GROWTH-INDUCED BIREFRINGENCE IN LPE-GROWN IRON GARNET FILMS

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ABSTRACT

The optical birefringence of various LPE-grown rare-earth and Bi-substituted iron garnet films are studied. In addition to the stress-induced birefringence, it is shown that there exists growth-induced birefringence Δn_f^G in the films containing light rare-earth or Bi ions. In Bi garnet films the Δn_f^G disappears on annealing and the thermal behavior of Δn_f^G is similar to that of the growth-induced uniaxial magnetic anisotropy constant K_u^G . Furthermore, the Bi content dependence of Δn_f^G is similar to that of K_u^G . As a result, it is suggested that the Δn_f^G and K_u^G originate in the same microscopic structure which is of an anisotropic character. It is pointed out that the positive sign and the magnitude of Δn_f^G in Bi garnet films are effective for the phase matching between TE and TM modes in a planar waveguide.

1 INTRODUCTION

Waveguide type optical devices are expected to play important roles in future optical communication and information processing systems. As for magneto-optical devices, nonreciprocal devices such as isolators and circulators are indispensable to prevent unstable laser oscillation due to the backward beam and to split the forward and backward beams [1].

Rare-earth iron garnet films are thought to be most promising materials because of their high magneto-optical constants, optically isotropic crystal structure and a variety of magnetic properties. Especially Bi-substituted iron garnet films are the materials of great interest [2,3]. The enhancement of Faraday rotation and the increase in refractive index and the Curie temperature occur with Bi substitution. Furthermore, it is known that the uniaxial magnetic anisotropy is induced in LPE-grown Bi garnet films. These studies have been extensively done by many groups, in particular, by Hansen et al. of Philips Research Laboratories [3].

Since the refractive index of these films is higher than that of the substrates such as GGG, these films are also suitable as waveguide materials. However, there remain several basic problems such as phase matching between TE and TM modes and microfabrication techniques of garnet films. As for the phase matching, it is known that the film birefringence $\Delta n_f (=n_{TM} - n_{TE})$ is effective to compensate for the phase mismatch due to the planar structure [4]. Furthermore, the magnitude of Δn_f can be controlled by the stress-induced birefringence Δn_f^S caused by the lattice mismatch between film and substrate [4~6]. In view of these considerations, studies on the birefringence of various rare-earth and Bi-substituted iron garnet films were carried out [7~14]. As a result, we found that there exists "growth-induced birefringence" Δn_f^G in most of these films and the sign of Δn_f^G is positive in Bi-substituted iron garnet films. This paper describes the relations of the optical birefringence with the lattice mismatch, annealing treatment, and Bi content in LPE-grown garnet films.

2 MODE CONVERSION AND BIREFRINGENCE

So far, many theoretical and experimental studies on waveguide type non-reciprocal devices or switches have been made in the last decade [1,15~24]. In these devices magneto-optical power conversion between TE (transverse electric) and TM (transverse magnetic) modes plays a crucial role. The power conversion between TE and TM modes in the Faraday configuration is expressed by the equation [17]

$$R(L) = \frac{K^2}{K^2 + (\Delta\beta/2)^2} \sin^2(\sqrt{K^2 + (\Delta\beta/2)^2} \cdot L), \quad (1)$$

where K is the coupling constant which equals the Faraday rotation coefficient, $\Delta\beta$ is the difference between the propagation constants for the two modes ($\Delta\beta = \beta_{TM} - \beta_{TE}$), and L is the propagation length. From eq.(1), it turns out that complete mode conversion occurs under the phase-matching condition, i.e. $\Delta\beta = 0$.

The effective refractive indices for TE and TM modes are defined by $N_{TE} = \beta_{TE}/k$ and $N_{TM} = \beta_{TM}/k$, where k is the propagation constant of light in vacuum ($k = 2\pi/\lambda$), and they have different dispersion curves to the film thickness. In an optically isotropic waveguide, $N_{TM} < N_{TE}$, i.e. $\Delta\beta_{iso} < 0$, because the phase changes suffered by the beam at the film-substrate and film-air interfaces are different between the TE and TM waves, and in general the velocity of the TM wave is slightly faster than that of the TE wave. The magnitude of $\Delta\beta_{iso}$ can be calculated from the

