OPTICAL ABSORPTION OF BI-SUBSTITUTED IRON GARNET LPE FILMS FOR 0.8 μm OPTICAL ISOLATOR

T. ISHIKAWA, T. MASUMOTO, K. ONO, and K. SHIROKI

Research Lab., Tokin Corp., Yatabe-machi, Tsukuba, Ibaraki 305, Japan

ABSTRACT

Bi-substituted gadolinium iron garnet grown by an LPE method on Ca,Zr,Mg-doped GGG substrate is a promising material for use in 0.8 μm wavelength band optical isolator. We have investigated the effect of Mg^{2+} -, Si^{4+} -, and In^{3+} -doping to the melt and film annealing on optical absorption.

The experimental result shows that optical absorption coefficient of as-grown films at 0.8 μm decreased with decreasing Mg^{2+} -doping and increasing Si^{4+} -doping levels. The absorption coefficients also decreased by annealing at 800 $^{\circ}\text{C}$ down to 52-55 cm^{-1} . The effect of annealing on optical absorption would be mainly caused by reduction of Fe^{3+} absorption at 0.7 μm . For Mg^{2+} -doped films annealed at 800 $^{\circ}\text{C}$ in N_2 atmosphere, high figure of merit of 27.3 deg/dB (1.65 dB loss for 45 degree rotation) was attained. The results of In^{3+} -doping are mentioned, too.

1. INTRODUCTION

In optical fiber communications systems using semiconductor lasers (e.g., LD) as a light source oscillation becomes unstable due to reflection of light from optical devices. To cope with this problem an optical isolator using thick film of Bi-substituted garnet which is grown by the liquid-phase epitaxial method (LPE method) has been developed and introduced into practical use [1]. The problem with the 0.8 μm band optical isolator is a low figure of merit (which is expressed in terms of Faraday rotation per insertion loss). The figure of merit of optical isolators can be improved in several ways. For instance, one way is to reduce optical absorption by substituting In^{3+} or Sc^{3+} for Fe^{3+} at the octahedral site of the crystal which is cause of absorption in the neighborhood of 0.8 μm . On the other hand, divalent and quadrivalent ions of impurities contained in the crystal can also be cause of absorption, since they give rise to Fe^{4+} and Fe^{2+} [2][3]. The figure of merit of isolators, therefore, may be improved by adding Ca^{2+} , Mg^{2+} or Si^{4+} to the crystal for purposes of electric charge compensation [4][5].

Optical absorption in the 0.8 μm band is situated by minimal values between the two absorption at 0.7 and 0.9 μm , which is caused by Fe^{3+} at the octahedral site (16a) of the garnet structure. So the absorption in this band is easily affected by Fe^{3+} at its octahedral site. The Faraday rotation coefficient for the garnet crystal decreases with $(X_a + X_d)$, where X_a and X_d represent the quantities of Fe^{3+} substituted at the octahedral and tetrahedral site, respectively; sites 16a and 24d [6]. In the Bi-substituted garnet, it is therefore conceivable that elements (e.g., In^{3+} , Sc^{3+}), which are preferentially substituted at its octahedral site, are more effective to improve the figure of merit than Ga^{3+} and Al^{3+} substituted for Fe^{3+} at both the tetrahedral and octahedral sites. In the case of electric charge compensation, on the other hand, Pt^{4+} contained in the crucible material, the flux component Pb^{2+} and other impurities may be incorporated in the garnet crystal, causing Fe^{3+} to Fe^{2+} and Fe^{4+} . This upsets the balance of electric charges in the crystal with the result that absorption in the neighborhood of 0.9 μm increases. Attempts have been made to compensate Fe^{2+} and Fe^{4+} for electric charges, using Ca^{2+} , Mg^{2+} , and Si^{4+} in order to reduce optical absorption in the band near 0.9 μm .

The present communication is a report of our results obtained in an investigation which was undertaken in order to improve the figure of merit of optical isolators. In this experimental study $(\text{YBi})_3(\text{FeIn})_5\text{O}_{12}$ and $(\text{GdBi})_3(\text{FeGaAlM})_5\text{O}_{12}$ ($\text{M} = \text{Mg}^{2+}$, Si^{4+}) were grown from $\text{PbO-Bi}_2\text{O}_3\text{-B}_2\text{O}_3$ flux by the LPE method, and an investigation was conducted using the crystals so grown to determine the effect of heat treatment on optical absorption, as well as the relationship between the quantities of additive impurities and electrical resistivity.

