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PII: S0167-577X(15)30441-9
DOI: http://dx.doi.org/10.1016/j.matlet.2015.08.085
Reference: MLBLUE19446

To appear in: Materials Letters

Received date: 29 June 2015
Revised date: 14 August 2015
Accepted date: 17 August 2015

Cite this article as: Zhe Chen, Lei Yang, Xiangyong Wang, Jun Wang and Yin Hang, Fabrication and characterizations of a erbium doped terbium gallium garnet crystal for faraday rotators, Materials Letters, http://dx.doi.org/10.1016/j.matlet.2015.08.085

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Fabrication and characterizations of a Erbium doped terbium gallium garnet crystal for Faraday rotators

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Abstract

High optical (Tb(1-x)Erx)3Ga5O12 (TEGG) single crystal has been grown by the Czochralski (Cz) technique. X-ray powder diffraction was carried out and lattice parameters were calculated, which showed that the single crystal belongs to cubic crystal system with high crystallinity. Observed optical characteristics and magneto-optical properties were compared with those of pure terbium gallium garnet (TGG) crystal at room temperature. The Verdet constant (V) dispersion of TEGG was evaluated in the visible and near-infrared region (VIS-NIR). The V of TEGG at 532nm comes up to 224.5 rad·m⁻¹·T⁻¹, obviously larger than that of TGG. The high thermal conductivity and laser induced damage threshold (LIDT) of TEGG further confirmed its great potential to be applied in the high power laser system.
Key word: Crystal growth, Crystal structure, Optical materials and properties, Magnetic materials

1. Introduction

Optical isolators (OIs) used to prevent optical feedback are the fundamental component of many contemporary optical-laser systems\cite{1-2}. In recent years, the requirement for Faraday isolators (FIs) has increased with continuous development of high-power laser-diodes and high-power fiber-lasers operating at the visible and near-infrared regions (VIS-NIR)\cite{3-5}. Yttrium iron garnet (Y$_3$Fe$_5$O$_{12}$:YIG) and Bi, Ce doped YIG crystals are the commonly used magneto-optical materials of optical isolators because of its large Faraday rotation angle, high transparency, and small absorption in the wavelengths of 1.2-5.0\textmu m\cite{6-8}. However, the conventional YIG crystals is not practical because of its very poor transmittance in the VIS-NIR, and terbium gallium garnet (TGG) is thought to be the better material among transparent magnetic materials due to its high transmittance in the VIS-NIR, high thermal conductivity and excellent size scalability.

The TGG single crystal can be easily grown by the Cz technique\cite{9-10} which can significantly reduce the intrinsic defects of the as-grown crystal, thus enhance the Faraday rotation in the gallium garnet material with rare earths\cite{11}. However, the comparative low Verdet constant of TGG (39-40 rad m$^{-1}$T$^{-1}$) leads to the development of magneto-active element (MOE) with superior magneto-optical properties. With the development of single crystals manufacturing techniques many of such media have been fabricated with good performance. For instance, terbium-scandium aluminum garnet (TSAG) and \{Tb$_3$][Sc$_{1.95}$Lu$_{0.05}$]Al$_3$O$_{12}$ single crystals\cite{12-13}. The good performance of FIs based on
such media in the high power laser system is provided by the MOE shortening and therefore, decreasing of the thermally induced radiation distortions, which is important for maintaining the high and effective performance of the optical system[14-15].

A number of papers show that the super-exchange interaction between two or more paramagnetic ions (Tb$^{3+}$ and other paramagnetic ions) can occur, significantly enhances the magneto-optical property in many optical media. Our research group has been fabricated terbium gallium garnet doped with cerium (Ce:TGG)[16], Praseodymi (Pr:TGG) [17], and neodymium(Nd:TGG) crystals with 20-30% larger of Verdet constant compared to that of TGG.

