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Growth, Characterization, and Properties of $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ garnet films prepared using two different substrate temperatures

ABSTRACT

We prepare highly Bismuth substituted iron garnet of composition type $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ by using low and high substrate temperatures (250 °C and 680 °C) during the deposition process inside the vacuum chamber of RF magnetron sputtering system. The crystallization process of this garnet type thin film materials are performed by means of optimized high temperature oven annealing process and conduct several characterization techniques to obtain and evaluate their properties. All the optimally annealed samples possess very promising and attractive properties. Comparatively low coercive force (below 15 Oe) is achieved in garnet films prepared at high substrate temperature of 680 °C, simultaneously with high MO quality and almost in-plane magnetization direction, which are the properties desired in various MO sensing, switching and imaging applications.

1. INTRODUCTION

High performance functional magneto-optic garnet materials are needed for modern optics and photonics applications. Bismuth-substituted iron garnets are very well known to be the best magneto-optical (MO) materials for various applications in integrated optics and nano-photonics due to their strong MO behaviour and fast magnetization switching response (Zvezdin and Kotov, 1997, Kang et al., 2007, Scott and Lacklison, 1976). A number of important application areas require MO thin films with very high MO quality and also low coercivity with either the in-plane-oriented easy axis of magnetization or having a strong in-plane magnetization component (Alam, Mohammad, 2012). Multiple variations in the material properties of garnets are required for different applications in integrated optics and in photonics, which can be engineered by adjusting the material composition, however every composition requires its own optimized deposition and annealing regimes.

We were the first group that synthesized a garnet material of composition type $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ using the sputter-deposition technique and oven-annealing, and achieved films that possessed very high MO performance simultaneously with strong in-plane magnetization component and magnetically-soft switching behaviour(Nur-E-Alam et al., 2010, 2011). Recently, we have demonstrated a significant enhancement of transverse magneto-optic Kerr effect (TMOKE) in magneto-plasmonic structures using sputtered films of composition type $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$. The magnitude of TMOKE observed in magneto-plasmonic crystals was about 13% (Pohl et al., 2013). Note that these garnet thin films were prepared in low temperature (250°C) atmosphere inside the RF magnetron sputtering chamber.

In this paper, we investigate the effects of substrate temperature (used during the deposition of garnet layers using RF magnetron sputtering) on the materials properties and compare the results achieved using either “cold (250°C)” or “hot (680°C)” substrates. The steps used in the investigations are as follows:

1. Target stoichiometry selection
2. Substrate preparation
3. Thin film garnet layer deposition
4. Garnet layer (as-deposited) crystallization
5. Garnet thin film characterization
6. Result evaluation

The experimental processes to deposit garnet layers onto the substrates are detailed in Section 2. Section 3 presents a short description of the techniques (experimental setups and processes) that were applied to characterize the sputtered garnets-type thin-film materials, and a summary of obtained results are discussed in Section 4. Finally, a short conclusion is drawn in Section 5.

2. THIN FILM GARNET LAYER GROWTH

Bi-substituted lutetium iron-aluminium garnet thin films (several batches from 650 nm to 1000 nm in thickness) were fabricated using RF magnetron sputtering technology. The sputtering target of nominal stoichiometry $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$was used to deposit amorphous garnet-type layers onto different substrates (GGG, Corning Eagle XG and silicon). As-deposited garnet layers were sputtered using two widely different substrate temperatures (250°C and 680°C as measured by a thermocouple sensor placed next to SiC heater) at high vacuum. Table 1 summaries the process parameters used to deposit the garnet amorphous layers onto the optical substrates.

As-deposited Bi-substituted MO garnet films (amorphous structure) have been subjected to crystallization using optimized annealing process within a conventional oven annealing system to obtain the garnet phase and achieve polycrystalline microstructure within the garnet layers (Vasiliev et al., 2009, Wo et al., 2009). Note that finding an optimized annealing regime for each garnet thin film material type was always a key factor necessary to achieve the best optical and MO properties in films, as well as to maintain high quality film surfaces. The effects of annealing heat treatment running with different temperatures and process durations on the optical and MO properties of garnet thin films have been observed and evaluated (discussed in Result section) to investigate the ways of optimizing the relevant process parameters. Table 2 summaries the optimized annealing regimes (temperature and process duration) obtained from many annealing trails used to crystallized this garnet type thin film materials. These are mostly reliable sources of data to reproduce high bismuth content garnet thin films of composition type $(BiLu)_3(FeAl)_5O_{12}$.

