Demonstration of Diode-Pumped Yb:LaF$_3$ and Tm,Ho:LaF$_3$ Lasers

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Abstract: Diode-pumped solid-state lasers using novel Yb:LaF$_3$ and Tm,Ho:LaF$_3$ crystals as laser gain materials are demonstrated herein. The Yb:LaF$_3$ and Tm,Ho:LaF$_3$ crystals were grown using the Bridgman method. By matching their absorption bands, continuous-wave laser operations were achieved for the first time. The Yb:LaF$_3$ laser obtained a maximum average output power of 1.19 W with dual wavelengths of 1028 nm and 1033 nm. The maximum average output power and slope efficiency of the Tm,Ho:LaF$_3$ laser were 574 mW and 18.5%, respectively. The Tm,Ho:LaF$_3$ laser exhibited two peaks at 2043 nm and 2048 nm. Both the Yb:LaF$_3$ and Tm,Ho:LaF$_3$ crystals were confirmed to be laser gain materials.

Keywords: Yb:LaF$_3$; Tm,Ho:LaF$_3$; continuous-wave laser

1. Introduction

Diode-pumped solid-state lasers (DPSSLs) have attracted wide attention owing to their advantages including high conversion efficiency, good beam quality, long life, and compact construction [1,2]. Over the past few decades, DPSSLs have been fully developed and have been extensively applied in scientific research, medical care, industry, and other fields, and they are now still moving forward to provide more diversified applications [3,4]. For promoting the advancement of DPSSLs, a major research topic is the study of laser gain materials.

For DPSSLs, the laser gain materials are formed by incorporating active ions into host materials whose characteristics greatly affect the physical, chemical, and mechanical properties of the laser gain materials. Fluoride crystals are of particular research interest as host materials because they have distinctive properties: (i) wide wavelength-transparent areas ranging from near ultraviolet to mid-infrared; (ii) a low phonon energy, which is beneficial for reducing non-radiative relaxation; and (iii) a high damage threshold [5–8].

Yb$^{3+}$ ions, as representative active ions emitting near 1 µm in lasers, have been successfully used to realize tunable lasers, quasi-continuous devices, femtosecond pulse operations, and high-power systems [9–12]. Yb$^{3+}$ ions have a simple energy level structure: the $^2F_{7/2}$ ground state and the $^2F_{5/2}$ excited state [13]. Different from other rare-earth ions, Yb$^{3+}$ ions have no other 4f electronic states; therefore, there is no excited-state absorption or fluorescence up-conversion that has an adverse influence on laser performance. Consequently, laser systems with Yb$^{3+}$-doped materials would have a
high conversion efficiency. After decades of development, Yb-doped fluoride crystals, such as Yb:CaF₂, have been comprehensively reported [14–17].

In recent years, thulium (Tm) and holmium (Ho) active ions have been investigated because of their luminescence emissions near the 2 μm region in lasers, giving them great application potential in human eye safeness and high atmospheric transmission [18–24]. Tm and Ho co-doped materials are promising candidates to directly obtain 2 μm region lasers with a compact structure. The around 790 nm laser diode (LD) acts as a pumped source to excite Tm³⁺ ions, then, according to the energy transfer between Tm³⁺ ions and Ho³⁺ ions, Ho³⁺ ions ultimately produce 2 μm laser radiation [25,26]. Therefore, Tm,Ho co-doped materials could be operated as laser gain materials for the DPSSL system. Thus far, studies on Tm,Ho co-doped fluoride crystals have focused on Tm,Ho:YLF, Tm,Ho:KYF and Tm,Ho:LLF [8,19–21,27–29].

In this work, we report two novel fluoride crystal materials: Yb:LaF₃ and Tm,Ho:LaF₃. Yb:LaF₃ and Tm,Ho:LaF₃ crystals are typical rare-earth-ion-doped fluoride crystals, so they possess the luminescent properties of rare-earth ions and the excellent host advantages of fluoride crystals. Moreover, La ions are of +3 valence, the same as Yb, Tm, and Ho ions; therefore, there is no charge imbalance in the process of ion doping. However, to date, Yb:LaF₃ crystals have not achieved laser output as solid-state laser gain materials, and Tm,Ho:LaF₃ crystals have not been reported [30].

