Continuous-wave and mode-locked laser operation of $Yb:NaLu(WO_4)_2$

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Abstract: Highly Yb-doped tetragonal crystals of $NaLu(WO_4)_2$ with disordered structure were grown by using a $Na_2W_2O_7$ flux. Continuous-wave laser operation yielded an output power of 650 mW and 90-fs pulses were generated by passive mode-locking.

OCIS codes: (140.5680) Rare earth and transition metal solid-state lasers; (140.4050) Mode-locked lasers

1. Introduction

We recently demonstrated tunable laser operation of Yb^{3+} in tetragonal $NaT_{1-x}Yb_x(WO_4)_2$, T=La and Gd, Yb:NaTW, crystals with local disorder, see [1] for a review. The spectral bandwidths of Yb^{3+} in these hosts are determined both by inhomogeneous broadening and vibrational coupling. The NaYW, NaLaW, and NaGdW Yb-hosts melt congruently while NaYbW decomposes [2]. This sets an upper doping limit for crystals grown by the Czochralski (Cz) method, e.g. for $NaLa_{0.85}Yb_{0.15}W$ [3]. Higher doping is, however, essential for application of power scaling concepts like the thin-disk laser design, especially in cases when the absorption cross sections are modest. For double tungstates exceptionally high cross sections are found in the monoclinic ordered phases (KGdW, KYW, and KLuW) but not in the tetragonal ones. Here we report on the growth of heavily doped Yb:NaLuW crystals and laser operation of 10 mol % Yb^{3+}-doped NaLuW in the continuous-wave (CW) and mode-locked regime. Besides the additional line broadening mechanism, important for tunable and mode-locked laser operation, such tetragonal crystals with disordered structure, being uniaxial, have better opto-mechanical properties for processing of thin active elements and epitaxial structures in comparison to the monoclinic potassium double tungstates.

2. Crystal growth and structure of Yb:NaLuW

Crystals of Yb:NaLuW were grown by the top seeded solution growth (TSSG) method using a $Na_2W_2O_7$ flux. The solute/flux mixtures were melted in Pt crucibles and held for several days at ≈50ºC above the melting temperature for homogenization. As seeds, Pt wire and 001-NaGdW crystals were used. The seed rotation was 10-30 rpm and the cooling rate 0.02-1ºC/h. The crystal composition was determined by X-ray fluorescence (XRF) spectrometry. Table 1 summarizes the crystals grown, the growth conditions, and the Yb content. It is worth noting that the x=0.5 crystal was transparent and free of macrodefects. The Yb doping for the first NaLuW crystal was unintentional.

Table 1. Composition and growth conditions for $NaLu_{1-x}Yb_x(WO_4)_2$ crystals grown by TSSG using $Na_2W_2O_7$ flux.

x_{MELT}	solute/flux molar ratio	cooling interval [ºC]	cooling rate [ºC/h]	$x_{CRYSTAL}$	$[Yb]_{CRYSTAL}$ $[10^{21}\ cm^{-3}]$
≈0	1:4	877-845	0.03	0.0056	0.038
0.1	1:6	834-824	0.02	0.111	0.746
0.5	1:3.5	917-903	0.04	0.508	3.40

Single crystal X-ray diffraction (XRD) was used for crystallographic characterization and independent evaluation of the composition. It was established that TSSG grown Yb:NaLuW crystals have the same space group (SG) $I\bar{4}$ as Cz-grown NaTW, T=Y, La, Gd, i.e. Yb can occupy two non-equivalent crystallographic sites, 2*b* and 2*d* [4]. However, the occupancy factors (OF) of Na and T ions in these sites are characteristic for the specific host. This is supported by the presence of non negligible systematic XRD absence exceptions with respect to the SG $I4_1/a$. Furthermore, crystals grown by the TSSG method have a composition very close to the nominal while crystals grown by the Cz method are Na deficient which is compensated by excessive incorporation of T cations.