3. CHARACTERIZATION OF GARNET LAYERS

A Beckman Coulter DU 640B UV/Visible spectrophotometer was used to obtain the transmission spectra in the garnet layers (as-deposited and annealed), and the measurement set-up is shown in Fig. 1 with a schematic diagram of sample chamber and measurement process. The calibration process for this system was performed by taking a blank transmission spectrum without any sample before performing the transmission measurement of the sample. When the light passes through the sample, a certain amount of light is absorbed and some light is reflected by the sample (and in some samples, some light is also scattered by the film layers), and the remaining power of the incident light is captured as transmitted light for spectral analysis. In this work transmission spectra measurement for all the batches of garnet layers (as-deposited and annealed) were one of the important tasks as the physical layer thicknesses and the absorption coefficients of garnet thin film materials were derived using the obtained transmission spectra by the spectrophotometer to the specialized thickness-fitting software. The film thicknesses were definitely monitored during the deposition processes using in-situ laser reflectometry.

The measurements of specific Faraday rotation and hysteresis loop of Faraday rotation were performed with a well calibrated measurement setup using Thorlabs PAX polarimeter and a custom-made electromagnet. Fig.2 Schematic of the set-up of Faraday rotation and Faraday rotation hysteresis loop measurement system that used in the materials characterization lab at the Electron Science Research Institute in Edith Cowan University. This measurement system is based on using linearly polarized laser light sources (mostly the sources working in the visible spectral range). The Thorlabs PAX polarimeter (used for Faraday rotation measurements) had a high dynamic range of 70 dB, a broad wavelength range, and an absolute accuracy of ± 0.2° (Alam, Mohammad, 2012). The sample to be characterized was usually placed into the middle of electromagnet gap and the polarized light is allowed to pass through the semitransparent garnet thin films along the direction of external magnetic field applied.

Magnetic hysteresis loop is the relationship between the magnetic flux induced within a magnetized material and the external magnetic field strength variations, which allows characterization of the magnetic switching behavior of garnet thin films. Hysteresis loop measurements in garnet thin films were performed using the same setup as shown in Fig. 2. The measurements of hysteresis loops of Faraday rotation in garnet thin films were performed in the presence of external magnetic field applied in the direction perpendicular to the film plane and parallel to the light propagation direction.

The MO figure of merit (doubled the ratio of the specific Faraday rotation to the material absorption coefficient at each wavelength, $2\Theta_F/\alpha$) for the annealed samples, was calculated using the measured Faraday rotation data. The specific Faraday rotation measured in films at the saturated (or sometimes, remnant) magnetization state and α is the optical absorption coefficient measured at the same wavelength. MO figure of merit allows comparing the expected performance of new or existing materials quickly and quantitatively which is very important and essential in new MO materials development for magneto-optic device applications (Challener, 1995, Steel et al., 2000).

A transmission mode polarization microscope (Leitz Orthoplan) was used to observe the magnetic domain patterns in the annealed thin garnet films.

The domain structures of the garnet thin films were observed in the absence of external magnetic fields, in demagnetised samples. EDX microanalysis performed at the Gwangju Institute of Science and Technology (GIST) characterization labs to confirm the presence of all elements expected to be present within the garnet films

4. RESULTS AND DISCUSSION

Crystallized garnet films annealed using temperatures above 620°C (prepared on GGG substrates) showed the best optical and MO properties across the visible spectral region. The derived absorption spectra of samples (as-deposited and annealed) prepared onto GGG substrates using 250 °C and 680 °C deposition are plotted in Fig. 3(a). It is important to note that actual substrate surface temperatures were estimated to be about 30% lower than these measured by thermocouple sensor, which is the likely reason that even samples grown on “hot” substrates were not crystallized in-situ.

Derivations of these absorption spectra were performed using the transmission spectra of samples and fitting software, which correctly accounts for the effects of refractive index dispersion as well as reflection spectrum. All films (sputtered at either 250 °C or 680°C) had rather low optical absorption and significantly high Faraday rotation, which led to obtaining high MO figures of merit. The maximum measured values of Faraday rotation per unit film thickness of the garnet films prepared at 250 °C were very close to these obtained in films prepared at 680 °C substrate temperature, but the lower absorption losses achieved in films prepared at higher temperature compared to other samples helped obtain a better MO figure of merit (42.8° at 635 nm) than that of the sample sputtered at 250°C. The best obtained values of MO quality factors of $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ garnet layers deposited onto GGG (111) substrates using 250 °C and 680 °C substrates temperature are shown in Fig. 3(b).