Yb:LaF₃ and Tm,Ho:LaF₃ crystals were prepared via the Bridgman growth method. Yb:LaF₃ crystals with a doping concentration of 1% had a maximum absorption coefficient of 0.25 cm⁻¹ at 973 nm. Matching the strongest absorption, Yb:LaF₃ continuous-wave (CW) lasers were demonstrated with a maximum average output power of 1.19 W. When the output mirror had a transmittance value of 5%, a Tm,Ho:LaF₃ CW laser was built and the maximum average output power was 416 mW. Employing an output mirror with a transmission value of 2%, the Tm,Ho:LaF₃ CW laser obtained a maximum average output power of 574 mW. The novel crystals, Yb:LaF₃ and Tm,Ho:LaF₃, were used as laser gain materials to demonstrate CW laser operations as DPSSLs.

2. Spectral Properties of Materials

The growth method of the Yb:LaF₃ crystals (Pujing Company, Suzhou, Jiangsu, China) was the Bridgman method which has the advantage of inhibiting the volatilization of fluoride. The Yb:LaF₃ crystals with dimensions 3 × 3 × 6 mm³ had a Yb doping concentration of 1%. As seen from Figure 1a, the crystals had a relatively high absorption coefficient over a wide wavelength range. The maximum absorption coefficient was 0.25 cm⁻¹ at 973 nm. Figure 1b shows the broad emission band whose full width at half-maximum (FWHM) was 52.4 nm with Gauss fitting (the red curve), revealing Yb:LaF₃ crystals to be a candidate for wavelength-tunable lasers.

**Figure 1.** (a) Absorption spectrum of Yb:LaF₃ (1 atom % of Yb); (b) emission spectrum with an 896 nm excitation source.

The Tm,Ho:LaF₃ materials were also prepared using the Bridgman growth method. The doping concentration was 5 atom % of Tm and 0.5 atom % of Ho ions, and the dimensions were
3 × 10 mm³. The absorption and emission spectrum information at room temperature are shown in Figure 2. It can be seen that the Tm,Ho:LaF₃ materials had several peaks in the absorption spectrum. Among them, a special absorption peak is presented deliberately in the inset of Figure 2a. This peak had a center wavelength of 792.5 nm, which illustrated that the Tm,Ho:LaF₃ materials could be stimulated with a commercial LD. In general, LDs have the disadvantage of wavelength drift caused by changes in the LD output power and temperature. The Tm,Ho:LaF₃ crystals had a FWHM of 34.8 nm, so they could avoid absorption instability caused by the wavelength drift of LDs. The emission spectrum had a broad fluorescence range mainly centered at 1925–2075 nm.

![Figure 2](image2.png)

**Figure 2.** Spectral information for Tm,Ho:LaF₃ materials: (a) absorption spectrum, (b) emission spectrum.

3. **Continuous-Wave Laser Experiment Operation**

The respective properties of CW lasers were researched with the Yb:LaF₃ and Tm,Ho:LaF₃ samples by building laser systems, which are presented in Figure 3. Two commercial LDs were used as the pump source. Matching their absorption wavelengths, an LD with a center wavelength of 974 nm was applied to the Yb:LaF₃ laser, and the Tm,Ho:LaF₃ laser was used with an LD with a central wavelength of 793 nm. The LDs both had a fiber core diameter of 200 μm and a numerical aperture of 0.22. The pump source laser they sent was focused by a 1:1 optics coupling system. In order to effectively dissipate the heat, the Yb:LaF₃ and Tm,Ho:LaF₃ samples were both wrapped with indium foil and mounted in a water-cooled system where the water was maintained at 20 °C temperature. Two mirrors (M1 and M2) formed a laser cavity with a length of 35 mm. One of them (M1) had a curvature of 100 mm, and the other (M2) was a flat mirror. For the Yb:LaF₃ experiment, M1 was the input mirror having high transmission at the pump wavelength and high reflection at 1030–1090 nm. M2 was the output mirror, having transmission values of 1%, 2%, or 5% at 1030–1090 nm. M3 was the same type of mirror as M1 and was used to separate the pump laser and the Yb:LaF₃ laser. The pump laser of 974 nm was transmitted in Direction 2 (D2), and the Yb:LaF₃ laser was reflected in Direction 1 (D1). For the Tm,Ho:LaF₃ laser, M1 had high transmission at a 793 nm wavelength and high reflection at 1.9–2.1 μm. There were two kinds of mirrors (M2) with transmission values of 2% and 5% at 1.9–2.1 μm. M3 was a beam splitter mirror. At a 45° angle, the 793 nm laser was reflected in D1, whereas the target laser was obtained in D2. The Yb:LaF₃ and Tm,Ho:LaF₃ samples were each placed near the M1 mirrors.

