

# Magneto-optical Indicator Garnet Films Grown by Metal-organic Decomposition Method

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Bi-substituted yttrium iron garnet,  $Y_{3-x}Bi_xFe_5O_{12}$  (Bi:YIG,  $x = 1, 1.5$ ), films were prepared on  $Gd_3Ga_5O_{12}$  (111) substrates by metal-organic decomposition (MOD) method to use as MO indicator films. Those films had the in-plane magnetic anisotropy and Faraday rotation as large as that of the single crystal and do not show prominent magnetic domain structure, which is often observed in single crystalline garnet films grown by liquid phase epitaxy method. Disappearance of magnetic domain structure can be attributed to a granular structure with a grain size of  $\sim 50$  nm in Bi:YIG thin films. The characteristics are suitable for MO indicator films to visualize a stray magnetic field strayed from a specimen. In this paper, MO imaging of Nb films with groove patterns is demonstrated.

**Key words:** magneto-optical imaging, magneto optical indicator film, Bi-substituted garnet, superconductor

## 1. Introduction

Magnetic imaging technique utilizing magneto-optical (MO) effect has been attracting attentions because it has comprehensive advantages of a fast measurement, a high resolution, a wide range of operating temperature, a quantitative evaluation of magnetic fluxes, etc., compared with recently developed magnetic microscopes such as a magnetic force microscope, a SQUID microscope etc.<sup>1)</sup> Bi substituted rare-earth iron garnet films have been used as an MO indicator film that makes magnetic flux distribution visible by utilizing its huge Faraday rotation in the visible to infrared range. In addition to a high sensitivity of approximately 1 Oe, to obtain a high resolution that is high enough to be within the optical limit in MO imaging, high quality garnet thin films with a thickness as thin as 1  $\mu$ m are required, since the magnetic flux generated by the samples rapidly diminishes with distance from the samples<sup>2)</sup>. Moreover, the zigzag-shaped magnetic domain structure often observed in single crystalline garnet films is a problem which should be solved, because it disturbs the MO observation, as discussed later.

Bi substituted garnet films have been prepared by several techniques, such as a liquid phase epitaxy (LPE)<sup>3)</sup>, an RF magnetron sputtering<sup>4)</sup> and a thermal decomposition method<sup>5-8)</sup>, etc. In those technique, MOD methods are advantageous not only for the homogeneity of the thin film, the controllability of composition, and the formation over a large area, but also for the good productivity, since MOD solutions have good stability for more than several years<sup>9)</sup>. In this paper, we report on Bi-substitute yttrium iron garnet thin films for MO indicator films without zigzag-shaped magnetic domains by a metal organic decomposition (MOD) method.

## 2. Experiments

### 2.1 MOD process

MOD liquids used in this experiment, which are consisting of solutions made from Bi, Y and Fe carboxylates with desired chemical compositions for  $Y_{3-x}Bi_xFe_5O_{12}$  (Bi:YIG,  $x = 1.0$  and  $1.5$ ), were made by Kojundo Chemical Laboratory Ltd. The total concentration of carboxylates in those MOD solutions was fixed at 3%. It should be noted that these MOD solutions are stable and precipitation or other changes have not been observed over two years.

These solutions were spin-coated on the gadolinium gallium garnet,  $Gd_3Ga_5O_{12}$ , (111) substrates in a 2 step process: 500 rpm for 5 s and 3000 rpm for 30 s, followed by drying at 150°C for 2 - 60 min using a hot-plate. In order to decompose organic materials and to obtain amorphous oxide films, samples were pre-annealed at 450°C for 15 min. These processes, i.e. spin-coating, drying, and pre-annealing, were repeated to obtain designed thickness. The thickness of films obtained in the present study was 200 - 800 nm. Finally, the precursor films were annealed for crystallization in a furnace at 500 - 800°C for 1-3 hour. All thermal treatments were performed in air. Detail of garnet films by MOD method have been described in Ref.9.

### 2.2 MO measurement

Values of Faraday rotation were evaluated by MO microscope using a polarization modulation technique<sup>10)</sup>, while MO images shown in this paper were measured with a conventional crossed polarized technique. Halogen lamp was used as a light source, and a wavelength of 500 nm was selected using an interference filter.

