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Tunable quasi-cw two-micron lasing in diode-pumped crystals of mixed Tm^{3+}-doped sodium – lanthanum – gadolinium molybdates and tungstates

F.A. Bol'shchikov, E.V. Zharikov, D.A. Lis, N.G. Zakharov, P.A. Ryabochkina, K.A. Subbotin, O.L. Antipov

***Abstract*. Two-micron lasing is obtained for the first time on the $^3F_4 \rightarrow ^3H_6$ transition of Tm^{3+} ions in diode-pumped crystals of mixed sodium – lanthanum – gadolinium tungstate $Tm:NaLa_{1/2}Gd_{1/2}(WO_4)_2$ ($C_{Tm} = 3.6$ at %) (3.6Tm:NLGW) and molybdate $Tm:NaLa_{1/3}Gd_{2/3}(MoO_4)_2$ ($C_{Tm} = 4.8$ at %) (4.8Tm:NLGM). For the 3.6Tm:NLGW crystal, the quasi-cw laser output power exceeded 200 mW and the slope efficiency (with respect to absorbed pump power) for the π- and σ-polarisations at wavelengths of 1908 and 1918 nm was 34 % and 30 %, respectively. The laser wavelength of this crystal was continuously tuned within the spectral range of 1860 – 1935 nm. For the 4.8Tm:NLGM crystal, the slope efficiency for the π- and σ- polarisations at wavelengths of 1910 and 1918 nm was 27 % and 23 %, respectively, and the laser wavelength was tunable within the spectral range of 1870 – 1950 nm.**

Keywords: *tunable lasing, Tm^{3+} ions, laser crystals, molybdates, tungstates.*

Two-micron laser radiation is widely used in medicine and monitoring of various gases (for example, NO_2, CO_2, NH_3) in the atmosphere, as well as in scientific investigations and special engineering. Therefore the search for new active media emitting in this wavelength region is an important problem. At present, solid-state hosts and fibre waveguides doped with Tm^{3+} [1 – 3] or Ho^{3+} [4, 5] ions are widely used as sources of two-micron laser radiation. In the process of two-micron lasing in active media pumped to the $^3H_4$ level, the upper $^3F_4$ laser level of Tm^{3+} ions is populated as a result of cross-relaxation ($^3H_4 \rightarrow ^3F_4$, $^3H_6 \rightarrow ^3F_4$). This scheme of population of the $^3F_4$ level of Tm^{3+} was proposed by the authors of [6] to obtain two-micron lasing under lamp pumping. Today, this scheme is extensively used for pumping thulium lasers by commercial laser diodes with $\lambda_p \approx 800$ nm.

Of interest for developing compact diode-pumped two-micron mid-power lasers are the Tm^{3+}-doped double tungstate and molybdate crystals with the scheelite structure [7 – 19]. Due to the disordered crystal structure, the absorption and luminescence bands of intermultiplet transitions in these crystals are strongly inhomogeneously broadened. Such absorption bands of the dopant ion better overlap with the emission spectra of diode pump sources, while the wide luminescence bands offer the possibility of continuous wavelength tuning, as well as the potentiality of generating ultrashort mode-locked pulses.

The laser emission of Tm^{3+} ions in the double tungstate crystals $NaGd(WO_4)_2$ (Tm:NGW) under diode pumping was reported for the first time in [7]. A little later, the authors of [8] obtained tunable laser radiation from crystals of the same composition under pumping by a $Ti^{3+}:Al_2O_3$ laser. Two-micron lasing in the $Tm^{3+}:NaLa(WO_4)_2$ (Tm:NLW) crystals pumped by a $Ti^{3+}:Al_2O_3$ laser was obtained in [9]. The authors of [16, 17] reported lasing of Tm^{3+} ions on the $^3F_4 \rightarrow ^3H_6$ transition in $NaLa(MoO_4)_2$ (Tm:NLM) and $NaGd(MoO_4)_2$ (Tm:NGM) double molybdate crystals pumped by a $Ti^{3+}:Al_2O_3$ laser.

