Reprinted from Ferrites: Proceedings of The Sixth International Conference on Ferrites (ICF 6)
Tokyo and Kyoto, Japan 1992

PROPERTIES OF NONSTOICHIOMETRIC Bi-YIG SPUTTERED FILMS

E. Komuro, T. Hirano*, T. Namikawa and Y. Yamazaki

Tokyo Institute of Technology, Nagatsuta, Midori-ku, Yokohama 227, Japan

*TOPPAN Printing Co., Ltd., Sugito, Saitama 345, Japan

Abstract- Bi-YIG films are prepared by rf-sputtering using targets, which have various Fe contents, to investigate the range in which garnet structure is obtained. Annealing temperature is varied from 500 to 1000°C, and X-ray diffraction patterns and M_s are measured for each temperature. In the wide range of Fe content from about 3.9 to 5.4, the garnet structure can be obtained at the annealing temperature of 650°C. For the Fe content less than 5, M_s increases linearly, and drops suddenly for the Fe content over 5. According to θ_F/M_s value, the film of Fe content 3.9 contains higher content of Y in the garnet structure than other high θ_F/M_s films. The lack of Bi in the garnet structure at the Fe content 3.9 film weakens the enhancing Faraday rotation.
KEYWORDS: Bi-YIG, GARNET, NONSTOICHIOMETRIC COMPOSITION, ANNEALING TEMPERATURE, FARADAY ROTATION

I. INTRODUCTION

Bismuth substituted iron garnet films are very attractive material for magneto-optical applications. It was reported that in spite of the considerable deviation from the stoichiometric composition of garnet, a single phase of garnet structure could be obtained by sputtering method[1][2]. It is worthwhile to determine the composition range in which the garnet structure can be obtained, and to clarify the film structure in order to get high quality films for the use as magneto-optical media.

In this paper, we prepare Bi-YIG sputtered films using targets which have various Fe contents, and analyze the structural , magnetic and magneto-optical properties of the films.

II. EXPERIMENTAL

The films were prepared on vitreous SiO_2 and Corning 0317 glass substrates by rf-sputtering under the conditions given in Table 1. The targets were prepared first by mixing oxide powders, then it was fired at 800°C for 4h and ground with a mill. Then it was pressed at 200kg/cm^2 to form a disk. The disk diameter of the target was 100mm and the spacing between the target and the substrate was 40mm. The as-deposited amorphous films were annealed in air at 500-1000°C for 4h to crystallize the films. Annealed films were examined by X-ray diffraction with Cu-Kα source. The film thickness was measured using a surface step analyzer (DEKTAK). The chemical composition of the prepared films was determined by inductively coupled plasma spectroscopy. Saturation magnetization was measured using a VSM. The magnetic field was applied up to 2kOe. The Faraday rotations were measured by the polarization modulation method. These optical measurement was carried out in visible wavelength region.

Table 1 Sputtering conditions

Target	$Bi_2YFe_xO_{4.5+1.5x}$ (x=1, 2, 3, 4, 5, 6)
Sputter gas	Ar, 6.7 Pa
Substrate	Corning 0317, SiO_2(Vitreous)
Substrate temperature	400°C
rf power density	2.5W/cm^2
Deposition rate	4.3nm/min

III. RESULTS AND DISCUSSION

Film composition

Film compositions are given in Table 2. We show two kinds of film composition in Table 2. One is that we presume the sum of cation in films is equal to the sum of cation in targets(a). Comparing the film composition (a) with target composition, content of the Y in the films is more than that of the targets, while the Bi content is less than that of the targets. But the contents of Bi and Y in the films are almost constant, and the Fe content in the films is a little more than the Fe content of targets. The other is that we assume cation : anion = 8 : 12 (b), because the ratio taken is for garnet structure. We will use (b) as the film composition in this paper.

