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Magneto-optical property of terbium-lutetium-aluminum garnet crystals

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Mixed terbium lutetium aluminum garnet $Tb_{2.2}Lu_{0.8}Al_5O_{12}$ (LuTAG) single crystal was grown by Czochralski technique successfully. The structure had been analyzed by X-ray diffraction. The paramagnetic behavior was observed in magnetic measurement. Magneto-optical properties and thermal conductivity of LuTAG had been studied in detail and compared with these of TGG sample. The crystal exhibited a high thermal conductivity and very high transmittance, particularly in visible and near-infrared region, indicating terbium-lutetium-aluminum garnet could be a potential magneto-optical material using in high-power laser system.

1. Introduction

Since M. Faraday first found Faraday effect in 1845, magneto-optical properties generally have been studied by contemporaneous and later scientists. Materials with Faraday effect can be used in optical communication, fiber optic current sensor, ring laser gyroscope, magneto-optic switch and so on. As its important utilization in laser system, magneto-optical material caused considerable interest of researchers and engineers with the rapid advance in of high power laser system in recent years.

Terbium based material is a very excellent magneto-optical material on account of having low lying energies of allowed magneto-optically active transitions and a relatively large region of optical transparency [1,2]. Therefore, it has been studied and developed for many years until today and accumulated many theoretical basis and practical experiences. Terbium gallium garnet (TGG) is a currently common material for practical application in optical systems of optical isolation. With the rapid development of femtosecond lasers and ultrafast magnetic dynamics, TGG crystals become an important and hot research field again [3–5]. This illustrates that the terbium based materials acting as magneto-optical material still have great research value.

TGG is a mainstream applications material with good magneto-optical properties. But recent researches reported that Terbium aluminum garnet (TAG) has a higher Verdet constant than TGG [6]. TAG can be considered the best magneto-optical material because of it large Verdet constant, high transmittance and low density. However, the problem of TAG is incongruent melting. This makes it hard to be grown and limited its practical applications. To solve this problem, Mikio Geho et al. created a hybrid laser floating zone method to grow TAG [7]. But the size of the as-grown crystal is small. Others try to grow TAG with doping to change its incongruent melting growth habit [8–10] in order to get large size crystal, such as terbium scandium aluminum garnet (TSAG), terbium aluminum gallium garnet (TAGG) etc.

In scintillation crystals researches, garnet structure oxide crystal material is a powerful candidate of scintillation materials because of its good chemistry stability and emission stability, high mechanical strength, and excellent luminescence property when rare earth elements doped. Among garnet structure oxide crystals, lutetium aluminum garnet has been widely researched in scintillating device. There are many studies about terbium lutetium aluminum garnet which have been reported in scintillation field [11,12]. It can be easily grown into large size single crystal by the Czochralski method. Along the way, we try to improve the content

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of terbium in LuTAG to get a new magneto-optical material. We try to grow LuTAG with the stoichiometric ratio of $Tb_{2.2}Lu_{0.8}Al_5O_{12}$ by Czochralski technique and study the magneto-optical relative properties. The results show it has magneto-optical function with good transmissivity and high thermal conductivity. So $Tb_{2.2}Lu_{0.8}Al_5O_{12}$ can be a potential paramagnetic material for magneto-optical devices in high power laser system. In this work, we report the growth, magnetic susceptibility, FR magneto-optical characteristics of $Tb_{2.2}Lu_{0.8}Al_5O_{12}$ for the first time.

2. Experimental

$Tb_{2.2}Lu_{0.8}Al_5O_{12}$ single crystals were fabricated using Czochralski technique (Cz-technique). Firstly, the polycrystalline materials of LuTAG were prepared using the conventional solid-state reaction methods. High-purity Lu_2O_3 (99.995%, Guangli Ganzhou), Tb_4O_7 (99.99%, Guangli Ganzhou) and Al_2O_3 (99.999%, Sinopharm Chemical Reagent Co., Ltd) were used as raw materials. The raw materials were weighted according to the stoichiometric ratio and mixed in the mortar. After grinded for 1 h, the mixed powders were pressed into disks with the size of 50 mm in diameter and 10 mm in thickness under a pressure of 50 MPa. All the disks were sintered at 1550 °C for 20 h in the air, resulting in the LuTAG polycrystalline. Then, LuTAG single crystal fabricated using Cz-technique with the single-crystal lifting furnace (Cyberstar, France). All the polycrystalline disks were crushed into powders and poured into an iridium (Ir) crucible. The detailed growth parameters were as follows. The grown direction was $\langle 111 \rangle$ with the pulling speed of 3–5 mm/h and rotation speed of 35–45 rpm. In order to reduce the volatilization of gallium oxide, the growth atmosphere was high purity nitrogen. The diameter of the crucible was 60–80 mm. Finally, LuTAG single crystal with φ25 mm × 30 mm were obtained.

