Passive *Q*-switching of the diode pumped Tm³⁺:KLu(WO₄)₂ laser near 2-µm with Cr²⁺:ZnS saturable absorbers

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Abstract: Single pulse energies as high as 145 μ J were generated with a passively *Q*-switched diode-pumped Tm:KLu(WO₄)₂ laser using polycrystalline Cr²⁺:ZnS as a saturable absorber. The maximum average power reached 0.39 W at a pulse repetition rate of 2.7 kHz with pulse durations in the 25 – 30 ns range. The maximum peak power amounted to 6 kW. The obtained results agree well with theoretical analysis.

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OCIS codes: (140.3540) Lasers, Q-switched; (140.3070) Infrared and far-infrared lasers; (140.3480) Lasers, diode-pumped.

References and links

- 1. A. Godard, "Infrared (2–12 µm) solid-state laser sources: a review," C. R. Phys. 8(10), 1100–1128 (2007).
- M. Mond, E. Heumann, G. Huber, S. Kuck, V. I. Levchenko, V. N. Yakimovich, V. E. Shcherbitsky, V. E. Kisel, and N. V. Kuleshov, "Passive *Q*-switching of a diode-pumped Tm:YAG laser by Cr²⁺:ZnSe," Conference on Lasers and Electro-Optics Europe, CLEO/Europe (2003), paper CA7–5-WED, CD ROM.
- L. E. Batay, A. N. Kuzmin, A. S. Grabtchikov, V. A. Lisinetskii, V. A. Orlovich, A. A. Demidovich, A. N. Titov, V. V. Badikov, S. G. Sheina, V. L. Panyutin, M. Mond, and S. Kück, "Efficient diode-pumped passively *Q*switched laser operation around 1.9 μm and self-frequency Raman conversion of Tm-doped KY(WO₄)₂," Appl. Phys. Lett. **81**(16), 2926–2928 (2002).
- M. S. Gaponenko, I. A. Denisov, V. E. Kisel, A. M. Malyarevich, A. A. Zhilin, A. A. Onushchenko, N. V. Kuleshov, and K. V. Yumashev, "Diode-pumped Tm: KY(WO₄)₂ laser passively *Q*-switched with PbS-doped glass," Appl. Phys. B **93**(4), 787–791 (2008).
- B. Yao, Y. Tian, G. Li, and Y. Wang, "InGaAs/GaAs saturable absorber for diode-pumped passively *Q*-switched dual-wavelength Tm:YAP lasers," Opt. Express 18(13), 13574–13579 (2010).
- 6. W. T. Silvast, Laser Fundamentals (Cambridge University Press 1996).
- S. A. Payne, L. L. Chase, L. K. Smith, W. L. Kway, and W. F. Krupke, "Infrared cross-section measurements for crystals doped with Er³⁺, Tm³⁺, and Ho³⁺," IEEE J. Quantum Electron. 28(11), 2619–2630 (1992).
- M. C. Pujol, X. Mateos, A. Aznar, X. Solans, S. Suriñach, J. Massons, F. Díaz, and M. Aguiló, "Structural redetermination, thermal expansion and refractive indices of KLu(WO₄)₂," J. Appl. Cryst. **39**(2), 230–236 (2006).
- V. Petrov, M. Cinta Pujol, X. Mateos, Ò. Silvestre, S. Rivier, M. Aguiló, R. M. Solé, J. Liu, U. Griebner, and F. Díaz, "Growth and properties of KLu(WO₄)₂, and novel ytterbium and thulium lasers based on this monoclinic crystalline host," Laser Photonics Rev. 1(2), 179–212 (2007).
- X. Mateos, V. Petrov, J. Liu, M. C. Pujol, U. Griebner, M. Aguiló, F. Díaz, M. Galan, and G. Viera, "Efficient 2μm continuous wave laser oscillation of Tm³⁺:KLu(WO₄)₂," IEEE J. Quantum Electron. 42, 1008–1015 (2006).
- O. Silvestre, J. Grau, M. C. Pujol, J. Massons, M. Aguiló, F. Díaz, M. T. Borowiec, A. Szewczyk, M. U. Gutowska, M. Massot, A. Salazar, and V. Petrov, "Thermal properties of monoclinic KLu(WO4)2 as a promising solid state laser host," Opt. Express 16(7), 5022–5034 (2008).
- S. Vatnik, M. C. Pujol, J. J. Carvajal, X. Mateos, M. Aguiló, F. Díaz, and V. Petrov, "Thermo-optic coefficients of monoclinic KLu(WO₄)₂," Appl. Phys. B 95(4), 653–656 (2009).
- R. Sole, V. Nikolov, and X. Ruiz, "Jna. Gavalda, X. Solans, M. Aguilo, and F. Diaz, "Growth of β-KGd_{1-x}Nd_x(WO₄)₂ single crystals in K₂W₂O₇ solvents," J. Cryst. Growth 169, 600–603 (1996).

