Armenian Journal of Physics, 2015, vol. 8, issue 1, pp. 21-29

Dependence of Magneto-Optical Properties of Bi-YIG Thin Films on Post-Annealing Temperature

O.A. Galstyan

Institute of Radiophysics and Electronics NAS of Armenia, Ashtarak 0203, Armenia
e-mail: ogsengalstyan@gmail.com

Received 25 December 2014

Abstract - Post-annealing effect on the magneto-optical and structural properties of $Bi_xY_{3-x}Fe_5O_{12}$ (x=1÷3) thin films prepared by MOD method have been investigated. For the deposition of films with x=2÷3 on glass substrate we used $Bi_1Y_2Fe_5O_{12}$ as a buffer layer. The highest Faraday rotation measured for $Bi_2Y_1Fe_5O_{12}$ thin films was about -6.6°/μm at 520 nm wavelength. These films were post-annealed at 620ºC. From X-ray diffractometry data analysis the lattice constant of film was measured to be about 12.5 Å.

Keywords: Garnet film; buffer layer, magneto-optics, Faraday effect, metal-organic decomposition, low-temperature annealing

1. Introduction

Thin films of bismuth-substituted yttrium iron garnet (Bi-YIG) have great potential for magneto-optical (MO) devices, since they have a large Faraday rotation and high transmittance in visible and near-infrared wavelength ranges [1]. It was shown by Hansen *et al.* in Ref. [2] that the Faraday rotation (FR) angle of YIG materials dramatically can be increased with further substitution of Yttrium ions (Y^{3+}) with Bismuth (Bi^{3+}).

So far many different growth techniques such as liquid phase epitaxy (LPE) [3], radio-frequency magnetron sputtering method [4], pulsed laser deposition (PLD) [5], the sol-gel method [6], the metal-organic chemical vapor deposition [7] and the metal-organic decomposition (MOD) method [8] have been used for the production of garnet thin films.

In this work we used MOD method for the deposition of garnet layers with different levels of concentration of Bi doped into the chemical structure of YIG. As a substrate we used amorphous glass substrates. We discussed the influence of post-annealing temperature on magneto-optical and structural properties of films. We found appropriate routines of MOD process to obtain thin films with high FR angles. FR angle is the most important parameter for the MO devices when the films are used as indicators [9]. It is very hard to obtain the crystallization of films for $Bi_xY_{3-x}Fe_5O_{12}$ ($x \geq 2$) layers when they are deposited directly on glass. To overcome this issue we used $Bi_1Y_2Fe_5O_{12}$ as buffer layers [10].

Prepared samples were characterized by their magneto-optical activity and X-ray diffractometry (XRD).

2. Experiment

It is easier to deposit garnet films on lattice matched substrates because morphological instabilities that are caused by misfit stress can be avoided. The use of non-garnet substrates such as glass is very important especially when considering the large size of its fabrication and availability. We succeed in the preparation of $Bi_1Y_2Fe_5O_{12}$ films by MOD method with different thicknesses directly deposited on glass substrates [11]. When concentration of Bi enhances it becomes difficult to obtain films with high quality using glass substrate. One of the techniques that can be applied is the magnetic buffer layer deposition technique [10].

We fabricated four sets of Bi-YIG thin films. Each set represents thin films with specific concentration of doped bismuth ($Bi_xY_{3-x}Fe_5O_{12}$, x=1, 2÷3) that were prepared by using different post-annealing temperatures. Preparation parameters of samples are shown in Table 1.

MOD method starts either with the preparation of MOD solution or by using commercially available one. As substrates we used glass with thickness of about 0.5mm. Buffer layers were used for the films with $x \geq 2$ concentration of doped Bi. We deposited solutions on glass by spin coating at 3000 rpm for 30 seconds. The total concentration of carboxylates was 3%. The deposited solution was dried at 70 °C for 30 minutes. Next step in the process was pre-annealing at 450 °C for 30 min. which could be skipped but as it was shown in ref. [12], it had huge impact

Table 1.Preparation conditions and Faraday rotation values of films.