Table 2 Film compositions

Sample No.	Target			Film						
				(a)			(b)			(Bi+Y)/Fe
	Bi	Y	Fe	Bi	Y	Fe	Bi	Y	Fe	
1	2	1	1	1.7	1.3	1.0	3.4	2.7	1.9	3.21
2	2	1	2	1.4	1.2	2.4	2.2	1.9	3.9	1.05
3	2	1	3	1.7	1.0	3.3	2.2	1.4	4.4	0.82
4	2	1	4	1.5	1.1	4.4	1.7	1.2	5.1	0.57
5	2	1	5	1.5	1.1	5.4	1.6	1.0	5.4	0.48
6	2	1	6	1.5	1.0	6.5	1.3	0.9	5.8	0.38

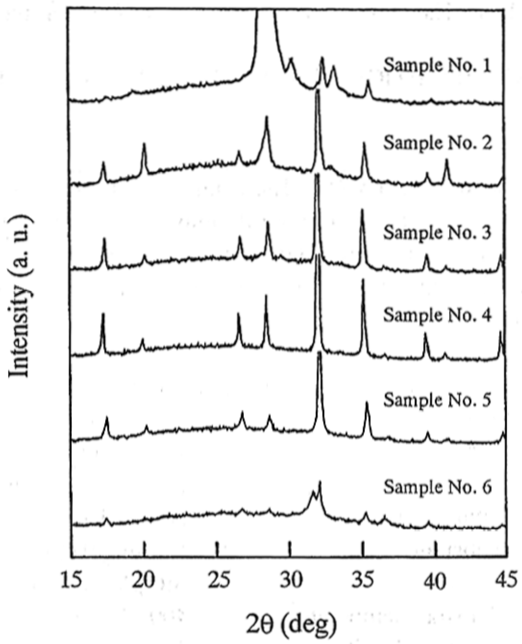

Fig.1 X-ray diffraction patterns of the films for various Fe contents.

The values of (Bi+Y)/Fe are 0.38–3.21, indicating considerable deviation from stoichiometric garnet (3/5=0.6) for almost all samples.

Film structure

The X-ray diffraction patterns of the samples are shown in Fig. 1, where 2θ is scanned from 15° to 45°. Sample number in Fig. 1 is consistent with that in Table 2. All samples were annealed at 650°C for 4h. For Sample 1, garnet peaks are scarcely observed. For Sample 2, while a impurity phase appears a little, almost all peaks are assigned to garnet. All peaks of Sample 3, 4 and 5 are assigned to garnet. Then, for Sample 6, the intensity of garnet peaks becomes weak and a impurity phase appears. In the wide range of Fe content from about 3.9 to 5.4, the garnet structure can be obtained at the annealing temperature of 650°C.

At the annealing temperature of 500°C, Sample 2, 3, 4 and 5 had no X-ray diffraction peaks of garnet. At 600°C, garnet peaks appeared in Sample 2, 3 and 4. And Sample 5 had a garnet diffraction pattern at 650°C. A side-band appeared at 700°C for Sample 2 and 3, and at 800°C for Sample 4 and 5. The diffraction patterns of all samples show a impurity phase for the films annealed over 800°C. From these results, for the films which had the compositions far from the stoichiometric garnet like Sample 2 and 3, a impurity phase appeared at lower temperature than the films which had the composition near the stoichiometric garnet like Sample 4. The excess Bi or Y seemed to form other phases.

Magnetic properties

Figure 2 shows the saturation magnetization of the films as a function of the annealing temperature. M_s appears at 600 or 650°C and disappears at 900°C for all samples. In the region of annealing temperature 600 to 800°C, M_s decreases gradually for Sample 2, 3, 4 and 5. Considering the result of X-ray diffraction patterns, garnet structure decomposes and other non-magnetic phases are formed as annealing temperature increases.

