



Growth and continuous-wave laser operation of disordered crystals of $\text{Yb}^{3+}:\text{NaLa}(\text{WO}_4)_2$ and $\text{Yb}^{3+}:\text{NaLa}(\text{MoO}_4)_2$

J. Liu¹, J. M. Cano-Torres², C. Cascales², F. Esteban-Betegón², M. D. Serrano², V. Volkov², C. Zaldo², M. Rico¹, U. Griebner¹, and V. Petrov^{*,1}

¹ Max-Born-Institute for Nonlinear Optics and Ultrafast Spectroscopy, 2A Max-Born-Str., 12489 Berlin, Germany

² Instituto de Ciencia de Materiales de Madrid, CSIC, Calle Sor Juana Inés de la Cruz 3, Cantoblanco, 28049 Madrid, Spain

Received 10 January 2005, revised 30 January 2005, accepted 31 January 2005

Published online 1 February 2005

PACS 42.55.Rz, 42.70.Hj, 81.10.Fq

* Corresponding author: e-mail petrov@mbi-berlin.de, Phone: +49 30 6392 1272, Fax: +49 30 6392 1289

Single crystals of disordered $\text{NaLa}(\text{WO}_4)_2$ and $\text{NaLa}(\text{MoO}_4)_2$ doped with Yb^{3+} are grown by the Czochralski method from the melt. Continuous-wave laser operation with Ti:sapphire laser pumping is demonstrated at room temperature without

special cooling. Tunability from 1017 to 1057 nm and from 1015 to 1053 nm is achieved for $\text{Yb}:\text{NaLa}(\text{WO}_4)_2$ and $\text{Yb}:\text{NaLa}(\text{MoO}_4)_2$, respectively. A maximum output power of 205 mW is obtained with $\text{Yb}:\text{NaLa}(\text{WO}_4)_2$.

© 2005 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

Double tungstate and molybdate compounds with the general formulae $\text{MT}(\text{WO}_4)_2$ and $\text{MT}(\text{MoO}_4)_2$ where M is a monovalent alkali cation (Li–Cs) and T is a trivalent cation (Y, La or rare earth Ln) exhibit ordered phases with separate sites for M and T cations and disordered phases where M and T cations are randomly distributed over the same cationic sublattice [1]. Some of the optically passive ordered phases like the monoclinic $\text{KGd}(\text{WO}_4)_2$, $\text{KY}(\text{WO}_4)_2$, and $\text{KLu}(\text{WO}_4)_2$ are established laser hosts with very large absorption and emission cross sections of the active dopant. The sodium compounds $\text{NaT}(\text{WO}_4)_2$ and $\text{NaT}(\text{MoO}_4)_2$ represent disordered phases with tetragonal structure at room temperature. For T = Y, La, Ce–Er, they exhibit also a congruent melting character. Hence their growth with active Ln-dopants by the Czochralski method is very attractive for the synthesis of novel crystalline laser materials.

The revived interest in tetragonal sodium double tungstates and molybdates is due to their potential to ensure larger tunability and bandwidths in mode-locked diode-pumped solid-state lasers in comparison to ordered crystals. This is especially true for doping with Yb^{3+} which, due to the stronger electron–phonon coupling to the lattice, exhibits intrinsically broader linewidths than the Nd^{3+} ion. Hence, the requirements to the pump laser diodes are reduced and Yb-doped disordered hosts hold a greater promise for the generation of mode-locked pulses shorter than 100 fs. It

should be added that Yb^{3+} possesses longer energy-storage lifetime and smaller quantum defect than Nd^{3+} , and that it can be pumped by the optically more robust InGaAs laser diodes operating in the 900–1000 nm spectral range [2]. Finally, the relatively simple two-manifold structure of Yb^{3+} prohibits excited state absorption, up-conversion and cross-relaxation processes.

The sodium lanthanum crystals $\text{NaLa}(\text{WO}_4)_2$ (NaLaW) and $\text{NaLa}(\text{MoO}_4)_2$ (NaLaMo) were studied in the past as room temperature hosts only for Nd^{3+} lasers. Lamp-pumped pulsed laser operation of Nd:NaLaW was reported for the $4\text{F}_{3/2}-4\text{I}_{11/2}$ transition at 1063.5 nm [3, 4] and for the $4\text{F}_{3/2}-4\text{I}_{13/2}$ transition at 1335.5 nm [5]. Lamp-pumped operation of Nd:NaLaMo was demonstrated not only in the pulsed regime at 1059.5–1065.3 nm [6, 7] and 1338–1344 nm [5] but also in the continuous-wave (cw) regime at 1065.3 nm [8]. In addition NaLaMo, which is an efficient Raman active medium [9], when doped with Nd^{3+} , was shown to be an efficient self-converting Raman crystal both in the picosecond [10] and in the nanosecond [11] regime.