Set of samples	Buffer layer Bi_xY_{1-x}Fe5O12	Concentration of Bismuth Bi_xY_{1-x}Fe5O12 (x=)	Sample	Post-annealing temperature (°C) and duration (hours)		Faraday rotation (°/μm)
Set 1	No	1	1	600	1	-0.6
			2	620		-0.8
			3	640		-1
			4	660		-1.2
			5	680		-1.6
			6	700		-2
			7	**750**		**-2.5**
Set 2	Yes	2.0	8	600	3	-4.2
			9	**620**		**-6.6**
			10	640		-5.6
			11	660		-5.6
			12	680		-5.5
			13	700		-4.5
			14	750		-4
Set 3	Yes	2.5	15	600	3	-2.4
			16	**620**		**-3.6**
			17	640		-3.3
			18	660		-2.5
			19	680		-2.6
			20	700		-2.4
			21	750		-2
Set 4	Yes	3.0	22	600	3	-
			23	620		-
			24	640		-
			25	660		-
			26	680		-
			27	700		-
			28	750		-

on the quality of films. During pre-annealing carboxylates were decomposed into metal oxides and films were brought to amorphous phase [12]. This cycle of thermal treatments was implemented only once for $Bi_1Y_2Fe_5O_{12}$ buffer layer (see Fig. 1). Afterwards films were post-annealed at 750°C for 1 hour in order to crystallize the buffer layer. Then process of the deposition of the film layer started. $Bi_xY_{3-x}Fe_5O_{12}$ (x=2÷3) solution was deposited, dried and pre-annealed for 10 times to achieve appropriate thickness of the films. Finally films were post-annealed in a furnace at different temperatures (Table 1). All thermal treatments were performed in air. Fig. 1 shows schematic diagram of MOD process. It was measured by cross sectional scanning electron microscope (SEM) that the thickness of the film was about 40 nm for a single coating. The thickness of the prepared films was about 0.4 µm.

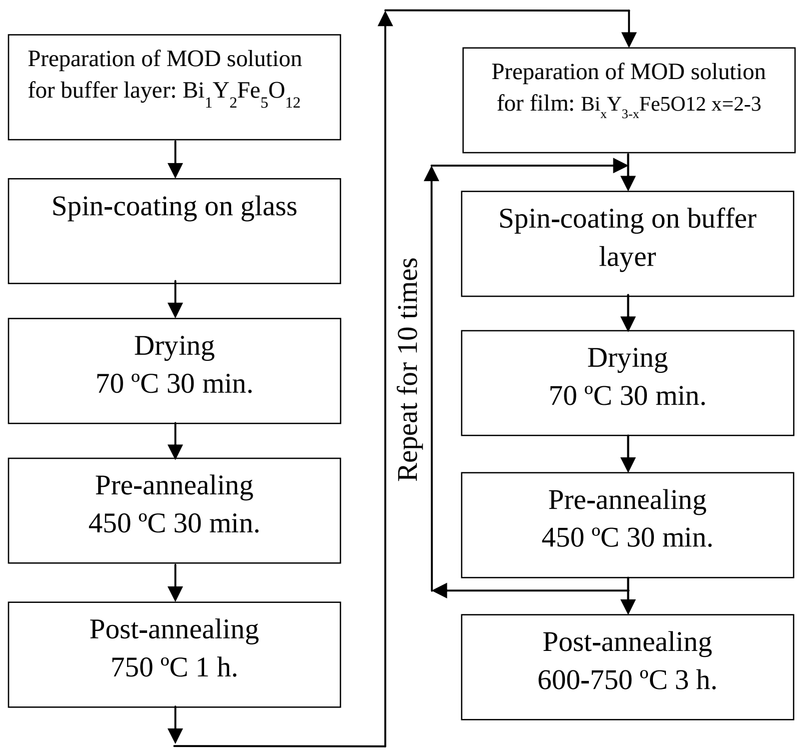

Fig.1. Schematic diagram of MOD process.

Magneto-optical properties of films can be described by Faraday rotation angle measurement. FR occurs when linearly polarized light propagates through magneto-optically active medium under the influence of parallel external magnetic field. A linearly polarized light can be represented as a sum of left and right circularly polarized lights LCP and RCP. FR is the phase difference between the RCP and LCP waves. FR is defined by the expression

$$\theta_F = \frac{\pi(n_+ - n_-)d}{\lambda}, \quad (1)$$

where λ is the wavelength, d is the thickness of a material, $n_\pm$ are refractive indices of left and right circularly polarized lights.

Using photodiode we measured change of light intensity depending on the rotation of polarization plane of light when it propagates in magnetically saturated films. The experimental setup and the technique of measurement of FR was described in our previous publication [10].

To check the crystallinity of films, we used X-ray diffractometry (XRD). Diffraction peaks corresponding to different crystallized phases were observed.