MO images of superconducting Nb films with groove patterns were measured by the MO microscope

using the Bi:YIG film directly put on the sample placed in a cryostat combined with an electro-magnet. The Bi:YIG film with Bi composition of  $x = 1.0$  and a thickness of 800 nm was used in this measurement. Pt mirror layer with a thickness of 100 nm was deposited on the garnet film by a magnetron sputtering method in order to obtain high reflectivity and to avoid scattering of the light due to the pattern fabricated on the sample. The Nb samples of 0.9 mm  $\times$  0.9 mm in size with groove patterns with a width of 2  $\mu$ m, a pitch of 4  $\mu$ m and a depth of 0.3 – 0.4  $\mu$ m were prepared by photolithography and chemical etching technique<sup>11</sup>.

### 3. Results and Discussion

#### 3.1 MO properties of garnet films

Figure 1 shows Faraday rotation spectrum of a 400 nm-thick Bi:YIG thin film with  $x = 1.0$  prepared on a GGG (111) substrate. This spectrum was measured with a magnetic field of 1.7 T. The spectral feature in Fig.1 is in good agreement with those reported for single crystals<sup>12</sup>.

Figure 2 shows a magnetic field dependence of Faraday rotation of Bi:YIG films with mirror layer measured by MO microscope<sup>3</sup> from substrate side with a light with a wavelength of 500 nm. It shows that the film has an easy-axis of magnetization in plane and reaches a saturation value at the magnetic field of  $\sim 1$  kOe for both cases of  $x = 1.0$  and 1.5. The saturation values of Faraday rotation depends on annealing temperature for the crystallization in MOD process, and those for  $x = 1.5$  are larger than those for  $x = 1.0$ , as shown in Fig.3. No magnetic domain structures were observed in the MO images, in spite of the fact that the Bi:YIG films are of good crystalline quality with well-defined [111] orientation and an appreciable Faraday rotation. We considered that absence of magnetic domain structure in the observed MO images is due to size of the magnetic domains which is smaller than optical resolution. We therefore measured the surface morphology using an atomic force microscope (AFM). As shown in the AFM image of Fig.4, the Bi:YIG thin film has a granular like structure with a grain size of  $\sim 50$  nm as we expected. Although the size of magnetic domain has not been elucidated by magnetic force microscopy, we suppose that it might be as small as the grain size, less than the optical resolution of our microscope. This property may be nonmagnetic materials in the grain boundary formed during the MOD process. However, in order to find the reason and control the property, further study is required.

Bi:YIG prepared on GGG (111) substrate usually has an easy-axis of magnetization perpendicular to the plane because the magnetic anisotropy due to the magneto-crystalline anisotropy with the easy axis of magnetization along [111] as well as to stresses induced by the lattice mismatch and the difference in the thermal expansion ratios between the film and the substrate. In the case of our samples, it is considered that the anisotropy due to stresses is small, because

above-mentioned stresses are released by taking the three dimensional growth mode of the Bi:YIG thin film, resulting in the granular-like structure. Thus, the easy axis of magnetization lies along in-plane direction due to a demagnetizing field of the Bi:YIG thin film despite its crystal anisotropy along [111] direction. This is the reason why the Bi:YIG thin film on GGG (111) substrate by the MOD method has the easy axis of magnetization in plane direction.

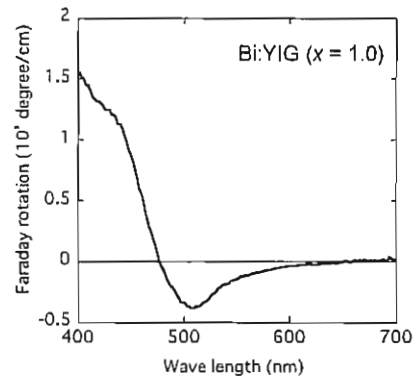


Fig.1 Faraday rotation spectrum of Bi:YIG film ( $x = 1.0$ ) on GGG substrate.

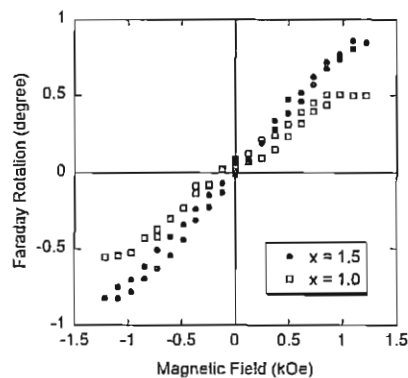


Fig. 2 Magnetic field dependences of Faraday rotation of  $Y_{3-x}Bi_xFe_5O_{12}$  films measured with  $\lambda = 500$  nm.