Here we report on the cw laser performance of $\text{Yb}:\text{NaLaW}$ and $\text{Yb}:\text{NaLaMo}$ crystals at room temperature. Note that the isostructural $\text{NaGd}(\text{WO}_4)_2$ or NaGdW , was the first and so far only disordered laser crystal of this type for which room-temperature cw laser operation could be demonstrated with Yb^{3+} doping [12].

© 2005 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

Yb-doped NaLaW crystals were grown for the first time in nitrogen atmosphere [13] but only unpolarized spectra were reported in this initial work. The growth of Yb:NaLaMo, also in nitrogen atmosphere, has been reported only very recently [14]. The Yb:NaLaW [15] and Yb:NaLaMo crystals for the present work were grown in air by the Czochralski method. In both cases the melts were held in Pt crucibles. Typical growth conditions were about 10 rpm for the rotation rate and 1 mm/h for the pulling rate. The crystallization temperature decreased with increasing Yb concentration. We succeeded in growing Yb:NaLaW crystals with sufficient optical quality for Yb concentrations as high as 15 mol% in the melt. Deterioration of the crystal quality for higher Yb content and the inability to grow crystals with more than 40 mol% Yb is associated with the incongruent melting character of $\text{NaYb(WO}_4)_2$. Yb:NaLaMo crystals were grown for 2–10 mol% Yb content in the melt. The fluorescence lifetime of Yb:NaLaW is 220 μs [15] and the fluorescence lifetime of Yb:NaLaMo amounts to 280 μs [14]. For comparison, the lifetime of Yb:NaGdW is 320 μs [14].

The Yb:NaLaW sample used was grown with 5 mol% Yb in the melt but the amount incorporated into the crystal was $1.1 \times 10^{20} \text{ cm}^{-3}$ (segregation coefficient $K \approx 0.6$). The tetragonal and hence disordered structure was confirmed by X-ray diffraction: the lattice cell parameters were $a = 5.3519(3) \text{ \AA}$ and $c = 11.640(1) \text{ \AA}$. The segregation coefficient for Yb:NaLaMo was 0.3–0.4. The sample used here was grown with 8 mol% Yb in the melt and the density in the crystal was $1.3 \times 10^{20} \text{ cm}^{-3}$. Its tetragonal structure ($a = 5.3387(4) \text{ \AA}$ and $c = 11.729(2) \text{ \AA}$) and disordered character were verified, too. In both hosts the main Yb^{3+} absorption line near 977 nm is stronger for the π -polarization. Figure 1 shows the optical densities for $E \parallel c$ (π) and $H \parallel c$ (σ). For the Yb:NaLaW sample the c -axis was in the surface plane and it was out of plane for the Yb:NaLaMo sample, as can be seen from the presence of a weak shoulder at 935 nm for the π -polarization spectrum in Fig. 1. The maximum small signal absorption at Brewster angle was $\alpha_0 = 50\%$ and 52% for the 3.4 mm thick Yb:NaLaW and the 2.5 mm thick Yb:NaLaMo sample, respectively.

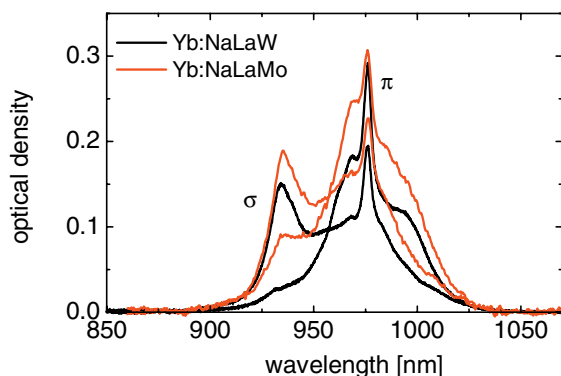


Figure 1 (online colour at: www.pss-rapid.com) Optical density measured at normal incidence for the Yb:NaLaW and Yb:NaLaMo samples used.

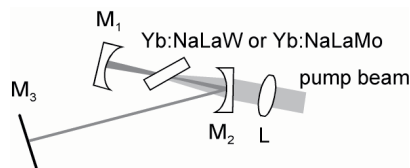


Figure 2 Schematic of the laser set-up.

The three-mirror astigmatically compensated laser cavity, see Fig. 2, consisted of a $\text{RC} = -5 \text{ cm}$ end mirror M_1 , a $\text{RC} = -10 \text{ cm}$ folding mirror M_2 through which the Ti:sapphire laser pump beam was focused by an $f = 6.28 \text{ cm}$ lens L , and a plane output coupler M_3 of transmission T_{oc} . The total cavity length was 65 cm. The pump power incident on the crystals was limited to 1.9 W. The crystal samples were uncooled. The pump beam was focused to a spot of about 22 μm (Gaussian waist).

