Thin Solid Films 520 (2012) 3647–3650

Contents lists available at SciVerse ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Green and blue magneto-optical photonic crystals

A.M. Grishin *, S.I. Khartsev

Department of Condensed Matter Physics, KTH Royal Institute of Technology, SE-164 40 Stockholm-Kista, Sweden

ARTICLE INFO

Article history:
Received 27 April 2011
Received in revised form 17 December 2011
Accepted 21 December 2011
Available online 30 December 2011

Keywords:
Bismuth/rare-earth iron garnet
All-garnet heteroepitaxial film
Faraday rotation

ABSTRACT

A series of one-dimensional heteroepitaxial all-garnet magneto-optical photonic crystals (MOPCs) were pulsed laser deposited to operate at 550 and 470 nm wavelength. We explored the concept of blue shift of the optical absorption edge of ferric ions by substituting Fe with Ga on the tetrahedral sites as well as Bi and Y, respectively, with Ca and Ce at the dodecahedral coordinated positions. 17-layered [$Y_2Ce_1Fe_5O_{12}$/$Gd_3Ga_5O_{12}$] MOPC with a total thickness of 968 nm demonstrates superior magneto-optical performance: Faraday rotation $\Theta_{Fmax} = +2.0°$ and transmittance as high as 0.35 at the resonance wavelength of 470 nm.

1. Introduction

Bismuth-substituted rare-earth iron garnet $RE_{3-x}Bi_xFe_5O_{12}$ which is transparent in visible and near infrared light is a foremost material for various magneto-optical applications [1]. Specific Faraday rotation (FR) linearly grows with increasing number x of Bi atoms: $\theta_F = -2.06 \times x$ deg/μm at 633 nm [2,3]. However, Bi^{3+} ion radius, as large as 1.25 Å [4,5], leads to a severe increase of lattice constant a hence obstructs the synthesis of garnet crystals with a high bismuth content. Long time it was believed that the maximum lattice constant for any iron-unsubstituted garnets is 12.540 Å [6,7]. Based on this assumption, one could use experimentally obtained linear relation [8]:

$$a(x) = a(0) + k \times x = \begin{array}{r} 12.375\,\text{Å} + 8.5 \times 10^{-2}\,\text{Å} \times x \quad \text{for} \quad Y_{3-x}Bi_xFe_5O_{12}, \\ 12.476\,\text{Å} + 5.8 \times 10^{-2}\,\text{Å} \times x \quad \text{for} \quad Gd_{3-x}Bi_xFe_5O_{12}, \\ 12.289\,\text{Å} + 11.0 \times 10^{-2}\,\text{Å} \times x \quad \text{for} \quad Lu_{3-x}Bi_xFe_5O_{12} \end{array} \quad (1)$$

to calculate the maximum permitted bismuth content $x_{max} = 1.9$, 1.1, and 2.3 for Bi-substituted yttrium, gadolinium and lutetium iron garnets, correspondingly. It seemed it is impossible to grow completely substituted $Bi_3Fe_5O_{12}$ garnet.

A "light at the end of the tunnel" appeared when V.J. Fratello et al. had synthesized unsubstituted $Nd_3Fe_5O_{12}$ and $Pr_3Fe_5O_{12}$ garnets with the lattice parameters as large as 12.595 Å and 12.646 Å, respectively [9,10]. The authors contrived low temperature liquid phase epitaxy technique to grow thick films on the large lattice parameter $Sm_3Sc_yGa_{5-y}O_{12}$ substrates and even bulk 60 mm^3 crystals using cracked film pieces as a seed. Using N. Kimizuka calculations of the heat of the garnets formation [11], they understood the growth temperature must be lowered to gain negative Gibbs free energy of garnet phase compared to the orthoferrite one. There were no reports on theoretical analysis of the same strategy for $Bi_3Fe_5O_{12}$ garnet. Also, nobody succeeded to grow iron-unsubstituted $RE_{3-x}Bi_xFe_5O_{12}$ films with $x > 1.3$–1.5 from a liquid phase onto available garnet substrates.

