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Fabrication, optical properties and LD-pumped 2.7 μm laser performance of low Er^{3+} concentration doped Lu_2O_3 transparent ceramics

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ABSTRACT

Transparent Er:Lu_2O_3 ceramics were fabricated by the solid-state reaction method and vacuum sintering followed by hot isostatic pressing. The micrograph of the Er:Lu_2O_3 transparent ceramics exhibited a pore-free structure with the average grain size of ~2 μm. The in-line transmittance at 600 nm and 1300 nm were about 80% and 83%, respectively. The absorption, emission spectra and the decay lifetimes of Er:Lu_2O_3 ceramics were investigated. The 3 at.% Er:Lu_2O_3 ceramics were pumped by 967 nm InGaAs laser diodes. A maximum output power of 189 mW at 2.7 μm was obtained with a corresponding slope efficiency of 2.16%.

1. Introduction

Efficient laser sources operating 2.7–3 μm has attracted considerable attention for its potential applications such as military countermeasures, remote sensing, atmosphere pollution monitoring, and medical treatments due to strong water absorption in this waveband [1–5]. In addition, 2.7–3 μm is also a preferable pumping source for 3–13 μm optical parameter oscillation (OPO) lasers [6]. To obtain laser generation in 2.7–3 μm region, optical parametric oscillation (OPO) incorporated with the near-infrared lasers are usually used. However, the complexity associated with multistage approach makes design of a rugged, reliable and efficient configuration difficult. A promising alternative is provided by compact diode-pumped solid-state erbium lasers utilizing the $^4I_{11/2} \rightarrow {}^4I_{13/2}$ laser transition emitting between 2.7 and 3 μm pumped by the well-developed ~970 nm InGaAs laser diodes (corresponding to $^4I_{15/2} \rightarrow {}^4I_{11/2}$ transition in Er^{3+}) [7–9]. Due to the fact that the lifetime of the upper laser level $^4I_{11/2}$ is less than that of the lower level $^4I_{13/2}$ of Er^{3+} ions, the 3 μm lasing is commonly believed to be self-terminating [10–12]. It enables to overcome this bottleneck effect by concentration dependent upconversion process ($^4I_{13/2} + {}^4I_{13/2} \rightarrow {}^4I_{9/2} + {}^4I_{15/2}$), which have been successfully used to realize laser operation at room temperature in 50 at.% Er:YAG and 38 at.% Er:YSGG [13,14]. With regard to these heavily erbium doped 3 μm lasers, the accompanying issues, such as deteriorated thermal effect, decreased thermal conductivity and increased losses associated with $^4I_{11/2}$ upconversion, will have negative effect on the laser performance [15–18].

It is well known that host materials play an important role in the optical performance (i.e. lifetime, radiative transition probability, quantum efficiency, etc.) of the rare-earth ions, thus considerable research has been carried out to find efficient host materials [19–23]. Er^{3+} doped garnets [24–27] crystalline materials have been widely studied as 3 μm laser sources. Recently, Er^{3+} doped sesquioxides with relatively low concentration have attract much interests due to the relatively low maximum phonon energy, appropriate lifetime ratio of upper to lower laser level as well as their large heat conductivity which exceed that of YAG by up to 50% [28–30]. Lu_2O_3 is a kind of attractive sesquioxide with cubic structure. It possesses extremely high density (9.42 g/cm^3), high thermal conductivity (12.5 W/mK), and a wide band gap (~6.4 eV), which favors its applications as promising laser gain media. However, the high quality Lu_2O_3 single crystal is difficult to fabricate because of its high melting point (~2490 °C). Compared with single crystals, polycrystalline transparent ceramics have many advantages such as larger doping concentrations with controllable distribution in the volume of material, increased compositional versatility, larger volumes, increased mechanical and thermal properties [31]. Sanamyan et al. realized 2.7 μm laser operations of diode-pumped 2 at.% Er:Y_2O_3 ceramic with cryogenic cooling (77 K) [32]. Under room temperature, efficient lasing in

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7 at.% Er:Lu_2O_3 and 5 at.% Er:CaF_2 were reported by Li et al. [29]and Šulc et al. [33] respectively. Low Er^{3+}concentration doped materials may provide more favorable conditions for the cooperative lasing at 3 μm and 1.6 μm in the same bulk gain medium, which have been successfully realized in erbium cascade fiber laser [34]. The lower laser level $^4I_{13/2}$ will be depopulated by simultaneous 1.6 μm lasing, in turn compensating the self-terminating effect for 3 μm laser action.

