

## Enhancement of Magneto-Optic Figure of Merit of Bi:YIG Films in Blue Wavelengths by La Substitution

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This paper reports the first success in enhancement of the magneto-optic figure of merit of garnet films in blue wavelengths by La substitution. This has been achieved through the increase of the Faraday rotation and the reduction of optical absorption in the wavelengths. The maximum of the absolute value of the Faraday rotation angle observed at about 500 nm for the films with no La substitution shifted toward shorter wavelength as La content increased, that leads the increase of the absolute value of the Faraday rotation angle in the wavelengths shorter than about 480 nm. The optical absorption coefficient at the wavelengths shorter than 500 nm decreased as La content increased. As a result the magneto-optic figure of merit increased in the wavelengths shorter than 470 nm as La content increased and was about two times larger for the film with La content of 0.6 in formula unit compared to that of the film without La substitution. This might be attributed to the shift of the centers of the charge transfer and crystal field transitions by La substitution.

### I. Introduction

Because magneto-optic recording employs diffraction limited optics, the recording density is inversely proportional to the wavelength of light source. For example, if one uses a blue laser with wavelength of 400 nm as a light source instead of the infrared semiconductor laser of 800 nm currently being used, one can increase the areal recording density by a factor of four. Therefore efforts have been made in both sides of light source and media to realize blue-wavelength recording.[1]

In the field of media, however, materials developed to date such as amorphous rare-earth transition metal alloy, platinum cobalt multilayer, and garnet films do not exhibit large enough magneto-optic figure of merit in blue wavelengths.[1]

In this paper, we reports the first success in enhancement of magneto-optic figure of merit (FOM) of garnet films, which is defined by [1]  $\theta_F \exp(-\alpha d)$ , where  $\theta_F$  is the Faraday rotation angle,  $\alpha$  the optical absorption coefficient, and  $d$  the

film thickness, in blue-wavelengths (400 - 500 nm) by La substitution. This has been achieved through the increase in the Faraday rotation and the reduction in the optical absorption coefficient by La substitution.

### II. Experiments

Poly-crystalline La-substituted Bi:YIG films with chemical compositions of  $\text{La}_x\text{Bi}_{0.5}\text{Y}_{2.5-x}\text{Fe}_4\text{AlO}_{12}$  ( $x = 0, 0.2, 0.4, 0.6, 0.8, 1.0, 1.2$ ) have been prepared on glass (Corning 7059) substrates (45x45x1 mm<sup>3</sup>) by pyrolysis. Metal nitrates and distilled water were used for solutes and solvent, respectively, to prepare coating solutions of 0.2 mole garnet/liter. The substrates were plasma pretreated just before spinning the solution to improve wetting. Details of the film preparation were discussed elsewhere. [2,3] The films obtained were about 0.3  $\mu\text{m}$  thick.

The crystal structure, magnetic, optical and magneto-optical properties were investigated at room temperature by XRD, VSM with applied field of 20 kOe, optical

spectrometry and magneto-optical spectrometry every 25 nm in the wavelengths ranging 400 - 800 nm, respectively.

### III. Results and Discussion

#### A. Crystal structure

The films obtained were single phase garnets as shown in Fig.1. Figure 2 shows lattice constants (closed squares) obtained from the XRD diagrams, that linearly increase as La content increases. The solid line in the figure is calculated values using the lattice constant (1.2376 nm) of YIG and that (1.2767 nm) of LaIG ( $\text{La}_3\text{Fe}_5\text{O}_{12}$ ). [4] The calculation was performed by applying the Vegard's law,

$$a(x) = a(0)(a_{\text{LaIG}} - a_{\text{YIG}})(x/3)$$

where  $a(x)$  is the lattice constant of the film with La content of  $x$  in formula unit ( $\text{La}_x\text{Bi}_{0.5}\text{Y}_{2.5-x}\text{Fe}_4\text{AlO}_{12}$ ) to be calculated, and  $a(0)$  is the experimentally measured lattice constant of the film with no La substitution ( $\text{Bi}_{0.5}\text{Y}_{2.5}\text{Fe}_4\text{AlO}_{12}$ ).  $a_{\text{LaIG}}$  and  $a_{\text{YIG}}$  are the above literature values of the lattice constants of YIG and LaIG. The factor  $(x/3)$  was multiplied because the increase in lattice constant by an amount of  $(a_{\text{LaIG}} - a_{\text{YIG}})$  corresponds to  $x = 3$ . From the fact that the experimentally obtained values agree well with the calculated values, La ions are thought to substitute Y ions located in the dodecahedral sites. [4]

