Magneto-optical properties of Ca-substituted Bi-YIG sputtered films

Teruyoshi Hirano
Toppan Printing Co., Sugito, 345 Saitama, Japan

Tatsuru Namikawa and Yohtaro Yamazaki
Tokyo Institute of Technology, Nagatsuta, Midori-ku, 227 Yokohama, Japan

Ca-substituted Bi-YIG films ($Bi_2YFe_{5-x}Ca_xO_{12}$, $x = 0.0$, 0.3, 0.5, 1.0) were prepared by rf sputtering, and their magnetic and magneto-optical properties in the visible wavelength region were investigated. The films were deposited on Corning No. 7059 glass substrates at 400 °C. Sputter gas was Ar, 6.7 Pa, deposition rate 3.3 nm/min, rf-power density 2.5 W/cm^2. The films were annealed in air at 650 °C for 4 h to convert to polycrystalline films. Prepared films were examined by x-ray analysis and determined to be as garnet phase. The x-ray diffraction peaks of the films shifted to lower diffraction angle with increasing Ca content. The saturation magnetization of the films did not decrease with increasing Ca content. These results suggest that Ca^{2+} ions were partially substituted for *c*-site ions. The Faraday rotation of the films have maximum values at 470 nm. The magnitude of the Faraday rotation and absorption coefficients in the wavelength region from 450 to 550 nm decreased with increasing Ca content. And the absorption edge was shifted to short wavelength region with increasing Ca content. The reduction in the absorption coefficient was remarkable.

INTRODUCTION

Bismuth substituted yttrium iron garnet (Bi-YIG) is one of the most promising magneto-optical materials in which large Faraday rotations and low optical absorptions are required. Recently, Bi-YIG films have been applied in magneto-optical devices working in an infrared wavelength region.

In this paper, we describe the preparation and analysis of Ca-substituted Bi-YIG sputtered films $Bi_2YFe_{5-x}Ca_xO_{12}$, and their magnetic and magneto-optical properties in the visible wavelength region.

EXPERIMENTS

Ca-substituted Bi-YIG films were prepared on Corning No. 7059 glass substrates by rf sputtering under the conditions given in Table I. The targets were prepared by mixing the oxide powders of Bi_2O_3, Y_2O_3, Fe_2O_3, and CaO, and fired at 800 °C for 4 h, then crushed in a ball mill for 4 h. The obtained powder was pressed at 200 kg/cm^2 to form a disk of 3 mm thick and 100 mm in diameter. The distance from the target to the substrate was 40 mm. As deposited amorphous films were annealed in air at 650 °C for 4 h to convert to polycrystalline films. The phases of the annealed films were identified by x-ray diffraction analysis with Cu $K\alpha$. The film thickness was measured with a DEKTAK thickness meter. The film compositions were determined with an EDAX analyzer. The saturation magnetization (M_s) and coercive force (H_c) were measured with a VSM. The Faraday rotation (θ_F) was measured by a polalization modulation method. The optical absorption coefficient (α) were measured with a spectrophotometer.

TABLE I. Sputtering conditions.

Target composition	$Bi_2YFe_{5-x}Ca_xO_{12}$ ($x = 0.0$, 0.3, 0.5, 1.0)
Sputter gas	Ar, 6.7 Pa
Substrate	Corning No. 7059 glass
Substrate temperature	400 °C
rf power density	2.5 W/cm^2
Deposition rate	3.3 nm/min
Target-substrate distance	40 mm

RESULTS AND DISCUSSION

Figure 1 shows x-ray diffraction patterns of the annealed films for various Ca contents. In the region from $x = 0$ to 1.0, all diffraction peaks were assigned to the garnet structure. These peaks slightly shifted to lower diffraction angle with increasing Ca content. The peak shifts indicate an expansion of the unit cell. It suggests that the Ca ions substitute for cations in the Bi-YIG films.

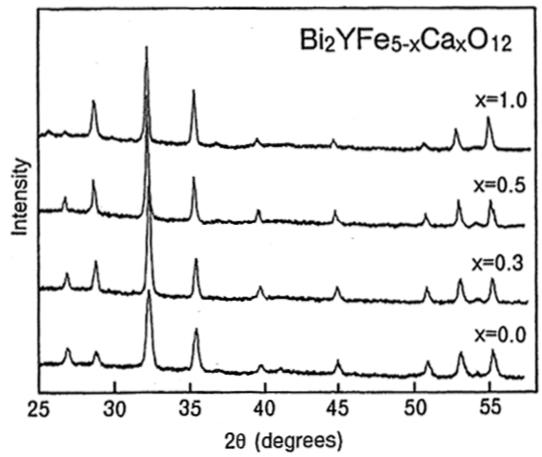

FIG. 1. X-ray diffraction patterns of Ca-substituted Bi-YIG films.