![Figure 3](image3.png)

**Figure 3.** Experimental setup of the continuous-wave (CW) laser. LD: laser diode; M1 and M2: two cavity mirrors; M3: beam splitter mirror; D1 and D2: direction 1 and 2.
First, the absorption efficiency was measured for the Yb:LaF$_3$ and Tm,Ho:LaF$_3$ crystals in non-lasing conditions. As shown in Figure 4, as the injection pump power was increased and the absorbed pump power was added linearly. The absorption efficiency of the Yb:LaF$_3$ crystal was found to be 22.9%, and the maximum injection pump power was 20.13 W. For the Tm,Ho:LaF$_3$ crystal, the absorption efficiency and the maximum injection pump power were found to be 21.9% and 15.85 W, respectively. Considering that a large amount of pump laser was not absorbed, the M3 mirror was used to split the pump laser and target laser.

![Figure 4. Variation of the absorbed pump power with the injection pump power.](image)

After the cavity mirrors were adjusted, the CW lasers of the Yb:LaF$_3$ crystal were obtained in D1. The relationship between the average output power and the absorbed pump power for different transmittance values is illustrated in Figure 5. When the transmittance of M2 was 1% and the absorbed pump power was 1.54 W, the Yb:LaF$_3$ laser was obtained. When the absorbed pump power was increased to 4.91 W, a maximum average output power of 904 mW was achieved, corresponding to a slope efficiency of 28.2%. With the highest transmittance of 5%, a maximum output power of 656 mW with a slope efficiency of 32.2% was obtained. The optimal average output power was 1.19 W, when the transmittance of output coupler was 2%. The highest slope efficiency was 37.1%.

![Figure 5. The average output power as a function of the absorbed pump power.](image)

The spectra of the Yb:LaF$_3$ lasers were recorded using a spectrometer (USB2000, Ocean Optics, Largo, Florida, FL, United States). For different transmittance values of M2, the experimental results are shown in Figure 6 at the maximum average output power. When M2 had transmittance values of 1% and 5%, the output spectra both exhibited a single peak. The central wavelengths were 1029 nm...
and 1018 nm, respectively. At a transmittance value of 2%, double peaks appeared with wavelengths of 1028 nm and 1033 nm. Because the sampling resolution of the spectrometer was 0.7 nm, the fine spectral components were not tested. However, our experimental results could reflect the information of the output spectra to a certain extent.

After the position of the cavity mirrors was carefully adjusted, Tm,Ho:LaF\(_3\) lasers were acquired in D2. The experimental results of the relationship between the absorbed pump power and the average output power are explained in Figure 7. For different output mirrors, it is obvious that the absorbed pump power and average output power were almost linear functions. At the absorbed pump power of 3.53 W, the maximum average output power was 574 mW and the slope efficiency was 18.5% under 2% transmittance by M2. When a higher transmittance of M2 of 5% was adopted, the maximum average output power and the slope efficiency were decreased to 416 mW and 14.2%, respectively. The power was tested using a power meter (30(150)A-BB-18, Ophir, Jerusalem, Israel).

The spectra of the Tm,Ho:LaF\(_3\) lasers, shown in Figure 8, were measured using a spectrometer (NIRQuest-512, Ocean Optics, Largo, Florida, FL, United States). The laser spectra exhibited two peaks with both of the two output mirrors used in the laser test. When the transmittance was 2% and the average output power was 574 mW, the spectra showed two peaks at 2043 nm and 2048 nm. At a transmittance of 5% and output power of 416 mW, the peaks were 2039 nm and 2041 nm.
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Conflicts of Interest: The authors declare no conflict of interest.