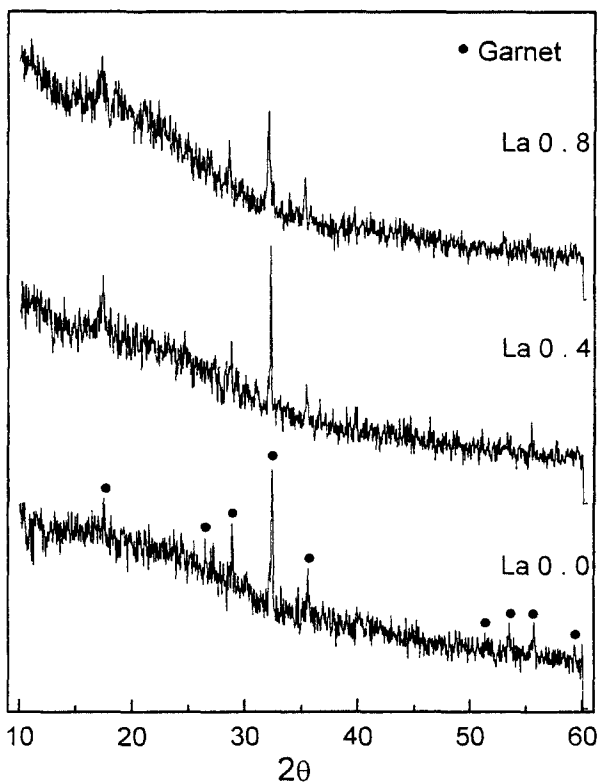


Figure 1. XRD diagrams of La-substituted Bi:YIG films.

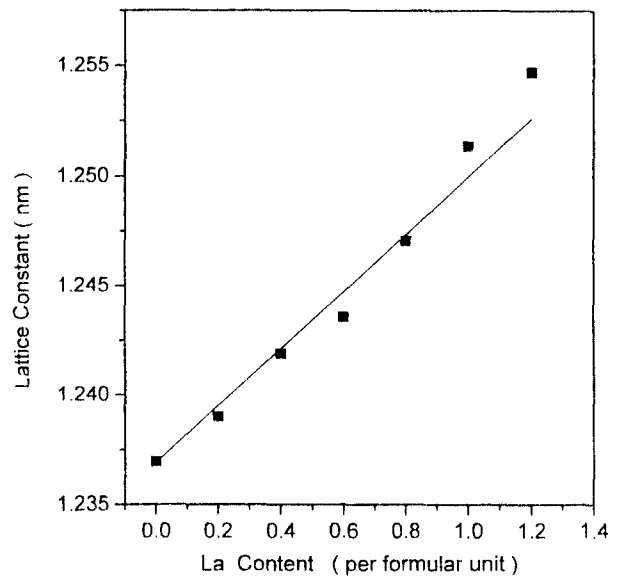


Figure 2. Lattice constants of La-substituted Bi:YIG films.

#### B. Optical and magneto-optical properties

Figure 3 shows the optical absorption coefficient spectra of the films obtained from transmittance spectra by ignoring surface reflection as a function of La content. As shown the absorption coefficient in the wavelength range of 400 - 500 nm decreases as La content increases. This might be due to the shift of the centers of the charge transfer transitions toward shorter wavelengths as follows.