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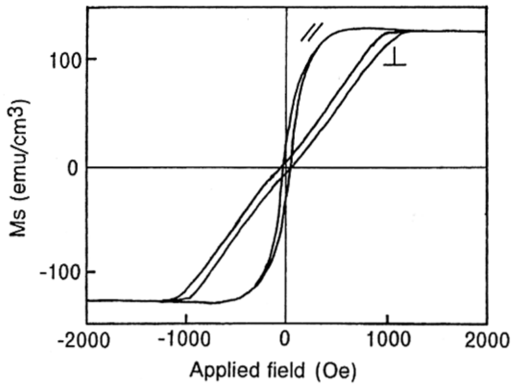

FIG. 2. Perpendicular and in-plane M-H curves of the film with $x = 0.5$.

Figure 2 shows the perpendicular and in-plane M-H curves measured of a film with $x = 0.5$. The curves indicate a slight perpendicular component of magnetization. Figure 3 shows the changes of M_s and H_c of the Bi-YIG films versus Ca content. The saturation magnetization of the films did not decrease with increasing Ca content. In Bi-YIG crystals, Fe ions occupy 16*a* and 24*d* sites, while Bi and Y ions occupy 24*c* sites. Then, considering the result that M_s did not decrease with Ca content, it is suggested that the Ca^{2+} ions are substituted for 24*c* ions. This assumption is consistent with the results on Ca substituted Bi-YIG reported by Roode *et al.*[1]

Figure 4 shows the spectrum of θ_F of the films for various Ca contents. The Faraday rotation of each sample has a maximum value at 470 nm. The magnitude of θ_F decreased with increasing Ca content. The origin of the large Faraday rotation in Bi-YIG systems is attributed to an electron orbit interaction between Fe^{3+} and Bi^{3+}.[2] Therefore it is expected that the reduction in θ_F with Ca content is caused by the substitution of Ca^{2+} ions for Bi^{3+} ions in the 24*c* sites. This result also supports the explanation of the M_s change with Ca content shown in Fig. 3.

Figure 5 shows α of Bi-YIG films in the visible wavelength region for various Ca contents. The absorption band of Bi-YIG below 550 nm was reported to be related to electron transitions in the Fe^{3+} ions.[3] The absorption increases with decreasing wavelength. It is noticeable that α is significantly lowered with increasing Ca content in the short wavelength region. Figure 6 shows the optical transmittance of the films near 500 nm for various Ca contents. It is clear that the absorption edge shifts to shorter wavelength region with increasing Ca content. The remarkable reduction in the optical absorption in the shorter wavelength region relates to the shift of absorption edge.

Figure 7 shows the relations between α at 470 nm and Fe, Ca contents. It is expected that a plot of α_{470} vs Fe content should be a straight line going through the origin. The dashed line in Fig. 7 is a line going through the origin. The plot obviously deviates from the line, especially as Ca content increases. This result indicates that the absorption at 470 nm is not proportional to Fe^{3+} concentration in the films. Therefore the origin of the reduction in α in this wavelength region must be discussed with regard to other absorption mechanism induced by the Ca^{2+} substitution.

Figure 8 shows the figure of merit θ_F/α_{470} of the films as a function of Ca content. It decreased with Ca content because the reduction of θ_F was larger than that of α_{470}.

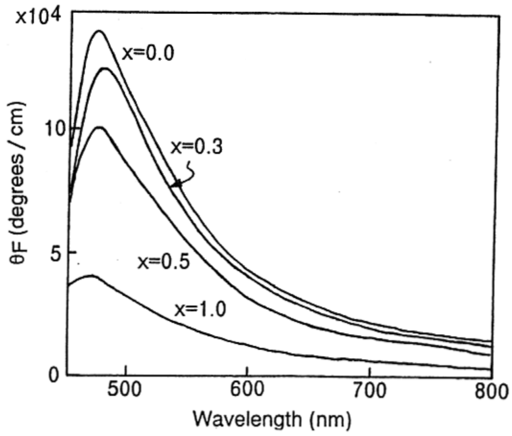

FIG. 4. θ_F as a function of wavelength for various Ca contents.