The magnetic films possessed very low coercive force values, as was observed during the measurements of hysteresis loop of Faraday rotation (made using a 532 nm polarized light source). Fig. 4 shows the obtained hysteresis loops of specific Faraday rotation in films prepared using 250°C (green color) and also 680°C (red color) substrate temperatures. The measured coercive force values were about 45 Oe for the garnet films sputtered at 250°C and 10 (±2) Oe for the films sputtered at 680°C. Both film batches showed very strong in-plane magnetization component (weak uniaxial magnetic anisotropy), which was confirmed by the almost-linear character of their magnetization curves below the saturation observed in the measured hysteresis loops of Faraday rotation.

The domain structures obtained in good-quality garnet films were observed in two demagnetized samples in absence of any externally applied magnetic fields (Fig. 5 (a, b)) confirmed that the good surface quality has been achieved in the films sputtered onto GGG substrates at different substrate temperatures. EDX microanalysis confirmed the presence of all elements expected to be present within films (measured element concentrations in atomic % are shown Fig.5(c)). Based on these measurement data, we derived the averaged composition of thin film to be $Bi_{1.68}Lu_{0.656}Fe_{4.294}Al_{1.184}O_{12}$ .which might be expected considering the sputtering target's nominal stoichiometry and possible Bi content loss occurring during layer growth.

5. CONCLUSION

Magneto-optic garnet thin films of composition type $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ have been prepared using two different substrate temperatures during the deposition process and the suitable annealing process parameters for films of this type were found, leading to obtaining high quality magneto-soft thin films with excellent MO properties. Significantly low coercive forces (less than 15 Oe), almost in-plane magnetization direction, and high MO performance have been observed in these garnet thin films, which are attractive for use in various existing applications such as MO sensing and imaging. The combinations of materials properties achieved in this garnet type material system are not only important for the development of garnet waveguides and non-reciprocal integrated-optics components but also highly attractive for new and forward-looking applications in photonics, optoelectronics, magnetic photonic crystals and magneto-plasmonics.

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Tables

Table 1. Sputtering conditions and process parameters used to synthesize the magneto-optic $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ garnet layers

Sputtering process parameters	**Values & comments**
Sputtering targets composition	$Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ (materials' purity 99.99%)
Target size	3´´ (diameter) with the material layer thickness of 1/8´´, bonded to a 1/8´´ Cu backing plate
Background Pressure	P(base) < 1-2·10^{-6}Torr
Process gas and pressure	Argon, P(Ar) = 1-2 mTorr
RF power density at target	Typically 3.7-3.8 W/cm^2 (170-175 W)
Substrate-target distance	18-20 cm
Substrate temperature during deposition	250 & 680 ^{0}C
Substrate stage rotation	36-40 rpm
Substrate types	Glass (Corning Eagle XG) and monocrystalline GGG (111)
Deposition rates	4-6 nm/min

Table 2.Summary of optimization of annealing temperature and annealing processes duration used to crystallize $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ garnet layer deposited at 250 °C and 680 °C substrates' temperature

Sample	**Annealing temperature (°C)**	**Annealing process duration (hr)**	**Comments**
Garnet layer deposited at 250 °C	650 (±10) °C	1	Annealed, good film surface quality and high Faraday rotation observed
Garnet layer deposited at 680 °C	630 (±10) °C	3	Annealed, good film surface quality and relatively high Faraday rotation observed

Figure Captions

Fig.1.Photograph of the set-up of a Beckman Coulter DU 640B UV/Visible spectrophotometer system used to perform the transmission spectra measurements in all batches of garnet thin film materials.

Fig.2.Schematic of the set-up of Faraday rotation and Faraday rotation hysteresis loop measurement system used to characterize the optimally annealed garnet films.

Fig.3. (a) Derived absorption spectra in films sputtered onto GGG substrates using 250 °C and 680 °C substrate temperatures. (b) The values of MO figure of merit at 532 and 635 nm obtained in both film batches after the optimized annealing process.