The optical absorption of magnetic garnets in the visible spectrum is mainly caused by the charge transfer transitions centered at ultraviolet region whose tails are extended up to visible spectrum. [5] The charge transfer transitions are the electronic transitions between the 2p orbit of ligand  $\text{O}^{2-}$  ion and the 3d orbit of  $\text{Fe}^{3+}$  ion. [6] (There is another theory that the charge transfer transitions are caused by the sublattice transitions between octahedral and tetrahedral  $\text{Fe}^{3+}$  ions. [5])

Because the charge transfer transitions are the transitions between the valence and conduction bands, if one increases the band gap, one can shift the centers and hence the tails of the transitions toward shorter wavelengths and decrease the absorption in the visible spectrum. One of the methods to increase the band gap would be to increase the ionic character of the bonds in garnets. The fractional ionic character of a bond in garnets can be given by [7]

$$f = 1 - \exp\left\{-\frac{1}{4}(X_A - X_B)^2\right\}$$

where  $X_A$  and  $X_B$  are the electronegativities of cation and anion involved. Since the electronegativity difference (2.34) in La-O bond is predicted to be larger than that (2.22) in Y-O bond. [7], the band gap of the film would increase as La content increases.

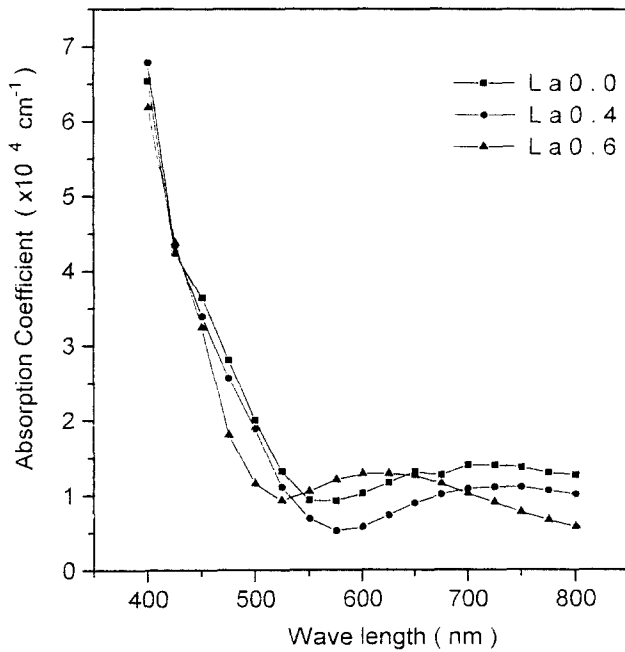


Figure 3. Optical absorption coefficient spectra of La-substituted Bi:YIG films.

The La substitution might also cause the shift of the centers of the charge transfer transitions toward shorter wavelengths. The crystal field transitions are centered at visible spectrum whose strength is about 1000 times weaker than that of the charge transfer transitions.[5] They are the electronic transitions between energy band splitted in 3d orbit of  $Fe^{3+}$  ions due to the surrounding nonspherical electrical field of  $O^{2-}$  ions originated from the difference in repulsive force of 3d electrons in  $Fe^{3+}$  ions located at octahedral or tetrahedral sites.[5] If one reduces the crystal field splitting, one can expect that the centers of the transitions shift toward shorter wavelengths.[5] This might happen by the La substitution, since one of the methods to reduce the crystal field is to increase the distance between the cations and anions through increasing lattice constant.[8]

Figure 4 shows the Faraday rotation angle spectra as a function of La content. The maximum of the absolute value of Faraday rotation angle observed at about 500 nm for the films with no La substitution shifts toward shorter wavelengths as La content increases. As a result it was found that the absolute value of the Faraday rotation angle in the wavelengths shorter than about 480 nm was increased as La content increased. This might also due to the shift of the centers of the charge transfer and crystal field transitions discussed above.

