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Stress- and growth-induced anisotropic birefringences in garnet films

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Résumé. — A partir de mesures de propagation guidée de lumière dans des films de grenat ferrimagnétiques substitués par des ions de terres rares et de bismuth, on montre que la biréfringence optique Δn présente des contributions importantes Δn_σ et Δn_G associées aux biréfringences induites par contraintes élastiques (photoélastique) et par croissance par épitaxie qui dépendent de la composition chimique, de la température de croissance T_G d'épitaxie et de l'orientation des substrats. La dépendance de Δn_σ avec T_G est directement associée aux variations des coefficients de ségrégation des différents ions en substitution dans le grenat, ce qui change leur taux de substitution et partant, les écarts des paramètres de maille entre les films et leurs substrats. Par contre Δn_G apparaît comme indépendant de T_G . Des comportements anisotropes de Δn_σ ont été par ailleurs observés dans les films de Bi-YIG de plan (110). Les différentes contributions à Δn ont été déterminées et sont discutées à partir de modèles théoriques.

Abstract. — From optical wave-guide experiment in rare-earth and bismuth-substituted iron garnet films, it is shown that the optical birefringence Δn presents important contributions Δn_σ and Δn_G arising from stress-induced (photoelastic) and growth-induced birefringences which depend on the chemical composition, growth temperature T_G and orientation of the film. The dependence of Δn_σ on T_G is directly associated with the variation of the segregation coefficients of the different ions substituted in the garnet which changes their content and therefore the lattice mismatch between the films and the substrates. On the other hand Δn_G seems to be independent on T_G . In (110)-oriented Bi-YIG films anisotropic behaviours have been observed for Δn_σ . The different contributions to Δn are determined and discussed from theoretical models.

Introduction.

Nonreciprocal device in integrated optics imply high value for the maximum TE-TM modes conversion ratio $R_m = K^2 / [K^2 + (\Delta\beta/2)^2]$ where K and $\Delta\beta$ are the coupling coefficient and the phase mismatch between the modes [1]. Full conversion is obtained ($R_m = 1$) when $\Delta\beta = 0$ which is not the case for most of epitaxial garnet films where an effective phase mismatch $\Delta n_e = n_{TE} - n_{TM}$ is observed even in optically isotropic films. Different methods have been proposed to achieve high mode conversion efficiencies in magneto-optic waveguides [2]. As shown in figure 1, $\Delta\beta = 0$ when the TE and TM mode presents a crossing

point which is the case in optically anisotropic films with different asymptotic values n_x and n_z of the effective modes indices. In addition to the linear magnetic birefringence or Cotton-Mouton effect, Hepner *et al.* have suggested the use the stress-induced birefringence $\Delta n_\sigma = n_{TE}^\sigma - n_{TM}^\sigma$ [3] to produce anisotropic film and therefore modes degeneracy. Conversion ratios higher than 97 % have been demonstrated by Courtois *et al.* [4] in films whose the stress-induced birefringence Δn_σ cancels the isotropic phase mismatch Δn_i such as $\Delta n_e = \Delta n_i + \Delta n_\sigma = 0$.

In the present paper we have determined the different contributions to the TE-TM modes phase mismatch in rare earth and bismuth-substituted YIG films grown

Fig. 1. — Effective TE and TM modes indices in optically anisotropic film.

from LPE method on (111)- and (110)-oriented $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ (GGG) substrates. It is shown that the phase mismatch can be described by four main terms associated respectively with isotropic Δn_i and anisotropic Δn_a contribution such as :

$$\left. \begin{aligned} \Delta n_e &= \Delta n_i + \Delta n_a \\ \Delta n_a &= \Delta n_\sigma + \Delta n_G + \Delta n_{CM} \end{aligned} \right\} \quad (1)$$

where Δn_G and Δn_{CM} are the growth-induced birefringence and the linear magnetic birefringence or Cotton-Mouton effect. As discussed later Δn_{CM} is one or two orders smaller than Δn_σ and Δn_G and therefore the main contributions to the anisotropic phase-mismatch arise from the photoelastic and the growth-induced birefringence.

The relative values of the different contributions are strongly dependent on the chemical composition, the growth temperature T_G and the film orientation. The important contributions to Δn_e from Δn_σ and Δn_G are determined and discussed from theoretical models.

Results.

Film of different compositions corresponding to the systems $(\text{YbPr})_3(\text{FeGa})_5\text{O}_{12}$ and $(\text{BiGdYbY})_3(\text{FeAl})_5\text{O}_{12}$ were grown from usual LPE method on (111)- and (110)-oriented GGG substrates respectively. The optical phase mismatch, the unit cell parameter $a_\perp$ of the films, the deformation difference $(u_\perp - u_\parallel) = (a_s - a_\perp)/a_s$, where a_s is the lattice parameter of the substrate, and the Curie temperature T_c have been determined for different growth temperature T_G as shown on figures 2 to 4. $a_\perp$ is determined from X-ray technique with accuracy better than 5×10^{-4} Å.