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 M. Segura, X. Mateos, M. C. Pujol, J. J. Carvajal, M. Aguiló, F. Díaz, V. Panyutin, U. Griebner, and V. Petrov, "Diode-pumped passively *Q*-switched Tm:KLuW laser with a Cr²⁺:ZnSe saturable absorber," Advanced Solid State Photonics, ASSP 2010, paper ATuA6, CD ROM.

1. Introduction

The eye-safe laser emission region around 2 μ m covered by Tm³⁺, Ho³⁺ and codoped (Tm³⁺-Ho³⁺) active media is important for medical applications, mainly due to the strong optical absorption by water, and remote sensing (LIDAR) of CO₂ and water in the atmosphere, as well as for pumping Optical Parametric Oscillators (OPO's) for conversion into the mid-IR [1]. The Tm ion, emitting on the ${}^{3}F_{4} \rightarrow {}^{3}H_{6}$ transition, is attractive because its absorption band around 800 nm matches the emission of AlGaAs laser diodes designed for Nd³⁺-ion pumping. Passive *Q*-switching (PQS) of such diode-pumped solid state lasers (DPSSL) by a saturable absorber (SA) is a common technique to generate short and high peak power pulses, mainly due to the simplicity and low cost of the cavity design. It has been applied to several Tm-doped laser materials such as YAG [2], KY(WO₄)₂ [3,4], and YAP [5] using Cr²⁺:ZnS end Cr²⁺:ZnS crystals, PbS quantum dots, and InGaAs/GaAs semiconductor based SAs.

Concerning the monoclinic double tungstates, PQS around 1.9 μ m with Cr²⁺:ZnS and Cr²⁺:ZnSe SAs was demonstrated using Tm³⁺:KY(WO₄)₂ and codoped Yb³⁺,Tm³⁺:KY(WO₄)₂ [3]. The best result of 116 mW output power at a repetition rate of 20 kHz with Yb³⁺,Tm³⁺: KY(WO₄)₂ and Cr²⁺:ZnS corresponds to a single pulse energy of 6.7 μ J and a pulse duration of 63 ns. Note that such high repetition rates do not allow one to fully utilize the long storage time of Tm which intrinsically limits the pulse energy. More recently, PQS of Tm³⁺: KY(WO₄)₂ has been achieved also using PbS-doped glass as SA [4]. In this set-up, up to 44 μ J of single pulse energy was produced at a repetition rate of 2.5 kHz but for the pulse duration only an upper detection limit of 60 ns was given.

Concerning other Tm^{3+} -doped crystals, semiconductor based SAs have been used for PQS of a Tm^{3+} :YAP laser in [5] achieving a maximum pulse energy of 28.1 µJ at a repetition rate of 43.7 kHz, however, the pulse duration, 447 ns, was rather long. The highest pulse energy (~400 µJ) from a diode-pumped PQS laser, to the best of our knowledge, has been achieved in Tm^{3+} :YAG using Cr²⁺:ZnSe as SA [2]: However, the pulse duration in this laser was also untypically long for PQS (about 300 ns), resulting in a peak power of about 1 kW.

A good choice of SA for PQS must fulfil the relation $I_{SA}\sigma_{SA}>I_g\sigma_0$ [6] where I_{SA} is the laser intensity in the SA, σ_{SA} is the absorption cross-section of the SA at the laser wavelength, I_g is the intensity of the laser beam in the gain medium and σ_0 is the stimulated emission crosssection of the gain medium. Also the SA must have a short upper level lifetime compared to that of the gain medium. Long gain upper lifetimes ensure high energy storage and consequently high energy output pulses. For instance, the upper lifetime of Tm:YAG (~10 ms [7]) is few times longer than in monoclinic double tungstates (see Table 1).

In this paper, we report on PQS of a DPSSL based on the monoclinic potassium lutetium tungstate KLu(WO₄)₂, doped with 3 at.% Tm³⁺, hereafter Tm:KLuW. In these biaxial crystals, three principal optical axes exist associated with the three refractive indices, $n_p < n_m < n_g$. The N_p principal optical axis is parallel to the *b* crystallographic axis. The other two axes of the optical ellipsoid, N_m and N_g , lie in the *a-c* crystallographic plane and the location of N_g with respect to the *c* crystallographic axis is at 18.5° in the clockwise direction when *b* is pointing towards the observer [8]. The relevant properties of Tm:KLuW are summarized in Table 1, see [8–12]. Among them are the high absorption and emission cross sections for the pump and laser radiation for the selected polarization along N_m and the possibility of high doping level.