values of film thickness d , refractive indices of film n_f , substrate n_s and air n_a . On the other hand, in an anisotropic case, the degeneracy of the effective refractive indices is expected under a certain condition. In a film with the birefringence of $\Delta n_f (=n_{TM} - n_{TE})$, the phase difference is expressed to a good approximation by

$$\Delta \beta = \Delta \beta_{iso} + k \Delta n_f \quad (2)$$

Here, the phase matching condition $\Delta \beta = 0$ is satisfied if $\Delta n_f = -\Delta \beta_{iso}/k$. Since the sign of $\Delta \beta_{iso}$ is negative, it is clear that the Δn_f with positive sign and proper magnitude is necessary for $\Delta \beta = 0$.

3 EXPERIMENTAL

Films of rare-earth and Bi-substituted iron garnets were grown on (111)-oriented pure and CaMgZr-substituted $Gd_3Ga_5O_{12}$ (GGG) substrates by liquid-phase epitaxy (LPE). A $PbO-B_2O_3$ or $PbO-B_2O_3-Bi_2O_3$ flux was used as a solvent. The compositions were determined mainly by means of electron probe X-ray microanalyser. The lattice mismatch perpendicular to the film surface $\Delta a^\perp (=a_s - a_f^\perp)$ was measured by X-ray double-crystal diffraction method. The a_s and $a_f^\perp$ are the lattice parameters of the substrate and film perpendicular to the surface, respectively.

Waveguiding experiments were performed using a $1.15 \mu m$ He-Ne laser beam and two prisms for coupling of the laser beam. The film thickness d and the refractive index n_f were determined by analyzing the mode coupling angles. Power conversion efficiencies $R(L)$ were measured by applying magnetic field parallel to the film plane. The values of K and $\Delta \beta$ were determined by analyzing the propagation-length dependence of $R(L)$. From eq. (2), we obtain the value of Δn_f using the calculated value of $\Delta \beta_{iso}$. Stress-induced birefringence Δn_f^S is estimated from the following relation:

$$\Delta n_f^S = 2B_{[111]}C_{44}\Delta a^\perp/a_f \quad (3)$$

where $B_{[111]}$ is the stress-optical constant for stress in the $[111]$ direction and C_{44} the elastic constant. Values of $B_{[111]} (=3 \times 10^{-13} \text{ cm}^2/\text{dyn})$ and $C_{44} (=7.65 \times 10^{11} \text{ dyn/cm}^2)$ of YIG were adopted in the calculation. The saturation magnetization $4\pi M_s$ was measured by using a vibrating sample magnetometer (VSM). The uniaxial magnetic anisotropy constants were determined by analyzing the ferromagnetic resonance (FMR) spectra. The Faraday rotation was measured by using a polarization modulation type Faraday rotation spectrometer (Jasco MOE-7).

4 RESULTS AND DISCUSSION

4-1 Dependence of birefringence on lattice mismatch

We first studied the dependence of Δn_f on lattice mismatch in YNdIG films which have in-plane magnetization [7]. Fig.1 shows the experimental values of Δn_f and the calculated Δn_f^S as a function of growth temperature T_g , where the saturation temperature of the melt was 1013°C . One can see here that there exists a negative gap ($\Delta n_f - \Delta n_f^S < 0$) between Δn_f and Δn_f^S even in misfit-free films ($\Delta a^\perp = 0$). However, for YIG films which were prepared under the similar condition, such a difference was negligibly small.

Lattice mismatch dependence of Δn_f was studied for other systems [9]. Figs.2 and 3 show the Δn_f and Δn_f^S as a function of $\Delta a^\perp$ in YSmIG and YGdIG films, respectively. In these cases, we also find that $\Delta n_f < \Delta n_f^S$.

Fig.1 Film birefringence Δn_f and stress-induced birefringence Δn_f^S in YNdIG films as a function of growth temperature [9].

Fig.2 Film birefringence Δn_f and stress-induced birefringence Δn_f^S in YSmIG films as a function of lattice mismatch $\Delta a^\perp$ [9].

Table 1 Physical and guided-wave properties of BiPrYbGaIG films at $\lambda = 1.15 \mu\text{m}$ [10]. T_g : growth temperature, d : film thickness, $\Delta a^\perp$: lattice mismatch, n_f : refractive index of film, α : absorption loss, K : coupling constant, $\Delta\beta$: phase mismatch between TE_0 and TM_0 modes, Δn_f : film birefringence, Δn_f^S : stress-induced birefringence, $4\pi M_s$: saturation magnetization.