The results of our first experiment on lasing in the crystal of mixed sodium – lanthanum – gadolinium tungstate $Tm:NaLa_{1/2}Gd_{1/2}(WO_4)_2$ with a thulium concentration of 3.6 at % (3.6Tm:NLGW) under diode pumping were presented in [20]. The crystals of mixed scheelite-like tungstates $Tm:NaLa_xGd_{1-x}(WO_4)_2$ and molybdates $Tm:NaLa_xGd_{1-x}(MoO_4)_2$ with various La – Gd ratios take an intermediate position between sodium lanthanum and sodium gadolinium tungstates (molybdates). The formation of a solid solution between lanthanum and gadolinium in the rare-earth sublattice of the scheelite structure leads to an additional disorder. Therefore, we can expect even larger broadening and smoothing of the thulium luminescence bands. We studied the spectral and luminescent properties of the concentration series of $Tm:NaLa_xGd_{1-x}(WO_4)_2$ and $Tm:NaLa_xGd_{1-x}(MoO_4)_2$ crystals [21], where $x = 0 - 1$, and, based on these investigations, predicted the possibility of obtaining laser emission on the $^3F_4 \rightarrow ^3H_6$ transition of Tm^{3+} ions in these crystals continuously tunable in a wide wavelength range. At the same time, as far as we know, the literature provides no information on lasing of thulium in double molybdate crystals with a variable La – Gd ratio, as well as on tunable

F.A. Bol'shchikov, P.A. Ryabochkina N.P. Ogarev Mordovian State University, ul. Bol'shevistskaya 68, 430005 Saransk, Russia; e-mail: ryabochkina@freemail.mrsu.ru;
E.V. Zharikov, D.A. Lis, K.A. Subbotin A.M. Prokhorov General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38, 119991 Moscow, Russia; e-mail: evzh@mail.ru, lisdenis@mail.ru, soubbot@lsk.gpi.ru;
N.G. Zakharov, O.L. Antipov Institute of Applied Physics, Russian Academy of Sciences, ul. Ul'yanova 46, 603950 Nizhnii Novgorod, Russia; e-mail: nikitagz@rambler.ru, antipov@appl.sci-nnov.ru

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diode-pumped lasing of thulium ions in any scheelite-like molybdate or tungstate crystals.

In this paper, we present the experimental results on almost single-mode laser radiation continuously tunable in the region of 2 μm that was obtained on the $^3F_4 \rightarrow {}^3H_6$ transition of Tm^{3+} ions in the 3.6Tm : NLGW and Tm : $NaLa_{1/3}Gd_{2/3}(MoO_4)_2$ crystals with a thulium concentration of 4.8 at % (4.8Tm : NLGM) under diode pumping.

The 3.6Tm : NLGW and 4.8Tm : NLGM crystals were grown by the Czochralski method from an iridium crucible in the atmosphere of $N_2 + (1-2)$ % vol O_2 at a pulling rate of 0.7 mm h^{-1}. The grown single crystals were annealed in air for four days at a temperature of 800 °C to relieve temperature stress and, in the case of the 4.8Tm : NLGM crystal, to remove the dark colour typical for scheelite-like molybdate crystals grown in an insufficiently oxidising atmosphere [19, 21].

To a first approximation, the orientation of crystals with respect to the optical axis (the main four-fold crystallographic axis) was determined by the orientation of the single-crystal seed, which was cut perpendicular to this axis. The orientation of the grown crystals was then refined using a DRON-3 diffractometer and optical conoscopy in crossed nicols on a MIN-8 polarisation microscope. The actual concentrations of components in the grown crystals were determined by microprobe analysis on a CAMECA SX 100 instrument.