The X-ray powder diffraction of LuTAG were performed using X-ray diffractometer (XRD, D8 Discover Davinci, Bruker AXS GMBH) with Cu Kα radiation in the 2θ range from 10° to 90° with a scanning speed of 10°/min in the normal routine. Lattice parameters were calculated using Rietveld refinement methods. The magneto-optic property was measured using the extinction method at room temperature in 632 nm with the self-made magneto-optical test system. The magnetic susceptibility was studied using physical property measurement system (PPMS-9 dxl, Quantum Design). The transmission spectroscopy was characterized using Ultraviolet Spectrophotometer (Cary 5000, VARIAN) in the wavelength range of 200–1600 nm at room temperature. The thermal conductivity was measured using Laser Thermal Conductance (LFA427, NETZSCH) in the temperature range of 4.2–500 K.

3. Results and discussion

3.1. Structure analysis

The XRD patterns of LuTAG single crystals were exibited in Fig. 1. It was clear to observe that the positions and intensities of all the diffraction peaks of LuTAG single crystal were in great consistence with the standard TAG crystal database (JCPDS76-0111). No secondary phases or any impurity peaks were observed. These confirmed the sample is high quality TAG single crystal. Lattice parameters were calculated using Rietveld refinement method and listed in Table 1. It was found that the grown crystal belongs to the cubic system with the Ia-3d space group. The calculated lattice parameter a = b = c = 12.02 Å was in good accordance with the standard database within a difference of 0.1%, revealing that introduction of Lu^{3+} does not change the essential structure of TAG crystals.

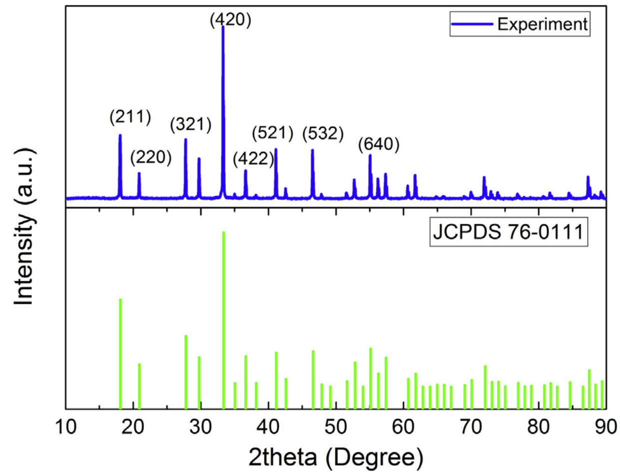

Fig. 1. Powder X-ray diffraction patterns of $Tb_{2.2}Lu_{0.8}Al_5O_{12}$ Crystal.

Table 1
Lattice parameters of LuTAG single crystals together with standard database.

Crystal	a	b	c	α	β	γ	V
Experiment	12.020	12.020	12.020	90	90	90	1736.6
Standard database	12.0	12.0	12.0	90	90	90	1728.0

3.2. Transmission spectrum

Transmittance is an essential parameter of the magneto-optical properties. With the develop of high-energy and high-average-power laser systems, the magneto-optical device that can be used in VIS-NIR region become more and more important. The demand for optical Faraday devices at wavelengths of 400–1100 nm is increasing rapidly. The transmittance spectra of LuTAG single crystal was shown in Fig. 2. The transmittance of LuTAG single crystal showed similar behavior in the energy range from 350 nm to 1600 nm. The optical transmittance of the grown LuTAG single crystal was almost 80% in the visible and near infrared region. The less than 20% absorption was considered to be arised from the inhomogeneously grown crystals, which can be improved by further optimize the growth conditions. The absorption peak centered at 488 nm was attributed to f-f transition Tb^{3+}:$^7F_6$-$^5D_4$ in the crystal [8], while the absorption of Tb^{4+} ion was not observed.

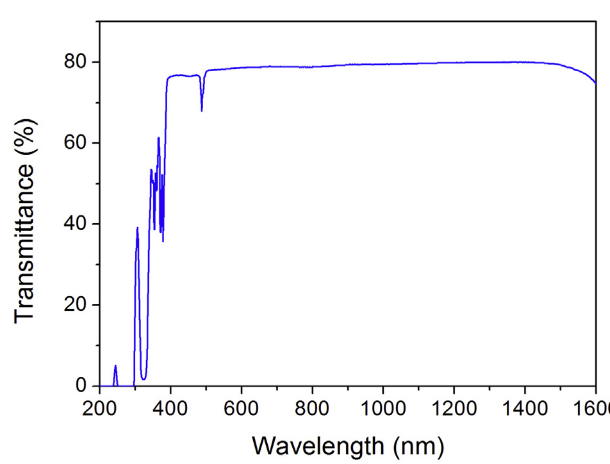

Fig. 2. Transmitting spectra of LuTAG single crystal.