BI-DYIG FILMS: ANALYSIS OF THE DY CONTRIBUTION TO THE FARADAY ROTATION.

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ABSTRACT

At \( \lambda = 633 \) nm, very accurate measurements of the Faraday rotation on epitaxial garnet films of the composition \( \{R_{3-x}Bi_{x}\} \{Fe_2\} \{Fe_3\}O_{12} \) with \( R = \text{Dy}, \ \text{Y} \) and high \( Bi \) content \( (x \approx 1.30) \) are reported, in the temperature range \( 10 - 620 \) K under magnetic field up to \( 20 \) kOe applied along the \( [111] \) direction. The analysis of the data in terms of the sublattice magnetization reveals, at low temperatures, a large contribution of the rare earth \( \{c\} \) sublattice induced by the \( Bi \) substitution. This enhancement of the "electric" dipole Faraday rotation is strongly temperature dependent and vanishes at room temperature. This result is compared with that observed for the same \( Bi \)-substituted rare earth iron garnets in the near infra-red band.

KEYWORDS: Faraday rotation, Rare-earth iron garnets, Bismuth substitution, Near infra-red.

Introduction

The oxide magnetic films present strong MO effects and are high in corrosion resistance and good in reproducibility of film preparation. However for low cost production, they have the disadvantage to be available only in polycrystalline form on glass substrates which causes important media noise induced by grain boundaries. In spite of this serious problem which is not fully solved, oxide films remain an interesting contender for the next generation MO recording media to succeed to the amorphous rare-earth-transition materials. Besides MO recording, oxide films are potential candidates to various devices such as isolator, optical amplifier, magnetic field sensor, etc... and different types of materials have been considered with a large attention paid to the Bi-rare-earth iron garnet films [1].

Among the different substitutions in Rare Earth Iron Garnets (REIG), diamagnetic \( Bi^{3+} \) and \( Pb^{2+} \) ions and some lighter rare-earth ions like \( Ce^{3+}, Pb^{3+} \) and \( Nd^{3+} \) enhance strongly the magneto-optical (MO) activity in the wavelength range 0.4-2 microns ([2][3][4] and ref. therein). It is worth noting that the large Faraday rotation (FR) is observed, in the Bi-substituted garnets, without a noticeable increase of the optical absorption.

In the visible range, relevant investigations of garnet films of composition \( Y_{3-x}Bi_{x}Fe_{5}O_{12} \) have been reported earlier by Hansen et al. [5]. Some years later, such studies have been extended to the infra-red range \( (\lambda = 1150 \) nm) in order to confirm that the \( Bi \) ions stimulate the MO activity of the \( Fe^{3+} \) ions at both the octahedral and tetrahedral sites of the garnet [2].

For different rare-earth iron garnets, the substitution of \( Bi \) has led to the following conclusion: the bismuth affects the iron transition only, the enhancement of the iron contribution to FR being independent of the nature of the rare-earth ion and that at \( \lambda = 633 \) nm [6].

In this article, we intend to probe the intrinsic MO properties of garnet films of the composition \( Dy_{3-x}Bi_{x}Fe_{5}O_{12} \) and \( Y_{3-x}Bi_{x}Fe_{5}O_{12} \) for \( x \approx 1.30 \) and \( \lambda = 633 \) nm to quantitatively analyse the effect of bismuth presence on the MO activity of the \( Dy^{3+} \) ion. It is worth noting that the behaviour of these films, at \( \lambda = 1150 \) nm, has been recently published [7].

The \( Dy^{3+} \) ion has been chosen because of its large negative magnetostriiction constant which induces a positive uniaxial anisotropy favorable for MO recording. It is briefly recalled that the magnetic anisotropy is caused by crystalline anisotropy and (or) stress induced anisotropy. Because the garnets have a very small magneto-crystalline anisotropy around the room temperature, only the stress-induced anisotropy contributes to magnetic anisotropy. We have sought to increase the amount of \( Bi \) using epitaxial thin films.