The optimum pump wavelength (976.6 nm) corresponded both for Yb:NaLaW and Yb:NaLaMo to the main absorption peak. At the maximum incident pump power (1.9 W) the absorption of the Yb:NaLaW crystal was bleached down to $\approx 20\%$ and that of Yb:NaLaMo down to $\approx 24\%$. Lasing had a recycling effect and, depending on the output coupler used, the absorption increased (see Table 1).

The maximum output power (205 mW) was obtained for Yb:NaLaW with $T_{\text{oc}} = 1.1\%$ (Fig. 3). Using output couplers with $T_{\text{oc}} = 3\%$ and 5.4% , the intracavity power decreased and the recycling effect was suppressed (see the α values in Table 1). Hence, these output couplers did not yield higher output powers although the slope efficiency slightly increased. In the case of Yb:NaLaMo P_{max} was almost the same for the three output couplers used. The same holds for the maximum pump efficiency η_0 for both crystals. In general Yb:NaLaW showed better laser performance in terms of threshold, efficiency and output power than the Yb:NaLaMo crystal although the small signal absorption was quite similar. At present, the reasons for this can be attributed only to the crystal quality.

Table 1 Parameters of the Yb:NaLaW and Yb:NaLaMo lasers achieved. P_{th} : absorbed pump power at threshold, P_{max} : maximum output power, η : slope efficiency with respect to the absorbed power, η_0 : maximum pump efficiency with respect to the absorbed pump power, λ_{t} : generation wavelength, α : crystal absorption for maximum incident pump power (1.9 W) in the lasing state. Note that T_{oc} is specified near 1030 nm. At 1025 nm the transmissions (1.2, 3.2 and 5.7%, respectively) are slightly higher.

sample	Yb:NaLaW			Yb:NaLaMo		
	T_{oc} (%)	1.1	3	5.4	1.1	3
P_{th} (mW)	100	110	200	150	160	240
P_{max} (mW)	205	181	132	113	120	109
η (%)	30.5	34.0	35.3	17.9	22.8	30.0
η_0 (%)	26.9	28.9	25.8	14.4	17.7	17.5
λ_{t} (nm)	1034	1029	1027	1030	1025	1023
α (%)	39.4	32.3	26.4	41.5	36.0	33.0

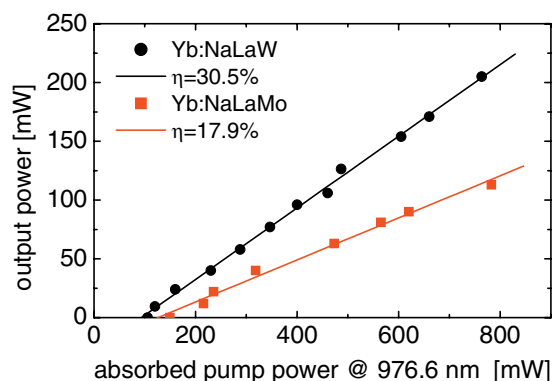


Figure 3 (online colour at: www.pss-rapid.com) Output power versus absorbed power of the Yb:NaLaW and Yb:NaLaMo lasers for E_{llc} and $T_{oc} = 1.1\%$.

The spectroscopic data available at present for the different Yb-doped sodium tungstates does not allow to attribute the differences observed in the laser performance to the emission or gain cross sections, or to different lifetimes because of insufficient accuracy and comparability of the data. The most essential difference that we observe in comparison to Yb:NaGdW [12] is the stronger bleaching of the absorption for lower small-signal absorption levels as in the present work. This results in lower thresholds and increased efficiencies with respect to the absorbed pump power but for the same incident pump power the absolute output power that can be achieved with higher small-signal absorption values can be about 50% higher [12].

The insertion of a two-plate Lyot filter did not affect the maximum output power but allowed tuning between 1017 and 1057 nm for Yb:NaLaW and 1015 and 1053 nm for Yb:NaLaMo (Fig. 4). Note that the reduction of the recycling effect additionally confines the achievable tunability in the cw regime while in a mode-locked laser this effect will be absent. Nevertheless we obtained slightly broader (FWHM = 25 nm) tunability both for Yb:NaLaW and Yb:NaLaMo in comparison to Yb:NaGdW [12].