Tb$^{3+}$, Er$^{3+}$ ions have been pointed out to show large effective magnetic moment among the rare earth ions and Er doped TGG crystal has not been grown or explored its magneto-optical property, we suspect that a lager Faraday rotation angle of Er$^{3+}$ doped TGG can be obtained due to the strong magnetic moment of Tb$^{3+}$-Er$^{3+}$ super interactions. So in this letter, we first gives the optical, magneto-optical characteristics measurements results of a new medium–Erbium doped TGG crystal (Er:TGG) by Czochralski method.

2. Material and methods

High-purity Tb$_3$O$_7$, Er$_2$O$_3$, Ga$_2$O$_3$ (5N) chemicals were mixed according to the designed Tb$_{3.3}$Er$_x$Ga$_5$O$_{12}$, then the mixture of powders(1-2% excess of Ga$_2$O$_3$) were pressed to sheet and sintered for 12 h at 1200-1400°C. Such formed materials were crushed, re-mixed and sintered again at 1200-1400°C for 24 h. Finally, we obtained polycrystalline materials. The crystals were grown by the Cz method in a iridium crucible with radio frequency (RF) induction heating. After that crystals growth were carried out under high purity N$_2$ (99.99%)
atmosphere. They were grown in the <111> orientation at a pulling rate of 1.0mm/h and a rotating rate of 10-15 rpm. Finally, Tb$_3$Er$_x$Ga$_5$O$_{12}$ and TGG crystals were obtained.

The TEGG and TGG were double-side polished for the measurements, as shown in the insert of Fig.1(a). The absorption spectrum was measured using a Perkin-Elmer Lambda 900 UV-VIS-NIR spectrophotometer at room temperature. The thermal conductivity of the sample with the best quality was measured by the well-known flash method on a Xenon Flash Apparatus (LFA447/1 NanoFlashR300, Netzsch, Germany).

The bulk laser damage threshold was measured using common testing conditions (Nd:YAG laser system, 1064nm wavelength, 12ns pulse duration, 1-on-1 test). The magneto-optical Faraday rotation of the crystal was measured at room temperature by the extinction method using the magneto-optical test system. Laser of 1064nm was used as the sources of the probe beam. The magnetic field could be adjusted from 0 to 1.2T continuously. Several length of the samples obtained from different part of the crystals to confirm the reproducibility of the materials and a 15 mm length TGG sample (CASTECH Corp) was used for comparison and calibration.

3. Experimental results

The Er:TGG crystal was analyzed by powder XRD and shows no impurity phase, as shown in Fig.1. X-ray powder diffraction patterns were indexed; The result indicated that doped with Er$^{3+}$ ion did not influence the crystal structure of the sample, which could be used to adjust the properties of TGG crystals. The lattice parameter of the crystal was calculated from the powder XRD data to be a=b=c=1.2346nm, which is slightly smaller than the pure TGG (a=b=c=1.2355nm). It can be explained by the reason that the radius of Er$^{3+}$ is
smaller than the radius of Tb$^{3+}$. The full width at half-maximum of the rocking curve for the crystal shown in the Fig.1(b) is about 20.2arcsec which shows the high crystallinity of the crystal.

Fig. 2 shows the transmission spectra of TEGG, The S1、S3、S4、S5 absorption bands centered at 410, 525, 660 and 980nm are assigned to the transitions from the $^4I_{15/2}$ ground state to the $^2H_{9/2}, ^2H_{11/2}, ^4F_{9/2}$ and $^4I_{11/2}$ states of Er$^{3+}$ ion, respectively. The absorption S2 at 484nm due to the Tb$^{3+}$:$^7F_6→^5D_4$ is so strong and the transitions of $^4G_{11/2}, ^2H_{9/2}$ of Er$^{3+}$ is covered and could not be observed. In the working wavelength of frequently used FLs around 830、1064、1330nm, TEGG has good optical transparency which can reach 80% upwards. we can conclude that TEGG has smaller absorption coefficient. This parameter is very important for FLs applications, since the use of lengthy crystals requires minimizing optical losses. So the TEGG can be used as a magneto-optical materials in VIS-NIR wavelength.