It is worth noting that, to our knowledge, this is the first report on \( DyBiG \) epitaxial films in the literature, since, surprisingly, the work of Hansen et al. [6]
performed on different Bi-substituted rare earth iron garnet at \( \lambda = 633 \text{ nm} \) contains no information on dysprosium garnets. It is mentioned that the optimization of the material parameters of Dy, Bi and Ga garnet films produced by classical rf diode sputtering has been analyzed in a previous paper [8].

**Experimental**

Garnet films of the composition of \( \text{Dy}_{3-x}\text{Bi}_x\text{Fe}_2\text{O}_{12} \) (DyBiIG) and \( \text{Y}_{3-x}\text{Bi}_x\text{Fe}_2\text{O}_{12} \) (YBiIG) were grown by liquid phase epitaxy onto (111) oriented cation-doped gadolinium gallium garnet (Gd\(_3\)Ga\(_5\)O\(_{12}\)) substrates with a lattice parameter equal to 12.498 Å. The amount of Bi achieved in the two preparations was determined by electron probe analysis and found equal to 1.27 ± 0.03 (DyBiIG) and 1.30 ± 0.03 (YBiIG). (see ref. [7]).

Very accurate FR measurements have been performed at 633 nm wavelength in the 10-620 K range and under magnetic field along the direction [111] up to 20 kOe using polarization modulation techniques. On one hand, a special attention was paid during the experiments to eliminate the influence of internal reflections and multilayer structures on the measurements (ref. given in [7]). On the other hand, all the data have been corrected of the window and substrate contributions.

Within the general formula \( \{\text{RE}_3\}\{\text{Fe}_2\}\{\text{Fe}_3\} \text{O}_{12} \), the trivalent Dy and Bi ions are distributed over the dodecahedrally coordinated (24c) sites, while the iron ions occupy the octahedral (16a) and tetrahedral (24d) sites. The magnetic properties result from the ferrimagnetic arrangement of the three corresponding sublattice magnetizations \( M_c, M_a \) and \( M_d \); \( M_a \) and \( M_d \) are strongly coupled antiferromagnetically and \( M_c \) is antiparallel to the resultant \( \text{Fe}^{3+} \) magnetization. The Curie points, \( T_c \), of different pure REIG's are the same as that of YIG. Furthermore in heavier rare-earth garnets, the macroscopic garnet magnetization which is given by \[ M_c - M_{111} \] vanishes at the compensation temperature \( T_{\text{comp}} \). In pure DyIG, \( T_{\text{comp}} \) is equal to (223 ± 1) K [9].

**Results and Discussion**

The figure 1 shows a lot of isothermal variation of FR versus applied field which are found linear in the whole temperature range; such a variation is observed, for the DyBiIG film, when \( H > 2 \text{ kOe} \) but above 3 kOe only for the YBiIG film. It is clear that for technological accessibility, a low saturation field is desirable for the garnet film. The spontaneous Faraday rotation, \( \text{FR}^S \), defined as the FR associated with the spontaneous ferrite magnetization was immediately deduced using a least squares refinement technique.

The temperature dependences of the FR\(^S\) that are displayed in Fig.2 confirm that the MO properties are strongly affected by the bismuth incorporation and that below room temperature. \( T_c \) was found to be (600 ± 5)K for the two studied films, this value is in perfect accordance with that deduced from the study of YBiIG film by Hansen et al. [10]. However this change \( \Delta T_c/\chi \) of 34 K/Bi/formula unit, observed in thin films, remains 10% below the value of the bulk [11].

According to the sign convention \( \text{FR (YIG)} > 0 \), the FR of the the DyBiIG film presents a large positive value (28000 deg.cm\(^{-1}\) at 6 K) and is weakly temperature dependent. Furthermore the absolute value of FR\(^S\) of this film is smaller than that of the BiYIG film at \( T < 300 \text{ K} \) as shown on Fig. 2. This result indicates that, at least at low temperatures, the contribution of the Dy sublattice is important as observed for Bi-free DyIG single crystal at \( \lambda = 1152 \text{ nm} \) [9].