Fig.4. Measured hysteresis loops of specific Faraday rotation at 532 nm in sputtered $Bi_{1.8}Lu_{1.2}Fe_{3.6}Al_{1.4}O_{12}$ garnet films on GGG substrates deposited at (a) 250 °C and annealed for 1 h at 650 °C, and (b) 680 °C substrate and annealed for 3 h at 630 °C.

Fig.5.Magnetic domain patterns observed in films sputtered onto GGG (111) at 250°C, annealed for 1 h at 650°C (a); deposited at 680°C and annealed for 3 h at 630°C (b); and EDX composition measurement data obtained from films grown at 680°C substrate temperature (c).

Author

Figures

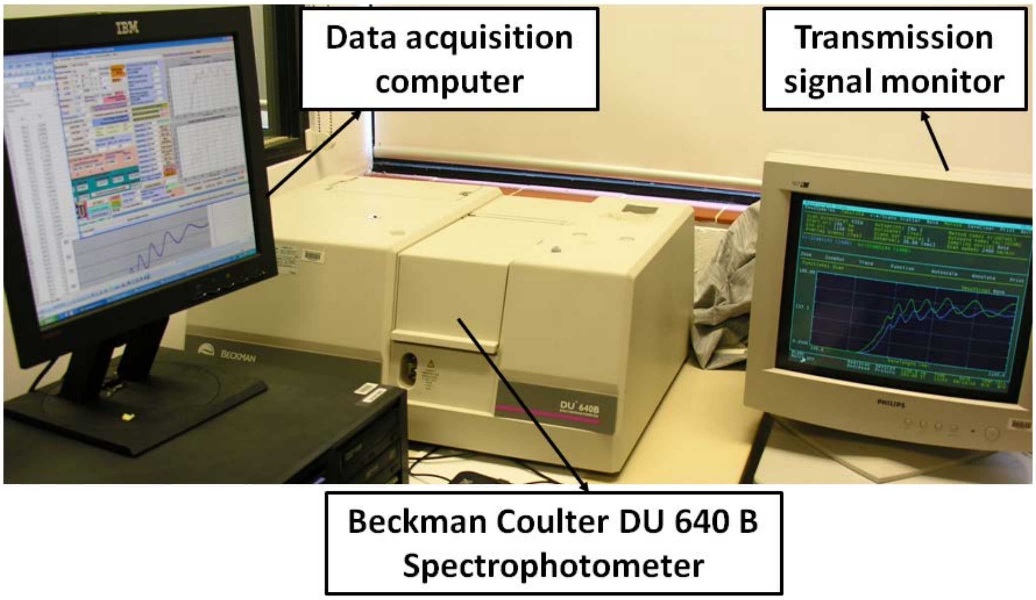


Fig. 1

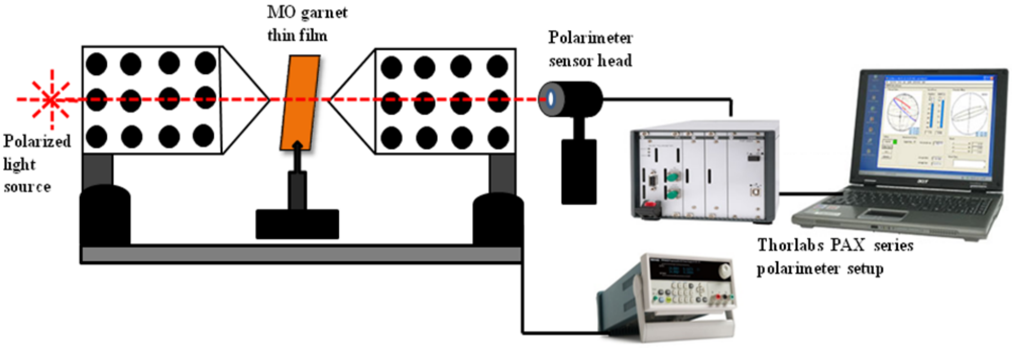


Fig. 2

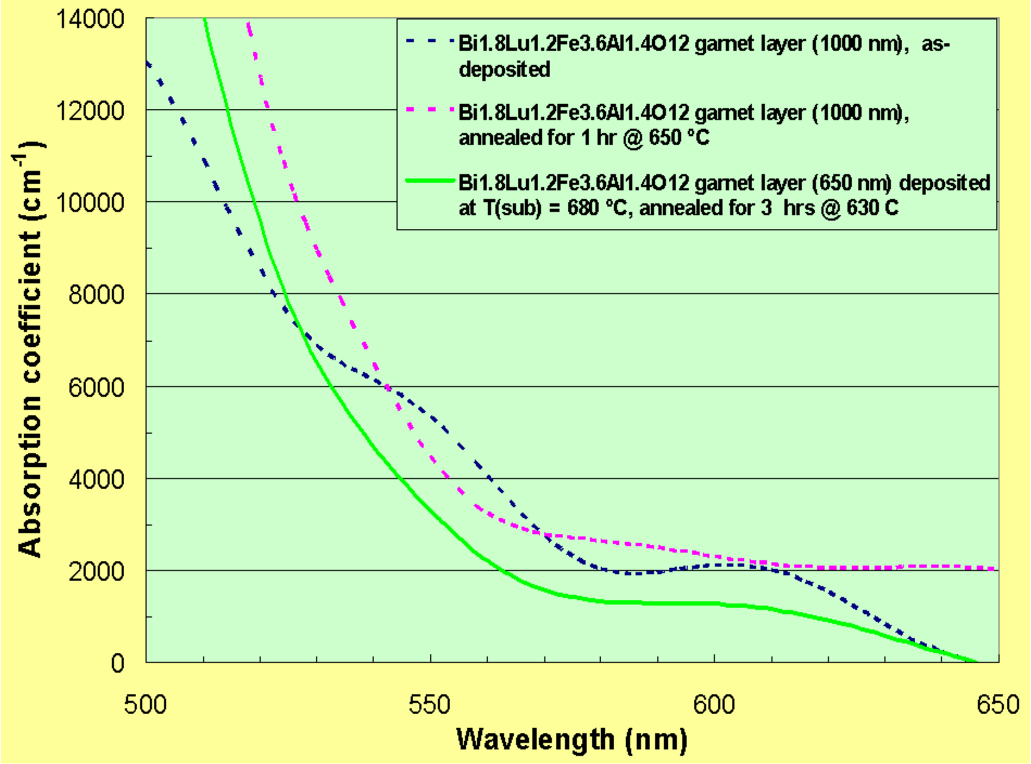

14000
12000
10000
8000
6000
4000
2000
0
Absorption coefficient (cm-1)
500
550
600
650
Wavelength (nm)
Bi1.8Lu1.2Fe3.6Al1.4O12 garnet layer (1000 nm), as-deposited
Bi1.8Lu1.2Fe3.6Al1.4O12 garnet layer (1000 nm), annealed for 1 hr @ 650 °C
Bi1.8Lu1.2Fe3.6Al1.4O12 garnet layer (650 nm) deposited at T(sub) = 680 °C, annealed for 3 hrs @ 630 C

Fig. 3(a)