2. EXPERIMENT

Using the LPE method, $(YBi)_3(FeIn)_5O_{12}$ and $(GdBi)_3(FeGaAlM)_5O_{12}$ ($M = Mg^{2+}, Si^{4+}$) were grown from $PbO-Bi_2O_3-B_2O_3$ flux on $Sm_3Ga_5O_{12}$ and $(GdCa)_3(GaZrMg)_5O_{12}$ substrates. The crystals were grown onto (111) oriented substrates 23 mm in diameter. The melt composition conducted was referred to the report by Klages et al. [7].

In order to determine the effect of In^{3+} the quantity of In^{3+} substituted in the garnet crystal was controlled by changing the value $R_6 = In_2O_3 / (Fe_2O_3 + In_2O_3)$ from 0 to 7% and to 13%, and the effect of Mg^{2+} and Si^{4+} was determined by varying the $R_6 = X / \{2(Fe_2O_3 + Ga_2O_3 + Al_2O_3) + X\}$ to control the quantities of Mg^{2+} and Si^{4+} substituted in the garnet crystal. The composition of the grown garnet crystal was determined by EPMA analysis. The optical absorption coefficient was measured spectrophotometrically, using films 20 to 30 μm thick. The laser beam was put into the specimen perpendicular to the surface. The optical absorption coefficient was calculated from transmittance, using an equation which takes account of multiple reflection [8]. The Faraday rotation coefficient was measured with an automatic Faraday rotation measuring instrument using a Faraday cell, and electrical resistivity measured by the DC conduction method.

3. RESULTS AND DISCUSSION

3-1. Effect of In^{3+}

The crystal growth temperature at $R_6 = In_2O_3 / (Fe_2O_3 + In_2O_3)$ varied from 0 through 7% to 13% was plotted against the Faraday rotation coefficient (measured at a wavelength of 0.8 μm), as shown in Fig. 1. As is obvious from this graph, there was a tendency that the Faraday rotation coefficient decreased with increasing R_6 . However, the Faraday rotation coefficient varied with the crystal growth temperature. Briefly, in the crystal growth temperature range of 755 to 745 $^{\circ}C$ the Faraday rotation coefficient decreased with increasing R_6 , while the Faraday rotation coefficient was comparable between $R_6 = 0$ and 7% in the crystal growth temperature range of 740 to 730 $^{\circ}C$. Fig. 2 plots the crystal growth temperature against the optical absorption coefficient. Optical absorption clearly decreased with increasing R_6 from 0 through 7% to 13%. The relationship between the crystal growth temperature and insertion loss per Faraday rotation of 45 degree is represented in Fig. 3. The insertion loss decreased from 6 to 4 dB/45 degree when R_6 was 0 or 7%, but there was little if any difference in insertion loss between $R_6 = 7\%$ and 13%.

Fig. 1. Faraday rotation coefficient vs. growth temperature for In-substituted films; $(YBi)_3(FeIn)_5O_{12}$

Fig. 2. Optical absorption coefficient at $\lambda = 0.8 \mu m$ vs. growth temperature for In-substituted films; $(YBi)_3(FeIn)_5O_{12}$

Fig. 3. Insertion loss at $\lambda = 0.8 \mu m$ vs. growth temperature for In-substituted films; $(YBi)_3(FeIn)_5O_{12}$

The maximum figure of merit of $(\text{YBi})_3(\text{FeIn})_5\text{O}_{12}$ at a wavelength of $0.8 \mu\text{m}$ was 11.3 deg/dB , which was small, compared to the previously reported value of 23 deg/dB [8]. This observation suggests that there is a limit to the attempt to improve the figure of merit of the garnet crystal by substituting In^{3+} for Fe^{3+} at the octahedral site.