In the present work, transparent Er:Lu_2O_3 ceramics were fabricated by the solid-state reaction method and vacuum sintering followed by hot isostatic pressing at 1750 °C for 4 h using high purity commercial Lu_2O_3 and Er_2O_3 powders as starting materials and 0.5 wt.% TEOS as sintering aid. The microstructures, absorption spectra, infrared luminescence spectra and decay curves of Er:Lu_2O_3 ceramics were systematically investigated. Room temperature CW laser operation of diode pumped 3 at.% Er:Lu_2O_3 ceramic laser at 2.7 μm was demonstrated.

2. Experimental details

The $(Lu_{1-x}Er_x)_2O_3$ (x = 0.03, 0.07, 0.15) ceramics were synthesized by the solid-state reaction method and vacuum sintering followed by hot isostatic pressing. The purity of starting materials were 99.999% for Lu_2O_3 and Er_2O_3 (Jiahua Advanced Material Resources Co., Ltd.). According to the nominal composition $(Lu_{1-x}Er_x)_2O_3$ (x = 0.03, 0.07, 0.15), above starting materials were weighted by appropriate stoichiometric ratio and tetraethyl orthosilicate (TEOS; Sigma–Aldrich, 99.999%) was introduced at 0.5 wt.% relative to the total powder mass in order to introduce SiO_2 as a sintering aid. The above starting materials were mixed with 99.99% ethanol (analytical pure; Merck, Darmstadt, Germany). The obtained slurry was then ball milled with high purity Al_2O_3 balls for 15 h using a planetary milling machine. After milling, the powder mixtures were dried at 55 °C for 48 h in an oven and then sieved through a 100-mesh screen. The powders were dry pressed in a stainless steel die at 15 MPa. After removing organic components by calcinating at 800 °C for 3 h, the green body pellets were further cold isostatically pressed (CIP) at 200 MPa for 5 min. After CIP, the relative density of the green body was ~50%. The green bodies were then sintered at 1800 °C for 4 h in a high temperature vacuum sintering furnace utilizing a tungsten heating elements under a vacuum of 5.0×10^{-3} Pa. The sintered samples were treated by hot isostatic pressing (HIP) at 1750 °C for 4 h and then annealed in air at 1400 °C for 15 h to completely remove internal stress and eliminate the oxygen vacancies. The as-prepared ceramics were mirror polished on both surfaces with different level of diamond slurries.

The phase compositions of the obtained specimen were identified by X-ray diffraction (XRD, Bruker-D2, Germany) equipped with a copper target X-ray tube (λ_{em} = 0.1541 nm), the morphologies of the starting powders and microstructural investigations of the sintered ceramics specimens were performed by scanning electronic microscopy (SEM, JSM-6510, JEOL, Japan) as well as optical transmission microscopy (Carl Zeiss, Axio Scope. A1, Germany). Mirror-polished ceramic samples on both sides were used to measure the optical transmittance on a UV–VIS–NIR spectrophotometer (Lambda 950 PerkinElmer, Waltham, MA, America). The emission spectra of the ceramics were measured using a spectrofluorometer (Edinburgh Instruments, FS980). Decay profiles were recorded with a Tektronix DPO 3054 digital storage oscilloscope in which the signal was fed from PbS detector. All emission spectra and decay curves were measured at room temperature.

To experimentally investigate the laser performance of Er:Lu_2O_3 transparent ceramics, the experiment configuration based on a simple plane-parallel cavity with a cavity length of about 10 mm is schematically shown in Fig. 1. A fiber coupled laser diode (LD) with center wavelength at 967 nm is used as pumping source. The delivery fiber has a core diameter of 105 μm and numerical aperture (NA) is 0.22. In the laser cavity design, we mainly considered good mode matching between the pump beam and the laser mode. The pump light was re-imaged into the Er:Lu_2O_3 ceramic with a spot size of about 210 μm in diameter by a simple telescopic lens system. A flat mirror coated high transmission (HT, T > 85%) at pump wavelength and high reflection (HR, R > 99.8%) at 2.7 μm was used as pump input mirror M1. And the output coupler M2 is also a plane mirror with transmittance of 1.5% at 2.7 and HR at 970 nm. The gain medium was a 3 at.%, $2 \times 3 \times 21$ mm^3 Er:Lu_2O_3 ceramic. End pumping was realized via the 2×3 mm face, which was cut and polished with flat and parallel but uncoated endfaces. The laser ceramic was wrapped with indium foil and mounted in copper block cooled by water at a temperature of 20 °C.