At (110 ± 5) K, there exists a compensation temperature \( T_{\text{comp}} \) causing a change of sign of FR\(^S\) as found in the infra-red range [7]. Note that in the Fig.2, we have changed the sign of FR\(^S\) values measured on YBiIG films in respect to the change of orientation of the iron sublattices in DyBiIG at \( T_{\text{comp}} \). Furthermore it
should be pointed out that the Bi free DyIG exhibits a higher $T_{\text{comp}}$ value ($220 \pm 2$ K). The shift of $T_{\text{comp}}$ towards lower temperatures with increasing bismuth content results from the presence of diamagnetic Bi$^{3+}$ ions in the $\{c\}$ sublattice.

At room temperature, the contribution of the Dy sublattice is very small and enters within our experimental accuracy as shown in Fig. 1; the main and important effect of the Dy presence is to induce a magnetic easy axis normal to the film. Near 300 K, FR amounts -2.65 deg.cm$^{-1}$ for both films, somewhat smaller than those (-5.5 deg.cm$^{-1}$) of the YBiIG films with the highest Bi content prepared by Okuda et al. [12] using ion beam sputtering.

In pure YIG, FR at $\lambda = 633$ nm is equal to +835 deg.cm$^{-1}$ at room temperature [13], it increases in both films by about -21000 deg.cm$^{-1}$ by substitution of one Bi atom/fu. Around 6 K, in Bi-free YIG, FR lies in the order of +320 deg.cm$^{-1}$, $\Delta FR/\Delta x$ is observed to be equal to -26300 deg.cm$^{-1}$/Bi/fu. and we can conclude that the MO intrinsic efficiency of Bi is strongly temperature dependent. A similar behaviour has been observed at $\lambda = 1152$ nm [7] where the Bi efficiency is 10 times smaller than at $\lambda = 633$nm.

Assuming that the contribution to the Faraday rotation of DyBiIG by iron ions is simply represented by that of BiYIG measured in the same experimental conditions [14], the total contribution to FR of the Dy$^{3+}$ sublattice is given by:

$$ FR (\text{Dy}^{3+}) = FR (\text{BiDyIG}) - FR (\text{BiYIG}) = \pm (C_e + C_m)|M_0| \quad (1) $$

where $C_e$ and $C_m$ are the respective magneto-optical electric "e" and magnetic "m" dipole coefficients. The upper (rep lower) sign is valid for $T < T_{\text{comp}}$ (rep $T > T_{\text{comp}}$). As in the $\{c\}$ sublattice, only the Dy$^{3+}$ ions are magnetic, $C_m$ is small (6.1 deg.cm$^{-1}$ $\mu_B^{-1}$ (references reviewed in [14]) and the magnetic contribution of the rare-earth sublattice can be neglected. The "e" dipole transitions contribution is then deduced from Eq.(1) and reported in Fig. 3. $M_0$ is known from magnetization measurements, and is generally reported in Bohr magneton per two formula units. At low temperatures the dodecahedral coefficient is increased by about -253 deg.cm$^{-1}$ $\mu_B^{-1}$ for a Bi substitution equal to 1.3. It remains roughly temperature independent up to about 160 K and decreases rapidly to zero near room temperature.

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garnet films has been studied. The comparison with the data obtained on films of the composition Bi$_3$Y$_2$Fe$_5$O$_{12}$ indicates that the replacement of yttrium by dysprosium gives rise to significant changes in the FR mainly at low temperatures. The observed data indicate that i) the bismuth affects strongly the "electric" FR of the Dy$^{3+}$ sublattice; ii) this Bi-induced change is temperature dependent; iii) to our knowledge, this is the first report on DyBiIG epitaxial films in the literature in the whole temperature range.

At the end of this paper, it is useful to discuss briefly the possible pollution by lead of our films, since the Pb$^{2+}$ contribution to FR was found to be comparable, in the visible range, with that of bismuth [15]. The amount of lead is too weak to affect the discussion developed in this paper: for the following points i) electron probe analysis has not revealed any trace of lead, ii) transparency of our films is very high (Pb$^{2+}$ ions increase the optical absorption much more than Bi, iii) monotonic decrease of FR with the temperature of the YBiIG films; (a small amount of lead would induce an additional minimum on the FR variation at low temperatures).

References