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Preparation and characterization of highly transparent Ce^{3+} doped terbium gallium garnet single crystal

Zhe Chen *, Lei Yang, Yin Hang, Xiangyong Wang

Key Laboratory of Materials for High Power Laser, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, No. 390, Qinghe Road, Jiading District, Shanghai 201800, China

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A Ce^{3+} doped terbium gallium garnet crystal (TCGG) with 30 mm in diameter and 25 mm in length has been grown by the Czochralski (Cz) method. The cation distributions in the TCGG crystal and thermal expansion coefficient have been investigated. Absorption spectrum was evaluated in the visible and near-infrared region (VIS–NIR) at room temperature, which indicated the crystal had low absorption coefficient at 500–1500 nm. The specific Faraday rotation of single crystal was measured at room temperature in 532, 633, and 1064 nm. The Verdet constant of the crystal at 633 nm comes up to 164.3 rad m^{-1} T^{-1}, 26.3% larger than that of TGG at 633 nm. The thermal conductivity and laser induced damage threshold (LIDT) were also measured. Overall, the TCGG single crystal studied here exhibits superior properties than the commercial TGG so far, therefore it has potential to cover the increasing demand for new and improved Faraday rotators in the VIS–NIR region.

1. Introduction

The Faraday effect causes a rotation of the plane of light polarization when a light beam passes through a material in the presence of an axial magnetic field. A device which exhibits this behavior is called a Faraday rotators (FRs) [1,2]. The basic design of a practical device is to use a rod of optical material placed in the axial magnetic field produced by permanent magnets, usually face magnetized toroids [3,4]. However, to make a so-called Faraday isolators (FIs), it is necessary to add a pair of polarizers to the FRs and to set the rotation angle to be 45° rotation is additive for reflectors passing back through the isolator giving 90° difference between the input and reflected polarization states at the input to the isolator. The input polarizer will therefore reject back reflections. An output polarizer is necessary to clean up the polarization state for transmission back through the isolator and thus to ensure good isolation even if the light is depolarized before returning through the device. FIs are elementary components employed in advanced optical communications, high-power laser machinery, etc. [5–8].

Yttrium–iron garnet, $Y_3Fe_5O_{12}$ (YIG), is so far the most commonly used crystal in FIs [9]. It is characterized by a high transparency in the IR region, a large Faraday rotation angle, and a low saturation magnetization. However, YIG cannot be used at shorter wavelengths (<1100 nm) due to its poor transparency [10]. So it is necessary to explore new magneto-optical crystals with highly transparent in the VIS–NIR region as optical isolators operating at wavelengths below 1100 nm in advanced optical data-storage systems, laser machinery, high-precision laser measurements, etc.

Materials under above consideration are based on rare-earth (RE) garnet single crystals [11]. These are optically isotropic due to the cubic symmetry, and their Faraday rotation is essentially paramagnetic, caused by the 4f–4f5d transition of RE^{3+} ions. Terbium–aluminum garnet, $Tb_3Al_5O_{12}$ (TAG) has been reported to show the best magneto-optical properties with high transparency and Verdet constant [12]. However, the incongruent melting nature of TAG has impeded the practicable use [13]. With the lack of TAG, FIs for high-power laser machinery are made with TGG which melts congruently. At increasing wavelengths, the low Verdet constant (39 rad m^{-1} T^{-1} at 1064 nm) [14,15] compared to TAG means that it required to use especially strong magnetic fields or larger size of the crystal can gain enough rotational angle to eliminate the back-reflection [16], increasing the instability of high power laser systems caused by decreasing of isolation ratio and the thermal birefringence effect [17].

Researchers found that the quantum based super-exchange interaction between Tb^{3+} and other paramagnetic RE ions can occur, greatly enhancing the Faraday effect. Recently, the study

* Corresponding author.
E-mail address: zhenzhe1201@sina.com (Z. Chen).

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of Ce^{3+} doped TAG ceramic has 199.55 rad m^{-1} T^{-1} of *V*, 16% larger than that of TGG [18]. $\{Tb_3\}[Sc_{1.95}Lu_{0.05}](Al_3)O_{12}$ (TSLAG) single crystal having a increment of 20% of Verdet dues to the doping of Sc^{3+}, Lu^{3+} ions [19]. Some fluoride single crystals with high concentration of efficient paramagnetic RE ions in UV–VIS region shows excellent magneto-optical properties [5,20].

Thus, it is very necessary to improve the Verdet constant of TGG by substituting Tb^{3+} by other RE non-Kramers ions, such as Ce^{3+}, Pr^{3+} and Nd^{3+}. Our group have successfully fabricated Ce^{3+}, Pr^{3+} doped TGG crystals [21,22]. However, some important properties of TCGG still need to investigate. So in this letter, we focus on the studies of the cation distributions, spectral, thermal, magnetic and magneto-optical properties of TCGG, and measured LIDT as well to evaluate its practical usage in the high-power laser application.

2. Experimental procedure

Nominal TGG and TCGG crystals were grown by the Cz technique under same growth conditions. As starting materials, commercial oxides of Tb_4O_7, CeO_2 and Ga_2O_3 of 5N purity were used. These were weighted in the corresponding nominal cationic ratios. Mixed powders were charged in a Ir crucible and molten with a 30 kW RF generator. [1 1 1] oriented TGG single crystals were used as seeds. The pulling and rotation rates were fixed to 3 mm/h and 12 rpm, respectively. Pure TGG with a size of $\Phi 25 \times 30$ mm^3 and 1.0 at% Ce^{3+} doped TGG crystal with $\Phi 30 \times 25$ mm^3 were obtained as shown in Fig. 1. They were with good optical quality and crack free. The TGG crystal in Ref. [21] is bought from CASTECH. Inc (CHINA), However, the TGG and TCGG used in this paper were grown in the same conditions, So it is a more convincing fact when compared with their properties.