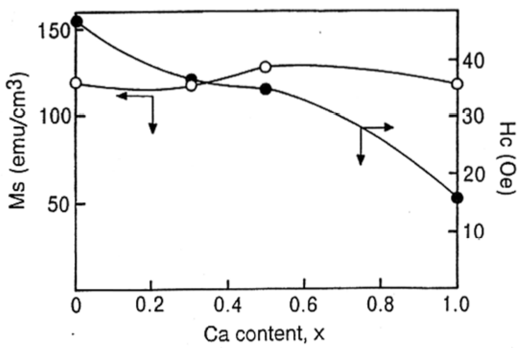

FIG. 3. Magnetization and coercive force as a function of Ca content.

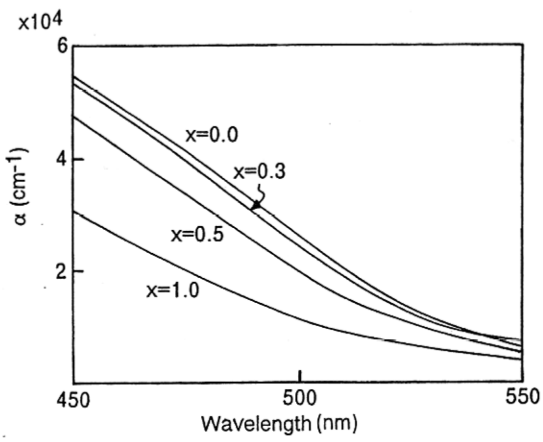

FIG. 5. Absorption spectra of the films for various Ca contents.

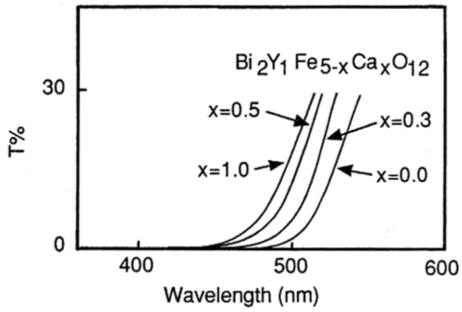

FIG. 6. Transmittance of the films for various Ca contents.

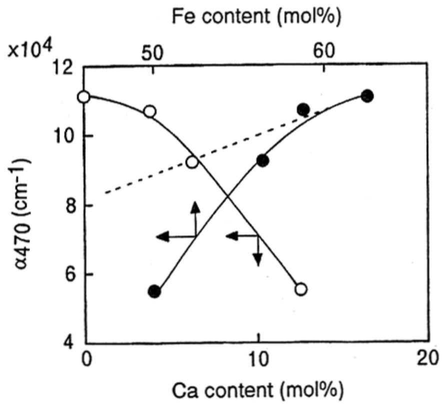

FIG. 7. Absorption coefficient at 470 nm vs Ca and Fe concentrations.

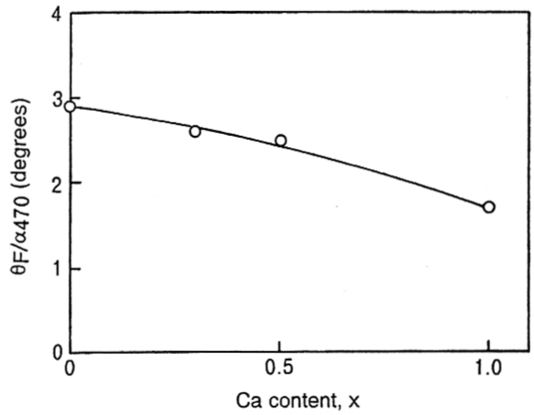

FIG. 8. θ_F/α_{470} vs Ca concentration.

CONCLUSIONS

Ca-substituted Bi-YIG polycrystalline sputtered thin films were prepared and the magnetic and magneto-optical properties were studied. The Faraday rotation of the films lowered with increasing Ca content, however, the magnetization did not change. This result suggest that the Ca^{2+} ions substitute to Bi^{3+} ions in 24*c* site. The absorption in the visible wavelength region substantially decreased with Ca content, and it was found that the absorption did not linearly correlate to the Fe^{3+} concentration in the films.

[1] W. H. Roode and C. A. Van de Pavert, J. Appl. Phys. **55**, 3115 (1984); Y. Yokoyama and N. Koshizuka, J. Magn. Soc. Jpn. **11**, 153 (1987); S. Geller, J. Appl. Phys. **5**, 30*S* (1960).
[2] S. Sinagawa, J. Magn. Soc. Jpn. **6**, 247 (1987).
[3] G. B. Scott, D. E. Lacklison, and J. L. Page, Phys. Rev. B **10**, 971 (1974).