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# Faraday effect improvement by $Dy^{3+}$-doping of terbium gallium garnet single crystal

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ABSTRACT

Highly transparent $Dy^{3+}$-doped terbium gallium garnet (TGG) single crystal was grown by Czochralski (Cz) method. Phase composition of the crystal was tested by XRD measurements. The distribution coefficient of $Dy^{3+}$ in the crystal was obtained. The optical and magneto-optical properties were analyzed in detail, and magnetic properties of the $Dy^{3+}$-TGG crystal were studied. The paramagnetic behavior is observed down to 10 K. The as-grown crystal exhibited high optical transmittance, particularly in the visible region. The Faraday rotation was investigated over visible and near-infrared regions (VIS–NIR) at room temperature. The Verdet constants increase at measured wavelengths and high thermal stability was found in $Dy^{3+}$-doped TGG, as compared to the properties of pure TGG, indicating that $Dy^{3+}$-doped crystals are preferable for magneto-active materials used in Faraday devices at VIS–NIR wavelengths.



## 1. Introduction

Faraday isolators (FIs) are the indispensable optical parts of currently used high-power-laser machinery and advanced optical communication systems [1,2]. FIs are passive unidirectional non-reciprocal devices utilizing the phenomenon of magneto-optic rotation to isolate the source from reflections in an optical system [3,4]. Magneto-active material, as a core part of FIs for high-power and high-energy Faraday devices, should possess three critical parameters: High Verdet constant, excellent thermal stability and size scalability to avoid laser-induced damage under high-energy pulse operation [5,6]. Yttrium–iron garnet, $Y_3Fe_5O_{12}$ (YIG) and Bi doped YIG materials with high transparency in the infrared regions are employed and characterized by very large Verdet constant (2200 and 1700 rad/Tm at 1310 and 1550 nm, respectively) [7–10]. However, poor transparency of YIG below the 1100 nm leads to the implementation of rare earth (RE) garnets in FIs working at VIS–NIR wavelengths.

RE garnets are complex oxides with chemical formula $A_3B_2C_3O_{12}$ ($A=RE^{3+}$ ion, B and $C=Ga^{3+}$, $Al^{3+}$ ions). The cubic symmetry of garnet crystals plays a crucial role in the magneto-optical properties of these compounds. The garnet structure contains three unequivalent crystallographic positions, namely, dodecahedral (c), octahedral (a) and tetrahedral (d) ones with 24A ions in the (c) sites, 16B ions in the (a) sites and 24C ions in the (d) sites. Hence, the structure can hold a wide variety of cations.

$Tb_3Ga_5O_{12}$ (TGG) crystals have a high Verdet constant of 36 rad/Tm at wavelength 1 μm, high thermal conductivity coefficient [11], and excellent size scalability [12] obtained by using Cz technology. These characteristics make TGG suitable for FIs in the wavelength ranges of 400–470 and 500–1100 nm. However, lower Verdet constant, as compared to that of $Tb_3Al_5O_{12}$ (TAG), means that higher magnetic field or larger crystal length can gain rotational angle high enough to eliminate the back-reflection, increasing the instability of high power laser systems caused by the isolation ratio decrease and thermal birefringence effect [13].

Thus, the present study aimed at the production of larger-Faraday-effect TGG-based material. The magneto-optical TGG properties arise from the transition $4f^8 \rightarrow 4f^7 5d^1$ of $Tb^{3+}$ ion. However, it has been found that garnet doping by paramagnetic ions can remarkably enhance the magneto-optical properties. The $Ce^{3+}$-, $Pr^{3+}$- and $Nd^{3+}$-doped TGG [14–16] and $Ce^{3+}$-doped TAG ceramics [17] possess 20–30% larger Verdet constant as compared to that of TGG. On the other hand, $Dy^{3+}$ ion has been pointed out to show the largest effective magnetic moment among the rare earth ions [18]. Thus, we suspect that larger Faraday rotation angle can be obtained in $Dy^{3+}$-doped TGG due to the strong magnetic moment of $Tb^{3+}$–$Dy^{3+}$ super interactions. In this work, the $Dy^{3+}$-doped TGG crystal growth by Czochralski method and magneto-optical characteristics are reported.

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## 2. Experimental procedure

High-purity $Tb_4O_7$, $Dy_2O_3$, and $Ga_2O_3$ (Meryer, 5 N) chemicals were mixed according to the designed $Tb_{2.98}Dy_{0.02}Ga_5O_{12}$, pressed into tablets and sintered at 1400 °C for 20 h in the air to obtain the polycrystalline material. The crystal was grown by the Cz method in Ir crucible with radio frequency (RF) induction heating. After that crystal growth was carried out under high purity mixed $N_2+CO_2$ (99.99%, the molar ratio of $N_2$ is 80%) atmosphere. It was grown in the (111) orientation at a pulling rate of 1.0 mm/h and a rotating rate of 8–12 rpm. Finally, pure and $Dy^{3+}$-doped TGG crystals were obtained, as shown in Fig. 1.