^{15.} W. Koechner, Solid State Laser Engineering (Springer, 2006).

Doping level (solution)	3 at.% Tm
Ion concentration	2×10^{20} at/cm ³
Pump wavelength, λ_p [10]	802 nm, <i>E</i> // <i>N</i> _m
Absorption cross section, σ_a [10]	$5.95 \times 10^{-20} \text{ cm}^2 \text{ for } E //N_{\text{m}}$
Absorption linewidth [10]	4 nm, <i>E</i> // <i>N</i> _m
Stimulated emission cross section, σ_0 (1920 nm) [10]	$1.38 \times 10^{-20} \mathrm{cm}^2$
${}^{3}F_{4}$ fluorescence lifetime, τ_{a} [10]	1.34 ms
Saturation intensity $(I = hc/\sigma_a \tau_a \lambda_p)$	3.1 kW/cm ²
Thermal expansion coefficient (10 ⁻⁶ K ⁻¹)* [11]	$\alpha_{\rm p} = 3.35, \alpha_{\rm m} = 11.19, \alpha_{\rm g} = 14.55$
Thermal conductivity at 300 K (W/m K)* [11]	$\kappa_{\rm p} = 2.36, \kappa_{\rm m} = 3.41, \kappa_{\rm g} = 3.59$
dn_p/dT , dn_m/dT , $dn_g/dT (10^{-6} \text{ K}^{-1})^* [12]$	-10.8, -1.6, -7.4
Refractive indices at 1946 nm* [8]	$n_{\rm p} = 1.96, n_{\rm m} = 2.01, n_{\rm g} = 2.06$

Table 1. Properties of Tm:KLuW (* undoped crystal).

2. Experimental setup

High optical quality Tm:KLuW crystals were grown by the Top Seeded Solution Growth Slow Cooling (TSSG-SC) method, according to the procedure described in [13]. For the laser experiments, we constructed an L-shape hemispherical resonator depicted in Fig. 1. The pump was delivered through the plane mirror (M1), antireflection (AR) coated for the pump wavelength (802 nm) and high reflection (HR) coated for the laser wavelength (1900-2400 nm). As output coupler (M3) we tested mirrors with transmission $T_{oc} = 5\%$ and 10% (1820-2050 nm) and radius of curvature $R_{oc} = -75$ mm. The bending mirror (M2) was plane, AR_s. $_{polyp-pol}(45^{\circ}, 790-820 \text{ nm})$ and $HR_{s-pol}(45^{\circ}, 1700-2280 \text{ nm}) + HR_{p-pol}(45^{\circ}, 1790-2120 \text{ nm})$. The pump source was a fiber-coupled (NA = 0.22, 200 µm core diameter) AlGaAs diode laser delivering up to 10 W at 802 nm (DILAS). The active elements were cut for propagation along the N_g direction with dimensions 2 × 3 × 3 mm³ along $N_p \times N_m \times N_g$. The AR-coated samples (both for pump and laser wavelengths) were mounted in a Cu holder with circulating water at 16°C for heat dissipation. The incident pump beam was focused to a 200 µm spot diameter on the crystal with a lens assembly of 20 mm focal length. The polycrystalline Cr²⁺:ZnS SA samples (IPG Photonics), were specified with low signal transmission (corrected for Fresnel reflections) of $T_0 = 78$, 85 and 92% at 1910 nm. The AR-coating reduced the reflection to about 1% per surface. The SAs were 2.2 mm thick, with lateral dimensions of 4.5 \times 9.3 mm². The output pulses were detected with a fast InGaAs photodiode with <35 ps risetime and measured with a LeCroy oscilloscope with 1 GHz bandwidth.



Fig. 1. Setup of the PQS Tm:KLuW laser. SA: Cr²⁺:ZnS saturable absorber, M1: dichroic pump mirror, M2: HR-laser, HT-pump mirror, M3: output coupler.

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3. Results

In our preliminary work on PQS of the Tm:KLuW laser in a linear cavity [14] we established that it is rather difficult to stabilize the pulse train, mainly due to the direct heating of the SA (in that case Cr^{2+} :ZnSe with $T_0 = 75\%$ at 1950 nm) by the residual non-absorbed pump. Thus, the maximum pulse energy achieved with Tm:KLuW was 16 µJ. To improve this performance, we designed the 3 mirror L-shaped cavity described before, with a length of $L_1 + L_2 = L_c$ and $L_1 = 30$ mm, in which the folding mirror transmits the non-absorbed pump.

