

Tunable laser operation of ytterbium in disordered single crystals of $\text{Yb:NaGd(WO}_4)_2$

M. Rico, J. Liu, U. Griebner, V. Petrov

Max-Born-Institute for Nonlinear Optics and Ultrafast Spectroscopy, 2A Max-Born-Str., D-12489 Berlin, Germany
petrov@mbi-berlin.de

M. D. Serrano, F. Esteban-Betegón, C. Cascales, C. Zaldo

Instituto de Ciencia de Materiales de Madrid, Consejo Superior de Investigaciones Científicas,
Calle Sor Juana Inés de la Cruz 3, Cantoblanco, E-28049 Madrid, Spain

Abstract: Lasing of Yb^{3+} in a disordered single crystal host, $\text{NaGd(WO}_4)_2$, is reported. Pump efficiencies as high as 20% and slope efficiencies as high as 30% are achieved for both σ - and π -polarizations with Ti:sapphire laser pumping. The emission of $\text{Yb:NaGd(WO}_4)_2$ is centered near 1030 nm. Tunability between 1016 and 1049 nm is obtained with a Lyot filter.

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1. Introduction

Solid-state lasers based on rare-earth doped crystals operating in the 1 μm spectral region are nowadays widely spread as compact and reliable sources for various applications due to the possibility for highly efficient diode pumping. The potential of the trivalent ytterbium ion, Yb^{3+} , as a dopant was recognized in the very early years of diode pumping because in comparison with Nd^{3+} this ion possesses longer energy-storage lifetime and smaller quantum defect, and can be pumped by the optically more robust InGaAs laser diodes operating in the 900-1000 nm spectral range [1]. The relatively simple two-manifold structure of Yb^{3+} has the advantage that excited state absorption, up-conversion and cross-relaxation processes are absent but the operation in a quasi-three-level laser scheme poses a significant design and implementation challenge. An additional advantage of the Yb^{3+} over the Nd^{3+} ion is related to the broader absorption and emission linewidths which on the one hand reduces the requirements to the pump laser diodes and on the other hand holds a greater promise for the generation of shorter light pulses by mode-locking in the high-power regime.

The monoclinic potassium double tungstates $\text{KR}(\text{WO}_4)_2$ with $\text{R}=\text{Y, Bi, Ln}$ ($\text{Ln}=\text{Sm} \dots \text{Lu}$) [2] occupy a special position among the laser crystals due to their extremely large absorption and emission cross sections which is related also to their strong anisotropy and biaxiality [3]. In addition, the separation of adjacent R^{3+} ions in these ordered hosts is relatively large and therefore less concentration quenching occurs for increased doping levels. Highly efficient laser operation has already been demonstrated for $\text{Yb}:\text{KY}(\text{WO}_4)_2$ and $\text{Yb}:\text{KGd}(\text{WO}_4)_2$ [4], and more recently for $\text{Yb}:\text{KLu}(\text{WO}_4)_2$ [5]. Continuous wave (cw) room-temperature laser operation was possible even with the 100% Yb-doped (stoichiometric) $\text{KYb}(\text{WO}_4)_2$ [6].

Sodium double tungstates $\text{NaR}(\text{WO}_4)_2$ and molybdates $\text{NaR}(\text{MoO}_4)_2$ where $\text{R}=\text{Y, La, Bi}$ and Ln ($\text{Ln}=\text{Ce}\dots\text{Lu}$) are also related to the Ca^{2+} ion substitution in the CaWO_4 tetragonal scheelite structure, space group C_{4h}^6 ($I4_1/a$), by a pair of Na^+ and R^{3+} ions [7]. For these compounds only weak phase transformations are observed upon cooling from high temperature. As a consequence the tetragonal crystalline symmetry is preserved at room temperature although some distortions towards lower symmetry space groups may appear as observed in the Bi-containing compounds [8,9].