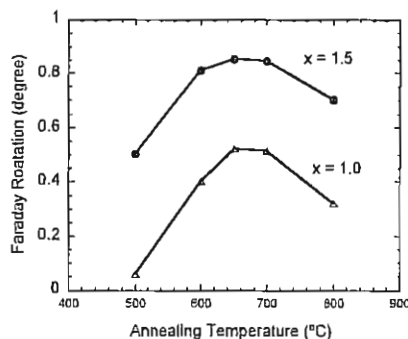


Fig. 3 Annealing temperature dependences of Faraday rotation of  $Y_{3-x}Bi_xFe_5O_{12}$  films measured with  $\lambda = 500$  nm.

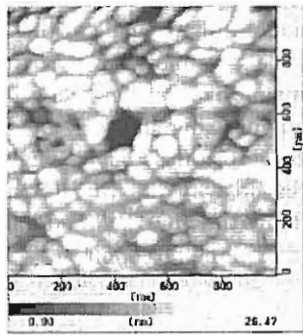


Fig. 4 AFM image of  $Y_2BiFe_5O_{12}$  films with thickness of 400 nm.

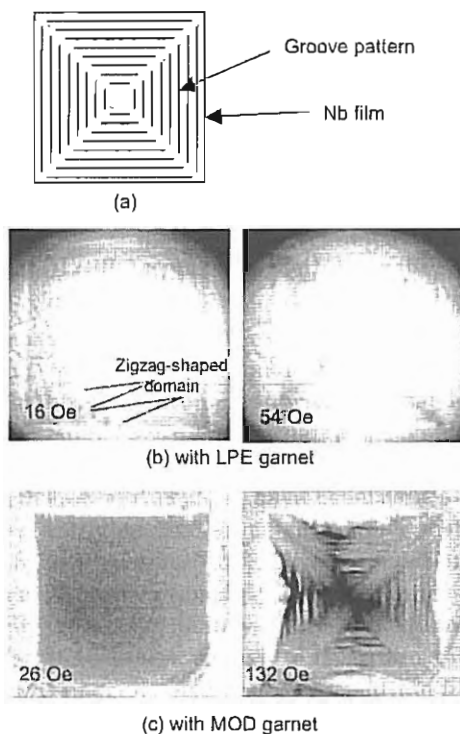


Fig. 5 (a) A schematic drawing and (b),(c) MO images of Nb film with groove patterns. A size of the Nb film is  $0.9 \text{ mm} \times 0.9 \text{ mm}$ , and a pitch of groove with a width of  $4 \mu\text{m}$  is  $4 \mu\text{m}$ . (b) and (c) were measured by using the LPE-grown and the MOD-grown garnet, respectively.

Noted that these properties, the thickness less than  $1 \mu\text{m}$ , the easy axis of magnetization in-plane and the invisible small magnetic domain size, are suitable for the MO indicator film.

### 3.2 MO imaging of Nb film

MO images of the Nb films with fine groove patterns were measured and compared with those measured with a LPE-grown single crystalline garnet film. The Nb sample with groove patterns is considered to be suitable to check the property of the MO indicator films because complicated magnetic flux

distribution is expected due to its groove structure.

Figure 5(a) shows a schematic drawing of the Nb sample and Figs. 5(b) and 5(c) are MO images measured with applied magnetic field indicated in those figures at a temperatures of 4 K which is sufficiently lower than the critical temperature of the Nb film,  $\sim 9 \text{ K}$ . Figures 5(b) and 5(c) show MO images measured with the LPE-grown and the MOD-grown garnet, respectively. Thickness of the LPE grown garnet and the MOD grown garnet used in this experiment is  $2 \mu\text{m}$  and  $800 \text{ nm}$ , respectively. In the case of the LPE grown garnet film, zigzag shaped magnetic domains appeared just after the beginning of the flux penetration into the Nb film persisted even in higher applied magnetic field, thus it makes difficult to recognize MO signals due to stray field from the Nb sample, although the magnetic sensitivity of the  $2 \mu\text{m}$ -thick LPE garnet is at least two times higher than that of the  $800 \text{ nm}$ -thick MOD garnet. On the other hand, a complicated flux structure in Nb film with strong fluxes penetrating perpendicular to the edges of the Nb film and weak fluxes along grooves is clearly observed, since no zigzag domain appeared with any applied magnetic fields.

## 4. Conclusion

Bi:YIG films with  $x = 1.0$  and  $1.5$  were prepared by the MOD method. MO properties of Bi:YIG films grown on GGG (111) substrates were investigated, and it was found that the easy axis lies along in-plane direction and no magnetic domain structure is observed in MO imaging. These properties are suitable as an MO indicator film.

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