Fig. 2. — Deformation dependence of the anisotropic birefringence $\Delta n_a = (n_{TM}^2 - n_{TE}^2)$ in (111)-oriented films.

Fig. 3. — Deformation dependence of the anisotropic birefringence Δn_a in (110)-oriented films.

Fig. 4. — Growth temperature dependences of films deformation and Curie temperature.

Therefore the accuracy for $(u_{\perp} - u_{\parallel})$ is better than 3×10^{-5} . For each chemical system the films were grown from the same melt solution at different T_G which corresponds to different compositions and film deformations. A light beam at 1.15 micron wavelength produced by an He-Ne laser was introduced into the films by using the prism method [5]. The angles of the incident light on the prism face, which depend on the excited m -order mode, were measured. Modes were excited under constant coupling value which implies that the optical gap between the film and the prism was taken into consideration for calculation in accordance with the method used in [6]. From the equations of the wave-guided propagation of light in anisotropic media [7], the refractive indices were determined for TE and TM mode separately and the effective phase mismatch or optical birefringence deduced from their difference $\Delta n_e = (n_{TM} - n_{TE})$ with accuracy better than one part to 10^4 .

The anisotropic birefringence Δn_a is deduced from equation (1) since the isotropic term Δn_i can be calculated. As shown on figures 2 and 3, Δn_a presents linear dependences on film deformation $(u_{\perp} - u_{\parallel})$.

The birefringence has anisotropic behaviour in (110)-oriented film (Fig. 3) since it depends on light propagation direction in the film plane. That is not the case in (111)-oriented films since it was found not to depend, in the experimental limits, on light direction. As seen in figure 2 the experimental points for YbPrFeGa-garnet system fit well a straight line going through the co-ordinate origin. Data are reported, for comparison, from results obtained in pure YIG and some rare-earth (Tb, Gd, Eu)-substituted YIG films grown previously on (111)-GGG substrates [7, 8].

In (110)-oriented Bi-based film under compression $(u_{\perp} - u_{\parallel} > 0)$ Δn_a presents different linear dependences on deformation when light is propagating along [001] and [110] direction. The straight lines are not going through the coordinate origin and cut equal segments on the ordinate axis which points to the presence of a non-photoelastic contribution to the birefringence. Strong deviation from linearity is observed in extension region $(u_{\perp} - u_{\parallel} < 0)$ and will be discussed later.

Discussion.

Two contributions to Δn_a are known to be independent on deformation : the magneto-optical one or Cotton-Mouton effect which is of the second order in magnetization components and the growth-induced one as observed recently by Ando *et al.* in (111)-oriented Bi-based iron garnet films [9]. The greatest linear magnetic birefringence at room temperature and 1.15 micron wavelength amounts from 10^{-5} to 10^{-4} ($\Delta n_{CM} = 6.5 \times 10^{-5}$ in pure YIG) which is between one and two orders of magnitude less than the birefringence measured in our Bi-ferrimagnetic films having zero deformation. Applied magnetic field up to 3 kOe did not change the value of Δn_a , so we can conclude that

the main contribution from non-photoelastic effects to the birefringence arises from a growth-induced birefringence ($\Delta n_a \simeq \Delta n_{\sigma} + \Delta n_G$).

• **STRESS-INDUCED BIREFRINGENCE.** — In (111)-films the stress-induced birefringence is described from the photoelastic constant P_{44} by the relation :

$$\Delta n_{\sigma} = n_0^3 P_{44}(u_{\perp} - u_{\parallel}) \quad (2)$$

where n_0 is the refractive index from isotropic films ($n_x \sim n_y \sim n_z \sim n_0$). If dependences in non-deformed films of n_0 and P_{44} on composition are small, direct proportionality of n_e on deformation can be expected as really observed from experiment in YbPrFeGa-system with P_{44} value equal to -0.052 ± 0.005 [10]. From data obtained previously in YIG : TbAl, YIG : GdGa, and YIG : EuAl [7, 8] similar proportionality is obtained with $P_{44} = -0.045 \pm 0.005$. This smaller value can be explained from the different experimental procedures used for their determination. As a whole P_{44} values obtained from our wave-guided measurements agree with those obtained by other methods [11-13].

This deformation increases by decreasing the growth temperature (Fig. 4) due to the segregation coefficient which becomes higher for the large ions as Bi^{3+} , Gd^{3+} , Pr^{3+} and becomes lower for the small ions as Al^{3+} and Ga^{3+} . Therefore, by decreasing T_G the content of large ions increases which induces an increase of the deformation $(u_{\perp} - u_{\parallel})$ from extension to compression with a sign change at $T_G = 915^\circ\text{C}$ for the Bi-YIG films. The dependence of the segregation coefficient on T_G explains also an increase of the Curie temperature at low T_G due to the lower content of Al^{3+} ions in the ferric sites and the higher content of Bi^{3+} in the dodecahedral sites which stimulates the super-exchange field in the garnets [14].