Pure bismuth iron garnet $Bi_3Fe_5O_{12}$ films (hereinafter BIG) were first prepared by reactive ion beam sputtering by T. Okuda [12] and later by robust pulsed laser deposition technique [13,14]. In laser plasma, due to a high kinetic energy and excited state of ablated species, film growth occurs at a very fast rate far away from the thermodynamic equilibrium. This enables synthesis of unique garnet compositions, like $Bi_3Fe_5O_{12}$ ($a = 12.627$ Å) [14] and $La_3Ga_5O_{12}$ ($a = 12.768$ Å) [15], which do not exist in the bulk form, at relatively low temperatures of 525 °C and 830 °C, respectively. Being deposited onto better matched $Gd_3(Sc,Ga)_5O_{12}$ crystal, BIG holds a record value of FR $\theta_F = -8.4$ deg/μm at 633 nm [12,14].

Further increase of FR became possible through a light localization in magneto-optical photonic crystals (MOPCs). Theoretical idea to enhance FR by placing MO material in a resonant cavity belongs to R. Rosenberg [16]. Due to the non-reciprocity, Faraday effect has a multiplicative character, i.e. FR increases by a factor of N, where N is a number of times light experiences reflections between Bragg mirrors. M. Inoue built the first MOPC composed of the $[SiO_2/Ta_2O_5]^6$ distributed Bragg reflectors and polycrystalline $(Dy,Bi)_3Fe_5O_{12}$ garnet microcavity in between [17]. The ultimate magneto-optical performance is reached in all-garnet heteroepitaxial MOPCs where FR accumulates both in MO-active layers in microcavity and Bragg reflectors [18]. Currently, all-garnet heteroepitaxial [$Bi_3Fe_5O_{12}/Sm_3Ga_5O_{12}$] MOPCs grown by rf-magnetron sputtering demonstrate the best MO performance achieved so far in visible and near infrared ranges. E.g. 21-layered 2.4 μm thick 980-$[Bi_3Fe_5O_{12}/Sm_3Ga_5O_{12}]^5Bi_3Fe_5O_{12}^2$

* Corresponding author at: KTH Royal Institute of Technology, SE-164 40 Stockholm-Kista, Sweden. Tel.: +46 8 790 4176; fax: +46 8 752 7850.
E-mail address: grishin@kth.se (A.M. Grishin).

0040-6090/$ – see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.tsf.2011.12.062

$[Sm_3Ga_5O_{12}/\ Bi_3Fe_5O_{12}]^5$ MOPC [19] showed net FR as high as −8.4° and transmittance of 0.68 at the resonance wavelength of 980 nm. That was 630% enhancement of FR compared to a single layer BIG film with the same thickness of 1.15 μm as a total thickness of BIG layers in the MOPC [20–22].

Multiple light reflections in transparent photonic crystals increase effective optical path length. Therefore, net Faraday effect in MOPCs is determined both by intrinsic FR and a transmissivity of crystal's constituents. Although at 537 nm BIG shows a peak value of FR as high as −28°/μm, the accompanying strong optical absorption hinders its practical application. It is a challenging task to engineer MO crystals high performing in green and blue light spectra.

It is well known that reduction of bismuth content and substitution of the iron ion by other trivalent diamagnetic ions cause a strong decrease of light absorption (e.g. [23–25] and references therein). Also, both high FR and transparency were observed in yttrium iron garnets substituted by cerium instead of bismuth [26–28]. In this paper we explore three different scenarios fabricating and comparing properties of pulsed laser deposited all-garnet MOPCs with Bi diluted by Ca, Fe diluted by Ga, and using instead of bismuth Ce-substitution for yttrium at the dodecahedral iron garnet sites.