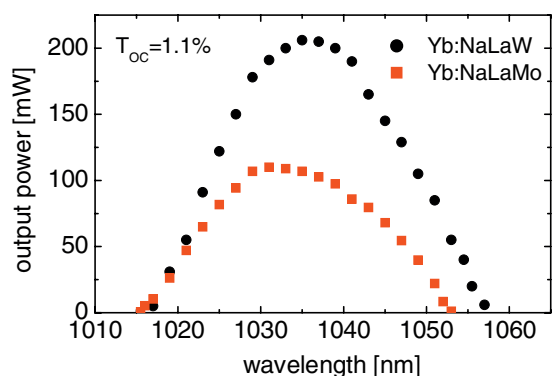


Figure 4 (online colour at: www.pss-rapid.com) Wavelength tunability of the Yb:NaLaW and Yb:NaLaMo lasers for an incident pump power of 1.9 W.

Yb:NaLaMo tends to generate at slightly shorter wavelengths in comparison to Yb:NaLaW: about 4 nm for the corresponding maxima in Fig. 4 or according to Table 1.

In conclusion, disordered crystals of NaLaW and NaLaMo doped with Yb were grown by the Czochralski method and laser operation demonstrated for the first time. The power levels and tunability achieved at room temperature in the cw regime are promising for the realization of mode-locked operation in the sub-100 fs regime.

Acknowledgements We acknowledge financial support through Projects MAT2002-4605-C05-05 and CAM-MAT-434-2004 (Spain) and DT-CRYS, NMP3-CT-2003-505580 (EU).

References

- [1] V. K. Trunov, V. A. Efremov, and J. A. Velikodnyj, *Kristallogichimija i svojstva dvojnyh molibdatov i vol'framatov*, (Nauka, Leningrad, 1986), in Russian.
- [2] W. F. Krupke, Ytterbium solid-state lasers – the first decade, *IEEE J. Sel. Top. Quantum Electron.* **6**, 1287 (2000).
- [3] N. S. Belokrinitskii, H. D. Belousov, V. I. Bonchkovskii, V. A. Kobzar-Zlenko, B. S. Skorobogatov, and M. S. Soskin, *Ukr. Fiz. Zh.* **14**, 1400 (1969), in Russian.
- [4] N. D. Belousov, V. A. Kobzar-Zlenko, and B. S. Skorobogatov, *Opt. Spectrosc.* **33**, 550 (1972) [transl. from *Opt. Spektrosk.* **33**, 1002 (1972)].
- [5] A. A. Kaminskii and S. E. Sarkisov, *Sov. J. Quantum Electron.* **3**, 248 (1973) [transl. from *Kvantovaya Elektron. (Moscow)* No. 3(15), 106 (1973)].
- [6] A. M. Morozov, M. N. Tolstoi, P. P. Feofilov, and V. N. Shapovalov, *Opt. Spectrosc.* **22**, 224 (1967) [transl. from *Opt. Spektrosk.* **22**, 414 (1967)].
- [7] G. M. Zverev and G. Ya. Kolodny, *Sov. Phys. JETP* **25**, 217 (1967) [transl. from *Zh. Eksp. Teor. Fiz.* **52**, 337 (1967)].
- [8] A. A. Kaminskii, G. Ya. Kolodnyi, and N. I. Sergeeva, *J. Appl. Spectr. (USSR)* **9**, 1275 (1968) [transl. from *Zh. Prikl. Spektrosk.* **9**, 884 (1968)].
- [9] A. A. Kaminskii, S. N. Bagaev, D. Grebe, H. J. Eichler, A. A. Pavlyuk, and R. Macdonald, *Quantum Electron.* **26**, 193 (1996) [transl. from *Kvantovaya Elektron. (Moscow)* **23**, 199 (1996)].
- [10] J. Viscakas and V. Syrusas, *Sov. Phys. – Collection* **27**, 31 (1987) [transl. from *Litovskii Fiz. Sbornik* **27**, 547 (1987)].
- [11] A. V. Gulin, V. A. Pashkov, and N. S. Ustimenko, *Proc. SPIE* **4350**, 36 (2001).
- [12] M. Rico, J. Liu, U. Griebner, V. Petrov, M. D. Serrano, F. Esteban-Betegon, C. Cascales, and C. Zaldo, *Opt. Express* **12**, 5362 (2004).
- [13] K. A. Subbotin, E. V. Zharikov, and V. A. Smirnov, *Opt. Spectrosc.* **92**, 601 (2002) [transl. from *Opt. Spektrosk.* **92**, 657 (2002)].
- [14] Yu. K. Voron'ko, E. V. Zharikov, D. A. Lis, A. A. Sobol, K. A. Subbotin, S. N. Ushakov, and V. E. Shukshin, *SPIE* **5478**, 60 (2004).
- [15] M. D. Serrano, F. Esteban-Betegon, and C. Zaldo, *J. Cryst. Growth* **274** (2005) in press.