

Highly transparent terbium gallium garnet crystal fabricated by the floating zone method for visible–infrared optical isolators



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ABSTRACT

Highly transparent terbium gallium garnet ($\text{Tb}_3\text{Ga}_5\text{O}_{12}$; TGG) single crystal having a large Verdet constant based on the visible and near-infrared region (VIS–NIR) Faraday rotator was grown by Floating Zone (FZ) growth machine. We successfully grew TGG single-crystal rods of 8–10 mm in diameter, which was suitable for the use in optical devices. The crystal showed a full-width at half-maximum as little as 18 arcsec by the X-ray rocking curve measurement. The Faraday rotation ($B = 0.55\text{T}$) was investigated at wavelength of 532, 632.8, 1064 nm at room temperature. The lower weak absorption coefficient, higher Verdet constant, thermal conductivity and laser induced damage threshold (LIDT) compared to the commercial TGG gives the great potential of using this new method to meet the increasing demand of VIS–NIR Faraday rotators (FRs).

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1. Introduction

Faraday isolators are the important parts of currently used for high-power-laser machinery and advanced optical communications [1–4], preventing reflected light from entering laser systems [5,6]. Bi-doped Yttrium iron garnet (YIG) is the key FRs material for the infrared region ($>1100\text{ nm}$) [7]. Recently, the demand for optical isolators that operate at shorter wavelengths (VIS–NIR) has increased greatly, where conventional YIG is not practical because of its very poor transmittance in this region [8,9]. Terbium gallium garnet (TGG) material (crystal or ceramic) has a high Verdet constant at a wavelength of $1\ \mu\text{m}$, a high thermal conductivity and excellent sizescalability [10–12], leading to the implementation of TGG in FR working at shorter wavelengths [13]. Although TGG bulk crystals could be grown by the Czochralski (Cz) technique for its congruent melting properties [14,15], its growth presents serious difficulties related with the decomposition and vaporization of Ga_2O_3 from the melt, resulting in the comparable low quality of the crystal [16]. Moreover, the TGG crystal grown by the Cz usually with some core area like the Nd:YAG, significantly reduce the optical homogeneity, the extinction ratio and beam quality after passing through the Faraday device of the crystal [17].

During our research into the crystals with high-vaporization nature, we realized that the FZ method has good potential to grow

this sort of materials. The several mm-diameter crystals are considered to be more attractive materials for mass production of miniature Faraday isolators [18]. This way, it can be easily fabricated by the FZ method and its growth cycle is relatively short compared to the Cz method. Moreover, crystals grown by FZ method have a typically rod-like shape and are thought to be suitable for optical device applications because the light can travel through the cylindrical shape without complicated processing [19]. The other advantages are high-purity growth due to crucible-free setup, effective heat inlet toward oxide crystals, diameter controllable growth. The current investigation centers on the feasibility study of the growth of TGG crystal by the FZ method, comparing its optical quality, micro-structure and magneto-optical property with commercial TGG crystal.

2. Experimental procedure

Single crystals of TGG were grown by the FZ technique. The starting materials were Tb_4O_7 , Ga_2O_3 (99.999%) mixed according to the corresponding nominal cationic ratios. First, the mixed powder was pressed into rods by a cold isostatic press under a pressure of 60 MPa. Then the rods were sintered in a high-temperature furnace equipped at $1400\ ^\circ\text{C}$ for 10 h in air. The sintered rods were about 70–80 mm long and 8–10 mm in diameter. Crystal growth was carried out by floating zone technique using four-halogen lamps (FZ-T-10000-H-VI-VP, Japan Crystal Growth System Co. Ltd.). The previously grown single crystal was set as a seed with

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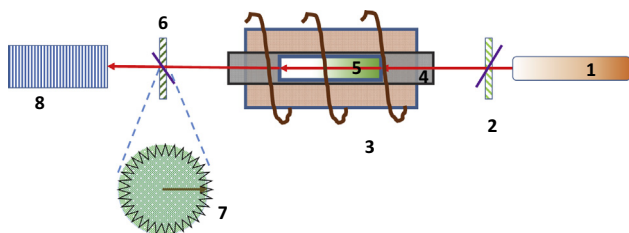


Fig. 1. The Vedert constant instrument for the TGG single crystal. 1. Light source, 2. 6. two polarizers, 3. magnetic field (550G), 4. quartz tube, 5. the crystal, 7. high accuracy goniometer and 8. laser light power.

the (111) plane in the growth direction. The sintered rods rotated at 8 rpm and the crystals were grown with a speed of 3–5 mm/h, under dry air flow with a rate $0.2 \text{ m}^3/\text{h}$.

The X-ray diffraction (XRD) analyses were performed using the Ultima IV (Rigaku, Japan) and the rocking curve for the crystal was measured by the PANalytical Empyrean XRD apparatus. The transmission spectrum and absorption spectrum were measured using a Perkin-Elmer Lambda 900 UV-VIS-NIR spectrophotometer (Japan).