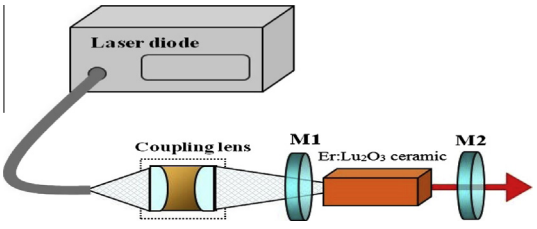

Fig. 1. Experiment configuration for the Er:Lu_2O_3 ceramic laser.

3. Results and discussion

Fig. 2 shows the XRD patterns of the $(Lu_{1-x}Er_x)_2O_3$ (x = 0.03, 0.07, 0.15) ceramics. The XRD patterns of the $(Lu_{1-x}Er_x)_2O_3$ (x = 0.03, 0.07, 0.15) ceramics match well with the referred standard JCPDs card (No. 12-0728: Lu_2O_3) selected in the International Centre for Diffraction Data (ICDD) database. No other impurities could be found. This indicates that the crystal formation of the samples is a single phase. As displayed inset in Fig. 2, the reflections monotonically shift toward small two theta degrees indicating that the lattices have an expansion with increasing Er^{3+} doping. This is induced by the substitution of Er^{3+} (coordination number CN = 6: r = 1.03 Å) for a smaller Lu^{3+} (CN = 6: r = 1.00 Å) in the lattices [35].

Fig. 3(a) shows the transmittance and photograph of $(Lu_{1-x}Er_x)_2O_3$ (x = 0.03, 0.07, 0.15). The thickness of all samples is polished to be 1.33 mm for measuring optical transmittance. With the increasing of Er^{3+} doping concentration, the transmittances of the ceramic samples remained about the same. The in-line transmittance at 600 nm and 1300 nm were about 80% and 83%, respectively. No obvious transmittance drop is observed from NIR and VIS wavelength region. Absorption bands centered at 657, 800, 979 and 1537 nm are attributed to the transitions of Er^{3+} ion from its ground state of $^4I_{15/2}$ to the excited states of $^4F_{9/2}$, $^4I_{9/2}$, $^4I_{11/2}$ and $^4I_{13/2}$, respectively. The intensity of absorption bands increases with the increase of Er^{3+} doping concentration. From the transmittance spectra and the thickness of the ceramics, the absorption coefficients of Er:Lu_2O_3 ceramics can be calculated. As the Er^{3+} doping concentration increased from 3 at.% to 15 at.%, the absorption coefficient at 979 nm increased from 1.14 to 3.40 cm^{-1}. Absorption band centered at 979 nm was selected as the pump wavelength in laser testing experiments. With broad absorption band in NIR region, Er^{3+}:Lu_2O_3 transparent ceramics are advantageous for lower dependency on pump wavelength and temperature control of a diode laser and are hopeful to be a kind of laser gain media for miniaturization of laser devices. The 2.7 μm laser performance pumped by 976 nm LD would be investigated in the Section 3.

Fig. 3(b) shows emission spectra of $(Lu_{1-x}Er_x)_2O_3$ (x = 0.03, 0.07, 0.15) ceramics under 980 nm excitation. Two main emission bands centered at 1537 nm and 2720 nm were observed which could be ascribe to the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ and $^4I_{11/2} \rightarrow {}^4I_{13/2}$ of Er^{3+}, respectively. With increasing the Er^{3+} doping concentration, the intensity of $^4I_{13/2} \rightarrow {}^4I_{15/2}$ decreases and the intensity of $^4I_{11/2} \rightarrow {}^4I_{13/2}$ meanwhile increases. These phenomena could be explained as a result of Energy Transfer Upconversion (ETU). In the heavy Er^{3+} doped materials, the distance between Er^{3+} ions decreases, which give rise to the increased interaction between Er^{3+} ions. The probability of ETU increases due to the increased interaction between Er^{3+} ions. Therefore, the intensity of $^4I_{11/2} \rightarrow {}^4I_{13/2}$ (2720 nm) increases due to the concentration dependent up-conversion process ($^4I_{13/2} + {}^4I_{13/2} \rightarrow {}^4I_{9/2} + {}^4I_{15/2}$).

Fig. 4 shows the micrographs of mirror-polished surface and fractured surface of Er:Lu_2O_3. Grain size, grain boundary phase and pores are usually considered as the main factors affecting transmittance of ceramics. As shown in Fig. 4, the grain size was homogeneous and the average grain size of Er:Lu_2O_3 was about 2 μm. Apparently, no grain-boundary phases and no obvious pore existed in the microstructure of the Er:Lu_2O_3 ceramic. As seen from the fractured surface figure (Fig. 3(b)), It could be seen that the