The active elements were cut from the crystals under study in the form of rods $\varnothing 3 \times 5$ mm in size so that the optical axis of the crystal was perpendicular to the rod axis. The faces of the active elements were AR coated for the pump ($\lambda_p = 795$ nm) and laser ($\lambda_{gen} = 1910$ nm) wavelengths. The residual reflection coefficients at the pump and laser wavelengths were 0.7 % and 0.2 %, respectively.

The optical scheme of a laser operating at the $^3F_4 \rightarrow {}^3H_6$ transition of Tm^{3+} ions in the 3.6Tm : NLGW and 4.8Tm : NLGM crystals is shown in Fig. 1. As a pump source in laser experiments, we used a 40-W laser diode array (*1*) kept at a temperature of ~ 26.5 °C, which corresponded to the emission wavelength of about 794 nm. To decrease the thermal load on the active element (*7*), the average pump power was decreased using an chopper (*5*), which formed pump pulses with a duration of 10 ms and a repetition rate of 5 Hz. The radiation of the laser diode array with an output fibre (*2*) (diameter 800 μm, numerical aperture 0.14) was focused inside the active element by lenses (*3*) and (*4*) with focal lengths of 5 and 3 cm, respectively. The distance between the lenses was 6 cm. Lens (*3*) was placed at a distance of 5 cm from the fibre end and, hence, served as a collimator lens. The active element was placed in the focal plane of lens (*4*).

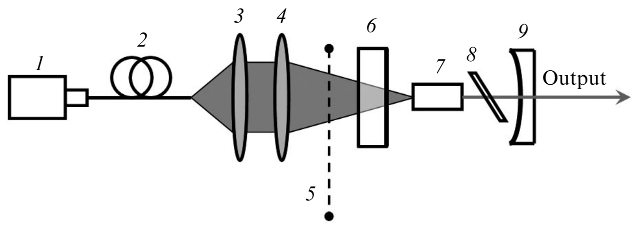

Figure 1. Optical scheme of lasers based on the 3.6Tm : NLGW and 4.8Tm : NLGM crystals.

For efficient cooling, the active element wrapped with indium foil was placed into a copper heat sink, whose temperature was kept constant at about 18 °C. In experiments, we used a 5-cm-long cavity formed by a plane dichroic mirror (*6*) [$T(\lambda_p) \approx 90$ %, $T(\lambda_{gen}) \approx 0.5$ %] and a plano-concave mirror (*9*) [$T(\lambda_{gen}) \approx 11$ %] with a 200-mm radius of curvature of the spherical face. To separate laser emission with the π- or σ-polarisation, we placed a glass plate (*8*) inside the cavity at the Brewster angle to the system axis. To obtain tunable lasing in the 3.6Tm : NLGW crystal, we used in the cavity a spectral-polarisation Wood filter instead of the glass plate. As this filter, we used a 5-mm-thick sapphire plate with a free spectral range of 80 nm. To obtain tunable lasing in the 4.8Tm : NLGM crystal, we used a 3-mm-thick quartz plate with a free spectral range of 160 nm.

The dependences of the average laser output power on the average pump power absorbed in the active element for the π- and σ-polarised laser radiation of the 3.6Tm : NLGW and 4.8Tm : NLGM crystals are shown in Fig. 2. Figure 3 shows the spectral positions of the laser lines of these crystals for both polarisations. The laser characteristics of these crystals for both polarisations are listed in Table 1. As was noted above, 2-μm lasing of diode-pumped Tm : NGW crystals was obtained in [2, 3]. The lasing slope efficiencies achieved in the present study for the 3.6Tm : NLGW crystals were 34 % for the π-polarisation and 30 % for the σ-polarisation, which exceed the corresponding values of [2, 3] for Tm : NGW crystals. The