The specific Faraday rotation of single crystals were measured at room temperature with the FRs test apparatus shown in Fig. 1 by the magneto-optic modulated double-frequency technique [20] (using a laser of 632.8 nm as the light source). The extinction ratio of the crystal was determined with a HP-Lightwave Multimetric 8153A and an Optical Head 81521B in a cross Nikol's configuration system with more than 60 dB.

The weak absorption coefficient of the TGG using the PCI technique [21] (photothermal common-path interferometer system manufactured by Stanford photon-thermal solution in Stanford, California, United States). The thermal conductivity 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 Q-switched Nd:YAG (1064 nm) laser operating in TEM00 mode. The laser was operated at the repetition rate of 1 Hz with the pulse width of 12 ns. To avoid surface damage of crystals, the laser beam with area 0.34 mm^2 was focused on the rear face of the crystal by a lens with a focal length of 5000 mm. The focal point was moved shot by shot (1-on-1 test) to avoid laser conditioning and the cumulative effect of pulse-to-pulse residual heat.

3. Result and discuss

The as-grown TGG 1 single crystal with the $10 \text{ mm} \times 30 \text{ mm}$ by the FZ method was shown in Fig. 2(a) and (b) is the commercial TGG 2 crystal (CASTECH INC China) used for further measurement in the same apparatus with TGG 1. X-ray powder diffraction patterns of the powder specimen is shown in Fig. 3. Compared with the JCPDS standard card, the XRD powder diffraction pattern of TGG 1 crystal is agree well with the standard patterns (JCPDS 88–0575) without any impurities peaks. The results of the rocking curve for the crystals are showed in Fig. 4. The full width at



Fig. 2. The as-grown single crystal with the $10 \text{ mm} \times 30 \text{ mm}$ by the FZ method (a) and (b) the commercial TGG 2 for further measurement in the same apparatus with TGG 1.

Table 1

The element analysis results from ICP-AES of TGG1 and TGG2.

Crystal	TGG1		TGG2	
	Tb	Ga	Tb	Ga
Measured (wt%)	47.82	34.72	49.57	33.88
Calculated molar number	0.301	0.498	0.312	0.486

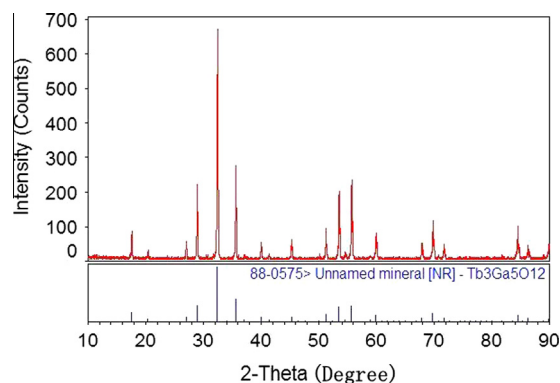


Fig. 3. X-ray powder diffraction patterns of powder specimen. The green peaks refer to the standard pattern while the red peaks refer to the as-grown TGG 1. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

half-maximum of the rocking curve for the TGG 2 is about 34 arcsec, while the TGG 1 presents 18 arcsec which showed the higher crystallinity of the crystal grown by the FZ method. The element analysis of the crystal is measured by the Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES) method. The element analysis results of the crystal are listed in Table 1, the TGG 1 can be expressed as $\text{Tb}_{3.01}\text{Ga}_{4.98}\text{O}_{12}$ according to the measured molar ratio, while the TGG 2 with $\text{Tb}_{3.12}\text{Ga}_{4.86}\text{O}_{12}$ resulting from the high vaporization of Ga_2O_3 (nearly 2.4%Ga ions deviation) from the melt of the Cz method.

The optical transmittance spectra of TGG 1 are shown in Fig. 5. The optical transmittance of the grown crystal was almost 80% in the visible and near infrared region. The absorption peak of the $\text{Tb}^{3+}:\text{7F}^6\text{-5D}^4$ located at about 484 nm was also detected, while the absorption of Tb^{4+} ion was not observed. Some minor absorption peaks below the UV cut-off edge may correspond to the intrinsic absorption of crystal. Therefore, it indicates that the as-grown single crystals in our work are suitable for application of apparatus used in the working wavelength 400–1000 nm (besides around 498).

The Verdet constant at 532, 632.8, 1064 nm of the TGG 1 and TGG 2 crystal was determined. The Verdet constant of TGG 1 was 200.8, 140.3, 45.2 $\text{rad m}^{-1} \text{T}^{-1}$ accordingly (slightly higher than the TGG 2, 190.3, 134.0, 39.2 $\text{rad m}^{-1} \text{T}^{-1}$ in turns). The extinction ratio is used to describe the optical homogeneity of the crystal. Extinction ratio can be defined as $10 * \log(P1/P0)$, where $P1$ is the optical power level generated when the light source is on, and $P0$

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