Material No.	T_g (°C)	d (μm)	$\Delta a^\perp$ (Å)	n_f	α (dB/cm)	K (rad/cm)	$\Delta\beta$ (rad/cm)	Δn_f	Δn_f^S	$4\pi M_s$ (G)
			10^{-5}					10^{-4}	10^{-4}	
62	810	3.32	7.8	2.225	14	-12.6	-20	0.2	-3.0	272
63†	808	4.00	1.0	2.231	16	-13.3	~0	2.3	-0.4	294
64	805	3.74	-3.2	2.234	22	-14.9	11	4.9	1.2	310
65	803	3.49	-10.1	2.239	28	-15.6	22	7.3	3.8	327

Saturation temperature $T_S = 822^\circ\text{C}$

†Composition: $\text{Bi}_{0.75}\text{Pr}_{0.34}\text{Yb}_{2.25}\text{Fe}_{3.58}\text{Ga}_{1.02}\text{O}_{12}$.

The effect of Ga substitution on the birefringence was studied in YNdGaIG films [9]. Fig.4 shows the lattice mismatch dependence of Δn_f and calculated Δn_f^S . We found no remarkable difference between the cases of YNdIG and YNdGaIG films. This means that a combination of rare-earth ions on the dodecahedral site is important in generating the gaps between Δn_f and Δn_f^S .

The birefringence of various Bi-substituted iron garnet films were also compared with the Δn_f^S calculated from the lattice mismatch [8~14]. Table 1 shows physical and guided-wave properties of BiPrYbGaIG films at $\lambda = 1.15 \mu\text{m}$ [10]. In these films we find that Δn_f is larger than Δn_f^S and the magnitude of the difference is the order of 10^{-4} . The similar result has been observed in (110)-oriented BiGdYbAlIG films by Ageev et al. [25]. From the annealing experiments described later, we considered that the difference is due to the growth-induced birefringence Δn_f^G like the growth-induced magnetic anisotropy K_u^G in LPE garnet films.

Table 2 shows guided-wave properties of various iron garnet films at $\lambda = 1.15 \mu\text{m}$ [6,9,10,26]. It turns out that the garnet films are classified into three groups from the values of Δn_f^G : (1) $\Delta n_f^G \sim 0$ (YIG, YTaAlIG), (2) $\Delta n_f^G < 0$ (YGdIG, YNdIG, YSmIG, YNdGaIG), and (3) $\Delta n_f^G > 0$ (BiNdYGaIG, BiPrYbGaIG, BiGdLuGaIG).

This classification suggests that there is a certain correlation between Δn_f^G and K_u^G . Because the K_u^G is known to arise when more than two rare-earth ions with different ionic radii occupy the inequivalent dodecahedral sites in the film growth process. Furthermore, it is noted that Bi-substituted iron garnet films have large K_u^G in general, although its detailed mechanism is not made clear yet [3].

Fig.3 Film birefringence Δn_f and stress-induced birefringence Δn_f^S in YGdIG films as a function of lattice mismatch $\Delta a^\perp$ [9].

Fig.4 Film birefringence Δn_f and stress-induced birefringence Δn_f^S in YNdGaIG films as a function of lattice mismatch $\Delta a^\perp$ [9].

Table 2 Physical and guided-wave properties of iron garnet films at $\lambda = 1.15 \mu\text{m}$. $\Delta n_f^G = \Delta n_f - \Delta n_f^S$: growth-induced birefringence.