In the tetragonal double tungstates and molybdates an almost random distribution of Na^+ and trivalent metal R^{3+} ions over two lattice sites occurs which holds also for the active laser dopant. As a consequence, a locally variable crystal field around the dopant ion is present and the linewidths of the electronic transitions for the rare earth elements are found to be broader than in ordered crystals (see Fig. 10 in ref.[10]). Such disordered crystals with their inhomogeneous line broadening [11] have an intermediate position with respect to their optomechanical and spectroscopic properties between ordered laser hosts [12] and glasses [13]. Moreover, the uniaxial sodium double tungstates and molybdates possess an important technological advantage over their monoclinic potassium analogues. Some of them (e.g. $\text{R}=\text{La, Nd}\dots\text{Er, Bi}$) melt congruently and can be pulled in large sizes by the Czochralski method.

The major part of the literature dealing with disordered sodium double tungstates or molybdates is devoted to their growth and their structural and spectroscopic properties. Pulsed laser emission at room temperature is known only for the Nd ion in $\text{NaR}(\text{WO}_4)_2$ and $\text{NaR}(\text{MoO}_4)_2$ with $\text{R}=\text{La, Bi, Gd}$ and Y [11,14], and also in the isostructural disordered $\text{KLa}(\text{WO}_4)_2$ [15], $\text{KLa}(\text{MoO}_4)_2$ [11,14], $\text{LiLa}(\text{MoO}_4)_2$ [11,14] and $\text{LiGd}(\text{MoO}_4)_2$ [11,14]. Room temperature cw laser operation has been demonstrated, however, only in four disordered Nd-hosts of this type: $\text{NaLa}(\text{MoO}_4)_2$ [16], $\text{LiGd}(\text{MoO}_4)_2$ [17], and more recently in $\text{NaGd}(\text{WO}_4)_2$ [18] and $\text{NaY}(\text{WO}_4)_2$ [19-21].

The present work is devoted to cw laser studies of Yb^{3+} -doped disordered $\text{NaGd}(\text{WO}_4)_2$ or shortly $\text{Yb}:\text{NaGdW}$. In order to determine the optimum pump conditions for the two polarizations we used in this initial experiment a tunable Ti:sapphire laser for pumping and in order to assess the mode-locking potential of $\text{Yb}:\text{NaGdW}$ tunable operation for the σ and π polarizations using a Lyot filter was studied.

2. Crystal growth and characterization of $\text{Yb}:\text{NaGdW}$

Single crystals of $\text{NaGd}_{1-x}\text{Yb}_x(\text{WO}_4)_2$ with $x=0.1$ in the melt were grown in air by the Czochralski method using a pulling equipment supplied with a crystal diameter control system. The raw materials, Gd_2O_3 (99.99%), Yb_2O_3 (99.99%), Na_2CO_3 (99.9%) and WO_3 (99.8%) from Alfa Aesar, were mixed in the appropriate ratios and the desired phase was synthesized in a two step process: firstly the mixture was heated up to 750°C at a rate of $50^\circ\text{C}/\text{h}$, held at this temperature for 18 h and then cooled down to room temperature. The product was ground and in a second step heated to 850°C for 24 h to complete the synthesis. The phase obtained was assessed by X-ray powder diffraction (XRPD) and then heated to 25°C above the melting temperature in a 75 cc Pt crucible. The melt was kept at this temperature for one hour for homogenization and then the optimum crystallization temperature was determined by monitoring the crucible weight when seeding with a (001)-oriented $\text{NaLa}(\text{WO}_4)_2$ seed. The rotation and pulling rates were 10 rpm and 2.3 mm/h, respectively. The grown crystals were cooled to room temperature at a rate of $10^\circ\text{C}/\text{h}$. The crystals obtained were colourless. The dimensions of the pulled boule were 20 mm in diameter and 50 mm in length. Some crystal cracking appeared likely upon cooling. The refractive index was measured for this composition only at 1047 nm: $n_o=1.948$ and $n_e=1.943$ indicate that the NaGdW host is a negative crystal with relatively low birefringence.