3. Results

Figure 2 presents values of FR for the films with different level of Bi substitution post-annealed at 600÷750°C. For the films with *x*=1 FR increases when they are post-annealed at higher temperatures. The largest FR was measured to be about -2.5°/µm. For the higher levels of Bi substitution (*x*=2, 2.5) the FR curves are different in comparison with *x*=1. We can notice that for *x*=2 and *x*=2.5 FR is increased for the films which were post-annealed at 620°C. But the decrease in FR for *x*=2.5 comparing to *x*=2 is interesting because FR should be increased for higher concentrations of substituted bismuth [2]. We suggest that films with *x*=2.5 are partially garnet which is confirmed by the XRD results. On the other hand, films with *x*=3 showed very weak magneto-optical properties and as it was expected XRD patterns showed no diffraction peaks.Thus, we conclude that *x*=3 films were amorphous.

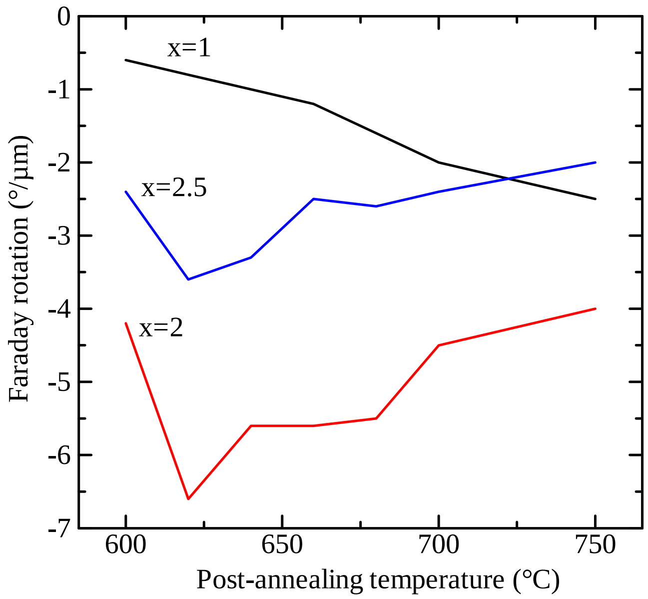

Fig.2. Dependence of FR on the post-annealing temperature of the films with different concentration of substituted Bi (x=1, x=2, x=2.5).

Figure 3 shows XRD patterns of the films that have the highest FR angles in each set. From these patterns we measured lattice parameters of samples. It was shown that by the substitution of Bi in YIG thin films lattice parameter increases. As publications show the lattice parameters are 12.38 Å for YIG and approximately 12.63 Å for fully substituted films BIG [13]. The increase of lattice constant can be explained by the larger ionic radius of Bismuth (Bi^{3+}) comparing to Yttrium (Y^{3+}). By comparing measured data with other publications and theoretical data for lattice constants we can make assumptions about the level of Bi in the crystal structure of films [14].

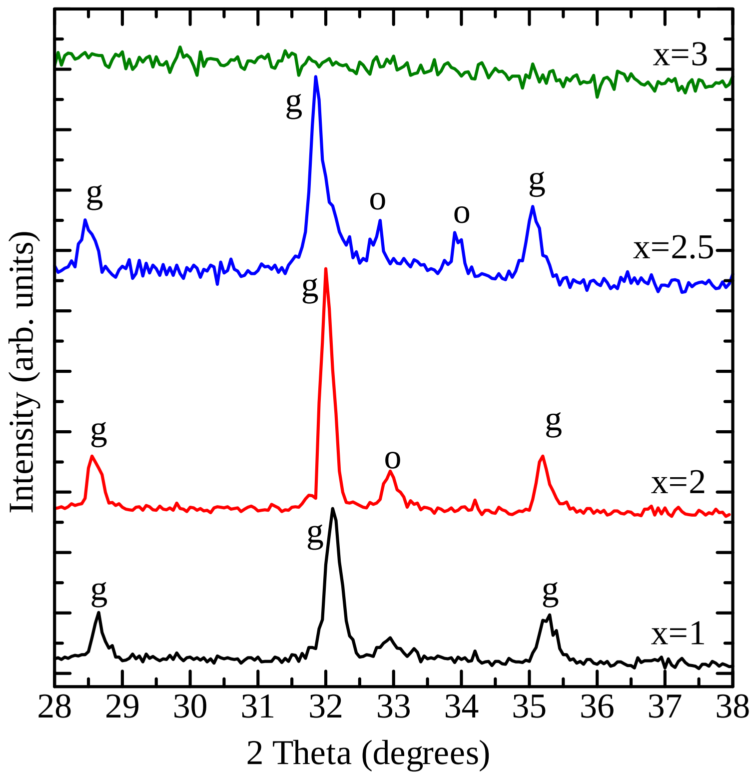

Fig.3. XRD patterns of samples with the highest FR from each set. Assignment of diffraction peaks are indicated as following: g-garnet phase, o-$YFeO_3$.