The transmission and absorption spectra were measured using a Perkin–Elmer Lambda 900 UV–vis–NIR spectrophotometer. The X-ray powder diffraction measurements were carried out by the Ultima IV (Rigaku, Japan) device. The temperature dependence magnetic susceptibility was measured using a PPMS6000 physical property measurement system (Quantum Design, USA). The Verdet constants at elected wavelengths were measured by a homemade instrument consisted of laser sources, two polarizers and electromagnet. The magnetic field can be continuously adjusted from 0 to 1.2 T. All measurements were performed at room temperature.

Thermal conductivity of the sample was measured with high quality by flash method on a Xenon Flash Apparatus (LFA447/1 NanoFlashR300, Netzsch, Germany). The bulk laser damage threshold was measured using common 1-on-1 test method (Nd:YAG laser system, 1064 nm wavelength, 12 ns pulse duration).

## 3. Results and discussion

### 3.1. Structural analysis and phase stability

The piece of as-grown crystal was ground into powder and its X-ray powder diffraction pattern is shown in Fig. 2. As compared with the JCPDS standard card, the XRD pattern of doped crystal agrees well with the standard patterns of TGG crystal (JCPDS 88-0575) without any impurities peaks. The result indicates that the $Dy^{3+}$ ion does not influence the crystal structure. The unit-cell parameter was determined with the help of X Pert High Score Plus program. The result shows that the crystal belongs to the cubic system with lattice parameter $a=1.2350$ nm and the value is slightly smaller than that of TGG. The reason may be that the $Dy^{3+}$ radius is smaller than that of $Tb^{3+}$.

**Fig. 1.** The picture of as-grown crystals by the Cz method. The left is the $Dy^{3+}$-TGG and the right is the pure TGG.

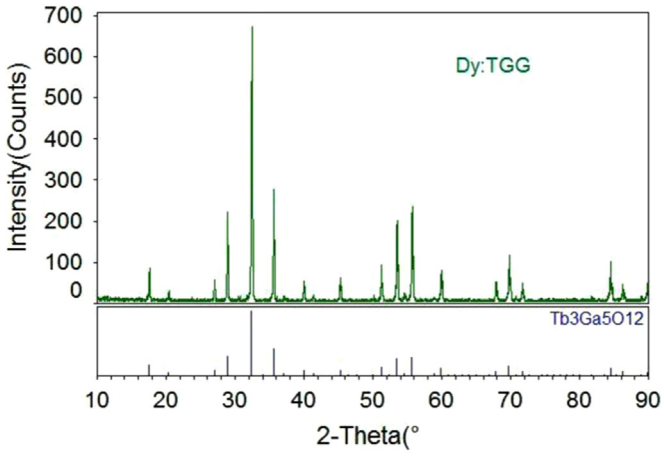


**Fig. 2.** X-ray powder diffraction patterns of the $Dy^{3+}$-TGG.

According to analysis in Ref. [19], as the segregated dopant disperses over melt homogenously, the concentration $C_s$ in the crystal at growth interface would change with the solid fraction of the grown crystal by

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

where $C_0$ is the initial concentration, and $k_{eff}$ is the effective segregation coefficient. In Fig. 3, the $Dy^{3+}$, $Tb^{3+}$ and $Ga^{3+}$ distributions are shown as a function of the solid fraction. As $g$ increases, $Dy^{3+}$ concentration in the crystal increases and $Tb^{3+}$ concentration decreases whereas $Ga^{3+}$ concentration seems to be independent on the solid fraction. The distribution coefficient $k_{eff}$ of $Dy^{3+}$ is nearly 0.84.

### 3.2. Transmittance spectrum

The transmittance spectra of $Dy^{3+}$-TGG and TGG crystals are shown in Fig. 4. For a good magneto-optical material, it is important to have a low absorption at specific wavelengths, such as 532, 633 and 1064 nm. In $Dy^{3+}$-TGG, there are four absorption bands centered at around 805, 905, 1094, and 1275 nm, which correspond to the transitions starting from the $^6H_{15/2}$ ground state to higher levels $^6F_{5/2}$, $^6F_{7/2}$, $^6H_{7/2}+^6F_{9/2}$ and $^6F_{11/2}+^6H_{9/2}$ in $Dy^{3+}$ ions, respectively. The absorption peak of pure TGG crystal centered at 484 nm is due to the $Tb^{3+}$:$7F^6 \rightarrow 5D^4$ [20]. It can be seen that $Dy^{3+}$-TGG has good optical transparency which can exceed 80%, even more higher than the pure TGG we grow, almost the same as the high-quality TGG crystal and ceramic obtained by the group of Yoshida [21]; however, the high transmittance region for $Dy^{3+}$-TGG is shorter due to the absorption of $Dy^{3+}$. This parameter is very important for FIs applications, since the use of lengthy crystals requires minimizing optical losses. Thus, the $Dy^{3+}$-TGG crystal can be applied as a magneto-optical material in VIS–NIR FIs devices.

### 3.3. Magnetic susceptibility

The magnetic susceptibility results of Dy:TGG are shown in Fig. 5. The $\chi(T)$ 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. 5 shows the reciprocal of $\chi(T)$, which exhibits a linear dependence on $T$ down, at least, to 10 K. 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 $\theta p$ is the Weiss