The crystal phase obtained was further studied by XRPD and the Yb content determined from ICP emission measurements. A tetragonal crystal structure was found. The lattice parameters derived from a Rietveld refinement of the XRPD profile were $a=5.2370(2)$ Å, $c=11.3636(6)$ Å [$V=311.66(3)$ Å³]. The molar Yb content in the crystal was 8% giving a

distribution coefficient of 0.8. Hence the Yb density in the crystal is $5.1 \times 10^{20} \text{ cm}^{-3}$. The laser sample prepared was a 3.273 mm thick plate with a clear aperture of $\approx 10 \times 15 \text{ mm}^2$ (Fig. 1) allowing polarization either parallel or perpendicular to the optical axis c to be used.

The resonant transition of Yb^{3+} is actually also magnetic-dipole allowed and three spectra have to be measured for a complete description in an uniaxial crystal: α ($E \perp c$, $H \perp c$), π ($E // c$) and σ ($H // c$). In the case of $\text{Yb}:\text{NaGdW}$, however, we recorded very similar optical densities for the α and σ polarizations (Fig. 1). The maximum absorption cross sections near 974 nm calculated for the π and σ polarizations are 1.51 and $1.21 \times 10^{-20} \text{ cm}^2$, respectively; very close to the estimations in [22]. For the Brewster angle geometry used in the laser the small signal absorption amounts to 94.4% and 89.4% for the π and σ polarizations, respectively.

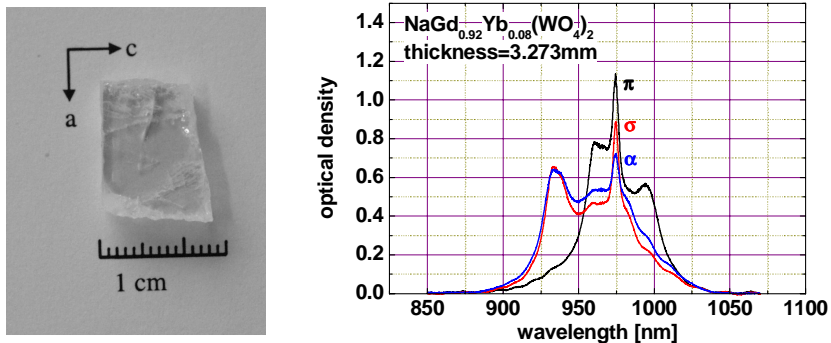


Fig. 1. The $\text{Yb}:\text{NaGdW}$ sample used in the present work (left) and the recorded optical density at room temperature (right) for the three polarizations $E // c$ (π), $H // c$ (σ), and $E \perp c$, $H \perp c$ (α).

The Yb segregation coefficient, crystal lattice parameters and optical absorption properties so far reported for our sample agree rather well with those previously reported for $\text{Yb}:\text{NaGdW}$ crystals grown in N_2/O_2 reductive atmosphere [22-25], despite the fact that the latter are supposed to contain W and O lattice vacancies and exhibit coloration in the visible and infrared spectral range. This suggests that the interaction of such lattice defects with the active Yb^{3+} ions can be neglected at least for the doping level considered here, $x < 10$ molar %. The maximum emission cross sections calculated near 996 nm amount to $\approx 2 \times 10^{-20} \text{ cm}^2$ for the π polarization and $\approx 1.5 \times 10^{-20} \text{ cm}^2$ for the σ polarization [22]. Nevertheless the reabsorption makes it difficult to predict which polarization will have better laser performance and calculations of the gain cross sections (subtracted emission and absorption cross sections assuming some inversion level) provide reliable information basically on the lasing wavelength at threshold only [22].

