Spectroscopic properties of Tm, Ho: LiLuF$_4$

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Received January 9, 2009

LiLuF$_4$ single crystals co-doped with thulium (5%) and holmium (0.5%), having large size, intact shape, and high optical quality, are successfully grown by the Czochralski technique. Absorption and fluorescence spectra of the crystal are measured. The absorption spectrum shows that the main absorption band locates at near 686 and 792 nm and the fluorescence spectrum peaks at 2.05 μm. At room temperature, the as-grown Tm, Ho: LiLuF$_4$ single crystals are end-pumped by a fiber-coupled laser diode system with pumping wavelength of 795 nm under CW operations. A power of 50 mw continuous laser output at 2.05-μm wavelength is achieved. Meanwhile an obvious green light is detected.

OCIS codes: 160.3380, 160.5690, 160.4760, 140.3380.

doi: 10.3788/COL20100801.0063.

During the last few years, interest in 2-μm lasers has increased for its large variety of applications requiring eye-safe lasers that propagate through the atmosphere. A 2-μm laser is considered to be an ideal source for medical applications and eye-safe detection, including laser ranging, laser differential instrument, and coherent Doppler velocimetry[1−5], etc.

Crystalline materials doped with Tm and Ho for obtaining 2-μm laser, which is based on Tm $^3H_4 \rightarrow ^3H_5$[6−8] and Ho $^5I_7 \rightarrow ^5I_8$[9,10] transitions, have attracted more and more attention from researchers. Previous researches indicated that Ho^{3+} $^5I_7 \rightarrow ^5I_8$ transition showed much more gain (gain = absorption cross section × life time) than that of Tm^{3+} $^3H_4 \rightarrow ^3H_5$ transition. But Ho^{3+} does not have an absorption band to match the emission of available laser diodes, hence there is a need to co-dope with Tm irons, which have the absorption band matching the laser diodes emission. At room temperature 2-μm laser emission of Tm and Ho co-doped crystals is based on Ho^{3+} $^5I_7 \rightarrow ^5I_8$ transition, and Tm ions play as sensitizing irons transporting pump energy to the metastable state of Ho ions[11−15].

LuLF crystal is an excellent host material for its relatively low phonon energy system and small absorption coefficient compared with other host materials. Theoretically LuLF crystal has lower laser threshold and up-conversion loss, which are advantageous for improving laser efficiency. As host material, LuLF crystals have other advantages such as no thermally induced birefringence, good resistance to optical damage, and the output of linearly polarized laser[16,17].

LuLF crystals co-doped with Tm (5 mol%) and Ho (0.5%) were grown by Czochralski method with medium frequency induction heating[18−20]. The absorption spectrum of the as-grown crystal was carried out on a JASCO V-570 UV/VIS spectrophotometer. Fluorescence spectrum was measured using a Nikon G250 spectrophotometer, pumped by a AlGaAs laser diode working at 808 nm, with a resolution of 0.5 nm in the wavelength range of 1800 − 2000 nm. After that, the diode-pumped laser performance of the Tm, Ho:LuLF crystal was studied.

The absorption spectra of as-grown Tm, Ho:LuLF crystal in the wavelength range of 200 − 2000 nm are illustrated in Fig. 1. There are five strong absorption bands in the spectra, which peaks at 356, 686, 792, 1208, and 1678 nm. The broad absorption bands are orderly attributed to the transitions of Tm^{3+} $^3H_6-^1D_2$, Ho^{3+} $^5I_8-^3H_6$, Tm^{3+} $^3H_6-^3F_3$, Tm^{3+} $^3H_6-^3H_4$, Tm^{3+} $^3H_6-^3H_5$, Ho^{3+} $^5I_8-^5I_6$, and Tm^{3+} $^3H_6-^3F_4$, respectively. It can be concluded that the Tm ions are dominant ions in the absorption progress because the concentration of Tm ions is much higher than that of Ho ions. In the measured range, the absorption peak at 686 nm is the strongest and the absorption at 792 nm takes the second place. The strong absorption at 686 and 792 nm is suitable for commercial applied InAs diode laser pumping. For further study in this wavelength range, we took the absorption spectrum of Tm, Ho:LuLF crystal in the wavelength range of 600 − 900 nm, as shown in Fig. 2.
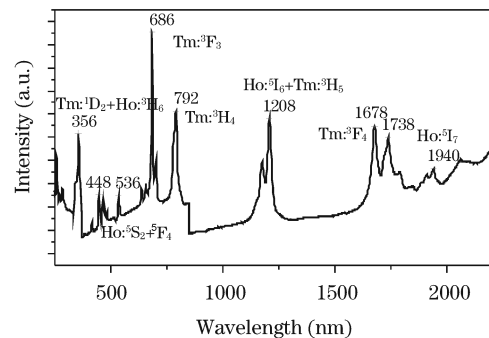