3-2. Effect of Mg^{2+} and Si^{4+}

Fig. 4 plots the crystal growth rate against the quantities of Pb^{2+} and Pt^{4+} contained in $(\text{GdBi})_3(\text{FeGaAlM})_5\text{O}_{12}$. In the case the difference between $[\text{Pb}^{2+}]$ and $[\text{Pt}^{4+}]$ was 0.15 to 0.2 atoms/f.u. Whereas there was a straight line relationship between crystal growth rate and quantities of Pb^{2+} and Pt^{4+} substituted, there was little difference between $[\text{Pb}^{2+}]$ and $[\text{Pt}^{4+}]$ when Mg^{2+} was substituted at a rate of 2% in terms of R_6 (Fig. 5). When R_6 was varied

Fig. 4. The content of Pb, Pt vs. growth rate for LPE films; $(\text{GdBi})_3(\text{FeGaAl})_5\text{O}_{12}$

Fig. 5. The content of Pb, Pt vs. growth rate for LPE films; $(\text{GdBi})_3(\text{FeGaAlMg})_5\text{O}_{12}$ ($R_6 = 2\%$)

from 0 to 3% with Mg^{2+} and Si^{4+} , crystals as-grown and annealed at 800°C in N_2 atmosphere yielded the optical absorption coefficient shown in Fig. 6. With the as-grown crystal, its optical absorption increased with R_6 for Mg^{2+} alone, but the reverse was the case with Si^{4+} . This may be accounted for by assuming that the as-grown crystal contained Fe^{4+} before its impurities were replaced. This assumption may be substantiated by the analytical values indicating that $[\text{Pb}^{2+}] > [\text{Pt}^{4+}]$. When the crystal was annealed in N_2 atmosphere at 800°C , the optical absorption decreased to $52\text{--}55 \text{ cm}^{-1}$, regardless of R_6 for Mg^{2+} and Si^{4+} . Fig. 7 shows the transmission spectra of as-grown and N_2 annealed crystals after substitution at $R_6 = 2\%$. What characterizes their transmission spectra is the fact that optical absorption of Fe^{3+} in the neighborhood of $0.7 \mu\text{m}$ was decreased with an increase in transmittance to match. This finding does not correspond with the reported results from Yokoyama et al.[4] and Kaneko et al.[5] indicating a significant

Fig. 6. Optical absorption ($0.8 \mu\text{m}$) vs. R_6 for substituted, as-grown and N_2 annealed films; $(\text{GdBi})_3(\text{FeGaAlM})_5\text{O}_{12}$ ($M=\text{Mg}^{2+}, \text{Si}^{4+}$)

decrease in absorption in the neighborhood of $0.9 \mu\text{m}$. Fig. 8 plots electrical resistivity of the garnet crystal against varying R_6 for Mg^{2+} and Si^{4+} . The electrical resistivity remained nearly constant when R_6 was varied from 0 to 3%. In short, correlation between electrical resistivity and optical absorption coefficient was not clarified. However, the increase in transmittance in the neighborhood of $0.7 \mu\text{m}$ and the lack of correlation between electrical resistivity and absorption coefficient are yet to be explained. It remains for further studies to answer these questions.

The Faraday rotation coefficient of the garnet film $(\text{GdBi})_3(\text{FeGaAlM})_5\text{O}_{12}$ at a wavelength of $0.8 \mu\text{m}$ ranged from 5800 to 6200 deg/cm , and the figure of merit at $R_6 = 2\%$ was 27.3 deg/dB (1.65 dB/45 deg).

Fig. 7. Transmission spectra of Mg^{2+} substituted $(\text{GdBi})_3(\text{FeGaAlM})_5\text{O}_{12}$ ($R_6 = 2\%$)

Fig. 8. Electrical resistivity vs. R_6 for substituted films; $(\text{GdBi})_3(\text{FeGaAlM})_5\text{O}_{12}$ ($M = \text{Mg}^{2+}$, Si^{4+})

4. CONCLUSION

Garnet films of $(\text{YBi})_3(\text{FeIn})_5\text{O}_{12}$ and $(\text{GdBi})_3(\text{FeGaAlM})_5\text{O}_{12}$ ($M = \text{Mg}^{2+}$, Si^{4+}) were grown by the LPE method in order to improve the figure of merit of optical isolators in the $0.8 \mu\text{m}$ band and tested under various sets of conditions. As a result, it was shown that the material under test yielded a figure of merit of 27.3 deg/dB ($1.65 \text{ dB} / 45 \text{ deg}$) when substituted with Mg^{2+} at a rate of 2% in terms of R_6 and annealed in N_2 atmosphere at a temperature of 800°C .

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