

Laser operation of Tm near 2- μm in the disordered double tungstate host NaGd(WO₄)₂

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Abstract: Lasing with an output power up to 300 mW is reported for Tm:NaGd(WO₄)₂, both with Ti-sapphire and diode laser pumping. This disordered crystal grown by the Czochralski method allowed tuning from 1813 to 2025 nm.

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1. Introduction

The use of the ${}^3F_4 \rightarrow {}^3H_6$ Tm³⁺-transition for all-solid-state lasers operating near 2 μm is attractive because AlGaAs laser diodes emitting near 800 nm can be used for efficient pumping and power scaling. The $4f^{12}$ configuration of Tm³⁺ is shielded from the crystal field of the host ligands by outer 5s and 5p electrons and the coupling to the vibronic environment is relatively weak. In some cases, however, the phonon broadening is larger and laser tuning from 1870 to 2160 nm and from 1850 to 2140 nm was demonstrated with the cubic oxides Tm:YAG and Tm:YSGG, respectively [1]. Further broadening of the spectral features can be expected for solids with multisites, defects or lattice disorder; the use of glasses, particularly at high power levels, is limited by their poor thermal properties. Recently there were some attempts to study such multisite fluorites for room temperature continuous-wave (CW) laser operation: Thus tuning between 1835 and 1970 nm was demonstrated with Tm:CaF₂ [2] while the initial results with Tm:KYF₄ (KYF) were modest and only narrow tunability (2010-2022 nm) was achieved [3].

The disordered tetragonal crystal NaGd(WO₄)₂ (NaGdW) was used as a host for Nd³⁺ as early as 1964 [4]. It belongs to a more general class NaT(WO₄)₂, where T is a trivalent cation (Al, Ga, In, Cr, Bi, Y, La or Ln³⁺=Ce-Lu). The spectral broadening in such double tungstates is related not only to the random distribution of Na and T cations but also to the existence of two lattice sites for the T cations because the refined crystalline structure has a symmetry $I\bar{4}$ [5]. Recently, we achieved tunability from 1016 to 1049 nm with Yb:NaGdW [6]. Here, we report on room temperature CW lasing of Tm:NaGdW, to the best of our knowledge for the first time with such a disordered crystal.

2. Crystal growth, structure, and spectroscopy of Tm:NaGdW

NaGdW crystals grown by the Czochralski method exhibit the acentric tetragonal space group $I\bar{4}$ where Na⁺ and Gd³⁺ reside in two non-equivalent $2b$ and $2d$ lattice sites, both with local S_4 point symmetry if all ligands of a given site are either only Na⁺ or Gd³⁺ [5]. The single crystal of NaGd_{1-x}Tm_x(WO₄)₂ was grown by the Czochralski technique with $x_{\text{melt}}=0.05$. The starting materials were 99.99% Gd₂O₃, 99.99% Tm₂O₃, 99.5% Na₂CO₃, and 99.8% WO₃. The stoichiometric composition was melted in a 75 cc Pt-crucible with addition of 1.3 mol % of Na₂W₂O₇ to slightly decrease the melting temperature of the mixture and to partially compensate for Na and W evaporation. The rotation and pulling rates were 10 rpm and 1 mm/h, respectively. The grown crystal was cooled down to room temperature at a rate of 10°C/h. The crystal dimensions were about 20 mm in diameter and 50 mm in length. Although the crystal was transparent, it exhibited some optical gradients with a period of about 1 mm along the growth axis. The tetragonal structure of the grown crystal was confirmed by X-ray powder diffraction. The Tm density in the crystal was $2.34 \times 10^{20} \text{ cm}^{-3}$ giving a segregation coefficient of 0.7.

Detailed spectroscopic results including absorption and photoluminescence studies at 5 and 300 K will be published elsewhere [7]. They allowed to identify the Stark levels of the multiplets. The 3H_6 splitting in Tm:NaGdW is relatively large which reduces the thermal population of the lower laser level for the ${}^3F_4 \rightarrow {}^3H_6$ transition.