3. Spectroscopy of Yb:NaLuW

Absorption and photoluminescence spectroscopy at 5 K provided further evidence of the coexistence of several Yb^{3+} optical centers in NaLuW. These spectra were used to derive the Stark levels of an average Yb^{3+} center. The 300 K emission cross sections were then calculated by the reciprocity method using the measured absorption cross sections (Fig.1). The peak values obtained, $\sigma_a(\sigma)=1.65\times10^{-20}$ cm^2 and $\sigma_a(\pi)=2.22\times10^{-20}$ cm^2 at 973.8 nm, as well as $\sigma_e(\sigma)=1.4\times10^{-20}$ cm^2 and $\sigma_e(\pi)=2.1\times10^{-20}$ cm^2 at 1000 nm are significantly larger than in NaLaW or NaGdW [1].

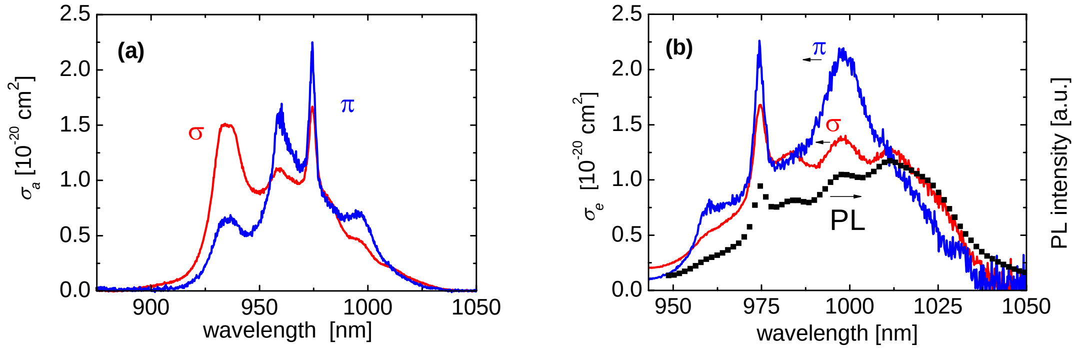

Fig.1: Measured absorption (a) and computed emission (b) cross sections of Yb:NaLuW. The recorded σ-photoluminescence (PL) is shown for comparison in (b).

Measurements of the upper laser level lifetime by the pinhole method gave (353±6) μs for the 10 mol % Yb^{3+} doped NaLuW sample.

4. CW laser operation of Yb:NaLuW

The initial CW laser experiments were performed with a standard astigmatically compensated Z-shaped four mirror cavity containing two folding mirrors with RC=-10 cm. The cavity length was ~140 cm. The 10 mol % Yb^{3+} doped NaLuW sample available for this experiment was 0.94 mm thick, uncoated, and its cut allowed pumping and lasing for σ-polarization. It was placed under Brewster angle between the two folding mirrors and no cooling was applied. The results, obtained by pumping in a single pass with a Ti:sapphire laser at 974 nm are summarized in Fig.2a. Up to the maximum pump power applied, which corresponded to an absorbed power of 1 W, the output power was linearly proportional to the absorbed power, i.e. no thermal effects were observed. The maximum output power of 471 mW was obtained with a T_{OC}=3% output coupler for an absorbed pump power of P_{abs}=932 mW. The maximum slope efficiency with respect to P_{abs} was obtained for T_{OC}=5%, η=68.2%. The oscillation wavelength λ_L slightly decreased with T_{OC}. Under lasing conditions, the actual crystal absorption varied between 53% and 60% for the three output couplers (decreasing with T_{OC}) but it was almost independent of the incident pump power. However, without lasing, the absorption dropped to 32% for the maximum incident power applied as a consequence of the bleaching.

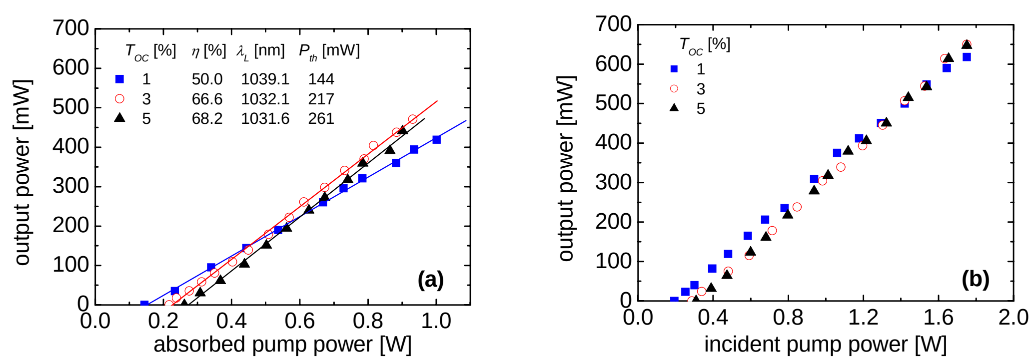

Fig.2: Characteristics of the CW Yb:NaLuW laser for σ-polarization in the case of single pass pumping (a) and when recycling the residual pump by retroreflecting it (b). The slope efficiency η, the oscillation wavelength λ_L, and the threshold absorbed power P_{th} are shown for different T_{OC}.