The chemical composition was analyzed by EPMA using the JEOL JXA-8621MX. The distributions of cation in single crystal were measured along the growth axis using an electron-probe 10 μm in diameter.

The optical transmittance spectra was measured on a V-570-type ultraviolet/visible/near-IR spectrophotometer (JASCO, Japan). The crystal structure was investigated by X-ray diffraction (XRD) on the Ultima IV (Rigaku, Japan). The temperature dependences magnetic susceptibility was measured using a PPMS6000 physical property measurement system (quantum Design). The Faraday rotation angles were measured with light source at three wavelengths 532, 633 and 1064 nm, two polarizers, and an electromagnet. The magnetic field intensity was adjusted from 0 to 1.2 T. All the characterizations were performed at the 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, 1064 nm wavelength, 12 ns pulse duration, 1-on-1 test).

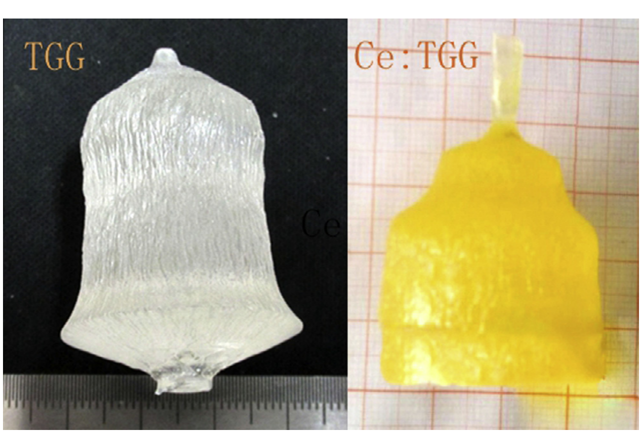

Fig. 1. The picture of as-grown crystals by the Cz method.

3. Results and discussion

3.1. Growth result and phase stability

According to the analysis in Ref. [23], 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 (g) of the grown crystal by

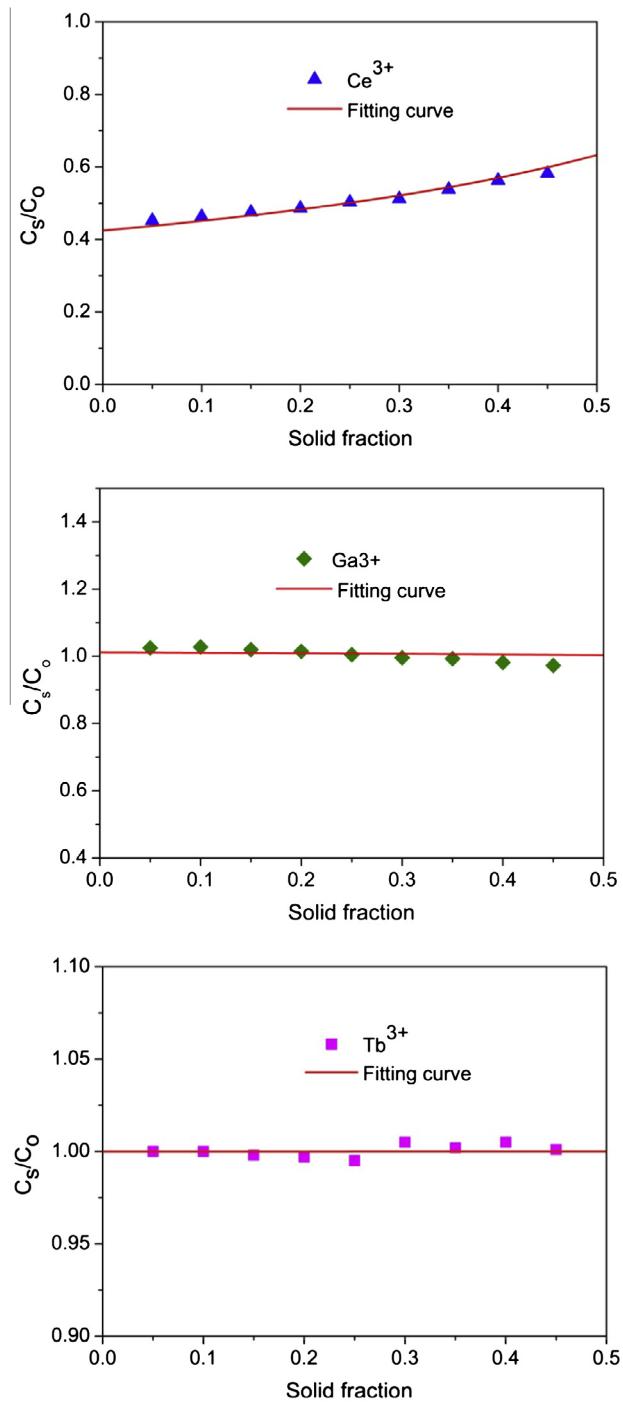

Fig. 2. The cation distributions as a function of the solid fraction g.