Material	d (μm)	$\Delta a^\perp$ ($\text{\AA}$)	n_f	K (rad/cm)	$\Delta\beta$ (rad/cm)	Δn_f	Δn_f^S	Δn_f^G	Ref.
		10^{-3}				10^{-4}	10^{-4}	10^{-4}	
$\text{Y}_3\text{Fe}_5\text{O}_{12}$	4.08	8.2	2.213	4.1	-29.5	-3.0	-3.1	0.1	9
$\text{Y}_{1.76}\text{Tb}_{1.22}\text{Pb}_{0.02}\text{Fe}_{4.71}\text{Al}_{0.29}\text{O}_{12}$	7.6	-1.1	2.19	6.7	1.8	0.3	0.4	0.1	6
$\text{Y}_{2.78}\text{Gd}_{0.22}\text{Fe}_5\text{O}_{12}$	4.77	-2.2	2.210	5.0	-7.0	0.1	0.9	-0.8	26
$\text{Y}_{2.85}\text{Nd}_{0.15}\text{Fe}_5\text{O}_{12}$	5.84	0.5	2.213	4.4	-8.5	-0.7	0.2	-0.7	9
$\text{Y}_{2.76}\text{Sm}_{0.24}\text{Fe}_5\text{O}_{12}$	3.65	-10.1	2.212	3.9	2.3	3.4	3.8	-0.4	9
$\text{Y}_{2.64}\text{Nd}_{0.36}\text{Fe}_{4.25}\text{Ga}_{0.75}\text{O}_{12}$	3.80	-8.0	2.177	2.0	-3.4	2.0	3.0	-1.0	9
$(\text{BiNdY})_3(\text{FeGa})_5\text{O}_{12}$	5.03	-32.5	2.207	-5.4	22.4†	15.0	12.3	2.7	9
$\text{Bi}_{0.75}\text{Pr}_{0.34}\text{Yb}_{2.25}\text{Fe}_{3.64}\text{Ga}_{1.02}\text{O}_{12}$	4.00	1.0	2.231	-13.3	0	2.3	-0.4	2.7	10
$\text{Bi}_{0.65}\text{Gd}_{0.87}\text{Lu}_{1.81}\text{Fe}_{3.96}\text{Ga}_{0.71}\text{O}_{12}$	4.13	5	2.244	-10.1	0.5	2.2	-2	4.2	26

† Mode order $m = 2$.

4-2 Annealing effects in Bi garnet films.

We studied the effects of annealing on the residual birefringence in Bi garnet films [8,11].

Fig.5 shows the Δn_f of as-grown films and annealed films as a function of $\Delta a^\perp$ for BiGdLuGaIG films grown by using a $\text{PbO-B}_2\text{O}_3\text{-Bi}_2\text{O}_3$ flux. The Δn_f of annealed films are extrapolated values to infinite time deduced from the annealing time dependence at 1150°C . It turns out that the values of Δn_f in annealed films coincide with those of the stress-induced birefringence Δn_f^S . That is, the difference between Δn_f and Δn_f^S disappears on annealing.

Fig.6 shows annealing time dependences of Δn_f , the uniaxial magnetic anisotropy constant K_u and the saturation magnetization $4\pi M_s$ in BiGdLuGaIG films. As seen here, the annealing time dependence is almost the same for Δn_f and K_u ; the time constants for the changes of Δn_f and K_u are much the same at $1100\sim 1150^\circ\text{C}$. From these results we concluded that both Δn_f^G and K_u^G are caused by the specific microscopic structure formed during the growth process.

4-3 Bi content dependence

The Bi-substitution of dodecahedral rare-earth ions in garnets affects their magnetic, optical and magneto-optical properties such as the Curie temperature T_C , uniaxial magnetic anisotropy constant K_u , refractive index n_f , and Faraday rotation θ_F [3]. Especially, since the growth-induced birefringence Δn_f^G is thought to be related with the growth-induced uniaxial anisotropy constant K_u^G , the Bi content dependence of these properties was studied for BiYGaIG and BiAlIG films [13,14].

Fig.5 Film birefringence Δn_f of as-grown (●), annealed (○) films and stress-induced birefringence Δn_f^S (—) as a function of lattice mismatch $\Delta a^\perp$ [11].

Fig.6 Film birefringence Δn_f (○), uniaxial magnetic anisotropy constant K_u (▲) and saturation magnetization $4\pi M_s$ (□) as a function of annealing time. The annealing temperature $T_a = 1150^\circ\text{C}$ [11].

Table 3 Physical properties of $Y_{3-x}Bi_xFe_{5-y}M_yO_{12}$ ($M=Ga,Al$) at $\lambda = 1.15 \mu m$ [13]. H_a : magnetic anisotropy field, θ_F : Faraday rotation.