The photoelastic contribution to Δn_a in (110)-films can be described for light propagating along the [001] and [110] directions by :

$$\left. \begin{aligned} \Delta n_{\sigma} [001] &= -n_0^3 P_{44}(u_{\perp} - u_{\parallel}) \\ \Delta n_{\sigma} [110] &= -n_0^3 \left(P_{44} + \frac{\Delta P}{4} \right) (u_{\perp} - u_{\parallel}) \end{aligned} \right\} \quad (3)$$

with $\Delta P = P_{11} - P_{12} - 2 P_{44}$.

From data of figure 3 the photoelastic coefficients in (110)-film are deduced such as : $P_{44} = 0.059 \pm 0.005$, $\Delta P = +0.085 \pm 0.005$ and $P_{11} - P_{12} = -0.083 \pm 0.05$.

• **GROWTH-INDUCED BIREFRINGENCE.** — The origin of the strain independent contribution to the birefringence has to be connected to non-cubic distortion of film crystal lattice induced during the growth process as observed previously in bulk TbIG by Dillon *et al.* [15]. It is known that such distortions are at the origin of the growth-induced uniaxial magnetic anisotropy K_u^G in (111)- and (110)-oriented epitaxial garnet films

Fig. 5. — Growth temperature dependences of anisotropic birefringences in (110)-oriented films.

as used for magnetic bubble stability. On the other hand a magnetocrystalline orthorhombic anisotropy K_0 has been observed in Bi-based (110)-oriented film only [10], similarly to the anisotropic behaviour for light propagation in our (110)-oriented film as shown in figure 3 with $\Delta n_a[001] \neq \Delta n_a[110]$.

The linear dependence of the birefringence on deformation has to mean that the magnitude of Δn_G is independent on T_G . There is no data to our knowledge on the behaviour of optical anisotropy when T_G is changing but the growth-induced magnetic anisotropy is known to decrease sharply when sufficiently high T_G is used. If it is assumed that Δn_G behaves similarly, one can explain the deviations from linearity for Bi-substituted films grown at the highest $T_G = 932^\circ\text{C}$ (extension region in Fig. 3).

The (111)-oriented YbPrFeGa films has a strong growth-induced magnetic anisotropy of easy-plane type as indicated by their magnetization saturation fields parallel (~ 10 Oe) and perpendicular (~ 10 kOe) to their plane independently of the sign of the deformation [16]. It is noteworthy that high Ku^G values are not followed in these compounds by appearance of noticeable Δn_G . From the tensor of growth-induced anisotropy one can obtain an expression for birefringence in (111)-oriented films such as :

$$\Delta n_G(\gamma_1 \gamma_2 \gamma_3) = \frac{\Delta C}{2n_0} \left[2R_{44}\gamma_1^2 + \left(R_{12} - R_{11} + \frac{5}{6}\Delta R \right) \gamma_2^2 + \frac{\Delta R}{6}\gamma_3^2 + \frac{\sqrt{2}}{3}\Delta R\gamma_1\gamma_2 \right] \quad (4)$$

with $\Delta R = R_{11} - R_{12} - 2R_{44}$, where ΔC is the difference of rare-earth ions concentration in non-equivalent dodecahedral positions, γ_i are direction cosines of the direction of crystal growth and R_{ij} are constants of growth-induced anisotropy tensor. For a crystal growth along [111] direction ($\gamma_1 = 1$; $\gamma_2 = \gamma_3 = 0$) Equation (4) is reduced to $\Delta n_G = R_{44} \Delta C$ and

this quantity must be equal or close to zero in accordance with experiment. The fact that $Ku^G \neq 0$ implies $\Delta C \neq 0$ and therefore it is conclude that $R_{44} = 0$ in our (111)-oriented films.

In (110)-oriented films the growth-induced birefringence is described for light propagating along the [001] and [110] directions by :

$$\left. \begin{aligned} \Delta n_G[001] &= n_0^{-1} R_{44}(\gamma_1^2 - \gamma_2^2) \Delta C \\ \Delta n_G[110] &= n_0^{-1} \left(R_{44} + \frac{\Delta R}{4} \right) (\gamma_1^2 - \gamma_2^2) \Delta C \end{aligned} \right\} \quad (5)$$

It is observed from experiment equal values of Δn_G independently of the light propagation direction in the film, which requires $\Delta R = 0$. The dependence of $\Delta n_a[001] - \Delta n_a[110]$ on deformation in (110)-film is given in figure 6. Since Δn_G is isotropic, such a difference is associated only with photoelasticity and therefore the absence of deviation from linearity in region of film extension supports the hypothesis made above concerning the decrease of Δn_G when T_G is going up to the melt saturation temperature.

Conclusion.

From optical wave-guide investigation methods it has been possible to separate the main contributions to the anisotropic modes phase-mismatch associated with stress-induced and growth-induced birefringences. In (110)-oriented Bi-based films the stress-induced birefringence has anisotropy similar to the magnetic orthorhombic anisotropy observed in these compounds.

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Fig. 6. — Deformation dependence of the birefringence anisotropy $\Delta n_a[100] - \Delta n_a[110]$ in (110)-oriented film.

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