



Improving characteristic of Faraday effect based on the Tm^{3+} doped terbium gallium garnet single crystal



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ABSTRACT

Highly transparent Tm^{3+} doped terbium gallium garnet single crystal was grown by the Czochralski (Cz) method for magneto-optical applications. The single-crystal X-ray diffraction confirms that the compound crystallize in the cubic systems with the structure $a = b = c = 1.2338$ nm. The temperature dependence of the magnetic susceptibility indicates that the Tm^{3+} :TGG crystal exhibits paramagnetic behavior over the experimental temperature-range 10–300 K. Transmittance spectra and the Faraday rotation have been investigated, which demonstrates that the as-grown crystal shows a high visible transparency and yields a larger Faraday rotation comparable to that of TGG crystal. The increasing of the Verdet constant at measured wavelength and high thermal property show the superior characteristics of Tm^{3+} doped TGG compared to the pure TGG, indicating that it has significant development for magneto-active materials used in Faraday devices at visible and near-infrared regions (VIS-NIR).

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1. Introduction

With continuous development of high-power laser-diodes and high-power fiber-lasers operating at VIS-NIR [1,2], Faraday isolators (FIs) used for such region gain great development in recent years. FIs are fundamental components used in advanced optical communications system to prevent harmful back-reflections [3,4], and to eliminate parasitic oscillations in amplifier systems or frequency instabilities in laser diodes [5,6]. As the core part of the FIs, the Faraday rotator obtained by magneto-optic materials primarily determines the performance of FIs. Yttrium-iron garnet, $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG) and Bi doped YIG materials with high transparency in the infrared regions is employed and characterized by a very large Verdet constant (2200 and 1700 rad/Tm at 1310 and 1550 nm, respectively) [7–10]. Terbium gallium garnet ($\text{Tb}_3\text{Ga}_5\text{O}_{12}$, TGG) is thought to be a suitable material for such requirement among transparent magnetic materials because of its favorable growth characteristics and high transmittance.

TGG can be grown large size single crystal for its congruent melting nature in comparison with TAG [11–13], however, as the rapid development of visible light communications the demand of

FIs operated at VIS-NIR are rapidly increasing, while the conventional TGG crystals are not practical due to its low V ($134\text{radT}^{-1}\text{m}^{-1}$ at 632.8 nm) compared to the $\text{Tb}_3\text{Al}_5\text{O}_{12}$, which means that FIs need a higher magnetic field intensity or larger length of the crystal to eliminate the back-reflection, increasing the instability of high power laser systems caused by some thermal-optics effect [14].

Some researchers have found that the quantum based super-exchange interaction between Tb^{3+} and other paramagnetic Re^{3+} ions can occur, greatly enhancing the Faraday effect. Recently, the study of Ce^{3+} doped TAG ceramic has $199.55\text{radT}^{-1}\text{m}^{-1}$ at 632.8 nm of V , 16% larger than that of TAG [15]. $\{\text{Tb}_3\}\{\text{Sc}_{1.95}\text{Lu}_{0.05}\}(\text{Al}_3)\text{O}_{12}$ (TSLAG) single crystal having a increment of 20% of V compared to TGG due to the doping of Sc^{3+} , Lu^{3+} , Al^{3+} ions [16]. Some fluoride single crystals with high concentration of efficient paramagnetic Re^{3+} ions in UV-VIS region shows excellent magneto-optical properties [17].

So in this letter, the current investigation centers on producing larger Faraday effect material based on TGG single crystal owing to its congruently nature compared with TAG. The magneto-optical properties in TGG arise from the transition $4f^8 \rightarrow 4f^7 5d^1$ of single Tb^{3+} ion. However, researchers have found that more than one single paramagnetism ions in garnets can remarkably enhance the magneto-optical property (Ce^{3+} , Pr^{3+} , Nd^{3+} doped TGG crystals [18–20] with 20–30% larger of Verdet constant compared to that of TGG). On the other hand, Tm^{3+} ion has been pointed out to show a

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large effective magnetic moment among the rare earth ions, Thus we suspect that a larger Faraday rotation angle of Tm^{3+} doped terbium gallium garnet can be obtained due to the strong magnetic moment of Tb^{3+} - Tm^{3+} super interactions. In this work, we report the growth of Tm^{3+} doped TGG crystal by Czochralski method for the first time, as well as its magneto-optical characteristic.

2. Experimental procedure

High-purity Tb_4O_7 , Tm_2O_3 , Ga_2O_3 (5N) chemicals were mixed according to the designed $\text{Tb}_{2.98}\text{Tm}_{0.02}\text{Ga}_5\text{O}_{12}$, then the mixture of powders (1–2% excess of Ga_2O_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 crystal was grown by the Cz method in a iridium crucible with radio frequency (RF) induction heating. After that crystal growth was carried out under high purity N_2 (99.99%) atmosphere. It was grown in the $\langle 111 \rangle$ orientation at a pulling rate of 1.0 mm/h and a rotating rate of 10–15 rpm. Finally, $\text{Tb}_{3-x}\text{Tm}_x\text{Ga}_5\text{O}_{12}$ crystal was obtained.

The transmission spectrum were measured using a Perkin–Elmer Lambda 900 UV-VIS-NIR spectrophotometer (United States) in transmission mode over the wavelength range of 300 nm–2000 nm. The X-ray powder diffraction measurement was carried out by the Ultima IV (Rigaku, Japan). A schematic of the experimental is setup for investigating the Faraday effects. A light sources (Xe-lamp and monochromator) passed through the sample located between a pair of Glan laser prisms. The transmitted light power was measured using a power meter. A polarization plane of laser light was rotated by the Faraday effect because of the magnetic field. Commercial TGG crystal (CASTECH) utilized for the calibration. All the measurements were performed at room temperature.