Fig. 1. Absorption spectrum of Tm, Ho:LuLF crystal in the wavelength range of 200 − 2000 nm.
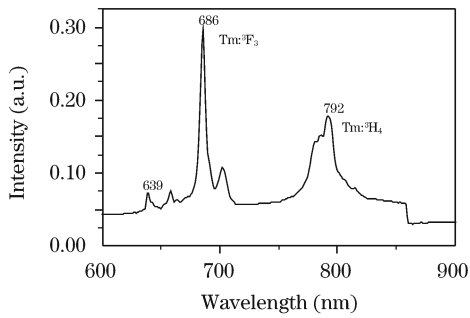
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Fig. 2. Absorption spectrum of Tm, Ho:LuLF crystal in the wavelength range of $600 - 900$ nm.
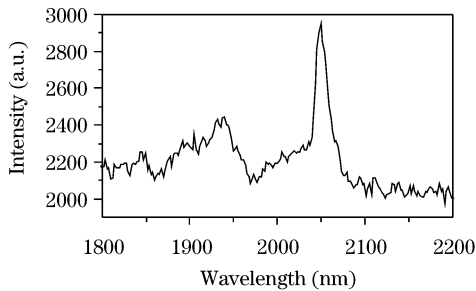

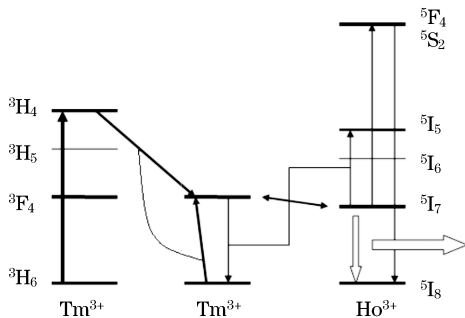Fig. 3. Fluorescence spectrum of Tm, Ho:LuLF crystal.

Fig. 4. Schematic of excitation mechanism and energy transfer process in Tm-Ho lasers.

From Fig. 2, we can see that there are two strong absorption bands in the absorption spectrum in the range of $600 - 900$ nm. The peaks center at 686 and 792 nm, and are due to transitions of Tm^{3+} $^3H_6 - ^3F_3$ and $^3H_6 - ^3H_4$, as explained above. The influence of Ho ions on the absorption peak at 686 and 792 nm can be ignored because Ho ions have no significant absorption at these wavelengths.

With the measured absorption spectrum and optical density, the absorption coefficient of Tm ions can be calculated by

$$a = \frac{-\ln(I/I_0)}{L} = \frac{2.303OD}{L}, \tag{1}$$

where OD is optical density, L is the thickness of the sample. The relation between absorption cross section and absorption coefficient is

$$\sigma = a/N, \tag{2}$$

where N is the number of absorption center per volume.

Using the above equations, we can obtain the absorption coefficient and absorption cross section of Tm,

Ho:LuLF crystal at 792 nm, which are 4.03 cm^{-1} and 5.58×10^{-21} cm^2, respectively, indicating a higher optical quality of our crystal compared with the results of Sudesh et al.[16], which were 3.73 cm^{-1} for absorption coefficient and 5.2×10^{-21} cm^2 for absorption cross section.

Fluorescence spectrum of Tm, Ho:LuLF crystals is shown in Fig. 3. There is only one significant emission band appearing at 2.05 μm, which can be attributed to the Ho $^5I_7 - ^5I_8$ transition. It indicates that pumped by a laser diode, Tm, Ho:LuLF crystals have the potential of producing 2.05-μm laser. By analyzing the absorption and fluorescence spectra of the Tm, Ho:LuLF crystal, we get the principal excitation mechanism and energy transfer process in Tm-Ho lasers, as shown in Fig. 4.

Tm^{3+} ions are excited from ground state of $^3H_6$ to excited state of $^3H_4$. When Tm ions are in the $^3H_4$ manifold, they can self-quench to the $^3F_4$ manifold. This self-quenching process makes it possible for every pump photon absorbed, and there are two ions may reach the $^3F_4$ manifold. Meanwhile, there is a fast energy transfer from Tm^{3+} $^3F_4$ manifold to Ho^{3+} $^5I_7$ manifold. This energy migration results a significant concentrations of Ho^{3+} ions in $^5I_7$ manifold. Finally, laser action takes place on the Ho $^5I_7 - ^5I_8$ transition. When the number of Ho^{3+} ions in $^5I_7$ manifold reaches the threshold value, up-conversion process ($^5I_7 - ^5S_2 + ^5F_4$) takes place. Accompany with the up-conversion process, there are transitions from $^5F_4$ manifold and $^5S_2$ manifold to $^5I_8$ manifold, on which 540-nm green light is produced. But we did not detect emission peak at 540 nm in our fluorescence spectrum in the range of $200 - 800$ nm.