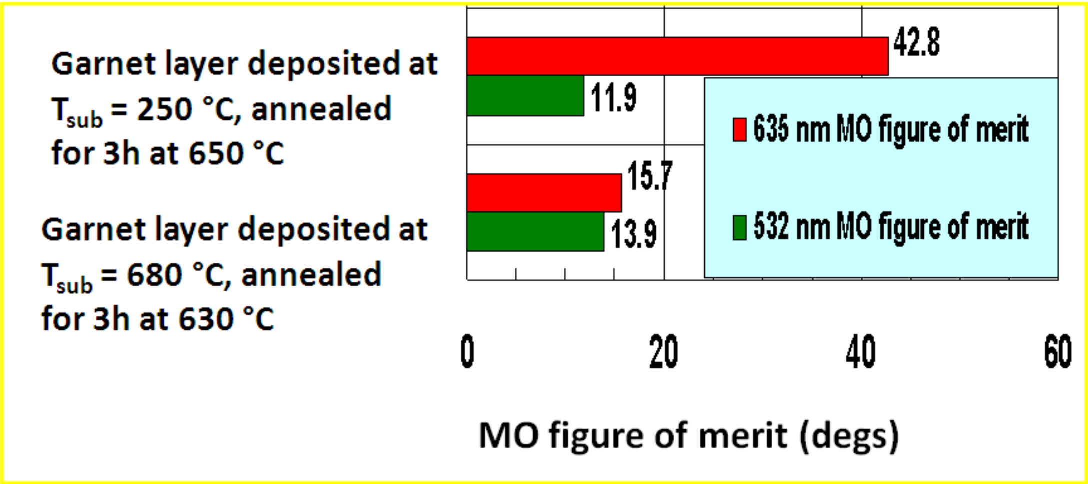

Garnet layer deposited at Tsub = 250 °C, annealed for 3h at 650 °C
Garnet layer deposited at Tsub = 680 °C, annealed for 3h at 630 °C
42.8
11.9
15.7
13.9
635 nm MO figure of merit
532 nm MO figure of merit
0
20
40
60
MO figure of merit (degs)

Fig. 3(b)

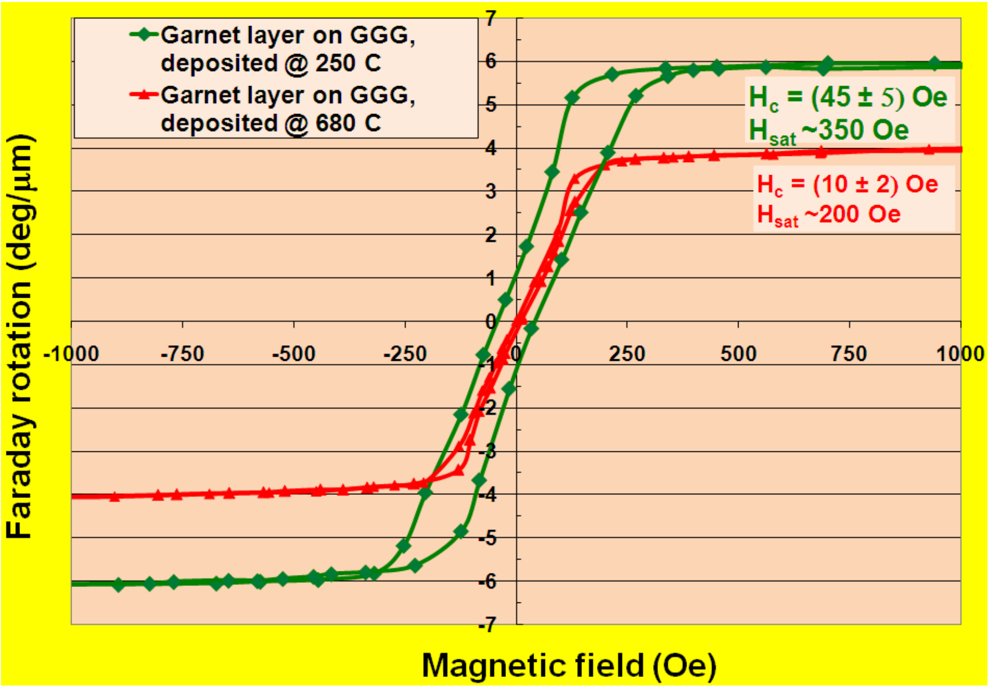
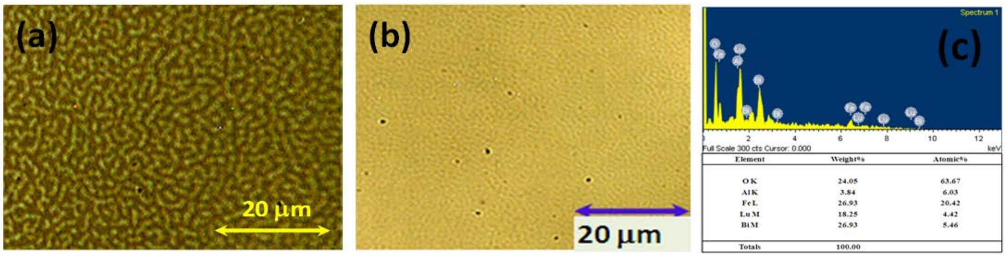


Fig. 4



Element	Weight%	Atomic%
O K	24.05	63.67
Al K	3.84	6.03
Fe L	26.93	20.42
Lu M	18.25	4.42
Bi M	26.93	5.46
Totals	100.00	

Fig. 5

Keywords: MO garnet, thin film, amorphous, annealing, substrates temperature, hysteresis loop, MO domain patterns, MO figure of merit, optical absorption, Faraday rotation.