References
1. Fan, T.Y.; Byer, R.L. Diode laser-pumped solid-state lasers. *IEEE J. Quantum Electron.* **1988**, *24*, 895–912. [CrossRef]
2. Byer, R.L. Diode laser-pumped solid-state lasers. *Science* **1988**, *239*, 742–747. [CrossRef] [PubMed]
3. Andriukaitis, G.; Ališauskas, S.; Pugžlys, A.; Baluška, A.; Tan, L.H.; Lim, J.H.W.; Phua, P.B.; Balskus, K.; Michailovas, A. Broadband 6-μm OPA driven by Yb:CaF$_2$ DPSSL system. In Proceedings of the Lasers and Electro-Optics, Optical Society of America, San Jose, CA, USA, 6–11 May 2012; CF3B.6.
4. Hugel, H. New solid-state lasers and their application potentials. *Opt. Laser Eng.* 2000, 34, 213–229. [CrossRef]

5. Hong, J.Q.; Zhang, L.H.; Hang, Y. Enhanced 2.86 μm emission of Ho3+,Pr3+-co-doped LaF3 single crystal. *Opt. Mater. Express* 2017, 7, 1509–1513. [CrossRef]

6. Li, S.M.; Zhang, L.H.; Zhang, P.X.; Hong, J.Q.; Xu, M.; Yan, T.; Ye, N.; Hang, Y. Spectroscopic characterizations of Dy:LaF3 crystal. *Infrared Phys. Technol.* 2017, 87, 65–71. [CrossRef]

7. Li, C.; Liu, J.; Jiang, S.Z.; Xu, S.C.; Ma, W.W.; Wang, J.Y.; Xu, X.D.; Su, L.B. 2.8 μm passively Q-switched Er:CaF3 diode-pumped laser. *Opt. Mater. Express* 2016, 6, 1570–1575. [CrossRef]

8. Galzerano, G.; Sani, E.; Toncelli, A.; Valle, G.D.; Taccheo, S.; Tonelli, M.; Laporta, P. Widely tunable continuous-wave diode-pumped 2-μm Tm:Ho:YLF laser. *Opt. Lett.* 2004, 29, 715–717. [CrossRef] [PubMed]

9. Krupke, W.F. Ytterbium solid-state lasers—The first decade. *IEEE J. Sel. Top. Quantum Electron.* 2000, 6, 1287–1296. [CrossRef]

10. Yu, H.; Liu, Y.Q.; Guo, J.; Zhang, L.Y.; Lin, H.; Liang, X.Y. Laser performance of a broadband wavelength tunable Ybgermanophosphate glass with direct diode pumping. *Chin. Opt. Lett.* 2017, 15, 071406. [CrossRef]

11. Lu, J.; Zou, X.; Li, C.; Li, W.K.; Liu, Z.Z.; Liu, Y.Q.; Leng, Y.X. Picosecond pulse generation in a monolayer MoS2 mode-locked Ytterbium-doped thin disk laser. *Chin. Opt. Lett.* 2017, 15, 041401.

12. Brenier, A.; Boulon, G. Overview of the best Yb3+-doped laser crystals. *J. Alloy Compd.* 2001, 323–324, 210–213. [CrossRef]

13. Zhu, J.F.; Gao, Z.Y.; Tian, W.L.; Wang, Z.H.; We, Z.Y.; Zheng, L.H.; Su, L.B.; Xu, J. Kerr-lens mode-locked femtosecond Yb:GdYSiO5 laser directly pumped by a laser diode. *Appl. Sci.* 2015, 5, 817–824. [CrossRef]

14. Šulc, J.; Ježková, H.; Doroshenko, M.E.; Basiev, T.T.; Konyushkin, V.A.; Fedorov, P.P. Tunability of lasers based on Yb3+-doped fluorides SrF2, SrF2-CaF2, SrF2-BaF2, and YLF. In Proceedings of the Advanced Solid-State Photonics, Optical Society of America, Denver, CO, USA, 1–4 February 2009. WB16.

15. Sévillano, P.; Machinet, G.; Dubrasquet, R.; Camy, P.; Doualan, J.L.; Moncorgé, R.; Georges, P.; Druon, F.; Descamps, D.; Cormier, E. Sub-50 fs, Kerr-lens mode-locked Ytterbium-doped thin disk laser. *Chin. Opt. Lett.* 2017, 15, 041401.

16. Singh, U.N. Tm:Ho:YLF and LuLiF laser development for global winds measurements. In Proceedings of the 13th International Conference on Fiber Optics and Photonics (Optical Society of America), Kanpur, India, 13–17 November 2013. AF3A.6.