2. Experimental details

The details of pulsed laser deposition of all-garnet heteroepitaxial MOPCs have been published elsewhere [18,29]. In brief, Compex 102 KrF eximer 248 nm laser was used to ablate targets placed at the distance of 55 mm from $Ga_3Gd_5O_{12}$ (GGG) single crystal substrates. Laser energy density and repetition frequency were 2–3 J/cm^2 and 20 Hz, respectively. Dense $Bi_2Ca_1Fe_5O_{12}$ (Ca:BIG), $Bi_3Fe_{4.25}Ga_{0.75}O_{12}$ (Ga:BIG), $Y_2Ce_1Fe_5O_{12}$ (Ce:YIG) and overstoichiometric $Bi_{3.15}Fe_5O_{12}$ composite oxide targets were prepared by conventional ceramic technology [14]. Meanwhile, we used $Ga_3Gd_5O_{12}$ single crystal as a target for MO non-active layers in Bragg mirrors. The optimal oxygen background pressure and substrate temperature were found to be 6.7 Pa and 600–620 °C for [BIG/GGG], [Ga:BIG/GGG] and [Ca:BIG/GGG] heteroepitaxial film structures while for [Ce:YIG/GGG] multilayers they were, respectively, 9.3 Pa and 790 °C. All depositions were accomplished by postannealing in situ in oxygen at 67 kPa close to the deposition temperatures.

First, reference epitaxial films were deposited to optimize growth conditions and to determine refractive indices $n(\lambda)$ and deposition rates of GGG and iron contained garnets. Then, MOPCs for the resonance wavelength λ_{res} were grown with Bragg reflectors having thickness of $\lambda_{res}/4n(\lambda_{res})$ and a microcavity which thickness is a multiple of $\lambda_{res}/2n(\lambda_{res})$. All MOPCs were fabricated on GGG(001) substrates except [Ce:YIG/GGG] one grown onto GGG(111) crystal.

Four different photonic crystals were fabricated and tested: non-substituted 650-$[Bi_3Fe_5O_{12}/Gd_3Ga_5O_{12}]^6$ superlattice, Ga-substituted 550-$[Ga:BIG/GGG]^3Ga:BIG^2[GGG/Ga:BIG]^3$, Ca-substituted 550-$[Ca:BIG/GGG]^3Ca:BIG^2[GGG/Ca:BIG]^3$, and Ce-substituted 470-$[Ce:YIG/GGG]^4Ce:YIG^4[GGG/Ce:YIG]^4$ MOPCs. Superscripts 4 in Ce:YIG^4, and 2 in Ca:BIG^2 and Ga:BIG^2 indicate, respectively $4\times\lambda_{res}/4n(\lambda_{res})$ and $2\times\lambda_{res}/4n(\lambda_{res})$ thicknesses of microcavities [19].

Dispersion spectra of the transmittance and FR were recorded in our homemade MO-spectrometer in perpendicular magnetic field $H\sim2.4\times10^5$ A/m which was sufficient to saturate MO-films [30]. FR vs. H hysteresis loops were traced at $\lambda=678$ nm with a rate of 380 Hz. To quantify a gain of the Faraday effect in the photonic crystal, we also grew corresponding reference single layer MO-film, measured specific FR θ_F[°/μm], and compared it with θ_F calculated as a net FR Θ_F normalized to the total thickness of MO-active layers in the MOPC.

3. Results and discussion

The upper frame of Fig. 1 presents transmittance and FR spectra in the reference 0.78 μm thick BIG film. Specific FR peaks to $\theta_{Fmax}=-28.1°/\mu m$ close to the absorption edge where BIG becomes very opaque at wavelengths shorter than 560 nm. Therefore, traditional design of MOPC with a resonant BIG microcavity unsuits green and blue light operation. The only option to slightly enhance FR in BIG contained crystals is to use Tamm surface states confined to the photonic crystal interface [31]. They exhibit themselves by enhanced light intensity (light localization) thus lead to an increase of FR at the edges of the stop band. To utilize this effect we sintered 650-$[BIG/GGG]^6$ MOPC as a quarter wavelength superlattice. MOPC transmittance is shown in the lower frame of Fig. 1. It has a stop band centered at designed resonant wavelength 650 nm and a local peak of FR $|\Theta_{Fmax}|=5.8°$ ($|\theta_{Fmax}|=16.6°/\mu m$) at the short wavelength edge of the stop band at $\lambda=588$ nm. This is only 18% increase compared to $|\theta_F^{BIG}(588\ nm)|=14.1°/\mu m$ in the reference BIG film hence other approaches to enhance net FR are highly desired.

