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

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Abstract

Mixed terbium lutetium aluminum garnet Tb2.2Lu0.8Al5O12 (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.

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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
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$_5$O$_{12}$ 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$_5$O$_{12}$ 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$_5$O$_{12}$ for the first time.

2. Experimental

Tb$_{2.2}$Lu$_{0.8}$Al$_5$O$_{12}$ single crystals were fabricated using Czo-
chralski technique (Cz-technique). Firstly, the polycrystalline ma-
terials of LuTAG were prepared using the conventional solid-state
reaction methods. High-purity Lu$_2$O$_3$ (99.995%, Guangli Ganzhou),
Tb$_2$O$_3$ (99.99%, Guangli Ganzhou) and Al$_2$O$_3$ (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 polycrys-
talline disks were crushed into powders and poured into an
iridium (Ir) crucible. The detailed growth parameters were as fol-
 lows. The grown direction was (111) 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. Fin-
ally, 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 20 range from 10° to 90° with a scan-
ing 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 sec-
ondary 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.
In addition, no absorption appeared in the wavelength 510–1600 nm region, which is needed for Faraday isolators for optical communication systems [9].

3.3. Magneto-optical Faraday rotation

The Faraday effect causes a rotation of the plane of polarization which is linearly proportional to the component of the magnetic field in the direction of propagation. Faraday rotation is an interaction between the light and the magnetic field in a medium, which can be calculated by Eq. (1):

\[ \beta = VBd \]  

where \( \beta \) is the angle of rotation, \( B \) is the magnetic flux density in the direction of propagation, \( d \) is the length of the path where the light and magnetic field interact, and \( V \) is the Verdet constant for the material which is a proportionality coefficient varying with wavelength and temperature.

The magneto-optical property of LuTAG was measured by the self-made magneto-optical test system at room temperature with the wavelength of 632.8 nm. It indicates that the Faraday rotation of LuTAG is smaller than that of TGG, which is shown in Fig. 3. The Verdet constants of LuTAG and TGG are 98.82 rad/mT and 124.58 rad/mT, respectively. Considering the Faraday effect caused mainly by Tb\(^{3+}\) ions in the material, this phenomenon might be caused by the lower Tb\(^{3+}\) ions concentration in LuTAG than that of TGG.

3.4. Magnetic susceptibility

Magnetic susceptibility of LuTAG single crystal measured under the constant magnetic field of 0.1 T were displayed in Fig. 4. It was clear to found that LuTAG is paramagnetic materials. As the temperature increased, the magnetic susceptibility decreases rapidly in the temperature range from 10 K to 50 K, then decreased slowly in the temperature range from 50 K to 300 K. For instance, the magnetic susceptibility was 10 emu/g/T when the temperature is 10 K, while the magnetic susceptibility was 1 emu/g/T when the temperature is 150 K. This curve can be described by Curie–Weiss equation, Eq. (2):

\[ \chi = \frac{C}{T - \theta_p} \]

where \( C \) is Curie constant, \( \theta_p \) is Weiss temperature. The Weiss temperature of Tb\(_{2.2}\)Lu\(_{0.8}\)Al\(_5\)O\(_{12}\) is –7.46 K. The negative Curie–Weiss constant indicates the dominance of the antiferromagnetic interactions between Tb\(^{3+}\) ions [13]. The inset shows the directly proportional relationship between the reciprocal of magnetic susceptibility and temperature. This illustrates LuTAG exists Curie–Weiss behavior. Magnetic ions may be affected by crystal field to some extent.

Faraday rotation can be described by \( \beta = VBd \). The magnetic susceptibility can change with the temperature, which is the same to magnetic flux density \( B \). The magnetic flux density will become larger with decreased temperature. So the Faraday rotation would be smaller with the temperature rise [14].

3.5. Thermal conductivity

With the development of high power laser system, thermal properties of the material are very significant indexes for optical device. Therefore, we measured the thermal diffusivity of LuTAG and TGG crystal respectively by laser thermal conductance, and calculated the thermal conductivity by Eq. (3):

\[ k = \lambda \cdot \rho \cdot C_p \]

where \( k \) is thermal conductivity, \( \lambda \) is thermal diffusivity, \( \rho \) is the density of materials, \( C_p \) is specific heat at constant pressure. The result shows in Fig. 5. The two curves exhibited the same tendency, the thermal conductivity decreased with the temperature. But the LuTAG is about 20–30% larger than TGG at the same temperature. The thermal conductivity of LuTAG is 6.8 W/m·K at room temperature, and dropped to 3.5 W/m·K at 500 °C.

The results demonstrate that the thermal property of LuTAG can meet the demands of the high power laser system. With the development of high power laser system, the much higher demands will be put forward. The LuTAG will be a very potential material as high power Faraday devices.
4. Conclusion

Tb$_{2.2}$Lu$_{0.8}$Al$_5$O$_{12}$ single crystal was successfully grown by the Czochralski method. We studied Verdet constant of Tb$_{2.2}$Lu$_{0.8}$Al$_5$O$_{12}$ at 632.8 nm and compared with TGG single crystal. The result shows that Verdet constant of Tb$_{2.2}$Lu$_{0.8}$Al$_5$O$_{12}$ single crystal is smaller than that of TGG. But it has high transmittance in a wide range from visible to near-infrared regions and the thermal conductivity is much larger than TGG. LuTAG single crystal has great potential magneto-optical material for high-power laser system in the visible and near-infrared regions.

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