The variations of the M_s of the films annealed at 650°C are shown as a function of Fe content in Fig. 3. Dotted line represents the M_s of bulk YIG (140 emu/cm^3). Dashed-dotted line indicates the calculated value assuming that the M_s is proportional to Fe content. The calculated values agree with the measured values for the Fe content less than 5. And the M_s drops suddenly for the Fe content over 5. It is presumed that M_s increases in proportion to garnet volume until Fe content 5, and garnet structure decomposes suddenly over Fe content 5.

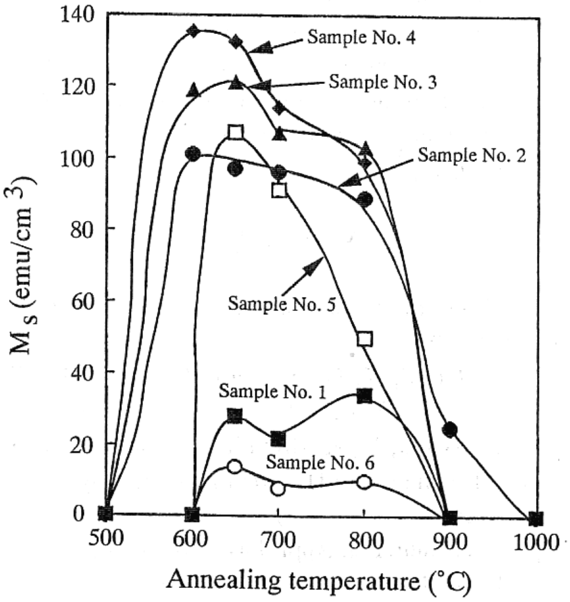

Fig.2 Magnetization of the films as a function of annealing temperature.

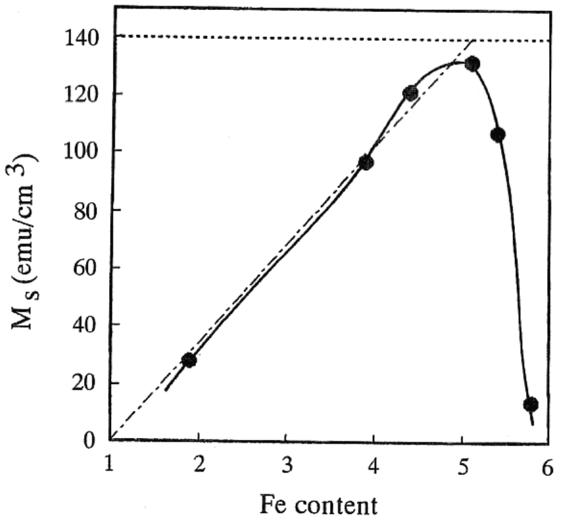

Fig.3 Saturation magnetization for the films having various Fe content annealed at 650°C.

Magneto-optical properties

The Faraday rotations measured at 520nm are shown in Fig. 4. The curve in Fig. 4 is similar to that in Fig. 3. By comparing the film of Fe content 3.9 and the film of Fe content 5.4, though these two films show almost the same value of M_s, the value of Faraday rotation for the film having Fe content 5.4 is about twice that for the film having Fe content 3.9.

Figure 5 shows θ_F/M_s as a function of Fe content. In the region of Fe content 4.4 to 5.4, the values of θ_F/M_s are constant. At Fe content 3.9, the θ_F/M_s is half

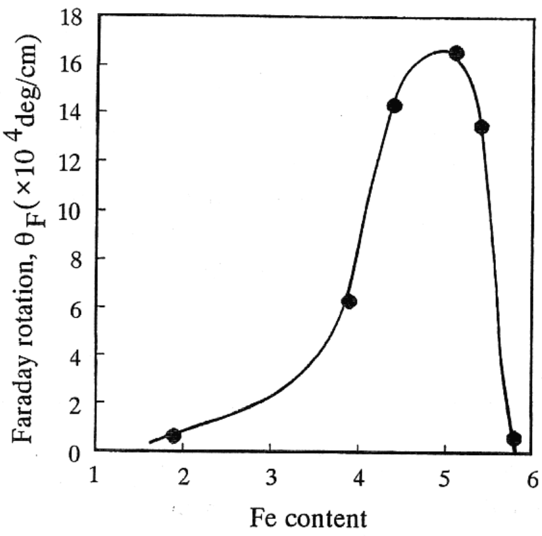

Fig.4 Faraday rotation of the films for various Fe content annealed at 650°C, measured at 520nm.