Samples with *x*=1 show X-ray diffraction peaks corresponding to the garnet phase. Lattice constant was measured to be about 1.245 nm which is in good agreement with data published for garnet thin films prepared by LPE method [2]. Very weak secondary phase which we assigned to Yttrium orthoferrite ($YFeO_3$) appeared for the film with *x*=2 [15]. $YFeO_3$ diffraction peaks became stronger for the film with *x*=2.5. We suggest that $Bi_{2.5}Y_{0.5}Fe_5O_{12}$is not fully crystallized and the concentration of Bi in the crystal structure is lower than 2.5. This suggestion can be confirmed by several results: appearance of $YFeO_3$ phases in XRD pattern, low intensity of the main (420) garnet diffraction peak comparing to the *x*=2, lower value of the lattice parameter comparing to the theoretical and other experimental data and finally low FR comparing to the film with *x*=2. No diffraction peaks appeared in the XRD spectra of *x*=3.We compared lattice parameters calculated from the XRD patterns represented in Fig.3 with data published in [2] which states that the lattice constant of $Bi_xY_{3-x}Fe_5O_{12}$ depends linearly on the content of bismuth. Our results show little decrease in the lattice constants for $Bi_2Y_1Fe_5O_{12}$ (about 0.05 Å) and $Bi_{2.5}Y_{0.5}Fe_5O_{12}$ (about 0.06 Å).

Figure 4 indicates the doped Bi level and post-annealing temperatures for which different phases of crystallization of films are detected by XRD spectrometry. Phases of crystallization were assigned as a result of the discussion which consists of the following 3 parts:

1. Faraday rotation angle value measurement,
2. Assignment of phases appeared in XRD spectra,
3. Calculation and comparison with published data of lattice parameters,

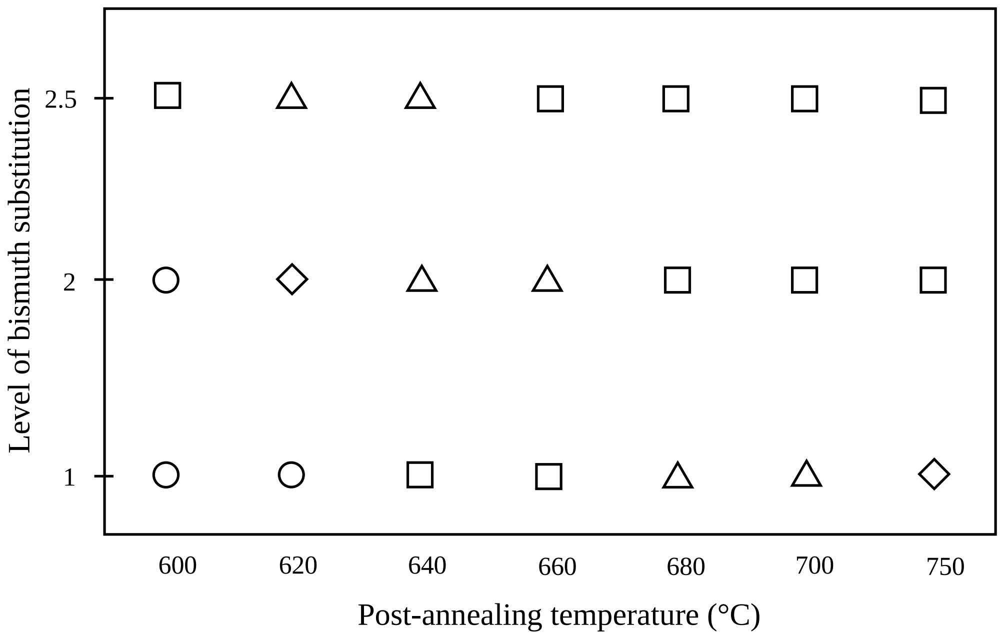

Fig.4. Crystallographic phases of Bi-YIG thin films depending on the level of doped Bi and post-annealing temperature. Assignment of phases is indicated as following: ◊-garnet phase, Δ-garnet partially, □-other phase, o-amorphous.