We used experimental data for the transmittance T in Ga and Ca substituted films to calculate the absorption coefficient α [cm^{-1}] = ln(1/T)/film thickness and present it as a normalized light absorption $(h\nu\times\alpha)^2$ vs. wavelength λ in Fig. 2. It is clearly seen that in bismuth contained iron garnets, substitutions of Fe by Ga and Bi by Ca shift absorption edge to shorter wavelengths compared to BIG. Now, getting more transparent MO-films let us check how high FR of blue light they produce compared to unsubstituted BIG.

Fig. 3 depicts transmittance T and FR spectra in the reference Ga:BIG(1.48 μm) film and 550-$[Ga:BIG/GGG]^3Ga:BIG^2[GGG/Ga:BIG]^3$ MOPC with a $\lambda_{res}/2n(\lambda_{res})=94$ nm thick Ga:BIG microcavity. At the resonance wavelength $\lambda_{res}=550$ nm, FR $|\Theta_{Fmax}|=8°$ ($|\theta_{Fmax}|=$

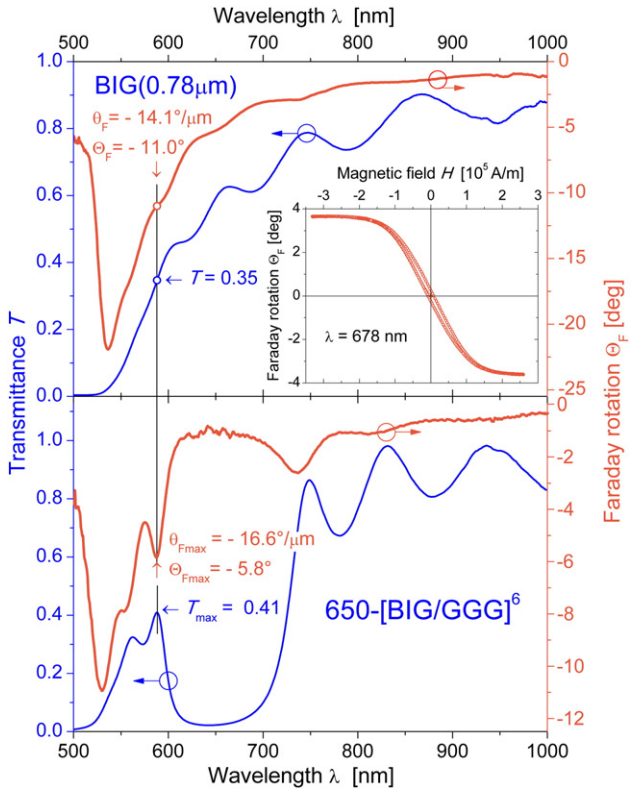

Fig. 1. Transmittance T and Faraday rotation (FR) Θ_F spectra of the reference 0.78 μm thick $Bi_3Fe_5O_{12}$ film (upper frame) and 650-$[Bi_3Fe_5O_{12}/Gd_3Ga_5O_{12}]^6$ magneto-optical photonic crystal (MOPC) designed for $\lambda_{res}=650$ nm. In MOPC, local maxima $|\Theta_{Fmax}|=5.8°$ and $T_{max}=0.41$ are reached at the short wavelength edge of the stop band at $\lambda=588$ nm. Inset shows FR vs. perpendicular magnetic field H hysteresis loop traced in BIG film at $\lambda=678$ nm.