Using broadband cavity mirrors, it was possible to pump the sample in a second pass with the residual pump light. In this case 650 mW were obtained both with T_{OC}=3% and 5% (Fig.2b). However, it was not possible to estimate the slope efficiency with respect to the absorbed pump power because the latter could not be reliably measured.

5. Mode-locking of the Yb:NaLuW laser

The mode-locking experiments were performed with a similar 10 mol % Yb^{3+} doped NaLuW sample which was 1.16 mm thick and uncoated. Its orientation allowed the use of π-polarization. The incident pump power measured in front of the Yb:NaLuW crystal was 1.45 W. The cavity was similar and included two SF10 prisms with a tip-to-tip separation of 32 cm in the arm containing the output coupler and an additional RC=-10 cm mirror in the other arm to increase the fluence on the semiconductor saturable absorber mirror (SESAM) which terminated the resonator. The total cavity length corresponded to a repetition frequency of 95 MHz. The SESAM used for mode-locking was grown by metalorganic vapor phase epitaxy (MOVPE). It was reflecting from 1000 to 1080 nm, the saturable absorption (a 10 nm thick single InGaAs quantum well implanted with As-ions and embedded in a GaAs layer) amounted to ~1%, and the relaxation time was 5 ps.

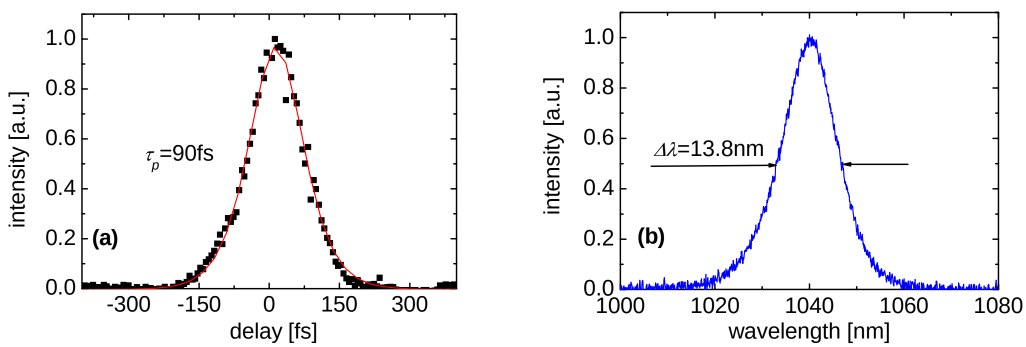

Fig.3: Autocorrelation trace with the corresponding fit assuming $sech^2$-pulse shape (a) and spectrum (b) of the femtosecond Yb:NaLuW laser with T_{OC}=1%.

Using an output coupler with T_{OC}=1% and depending on the alignment it was possible to obtain pulses as short as 90 fs (FWHM assuming a $sech^2$-pulse shape) at an average output power of 50 mW or longer pulses (170 fs) for an average power of 130 mW. The intensity autocorrelation trace with the corresponding fit and the spectrum of the shortest pulses centered at 1040 nm are shown in Fig. 3. The resulting time-bandwidth product is 0.344, which is only slightly above the Fourier limit for a $sech^2$-pulse (0.315). No tendencies for Q-switching instabilities were observed.

6. Conclusion

In conclusion, we demonstrated that high Yb-doping is possible in tetragonal crystals of NaLuW grown by the TSSG technique which possess disordered structure and additional inhomogeneous broadening of the spectral lines that makes them attractive for tunable and mode-locked laser operation in the 1 µm spectral range. Output powers as high as 650 mW were achieved in the CW regime with Ti:sapphire laser pumping and first realization of mode-locking using a SESAM yielded bandwidth-limited pulses as short as 90 fs.

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