Measurements of fine powders in an immersion liquid in [22] provided a value of $320 \mu\text{s}$ for the fluorescence decay time independent of the Yb-doping level in the 0.5 to 5% range. This value is close to the radiative lifetime of $397 \mu\text{s}$ calculated from the absorption spectra [22], and it is also comparable to the lifetimes estimated for the monoclinic potassium double tungstates [5]. The pump saturation intensity we calculated with it is equal to 42.3 kW/cm^2 (π) and 52.7 kW/cm^2 (σ), respectively. Note that both the emission and the absorption cross sections of $\text{Yb}:\text{NaGdW}$ are almost an order of magnitude lower than the cross sections observed for the $E // N_m$ polarization in the Yb-doped monoclinic double tungstates [4-6]. That is why it is normal to expect lower efficiencies and higher laser thresholds for $\text{Yb}:\text{NaGdW}$ than those found with the ordered double tungstates.

3. Experimental results

The experimental set-up used in the present work is shown in Fig. 2. The astigmatically compensated three-mirror cavity consisted of a total reflector M_1 for the laser radiation with 50 mm radius of curvature, a folding totally reflecting mirror M_2 with 100 mm radius of

curvature and a plane output coupler M_3 whose transmission was varied between 3 and 10%. Both M_1 and M_2 were highly transmitting for the pump radiation which ensured single side pumping and helped to avoid any feedback to the pump laser. The antireflection coated pump lens L (focal length 62.8 mm) and M_2 introduced only negligible reduction of the pump power so that the incident power P_{in} on the crystal could be conveniently measured outside the resonator. The total cavity length amounted to 73 cm.

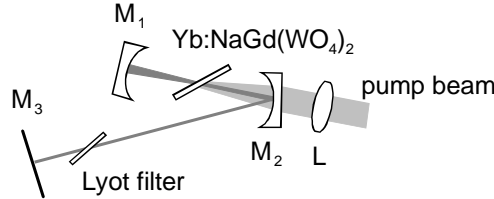


Fig. 2. Scheme of the three-mirror laser cavity of the Yb:NaGdW laser.

The pump and the laser spot diameters were almost equal in the position of the crystal and we estimated from the Brewster reflection a Gaussian pump waist of $w_{pump} \approx 22 \mu\text{m}$. Thus the pump beam size is $\pi w_p^2/2 = 7.6 \times 10^{-6} \text{ cm}^2$. The peak incident on-axis intensity for the maximum incident power of $P_{in} = 2 \text{ W}$ used in the present experiment is 260 kW/cm^2 . Averaging across the cross section by taking a factor of 0.5 means that we are approximately 3 times (π) and 2.5 times (σ) above the saturation intensity. For both polarizations, however, our measurements showed that the absorption depletion effect is partially compensated at high pump levels by the recycling effect (Fig. 3). Nonetheless, the absorbed power is always higher for the π -polarization.

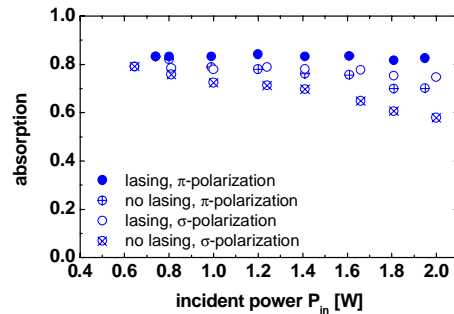


Fig. 3. Measured single pass absorption with lasing and with lasing interrupted in the arm containing the output coupler versus incident pump power P_{in} for $T_{oc} = 10\%$.

