measured for a 4 μm-thick film of the same composition and reported in Ref [10]. The coercivity of $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ garnet films of 1 μm thickness prepared onto GGG substrates was only about 45 Oe (green curve), and their saturation magnetization was about 350 Oe, as shown in Fig.4. The almost-linear character of the magnetization curves observed in $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ garnet films below magnetic saturation indicates that a significant component of the film's magnetization lies in the film plane [13]. The observed magnetic domain patterns in these two magneto-optic garnet thin-film materials were also totally different, as shown in the inset of Fig. 4.

A notably low coercive force and also rather high uniaxial magnetic anisotropy (evidenced by the large remnant magnetization) was observed in our all-garnet multilayer structure (Fig. 5). The best (among all samples from our deposition batch, which had different thermal treatment histories) Faraday rotation at 532 nm was measured in the multilayer structure which was annealed for 3h at 630 °C and was more than ± 3° (total angle), and the coercive force

measured in the same sample was about 100 Oe, with a value of saturation field of near 200 Oe.

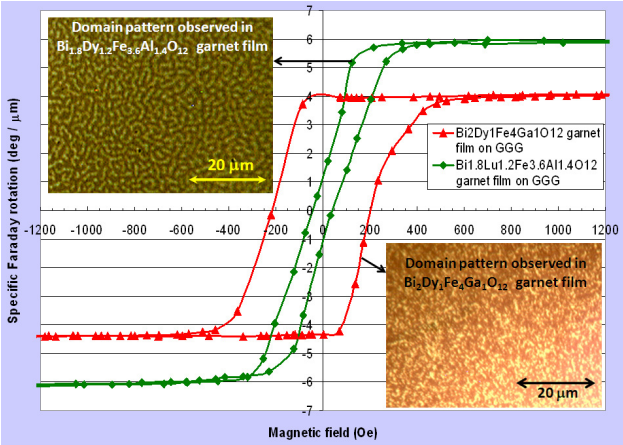

Fig.4. Measured hysteresis loops of Faraday rotation in a $Bi_2Dy_1Fe_4Ga_1O_{12}$ garnet thin film sputtered onto GGG substrate (red curve) and in $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ garnet thin film prepared onto GGG substrates. The film thicknesses were 1000 nm, and their magnetic domain structures observed visually using a transmission-mode polarization microscope (Leitz Orthoplan) are shown in inset.

We believe that within the garnet thin-film multilayer structures, the effects of magnetostatic coupling between the different garnet layers of two different magnetic behaviour types controlled the overall magnetic properties of the structure. The intermediate layer of garnet having almost in-plane magnetization might have reduced the overall coercivity compared to that of the top and bottom magneto-hard garnet layers, whilst the strong perpendicular magnetization of these two outer layers helped retained the magnetic memory properties in the behaviour of the multilayer structure.

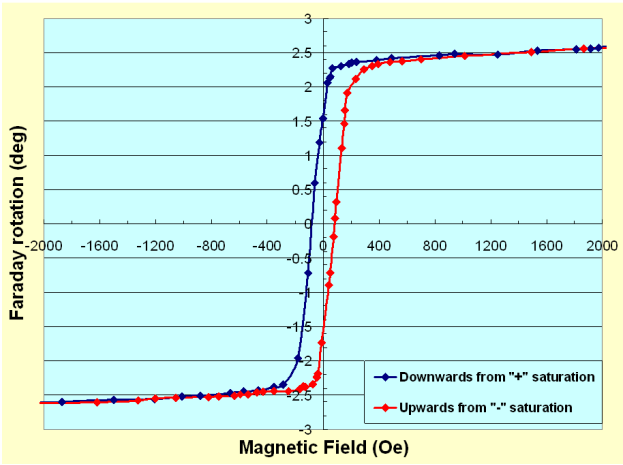

Fig.5. Measured magnetic hysteresis loop in [Sub (GGG) / (500 nm $Bi_2Dy_1Fe_4Ga_1O_{12}$) / (500 nm $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$) / (500 nm $Bi_2Dy_1Fe_4Ga_1O_{12}$)] multilayer all-garnet structure deposited onto GGG (111) substrate (and annealed for 3h at 630 °C after the deposition).

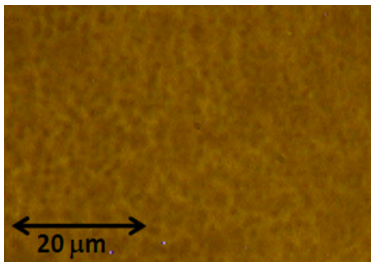

Fig.6. Magnetic domains observed in an all-garnet multilayer thin-film structure deposited onto a GGG substrate at T(sub) = 250 °C (annealed for 3 h at 630 °C) using a transmission-mode polarization microscope at high magnification (630 X).

Fig. 6 shows the magnetic domains obtained in a three-layer all-garnet structure prepared onto a GGG substrate applying an optimized annealing process. The image was taken after the structure was brought into a brief mechanical contact with a strong permanent magnet, which magnetized both outer garnet layers into the same saturated state, after which these layers' magnetization was at its remnant (monodomain) state. The image thus only revealed the magnetic domain pattern of the intermediate (magneto-soft) layer. A focused ion beam (FIB) cross-sectional image of this annealed garnet multilayer structure is shown in Fig. 7. This microstructure observation confirmed the successful crystallization of the entire multilayer structure having dissimilar high-bismuth-content MO garnet materials after a single optimised thermal annealing process. No significant incompatibilities were found between these two types of magnetic materials used in the multilayer structure, which is confirmed by the smooth interfaces between the layers.