Figure 5 shows the magneto-optic figure of merit spectra obtained from the absorption and Faraday rotation spectra. As shown the magneto-optic figure of merit in the wavelengths shorter than 470 nm tends to increase as La content increases and it was found that the magneto-optic figure of merit of the film with La content of 0.6 in formular

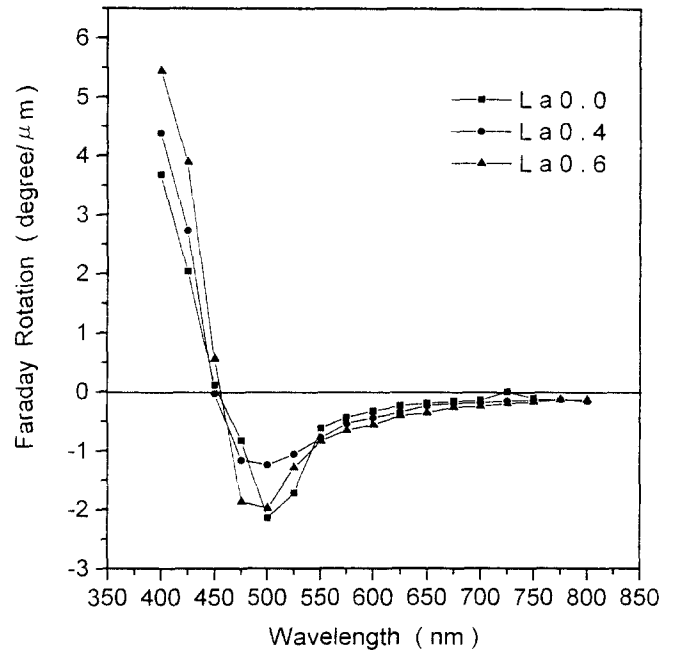


Figure 4. Faraday rotation per unit length spectra of La-substituted Bi:YIG films.

unit was about 2 times greater in the wavelengths than that of the film with no La substitution.

#### IV. Conclusions

Polycrystalline La-substituted Bi:YIG films have been prepared by pyrolysis. The XRD analyses revealed that La

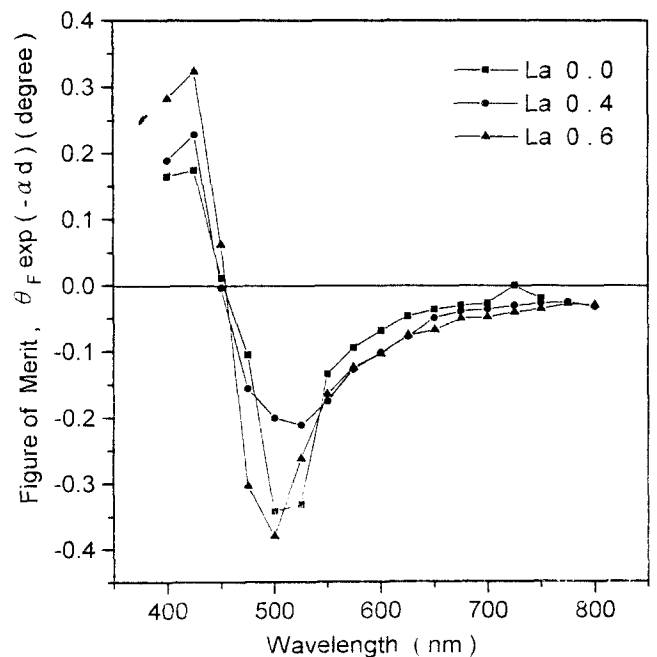


Figure 5. Magneto-optic figure of merit spectra of La-substituted Bi:YIG films.

ions substituted Y ions located at the dodecahedral sites. The optical absorption coefficient at the wavelengths shorter than 500 nm decreased as La content increased. The maximum of the absolute value of the Faraday rotation angle observed at about 500 nm for the films with no La substitution shifted toward shorter wavelength as La content increased, that leads the increase in the absolute value of the Faraday rotation angle in the wavelength shorter than about 480 nm. As a result the magneto-optic figure of merit increased in the wavelengths shorter than 470 nm as La content increased and was about two times larger for the film with La content of 0.6 in formula unit compared to that of the film without La substitution. This might be attributed to the shift of the centers of the charge transfer and crystal field transitions by La substitution.

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