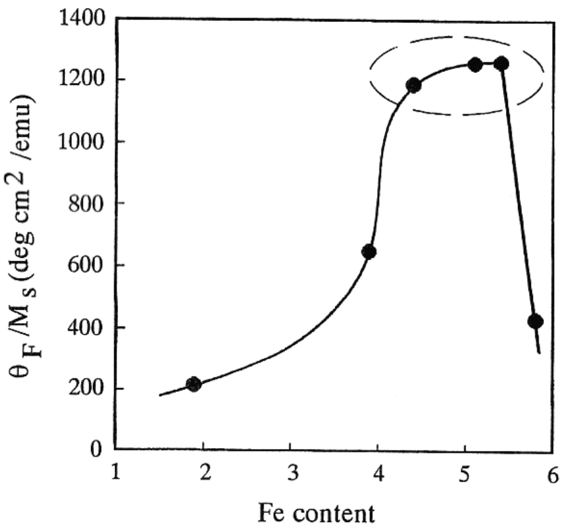

Fig.5 θ_F/M_s vs. Fe content for the films annealed at 650°C.

of the constant value in the region of Fe content 4.4 to 5.4. It is considered that in the film of Fe content 3.9, Bi and Y are contained in amorphous phase in the films. Y is incorporated more stably in the garnet structure than Bi because a special technique must be used to prepare highly Bi substituted YIG[3].

It can be assumed that there were no other magnetic phases except garnet phase in the films. Therefore, the value of θ_F/M_s represents the magnitude of Faraday rotation of the garnet phase formed in the films. It is noted that the value of θ_F/M_s for the film having Fe content 3.9 is lower than the value expected from the relatively large value of M_s shown in Fig. 3. On the other hand, the value of θ_F/M_s for the film having Fe content 5.4 is higher than the value expected from the M_s shown in Fig.3, where the Fe content exceeds the value of 5.1 at which the M_s and θ_F show the maximum values. Considering that Faraday rotation is much enhanced by Bi[4][5], this result suggests that the garnet phase formed in the films having the Fe content 3.9, contains less Bi ions than the garnet phase formed in the films having the Fe content around 5.

IV. CONCLUSIONS

Aiming at determination of the composition range in which the garnet structure is obtained, we prepared the films by rf–sputtering using $Bi_2YFe_xO_{4.5+1.5x}$ targets. Through the magnetic and magneto–optical measurements on various Bi–YIG sputtered films, the following results were obtained. In the wide range of Fe content from about 3.9 to 5.4, the garnet structure could be obtained at the annealing temperature of 650°C. It is presumed that M_s increases in proportion to the volume having garnet phase until Fe content of 5, and garnet structure decomposes suddenly for the Fe content over 5. All Bi were not incorporated in garnet phase as for the film having Fe content of 3.9, resulting in the small value of θ_F/M_s.

V. REFERENCES

[1] M. Gomi, T. Tanida and M. Abe: J. Appl. Phys., **57**, 3888(1985).
[2] M. Gomi, K. Utsugi and M. Abe: IEEE Trans. Magn., **MAG–22**, 1233(1986).
[3] T. Okuda, N. Koshizuka, K. Hayashi, T. Takahashi, H. Kotani and H. Yamamoto: J. Mag. Soc. Jpn., **11**, 147(1987).
[4] C. F. Buhrer: J. Appl. Phys., **40**, 4500(1969).
[5] J. M. Robertson, S. Wittekoek, Th. J. A. Pompa and P. F. Bongers: Appl. Phys., **2**, 219(1973).