Figure 4 also shows that for the samples with higher level of substituted bismuth the crystallinity can be improved with a lower temperature of post-annealing (620÷660 °C). Similar behavior of Bi-YIG thin films crystallization was published for the samples prepared with the sol-gel process [6]. We suggest that the increase of FR values came from better crystallinity of films which as we noted became possible with lower temperatures of post-annealing. FR values are in good agreement with intensity of main (420) XRD diffraction peaks. The Faraday Effect of the Bi-YIG thin film is due to the excited-state splitting of the Iron ion and by orbital mixing of the neighboring Bismuth ion Faraday Effect is amplified. Furthermore the orbital mixing of Bismuth ion was increased with better crystallization, which in its turn increases the FR [16, 17].

4. Conclusion

Polycrystalline Bi-YIG thin films were prepared by MOD method. Buffer layer deposition method was used to prepare films on glass substrates for films with high level of bismuth substitution. Magneto-optical measurements showed that FR for films with x=1 can be increased with high post-annealing temperatures (about 750 °C). For films with x=2 and x=2.5 FR increased when films were post-annealed at lower temperatures (about 620 °C). From XRD measurements we concluded that films with highest FR showed better crystallinity of garnet phase. Lattice parameters of films with x=1, 2 and 2.5 were measured to be 12.45Å, 12.5 Å and 12.54Å, respectively. The increase in the lattice parameter with a substitution level of bismuth is in agreement with previous publications for garnet films prepared with other methods.

References

1. T. Ishibashi, Z. Kuang, S. Yufune, T. Kawata, M. Oda, T. Tani, Y. Iimura, K. Sato, J. Appl. Phys. 100 (2006) 093903.
2. P. Hansen, K. Witter, W. Tolksdorf, Phys. Rev. B, 27 (1983) 6608.
3. T. Aichele, A. Lorenz, R. Hergt, P. Görnert, Cryst. Res. Technol 38 (2003) 575.
4. T. Boudiar, B. Payet-Gervy M.-F. Blanc-Mignon, J.-J. Rousseau and M. Le Berre H. Joisten, J. Magn. Magn. Mater. 284 (2004) 77.
5. T. Tepper, C.A. Ross, J. Cryst. Growth 255 (2003) 324.
6. K. Matsumoto, K. Yamaguchi, A. Ueno and T. Fujii, IEEE Transl J MagnJpn, 6 (1991) 15.
7. M. Okada, S. Katayama and Koji Tominaga, J. Appl. Phys. 69 (1991) 3566.
8. A. Azevedo, S. Bharthulwar, W. Eppler, M. Kryder, IEEE Trans. Magn. 30 (1994) 4416.

9. R. M. Grechishkin , M. Yu. Goosev, S. E. Ilyashenko, N. S. Neustroev, J. Magn. Magn. Mater. 157 (1996) 305.
10. T. Ishibashi, T. Yoshida, T. Kobayashi, S. Ikehara, T. Nishi, J. Appl. Phys. (2013) 17A926.
11. O. Galstyan, H. Lee, S. Lee, N. Yoo, J. Park, A. Babajanyan, B. Friedman, K. Lee, J. Magn. Magn. Mater. 366 (2014) 21.
12. H. Lee, Y. Yoon, S. Kim, H. K. Yoo, H. Melikyan, E. Danielyan, A. Babajanyan, T. Ishibashi, B. Friedman, K. Lee, J. Cryst. Growth 329 (2011) 27.
13. SörenKahl, Bismuth iron garnet films for magneto-optical photonic crystals, [dissertation]. Stockholm, Royal Institute of Technology, (2004).
14. T.Ishibashi, T. Kosaka, M. Naganuma and T. Nomura, Journal of Physics: Conference Series 200 (2010) 112002.
15. D. S. Schmool, N. Keller, M. Guyot, R. Krishnan, M. Tessier, J. Appl. Phys. 86 (1999) 5712.
16. H. Lee, T. Kim, S. Kim, Y. Yoon, S. Kim, A. Babajanyan, T. Ishibashi, B. Friedman, and K. Lee, J. Magn. Magn. Mater. 322, (2010) 2722.
17. S. Wittekoek, T. Popma, J. Robertson, P. Bongers, Phys. Rev. B 12 (1975) 2777.