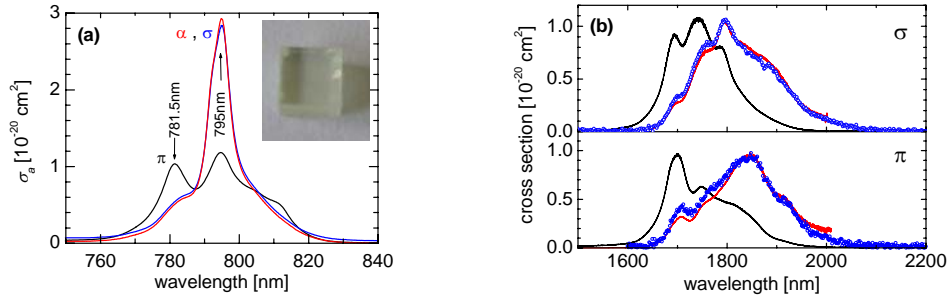


Fig. 1: ${}^3\text{H}_6 \rightarrow {}^3\text{H}_4$ ground state absorption cross sections σ_a of Tm:NaGdW for α , σ and π polarizations with an inset showing the $\approx 3 \times 3 \times 3 \text{ mm}^3$ sample used in the laser experiments (a). Measured ${}^3\text{H}_6 \leftrightarrow {}^3\text{F}_4$ ground state absorption cross sections (black lines) and calculated emission cross sections (red lines); the experimental photoluminescence excited at 795 nm (blue points) is shown for comparison (b).

Figure 1a shows the absorption cross sections σ_a for the ${}^3\text{H}_6 \rightarrow {}^3\text{H}_4$ transition recorded at 300 K. These bands match rather well the emission spectrum of AlGaAs diode lasers. The α and σ spectra are very similar. Their maximum cross sections exceed by a factor of more than 2 the cross section for the π polarization at the same peak wavelength, 795 nm: $\sigma_a(\sigma) \approx \sigma_a(\alpha) = 2.9 \times 10^{-20} \text{ cm}^2$ and $\sigma_a(\pi) = 1.18 \times 10^{-20} \text{ cm}^2$. The FWHM of the more intense σ and α absorption lines is about 8 nm which relaxes the requirements for pumping with diode lasers. Figure 1b shows the measured absorption and the calculated, using the reciprocity method, emission cross sections σ_e for the ${}^3\text{F}_4 \rightarrow {}^3\text{H}_6$ laser transition. They agree rather well with the experimentally recorded photoluminescence profiles. The maximum emission cross sections are $\sigma_e(\sigma) = 10.6 \times 10^{-21} \text{ cm}^2$ at 1796 nm and $\sigma_e(\pi) = 9.5 \times 10^{-21} \text{ cm}^2$ at 1847 nm. The lifetime obtained by the pinhole method amounts to $1.35 \pm 0.20 \text{ ms}$ for this Tm doping level.

3. Laser operation

The Tm:NaGdW laser was first pumped by a tunable Ti:sapphire laser with a linewidth of $\approx 0.2 \text{ nm}$ delivering an output power up to 3 W near 800 nm. The pump beam was focused by an $f=70 \text{ mm}$ lens to a spot with a Gaussian waist of $37 \mu\text{m}$. The astigmatically compensated cavity of the X-type had a total length of 90 cm. The uncoated a -cut sample (inset Fig.1a) was placed under Brewster angle between the two folding mirrors with $\text{RC}=10 \text{ cm}$ which determines the laser polarization and the pump polarization was always in the same plane. The sample was clamped in a Cu block and the temperature was maintained at 10°C by water cooling.

The optimum pump wavelength was 795.5 and 795.6 nm for the σ and π polarizations, respectively. Figure 2a shows the output power versus absorbed pump power P_{abs} for different output coupling T_{OC} . In the case of σ polarization, an output power of 295 mW was achieved for $P_{abs}=1054 \text{ mW}$ ($T_{OC}=3\%$). For π polarization, the maximum output reached 207 mW for $P_{abs}=740 \text{ mW}$ ($T_{OC}=1.5\%$). The higher output power for the σ polarization was mainly due to the higher absorption: The efficiency in terms of P_{abs} as well as the oscillation wavelengths were rather similar. The laser threshold in terms of P_{abs} was slightly lower for the π polarization. A systematic decrease of the laser wavelength from 1943 to 1895 nm with T_{OC} was observed which is related to the increased inversion rate. The slight roll-over with increasing power (Fig.2a) is an indication of thermal population of the lower laser level.

Figure 2b shows the tunability achieved inserting a 3-mm thick quartz plate whose optical axis was at 60° to the surface, for $P_{abs}=1.05$ and 0.74 W for the σ and π polarization, respectively. The laser wavelength was tunable from 1813 to 2025 nm in the case of σ polarization and from 1817 to 1998 nm for the π polarization. The tunability limits achieved at the zero level for the σ polarization correspond to $\approx 17 \text{ THz}$. Such a full tuning range is one of the largest found in Tm^{3+} -doped solid state lasers. While the tunability achieved with the ordered monoclinic Tm:KLu(WO₄)₂ using the same setup was only 10% narrower [8] it should be outlined that in that case the crystal quality was much better and the maximum output level was above 1 W. It can be expected that once the quality of Tm:NaGdW is also improved and the laser efficiency correspondingly increased, the tuning range will be even broader.

