Growth, spectroscopy and laser operation of in-band pumped Ho^{3+}-doped Yttrium Gallium Garnet

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Abstract: We report on the crystal growth of a 2.86 at.% Ho:YGG crystal, its structural and spectroscopic characterization, as well as on its first laser operation near 2.1 µm under in-band pumping by a Tm-fiber laser.

In the last years, solid-state lasers emitting in the 2 µm spectral range are gaining attention because of potential applications in different sectors, including surgery, remote sensing, spectroscopy, military, etc. [1]. Holmium (Ho^{3+}) lasers operating on their main transition near at ~2.1 µm ($^5I_7 \rightarrow ^5I_8$) are becoming increasingly important due to the availability of powerful and reliable Tm-fiber based pump sources emitting near 1.9 µm. Such in-band pumping into the upper $^5I_7$ state greatly reduces parasitic losses due to up-conversion or energy transfer among the Ho^{3+}-ions, and ensures a small quantum defect which is a prerequisite for high efficiency, low heat load and power scaling. Due to the close ionic radii, Y-containing hosts are best suited for Ho-doping and one of the most successful materials is the classical Ho:$Y_3Al_5O_{12}$ (YAG) with cubic symmetry [1]. Yttrium gallium garnet with chemical formula $Y_3Ga_5O_{12}$ (YGG) is another garnet with the same structure and similar properties as a host material. Although it was introduced for Nd doping simultaneously with YAG as early as 1964, YGG received much less attention. In fact, the only $^5I_7 \rightarrow ^5I_8$ Ho:YGG lasing was demonstrated using Tm as a sensitizer and a pump laser diode at 795 nm [2]. Here we report on the crystal growth of Ho:YGG, its spectroscopic properties, and continuous-wave (CW) laser operation using a Tm-fiber pump laser.

The Ho:YGG crystal shown in Fig. 1(a) was grown using the optical floating zone (OFZ) technique in an oxygen-rich environment. The raw materials, Ho_2O_3, Ga_2O_3, and Y_2O_3 with a purity of 99.99%, were pressed in ratios corresponding to 2.5% at.% Ho doping into two rods of identical diameters and subjected to a pressure of 60 MPa for 10 min. The rods were then sintered in air at a temperature of 1300°C for 10 hours and subsequently transferred to the OFZ furnace equipped with four xenon lamps. The rotation rates of the feed and seed rods were set at 15 rpm and the growth rate was fixed at 6 mm/hour. Once the growth process was finished, the furnace was cooled down to room temperature (RT) within 3.5 hours. No annealing was applied. The actual Ho^{3+} concentration was measured using Electron Probe MicroAnalysis (EPMA) to be 2.86 at.%. X-ray diffraction measurements confirmed the phase purity and structural integrity of the crystal. The Ho:YGG adheres to a typical cubic garnet structure with the general chemical formula $\{A\}_3[B]_2(C)_3O_{12}$ (sp. gr. O_h^{10}- Ia$\bar{3}$d, No.230) with a lattice constant a = 12.2796(9) Å. The most intensive Raman line, attributed to the symmetric stretching mode of the [GaO_4] tetrahedron, was observed at 757 cm^{-1} and this peak was overlapping with the one corresponding to the maximum phonon frequency of 783 cm^{-1}.

Figure 1(b) reveals a broad and structured absorption band around 1.9 µm. The maximum absorption cross-section σ_{abs} of Ho:YGG amounts to 3.7×10^{-21} cm^2 at 1914 nm, while at 1908 nm (pump wavelength) it drops to 2.6×10^{-21} cm^2. For the $^5I_7 \rightarrow ^5I_8$ Ho^{3+} electronic transition, relevant to laser operation slightly above 2 µm, the maximum stimulated emission cross-section (σ_{SE}) is 9.5×10^{-21} cm^2 at 2084 nm. There are two additional peaks: one occurring at 2111 nm with $\sigma_{SE} = 4.3 \times 10^{-21}$ cm^2, and the other at 2061 nm, with $\sigma_{SE} = 5.2 \times 10^{-21}$ cm^2.

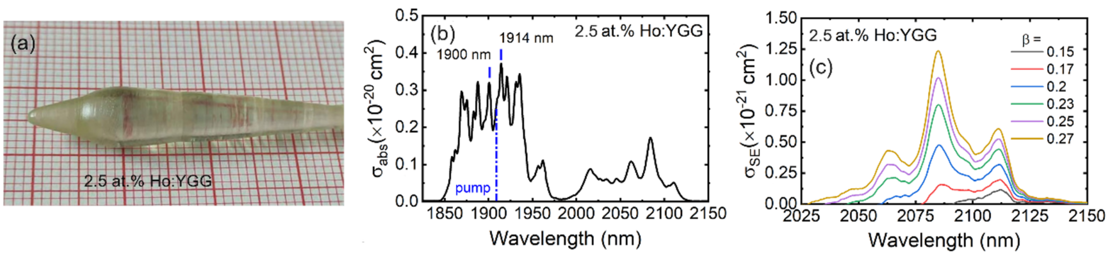

Fig. 1. (a) A photograph of the as-grown 2.86 at.% Ho:YGG crystal; **(b)** RT absorption cross-section of Ho^{3+} (the $^5I_8 \rightarrow ^5I_7$ transition) in YGG; **(c)** Gain cross-sections, $\sigma_{gain} = \beta\sigma_{SE} - (1 - \beta)\sigma_{abs}$, β – inversion rate.