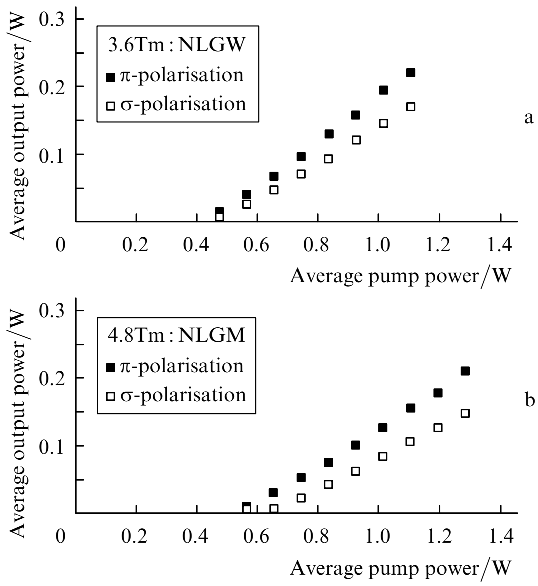

Figure 2. Dependences of the average laser output power on the average absorbed pump power for lasers based on the Tm : $NaLa_{1/2}Gd_{1/2}(WO_4)_2$ ($C_{Tm} = 3.6$ at %) (a) and Tm : $NaLa_{1/3}Gd_{2/3}(MoO_4)_2$ ($C_{Tm} = 4.8$ at %) (b) crystals.

Table 1. Laser parameters.

Crystal	Polarisation	Slope efficiency (%)	Total efficiency (%)	Laser wavelength/nm
NLGW : Tm	π	34	19	1908
	σ	30	16	1918
NLGM : Tm	π	27	15	1910
	σ	23	12	1918

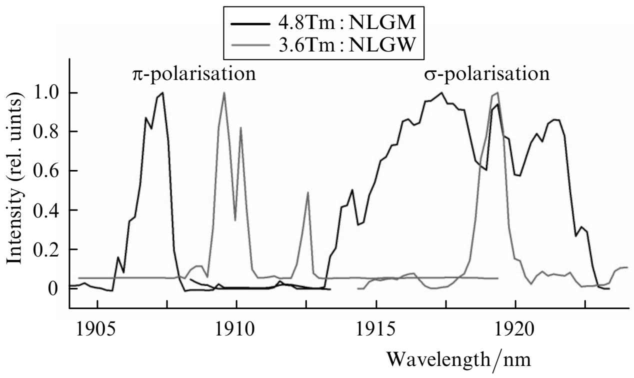

Figure 3. Spectral positions of the laser lines for the $Tm:NaLa_{1/2}Gd_{1/2}(WO_4)_2$ (C_{Tm} = 3.6 at %) (a) and $Tm:NaLa_{1/3}Gd_{2/3}(MoO_4)_2$ (C_{Tm} = 4.8 at %) (b) crystals.

available literature contains no information on diode-pumped 2-μm lasing of other scheelite-like molybdate and tungstate crystals doped with Tm ions.

Many practical applications need lasers with a continuously tunable wavelength. Recently [3, 4], 2-μm tunable lasing in Tm : NGW and Tm : NLW crystals was obtained only under pumping by a $Ti^{3+}:Al_2O_3$ laser. As far as we know, today there are no papers on tunable lasing in Tm : NGM crystals under diode pumping. In our experiments, we obtained tunable laser radiation from 3.6Tm : NLGW and 4.8Tm : NLGM crystals under diode pumping. The dependence of the average output power on the laser wavelength for the 3.6Tm : NLGW crystal at an average pump power of 1.0 W is shown in Fig. 4a. The tuning was achieved in the range of 1860 – 1935 nm with the half-width $\Delta\lambda$ of 60 and 48 nm for the π- and σ-polarisations, respectively. The wavelength dependence of the average output power for the laser based on the 4.8Tm : NLGM crystal at an average pump power of 1.5 W is given in Fig. 4b. In this case, the achieved tuning range was 1870 – 1950 nm with $\Delta\lambda$ = 58 and 46 nm for the π- and σ-polarisations, respectively. Thus, in this work, we for the first time achieved continuously tunable two-micron lasing in diode-pumped scheelite-like crystals doped with thulium ions.