CONTENT (atoms/f.u.)			d (μm)	$4a^{\perp}$ ($\text{\AA}$)	$4\pi M_s$ (G)	H_a (Oe)	θ_F (deg/cm)	n_f	Δn_f^G
x_{Bi}	y_{Ga}	y_{Al}		10^{-3}					10^{-4}
0	0	0	4.08	8.2	1,750	—	240	2.213	0.1
0.15	0.45	0	4.91	<1.0	942	280	— 63	2.191	<0.5
0.24	0.77	0	3.51	3.4	339	409	— 211	2.179	1.3
0.32	0.95	0	6.43	<2.0	89	2,170	— 258	2.171	2.4
0.36	1.16	0	4.20	<2.0	16	—	—	2.160	3.5
0.44	0	0.32	3.50	<2.0	1,120	212	— 893	2.237	3.7
0.61	0	0.47	4.10	<2.0	829	1,054	—1,280	2.241	8.1
0.76	0	0.57	3.85	<2.0	603	1,920	—1,390	2.242	12.6
0.82	0	0.61	3.89	<2.0	540	2,553	—	2.243	14.8

Table 3 shows physical properties of BiYGaIG and BiYAlIG films [13]. From the atomic absorption analysis, the impurity content of Pb or Pt was estimated to be 0.01~0.02 atoms/f.u..

Fig.7 shows Bi content dependence of K_u^G [13]. The K_u^G was obtained from the value of K_u by taking into account the stress-induced anisotropy constant K_u^S and neglecting the relatively small crystalline anisotropy constant K_1 .

Fig.8 shows Bi content dependence of refractive index n_f at $\lambda=1.15\mu m$. In general, it is known that the n_f increases with Bi content because of the optical red shift of the band gap. While, the n_f decreases by the incorporation of nonmagnetic ions, Ga^{3+} and Al^{3+} , into Fe^{3+} sites, because the absorption intensity of the Fe associated with charge transfer transitions and pair transitions decreases. The difference of Bi content dependence in n_f between two series of films is explained by these factors.

Fig.9 shows Bi content dependence of Δn_f^G at $\lambda=1.15\mu m$ [13]. It turns out that the Δn_f^G increases monotonically with Bi content. The Bi content dependence of Δn_f^G are different between BiYGaIG and BiYAlIG films as that of K_u^G . However, the difference is not so large compared with the case of n_f . The contribution of Al ions to Δn_f^G seems to be small compared with that of Bi ions. The different Bi content dependence of Δn_f^G between BiYGaIG and BiYAlIG films may be due to the difference in LPE-growth conditions such as melt composition and growth temperature. Recently, we have found that the magnitude of Δn_f^G is dependent not only on the growth conditions but also on the lattice size of the substrate [14].

Fig.7 Growth-induced uniaxial magnetic anisotropy constant K_u^G as a function of Bi content in BiYGaIG and BiYAlIG films [13].

Fig.8 Refractive index of film n_f as a function of Bi content in BiYGaIG and BiYAlIG films [13].

Fig.9 Growth-induced birefringence Δn_f^G as a function of Bi content in BiYGaIG and BiYAlIG films [13]

5 SUMMARY

Recent studies of growth-induced birefringence for LPE-grown iron garnet films, especially for Bi-substituted garnet films were reviewed. The specific properties of Δn_f^G are as follows : (1) Δn_f^G is found in rare-earth iron garnet films containing more than two rare-earth ions and in Bi garnet films. (2) The sign of Δn_f^G is negative in rare-earth garnets and is positive in Bi garnets. (3) It is confirmed in Bi garnet films that the Δn_f^G disappears by proper annealing and the annealing time constant is almost the same as that of K_u^G . (4) The Δn_f^G increases with Bi content, but the magnitude depends on various growth conditions.

From these results it is considered that the Δn_f^G and K_u^G originate in the same microscopic structure in garnets ; a non-equilibrium distribution of dodecahedral site ions and a consequent deformation of the microscopic structure. Especially, in the case of Bi garnets, the large ion radius and large electronic polarizability of Bi^{3+} may play an important role in generating the optical anisotropy.

From the applications of point of view, it is concluded that the positive sign and the magnitude of the Δn_f^G in Bi-substituted garnet films are favorable for the phase matching in a planar waveguide.

The authors are much indebted to Y.Yokoyama, A.Murata, N.Takeda and T.Takahashi for sample preparation and characterization.

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