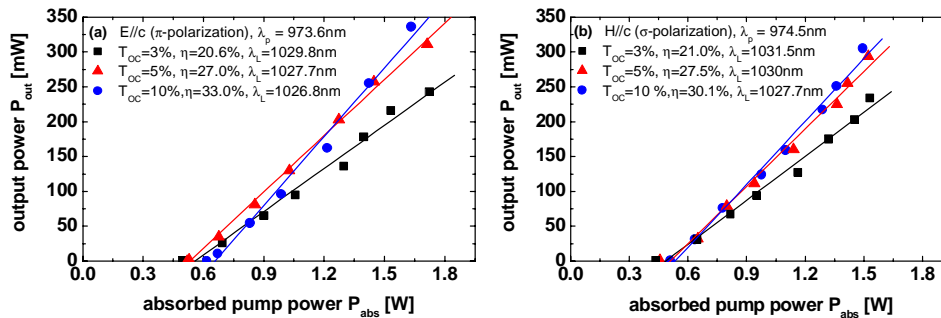


Fig. 4. Output power P_{out} versus absorbed pump power P_{abs} (symbols) of the cw Yb:NaGdW laser obtained for E//c (a) and H//c (b) at the optimum pump wavelength λ_p indicated. The linear fits give the calculated slope efficiencies η for three different output couplers (T_{oc}).

Both the thresholds (at $P_{\text{abs}} \approx 0.5$ W) and the slope efficiencies obtained with three different output couplers were essentially the same for the two polarizations (Fig. 4). In both cases the optimum pump wavelengths λ_p indicated in the figures are very close to the absorption maximum. Such a similar performance for E//c and H//c can be attributed not only to the different reabsorption effect but also to the comparable emission cross sections at the actual laser wavelengths (Fig. 4) which are far from the corresponding maxima. Thermal effects were not observed up to $P_{\text{in}}=2$ W although no special cooling was provided to the crystal.

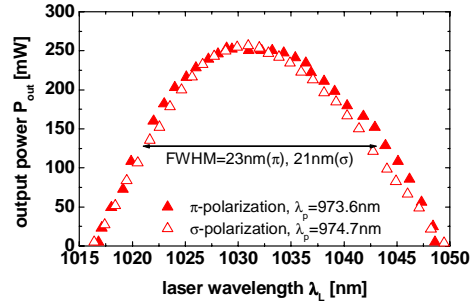


Fig. 5. Output power P_{out} versus laser wavelength λ_L for a maximum incident pump power of $P_{\text{in}}=2$ W and $T_{\text{OC}} = 5\%$ near 1030 nm. The tuning performance by a Lyot filter for both the π (filled triangles) and σ (empty triangles) polarizations is shown.

The tuning operation of the laser was investigated inserting a two-plate Lyot filter under Brewster angle in the M_2 - M_3 arm (Fig. 2) close to the output coupler. Under optimum alignment the power reduction with the filter inside the cavity did not exceed 10%. The tuning curve obtained for the π -polarization is slightly broader than for the σ -polarization (Fig. 5). Both curves seem to be slightly truncated at shorter wavelengths by the reflection characteristics of M_1 and M_2 . Tunability extending from 1016 to 1049 nm at the zero-level was obtained. The FWHM for the π -polarization corresponds to $\Delta\nu=6.5 \times 10^{12}$ Hz. For a sech²-shaped mode-locked pulse this means that sub-50-fs pulses can be expected from this laser in the mode-locking regime. This is about two times less than the shortest pulses typically achieved nowadays with femtosecond lasers based on the monoclinic double tungstates for similar T_{OC} values [26].

4. Conclusion

In conclusion cw and tunable room temperature laser operation of $\text{Yb}^{3+}:\text{NaGd}(\text{WO}_4)_2$ under Ti:sapphire laser pumping was demonstrated. The performance for the two possible polarizations was very similar achieving an output power of about 300 mW for 1.5 W of absorbed pump radiation. The optimum pump wavelength corresponds to the maximum absorption and is suitable for InGaAs laser diodes. Initial tuning experiments indicate interesting potential for mode-locking with this disordered Yb-host. We note that in comparison to the recent laser demonstration of $\text{Yb}:\text{CaF}_2$ [27] the disorder and the inhomogeneous line broadening in the sodium double tungstates is rather due to occupation with some probability of two possible sites (each with multiple environments) than to lattice defects.

The mode-locking experiments planned for the near future will reveal the actual advantages of this new Yb host. Besides this we intend to investigate the spectroscopic and laser properties for higher doping levels which could be interesting for highly absorbing thin layer laser designs.

Acknowledgments

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