We studied the laser performance of the as-grown Tm, Ho:LuLF crystal. Figure 5 shows the schematic of the laser setup. A commercial AlGaAs laser diode was used as a pump source with pump wavelength centering at 795 nm. Specimen for this measurement was an a-axis grown Tm (5%), Ho (0.5%):LuLF$_4$ single crystal rod, 2 mm in diameter and 10 mm in length. Both surfaces of the crystal were optical flat. On the pumping surface, there was a coating with high reflectivity (HR) at 2000 nm ($R > 95\%$) and high transmissivity at (HT) 795 nm ($T > 90\%$), while the output side was a mirror with a coating with 95% reflectivity at 2000 nm. Hence the surface on the pumping side and the output mirror formed a flat-flat laser cavity, and the axis of the cavity was parallel to the axis of the crystal. The pumping laser light was focused onto the crystal through collimating and focusing lenses. The crystal was coated with an indium paper and set on a copper crystal holder, which was cooled by a TE-cooler.

The pump source used in our experiment was centered at 795 nm under continuous-wave (CW) operations. To our knowledge, there are many reports about Ho,Tm doped crystals' laser performance pumped by a source under CW operations[21−24], but we have not read any report about Ho,Tm:LiLuF lasers pumped by a CW operation source. The absorbance of the crystal against the pump laser light is higher than 80%. The minimum input power was 1.83 W. An laser output at 2.05 μm with a power of 50 mW was obtained when the input power reached 5 W. The slope efficiency was 2%, and it could be increased by improving the crystal
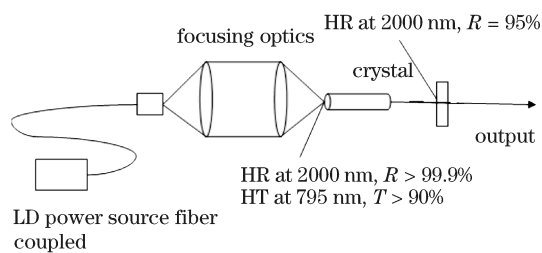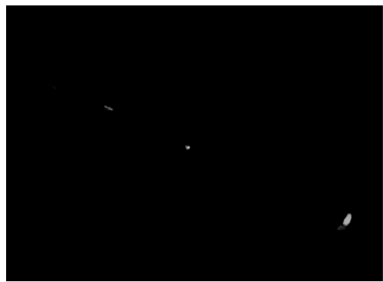
Fig. 5. Schematic of the laser setup.

Fig. 6. Picture of the green light emission.

quality and optimizing the laser set up and cooling system.

We observed a significant luminescence of green light when pumping light was focused on the crystal, which is shown in Fig. 6. As we discussed above, the green light emission was due to the transition of Ho^{3+} $^5S_2 - ^5I_8$. Tm, Ho:LuLF crystal is a quasi-three-level system, in which up-conversion effect takes place when the crystal works at room temperature. In the up-conversion process, ions can be excited onto energy levels which are higher than the $^5I_7$ manifold. Ions in those metastable states are easily self-quench to a lower state and the fluorescence is generated. In our experiment, we observed green light when the pumping laser light was focused on the crystal with the pumping power lower than the laser threshold value. It indicates that the up-conversion process can take place as long as the pumping power is focused on the crystal. The up-conversion process can deplete ions in the $^5I_7$ manifold and subsequently lower the laser gain.

In conclusion, the Tm (5%), Ho (0.5%) codoped LuLF crystal was grown by the Czochralski technique. The absorption and fluorescence spectra of the as-grown crystal were studied. We analyzed the excitation mechanism and energy transfer process in the Tm, Ho:LuLF crystal and then demonstrated our discussion by operating an laser experiment on the crystal. According to our analysis, there should be a fluorescence peaking at 540 nm, but we did not detect it in our fluorescence spectrum, and this may be due to that the pump power was too low. We also observed a significant green light emission in our experiment, in which the Tm, Ho: $LiLuF_4$ single crystal was end-pumped by a fiber-coupled laser diode system with pumping wavelength of 795 nm. We achieved a 50-mW continuous laser with wavelength of 2.05 μm.

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