Diode laser pumping of the same Tm:NaGdW sample, again mounted in a water cooled Cu holder with water temperature of 12°C , was studied in a nearly hemispherical cavity. The diode laser module containing a single 50 W commercial bar with 19 emitters and 30% fill factor was mounted in a conduction cooled packaging [8]. Only simple adapted beam shaping optics was used for the pump beam which was unpolarized. The nearly collimated beam had roughly a square cross section with a size of several millimeters. For the present experiment the pump wavelength changed only from about 800 nm near threshold up to about 801 nm at the maximum incident power of

9.6 W applied. The single-peaked pump spectrum had a FWHM of ≈ 2 nm. The laser diode output was collimated by a lens system with $f=34$ mm and 80% transmission, and focused by a lens system with $f=20$ mm and 88% transmission through the plane total reflector of the cavity. The nearly circular spot achieved had a cross section with a diameter of about $125 \mu\text{m}$ [8]. The output couplers used had a $RC=50$ mm.

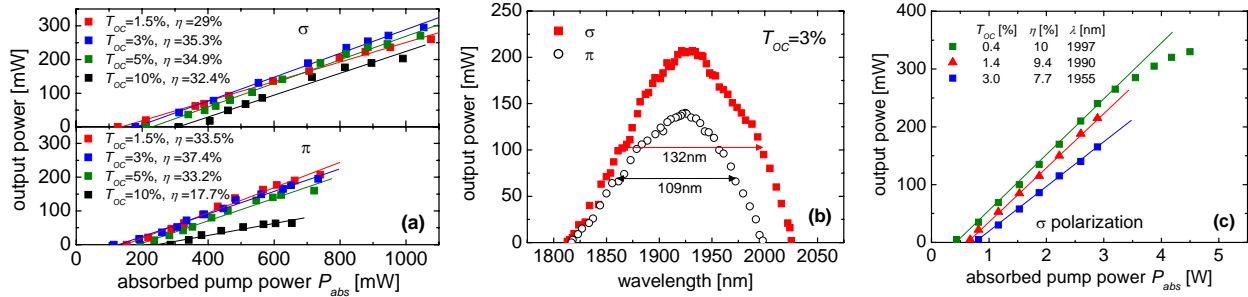


Fig.2: Input-output characteristics of the Tm:NaGdW laser (symbols) and fits for calculation of the slope efficiencies η (lines) (a). Tuning of the Tm:NaGdW laser using an intracavity Lyot filter for an incident pump power of 1.54 W (b). Input-output characteristics of the diode-pumped Tm:NaGdW laser (c).

The maximum output power achieved with the diode pumped Tm:NaGdW laser and a $T_{OC}=0.4\%$ output coupler was 330 mW when pumping with 9 W (incident power) corresponding to $P_{abs}=4.5$ W. The M^2 factor was of the order of 2. In terms of P_{abs} the threshold was 430 mW and the slope efficiency was 10%. The laser wavelength was 1997 nm in this case and got shorter for increasing T_{OC} as in the case of Ti:sapphire laser pumping. The increased oscillation wavelength and thresholds in comparison to Ti:sapphire laser pumping are a consequence of the lower gain and inversion rates. Since there were no polarization selecting surfaces in this cavity the laser selected the polarization only according to the higher gain and this was always the σ polarization. This is in agreement with the slightly better performance of this polarization in the Ti:sapphire laser pumped configuration. Roll-over in the power dependence can be clearly seen in Fig.2c. Also the oscillation wavelength increased with the pump level and reached, e.g. for $T_{OC}=0.5\%$, 2002 nm. The increase of the laser wavelength is a further indication of thermal effects which limit at present the performance of this laser. Note that reduction of the reabsorption losses at higher pump levels would, on the contrary, result in shorter oscillation wavelengths.

4. Conclusion

We achieved tunable laser operation of Tm^{3+} in NaGdW near $2 \mu\text{m}$ pumping it around 800 nm. The output powers obtained at room temperature in the CW regime (≈ 300 mW) are modest but normal for this initial attempt to grow Tm:NaGdW. The laser gain is slightly higher for the σ -polarization. The continuous tunability extends from 1813 to 2025 nm, one of the largest achieved with a Tm^{3+} -doped crystalline material, despite the relatively low efficiency obtained with this first sample of Tm:NaGdW. Considerable improvement in the laser performance can be expected after elimination of the internal refractive index gradients observed in the first grown crystal.

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