The emission cross-section was calculated by the reciprocity method and the Füchtbauer–Ladenburg (F-L) equation using the crystal field energy splitting data tabulated in [3]. Both methods gave a satisfactory agreement. The results were used to calculate the gain cross-section (σ_{gain}) for the quasi-3-level Ho laser system shown in Fig. 1(c). With a modest inversion rate of β = 0.15, the gain spectrum displays smooth features, ranging from 2090 to 2120 nm, with a single peak centered at 2111 nm. The Full Width at Half Maximum (FWHM) of the gain curve centered at 2085 nm (laser wavelength) is approximately 11 nm for β = 0.2. Further increase of the inversion rate results in an additional peak emerging at 2062 nm. The fluorescence from the $^5I_7$ energy level of Ho^{3+} decays following a single exponential law, with a time constant (lifetime) of 8.9 ms.

A cubic laser element with an edge of 3 mm was cut from the originally grown 2.86 at.% Ho:YGG crystal to enable light propagation along the [111] direction. The uncoated laser crystal was mounted in a passively-cooled Al holder with thermal contact on all for lateral sides. A plane-concave laser cavity was employed. The flat pump mirror (PM) was coated for high transmission (HT) at the pump wavelength and high reflectance (HR) at 2.05–2.43 µm. Concave output couplers (OCs) with transmission (T_{OC}) in the range 0.5% - 5% at the laser wavelength were used which were HT for the pump. A commercial Tm-fiber laser (IPG Photonics TLR series) was employed as a pump source, delivering up to 5.2 W of unpolarized output at a central wavelength of 1908 nm with a linewidth of 0.2 nm. The pump beam was focused into the laser crystal through the PM using a spherical focusing lens with a focal length of 150 mm. A 45° dielectric mirror, HR for the laser and HT for the pump radiation, was placed behind the OC (RoC = 50 mm) to separate the laser beam from the residual pump.

The input-output performance of the Tm-fiber-laser pumped CW Ho:YGG laser is summarized in Fig. 2(a). Under non-lasing conditions the single pass intrinsic absorption of the crystal was estimated to be ~59% at maximum pump level. The calculated low-signal absorption is ~62 %. Thus it can be concluded that there is no bleaching of the absorption and in lasing conditions it will not depend on the OC or the pump level. To calculate the absorbed power in Fig. 2(a) we used the value of 59% taking into account the Fresnel reflection at the front surface. In all cases the laser emission was unpolarized. A maximum output power of 976 mW was achieved for the maximum output coupling used. The slope efficiency (η) for T_{OC} = 5% reached 50.1% with respect to the absorbed pump power. This is much higher than the 13.5% reported with the same T_{OC} in [2] by using the co-doped Tm,Ho:YGG.

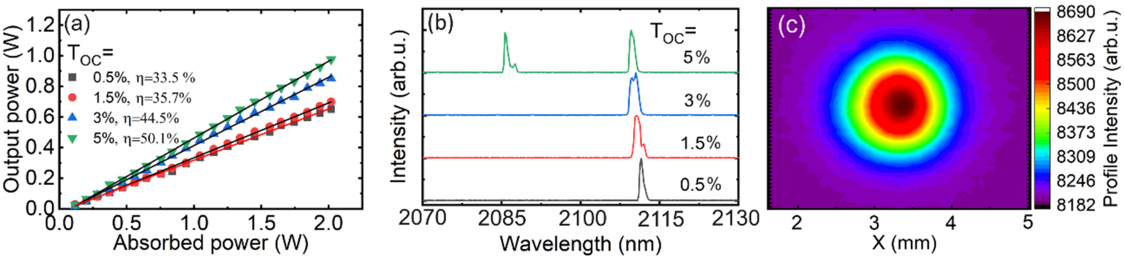

Fig. 2. Laser performance of the Ho:YGG crystal: **(a)** input-output power dependence and slope efficiency (η) for different OCs; **(b)** typical laser spectra recorded with different T_{OC}; **(c)** 2D spatial intensity profile captured at a distance of 40 cm from the OC.

The output spectra shown in Fig. 2(b) indicate a blue shift with the output coupler transmission, determined by the changing reabsorption effect in dependence on the steady-state inversion as expected for a quasi-3-level laser system. With a low-transmission OC (T_{OC} = 0.5%), the central wavelength was at 2111 nm. However, when the OC transmission reached 5%, simultaneous laser emission occurred at two wavelengths, 2085.3 and 2109.5 nm, with nearly equal powers. A circular beam profile was obtained using a Pyrocam PY-III-C-B camera (Ophir-Spiricon) at the maximum absorbed power with T_{OC} = 5%, see Fig. 2(c).

In summary, a Ho^{3+}-doped YGG crystal has been successfully grown by the optical floating zone method. The optical absorption, emission, and fluorescence lifetime, have been studied. We demonstrated the first laser operation of Ho^{3+}:YGG on the main Ho transition near 2 µm. A slope efficiency as high as 50.1 % was achieved under in-band pumping at 1908 nm.

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