3. Results and discussion

3.1. Structural analysis and phase stability

The $\text{Tb}_{3-x}\text{Tm}_x\text{Ga}_5\text{O}_{12}$ crystal which is shown in the insert of Fig. 1 was ground into powder and its X-ray powder diffraction pattern was shown in Fig. 1. Compared with the JCPDS standard card, the XRD powder diffraction pattern of doped crystal is agree well with the standard patterns of TGG crystal (JCPDS 88–0575) without any impurities peaks. The result indicates that the Tm^{3+} ion does not influence the crystal structure. The unit–cell parameters were determined with the help of X Pert High Score Plus computer

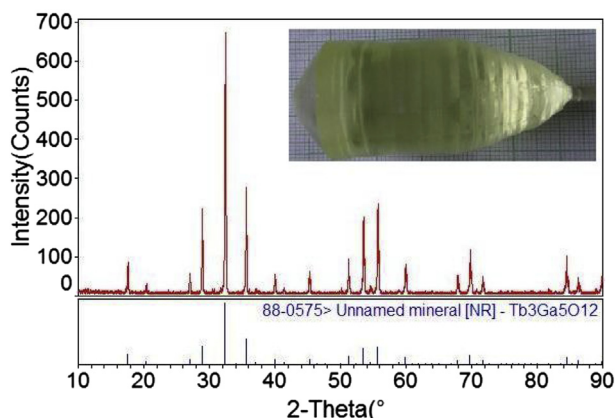


Fig. 1. X-ray powder diffraction patterns of the Tm:TGG, Inset: Tm:TGG crystal was obtained by the Cz method.

program. The result shows that the crystal belongs to the cubic system and its lattice parameters are $a = b = c = 1.2338$ nm, a slightly smaller than that of pure TGG. It may be the reason that the radius of Tm^{3+} is smaller than of Tb^{3+} .

As the segregated dopant disperses into melt homogenously, the solute concentration in the crystal C_s at growth interface would change with the solid fraction of the grown crystal by:

$$C_s = C_0 k_{\text{eff}} (1 - g)^{k_{\text{eff}} - 1} \quad (1)$$

where C_0 is the initial solute concentration, and k_{eff} is effective segregation coefficient. In the Fig. 2, the cation distribution of Tm^{3+} ion as a function of the solid fraction is shown, as g increase, Tm^{3+} concentration in the crystal increases and Ga^{3+} concentration decreases, whereas Tb^{3+} concentration seems not to depend on the solid fraction. The distribution coefficient k_{eff} of Tm^{3+} is nearly 0.50.

3.2. Transmittance spectrum

Fig. 3 shows the transmittance spectrum of Tm: TGG crystal. It is important for a good magneto-optical material to have a low absorption loss in some specific wavelength range, such as 532, 633 and 1064 nm. There are two absorption bands centered at around 660, 760 nm, which correspond to the transitions starting from the $^3\text{H}_6$ ground state of Tm^{3+} to higher levels $^3\text{F}_{2,3}$, $^3\text{H}_4$, respectively, while absorption band centered at 486 nm is mainly related to the energy level transition $^7\text{F}_6$ - $^5\text{D}_4$ of Tb^{3+} . It can be seen that Tm:TGG has good optical transparency which is around 80% in VIS-NIR. This parameter is very important for FIs applications, since the use of lengthy crystals requires minimizing optical losses. So the Tm:TGG crystal can be applied as a magneto-optical material in VIS-NIR FIs devices.

3.3. Magnetic susceptibility

The magnetic susceptibility results of Tm:TGG are shown in Fig. 4. The χ (T) decreases monotonously without any magnetic transition over the temperature range from 10 to 300 K under a constant magnetic field of 0.01 T. The inset of Fig. 4 indicates the reciprocal of $\chi(T)$, which exhibits a linear dependence on T down to 10 K at least. The linear part of the reciprocal of $\chi(T)$ can be described by the following Curie-Weiss equation which is defined as $\chi = C/(T - \theta_p)$, where C is the Curie constant and θ_p is the Weiss temperature. The effective magnetic moment and Curie-Weiss

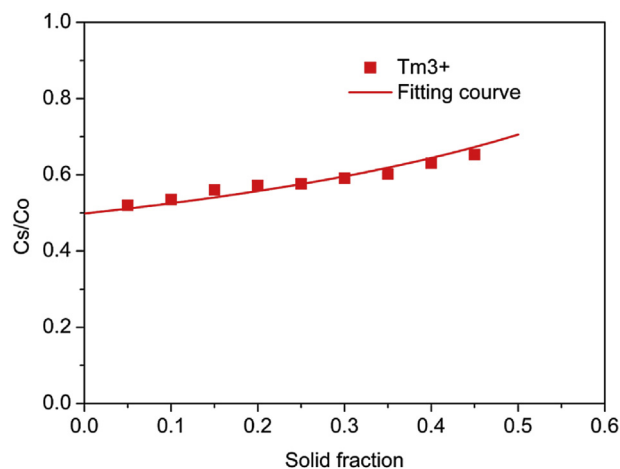


Fig. 2. The Tm^{3+} distribution as a function of the solid fraction in the Tm:TGG crystal.

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