17. Siebold, M.; Hornung, M.; Boedefeld, R.; Podleska, S.; Klingebiel, S.; Wandt, C.; Krausz, F.; Karsch, S.; Uecker, R.; Joehmann, A.; et al. Terawatt diode-pumped Yb:CaF2 laser. *Opt. Lett.* 2008, 33, 2770–2772. [CrossRef] [PubMed]

18. DeShano, B.R.; Cook, G.; Harris, T.R.; Jessen, H.P.; Cassanho, A. Efficient 2-micron Ho lasers based on fluoride crystal hosts. In Proceedings of the Laser Technology for Defense and Security XIV (Proc. of SPIE), Orlando, FL, USA, 4 May 2018. 1063712.

19. Zhang, X.L.; Ju, Y.L.; Wang, Y.Z. Diode-end-pumped room temperature Tm:Ho:YLF lasers. *Opt. Express* 2005, 13, 4056–4063. [CrossRef] [PubMed]

20. Sudesh, V.; Asai, K. Spectroscopic and diode-pumped-laser properties of Tm, Ho:YLF; Tm, Ho:LuLF; and Tm, Ho:LaF3 crystals: A comparative study. J. Opt. Soc. Am. B 2003, 20, 1829–1837. [CrossRef]

21. Singh, U.N. Tm:Ho:YLF and LuLiF laser development for global winds measurements. In Proceedings of the 13th International Conference on Fiber Optics and Photonics (Optical Society of America), Kanpur, India, 13–17 November 2013. AF3A.6.

22. Hong, J.Q.; Zhang, L.H.; Hang, Y. Optical characterization of Tm3+ in LaF3 single crystal. *Infrared Phys. Technol.* 2017, 82, 50–55. [CrossRef]

23. Hong, J.Q.; Zhang, L.H.; Zhang, P.X.; Xu, M.; Hang, Y. Ho:LaF3 single crystal as potential material for 2 μm and 2.9 μm lasers. *Infrared Phys. Technol.* 2016, 76, 636–640. [CrossRef]

24. Payne, S.A.; Chase, L.L.; Smith, L.K.; Kway, W.L.; Krupke, W.F. Infrared cross-section measurements for crystals doped with Er3+, Tm3+, and Ho3+. *IEEE J. Quantum Electron.* 1992, 28, 2619–2630. [CrossRef]

25. Louchev, O.A.; Urata, Y.; Saito, N.; Wada, S. Computational model for operation of 2 μm co-doped Tm, Ho solid state lasers. *Opt. Express* 2007, 15, 11903–11912. [CrossRef]
26. Walsh, B.M.; Barnes, N.P.; Bartolo, B.D. Branching ratios, cross sections, and radiative lifetimes of rare earth ions in solids: Application to Tm$^{3+}$ and Ho$^{3+}$ ions in LiYF$_4$. *J. Appl. Phys.* 1998, 83, 2772–2787. [CrossRef]

27. Sato, A.; Aoki, M.; Ishii, S.; Otsuka, R.; Mizutani, K.; Ochiai, S. 7.28-W, high-energy, conductively cooled, Q-Switched Tm,Ho:YLF laser. *IEEE Photonics Technol.* 2017, 29, 134–137. [CrossRef]

28. Ling, W.J.; Tao, X.; Dong, Z.; Zuo, Y.Y.; Li, K.; You, L.F.; Lu, F.P.; Liu, Q. Passively Q-switched mode-locked Tm,Ho:LLF laser with a reflection-type MoS$_2$ saturable absorber. In Proceedings of the 2017 International Conference on Optical Instruments and Technology: Advanced Laser Technology and Applications (Proc. of SPIE), Beijing, China, 28–30 October 2017. 106190Q.

29. Dinndorf, K.M.; Miller, H. Two micron diode-pumped laser operation of Tm,Ho:BaY$_2$F$_8$. In Proceedings of the Advanced Solid State Lasers (Optical Society of America), Boston, MA, USA, 31 January–3 February 1999. WB17.

30. Hong, J.Q.; Zhang, L.H.; Hang, Y.; Xu, M. Spectroscopic and thermal characterizations of Yb:LaF$_3$ single crystal. *Opt. Mater.* 2016, 60, 128–131. [CrossRef]