The measured Verdet constant dispersion is described by a single electronic transition, and the V constant dispersion is given as a function of the wavelength $\lambda$ in simplified form as: $V = \frac{E}{\lambda^2 - \lambda_0^2}$, whereas E includes all the constant terms, and $\lambda_0$ is the transition wavelength, which is associated with the 4f -4f 5d electronic transition. The results of the FR measurements are displayed in Fig. 3, The curves for TEGG and TGG are very close to each other as the wavelength increased, while the values for TEGG being higher in the 400-1000nm. It can be seen that TEGG possesses a higher V value, with an increment of 10%-20% independently of the considered wavelength in comparison with the reference TGG. The measured Verdet constants at wavelength of 532、633、830、1064 and 1330nm are 224.5、145.3、78.8、46.5、27.1 rad-m$^{-1}$T$^{-1}$, respectively.

Since the thermal management is a very important point in high power laser system,
thermal conductivity (Fig.4(a)) and LIDT of the crystal have been measured. The curves for TEGG and TGG are very close to each other, while the TEGG being slightly lower in comparison with the reference TGG at the measured temperature. The thermal conductivity of TEGG at room temperature was 7.35 Wm$^{-1}$K$^{-1}$, dropping to the 4.36 Wm$^{-1}$K$^{-1}$ at 500 °C. The LIDT of measured was about 13.63 J/cm$^2$, shown in the Fig.4(b). The LIDT value of TEGG is almost same as the pure TGG. These results further confirm the high quality of the TEGG crystal and its further application in the high power Faraday devices.

4. Conclusions

Highly transparent Er doped TGG single crystal has been successfully grown by the Cz-method. The optical and magneto-optical characterization leads to the conclusion that Er doped TGG and pure TGG are very similar, the properties of Er:TGG being better than those of TGG. It can be explained by that the magnetization depends on the splitting of the ground configuration induced by the spin–orbit (SO) interaction, crystal field (CF), exchange interaction and external magnetic field. It should be emphasized that the exchange interaction between magnetic rare-earth ions, although much smaller, will have effects under some conditions in paramagnetic gallium garnet.

The Verdet constant of TEGG is remarkably higher though the doping Er concentration is small. However, we can not obtain a higher concentration of Er$^{3+}$ doped TGG crystal in present Cz technology. Overall, TEGG is an attractive candidate to replace TGG crystals in FLs for high-power laser sources operating in the VIS-NIR wavelength range, especially around the visible region.

5. Acknowledgment
This work was supported by Nature Science Foundation of China (Nos.51472257)

References


FIGURE CAPTION

Fig.1. X-ray powder diffraction patterns of the TEGG (Fig.1.a), insert of Fig.1(a) was TGG and TPGG samples for further measurements and Fig.1(b) was the rocking curve for the TEGG crystal

Fig.2. The transmission spectra of TEGG crystals at VIS-NIR wavelength

Fig.3. Verdet constant dispersion of TEGG in comparison with that of TGG

Fig.4. Thermal conductivity(Fig.4.a) and LIDT of the TEGG crystal (Fig.4.b)
Fig. 1. X-ray powder diffraction patterns of the TEGG (Fig. 1.a), insert of Fig. 1(a) was TGG and TPGG samples for further measurements and Fig. 1(b) was the rocking curve for the TEGG crystal.

Fig. 2. The transmission spectra of TEGG crystals at VIS-NIR wavelength.

Fig. 3. Verdet constant dispersion of TEGG in comparison with that of TGG.
Highlights

1. Highly transparent Er:TGG crystal was first grown by the Czochralski method.
2. The Verdet constant of Er:TGG crystal is enhanced at VIS-NIR region.
3. The crystal obtains high thermal conductivity and laser induced damage threshold.
4. The crystal is suitable for new and improved Faraday rotators.