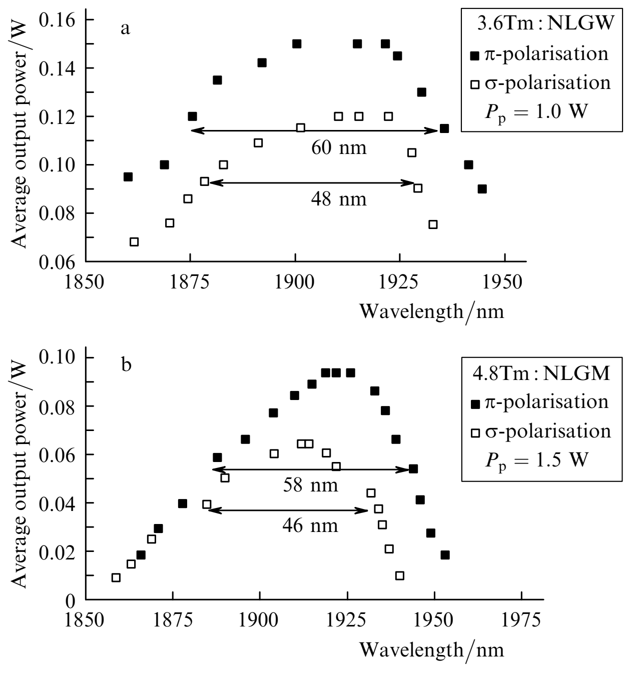

Figure 4. Wavelength dependences of the average laser output power on the average absorbed pump power for lasers based on the $Tm:NaLaGd_{1/2}(WO_4)_2$ (C_{Tm} = 3.6 at %) (a) and $Tm:NaLa_{1/3}Gd_{2/3}(MoO_4)_2$ (C_{Tm} = 4.8 at %) (b) crystals at different average pump powers.

The spatial intensity distribution of the laser beam of the 4.8Tm : NLGM crystal was studied using an IR Pyrocam III camera and is shown in Fig. 5. The correlation coefficient between the curve characterising the laser beam intensity distribution and the Gaussian distribution is 0.9, which testifies to the occurrence of almost single-mode lasing.

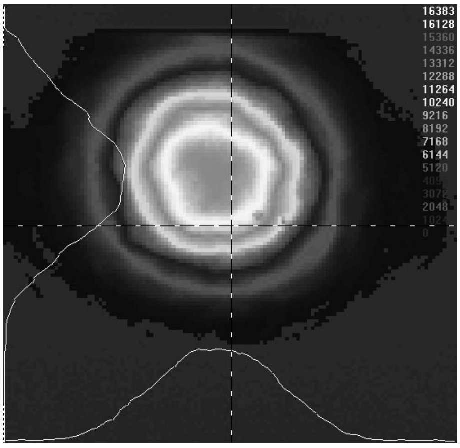

Figure 5. Transverse beam intensity distribution for a laser based on the 4.8Tm : NLGM crystal.

Thus, this paper for the first time reports the quasi-cw lasing in the mixed $Tm:NaLa_{1/3}Gd_{2/3}(MoO_4)_2$ crystals with a thulium concentration of 4.8 at % under diode pumping. The average output power of the laser based on the 4.8Tm : NLGM crystal exceeded 200 W with the slope efficiency of 27 % (π-polarisation) and 23 % (σ-polarisation). In these crystals, we obtained tunable lasing. The wavelength tuning range was 1870–1950 nm with half-widths of 58 and 46 nm for the π- and σ-polarisations, respectively. For the $Tm:NaLaGd_{1/2}(WO_4)_2$ crystals with a thulium concentration of 3.6 at %, the wavelength tuning range was 1860 – 1935 nm with half-widths of 60 nm (π-polarisation) and 48 nm (σ-polarisation).

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