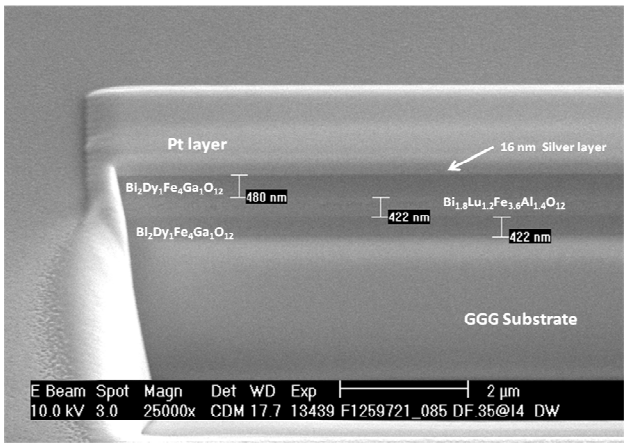

Fig.7. FIB cross-sectional image (imaged by electron beam) of a 3-layer all-garnet structure which was subjected to annealing for 10 h at 630 °C. The thicknesses of all layers are almost the same and are close to 500 nm each, even though some measurement bars in the figure are inaccurate and show a smaller thickness.

The experimental results achieved demonstrate the possibility of engineering the magnetic properties of multilayer all-garnet structures using combinations of materials with different magnetic behavior types.

IV. Conclusion

All-garnet multilayer structures incorporating two types of highly Bi- substituted doped iron garnet materials of high MO performance but having dissimilar magnetic properties have demonstrated that very promising magnetic properties can be engineered within multilayer structures. This is technologically interesting for a number of applications in photonics and integrated optics. This has been our first experimental work on modifying the magnetic properties of garnet materials with the goal of reducing the coercive force whilst maintaining high specific Faraday rotation and remanence. Further studies will be conducted in order to prepare garnet multilayer structures having different combinations of high-performance garnet materials of various optimized thicknesses as well as different orientations of magnetization vectors within the material layers. These types of nano-engineered garnet multilayer structures are very important for multiple applications in nano-photonics, especially for the design of optical sensors and integrated optical isolators.

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] G. B. Scott and D. E. Lacklison, "Magnetooptic properties and applications of bismuth substituted iron garnets," IEEE Trans. Magn. 12, 292-311, 1976.

[3] I. Nistor, C. Holthaus, I. D. Mayergoyz, and C. Krafft, "Determination of layer-specific magnetization and anisotropy fields in multilayer garnet films from FMR and VSM measurements," IEEE. Trans. Magn. 42, 10, 3261-3263, 2006.

[4] I. L. Lyubchanskii, N. N. Dadoenkova, M. I. Lyubchanskii, E. A. Shapovalov and Th. Rasing, "Magnetic photonic crystals," J. Phys. D: Appl. Phys. 36, R277–R287, 2003.

[5] M. Vasiliev, V. A. Kotov, K. E. Alameh, V. I. Belotelov and A. K. Zvezdin, "Novel Magnetic Photonic Crystal Structures for Magnetic Field Sensors and Visualizers," IEEE Trans. Magn. 44, 3, 323, 2008.

[6] M. J. Steel, M. Levy, and R. M. Osgood, "High Transmission Enhanced Faraday Rotation in One-Dimensional Photonic Crystals with Defects," IEEE Photon. Technol. Lett. 12, 9, 1171-1173, 2000.

[7] S. I. Khartsev and A. M. Grishin "[Bi3Fe5O12/Gd3Ga5O12]^m magneto-optical photonic crystals" Appl. Phys. Lett., 87, 12, p. 122504, 2005.

[8] S. I. Khartsev and A. M. Grishin "[Bi3Fe5O12/Sm3Ga5O12]^m magneto-optical photonic crystals" J. Appl. Phys.,101, 053906 (2007).

[9] M. Nur-E-Alam, M. Vasiliev and K. Alameh, "Nano-structured magnetic photonic crystals for magneto-optic polarization controllers at the communication-band wavelengths," Opt. Quant. Electron. 41, 661-669, 2009.

[10] M. Vasiliev, K. Alameh, V.A. Kotov, and Y.T. Lee, "Nanostructured Engineered Materials With High Magneto-Optic Performance for Integrated Photonics Applications" in Proc. IEEE Photonics Global @ Singapore, 2008 (IPGC 2008), ISBN 9781424439010, http://ieeexplore.ieee.org/stamp/stamp.jsp?arnumber=4781410&isnumber=4781297 .

[11] M. Vasiliev, P. C. Wo, K. Alameh, P. Munroe, Z. Xie, V. A. Kotov and V. I. Burkov, "Microstructural characterization of sputtered garnet materials and all-garnet magnetic heterostructures: establishing the technology for magnetic photonic crystal fabrication," J. Phys. D: Appl. Phys. 42, 135003, 2009.

[12] M. Vasiliev, M. Nur-E-Alam, V. A. Kotov, K. Alameh, V. I. Belotelov, V. I. Burkov and A. K. Zvezdin, "RF magnetron sputtered $(BiDy)_3(FeGa)_5O_{12}$:Bi_2O_3 composite garnet-oxide materials possessing record magneto-optic quality in the visible spectral region," Opt. Express 17, 19519, 2009.

[13] M. Nur-E-Alam, M. Vasiliev, V. A. Kotov, and K. Alameh, "Highly bismuth-substituted, record-performance magneto-optic garnet materials with low coercivity for applications in integrated optics, photonic crystals, imaging and sensing", Opt. Mat. Express 1, 2, 413, 2011.