In the present work we used polycrystalline Cr^{2+} :ZnS samples as SA. It was not possible to obtain Q-switching with the Cr^{2+} :ZnSe plates because the fluence was not high enough to bleach Cr^{2+} :ZnSe at the longer distance between SA and pump mirror in the folded cavity. Additionally, the initial transmissions of Cr^{2+} :ZnSe at 1910 nm were lower (50%, 76% and 78%) compared to the applied Cr^{2+} :ZnS samples. The estimated laser beam diameters were 140 and 350 µm at the Tm:KLuW crystal and SA position ($L_{SA} = 40$ mm), respectively. The absorption cross-section at 1920 nm of Cr^{2+} :ZnS SA is 4×10^{-19} cm² and the emission crosssection of the laser crystal at the same wavelength is 1.38×10^{-20} cm². Together with the spot sizes of the laser beam at the laser crystal and the SA this gives $I_{SA}\sigma_{SA}\sim 4.6 I_g\sigma_0$ that matches with the criteria exposed in the introduction.



Fig. 2. CW and passively *Q*-switched power characteristics of the Tm:KLuW laser with $T_{oc} = 10\%$ and $R_{oc} = -75$ mm output coupler and different SAs.



Fig. 3. PQS characteristics of the Tm:KLuW laser with a) $T_{oc} = 10\%$ for $T_0 = 92\%$, 85% and 78% Cr^{2+} :ZnS SAs, and b) $T_{oc} = 5\%$ for $T_0 = 92\%$.

The CW and PQS performance of the laser with Tm:KLuW crystal, naturally polarized along $N_{\rm m}$, is shown in Fig. 2 for $T_{\rm oc} = 10\%$. In all cases the cavity length was decreased from 74 mm in CW mode to 73 mm in PQS regime compensating the optical path in the SA

#156170 - \$15.00 USD (C) 2012 OSA Received 10 Oct 2011; accepted 3 Nov 2011; published 30 Jan 2012 12 March 2012 / Vol. 20, No. S2 / OPTICS EXPRESS A3397 (n~2.27 at 1.9 µm). Figure 3a shows the average output power, repetition rate, pulse energy and pulse duration (FWHM) obtained in stable PQS regime using $T_{oc} = 10\%$ and Fig. 3b the same parameters with $T_{oc} = 5\%$ for which only the $T_0 = 92\%$ SA ensured stable operation. For $T_{oc} = 10\%$, the SA with $T_0 = 92\%$ *Q*-switched the laser in almost any position in the second arm but the highest output power was obtained at the maximum possible separation from the output coupler, $L_{SA} = 40$ mm (350 µm spot size). For $T_{oc} = 5\%$, with the same SA most stable operation was achieved at $L_{SA} = 20$ mm (400 µm spot size). The maximum average output power amounted to 0.39 W with $T_{oc} = 10\%$ and 0.26 W with $T_{oc} = 5\%$ at incident pump powers of 4.2 and 3.5 W, respectively, the limits set by SA bleaching. At the same pump levels, the output power in the CW regime (SA removed), was 0.66 and 0.75 W for $T_{oc} = 10\%$ (Fig. 2) and 5% (not shown), respectively, which translates into CW to PQS conversion of 59% and 35\%, respectively. The estimated small-signal absorption of the Tm:KLuW crystal was 70\%, so that the net pump efficiency in the PQS regime was 13% and 11% for $T_{oc} = 10\%$ and 5%, respectively.



Fig. 4. Dependence of the pulse repetition rate (prr) and single pulse shapes on the pump level using $T_{\rm oc} = 10\%$ and ${\rm Cr}^{2+}$:ZnS SA with $T_0 = 92\%$.

The laser wavelength in PQS operation for $T_{oc} = 5\%$ was $\lambda_{\rm L} = 1918$ nm with 2 nm bandwidth, while the laser emission for $T_{oc} = 10\%$ was in the $\lambda_{\rm L} = 1917$ -1925 nm range. The instabilities of the pulse train reduce with when the pumping power increased, e.g. from $\pm 20\%$ at $P_{\rm inc} = 3.5$ W (Fig. 4a) to $\pm 10\%$ at $P_{\rm inc} = 4.2$ W (Fig. 4c) for $T_{\rm oc} = 10\%$. The pulse shapes corresponding to these cases are shown in Figs. 4b and 4d. With the $T_0 = 92\%$ SA, the pulse duration was slightly shorter for $T_{\rm oc} = 10\%$ in comparison to $T_{\rm oc} = 5\%$, decreasing typically from ~30 ns to ~24 ns. The shortest pulses (~10 ns) for $T_{\rm oc} = 10\%$ were obtained with the $T_0 = 78\%$ SA for a pulse energy of 50 µJ (Fig. 3a), the peak power is 5 kW.

The repetition rate at maximum power with $T_{oc} = 5\%$ was 2 kHz corresponding to a maximum single pulse energy of 127 µJ and maximum peak power of 4.4 kW. The maximum energy achieved with $T_{oc} = 10\%$ was 145 µJ at a repetition rate of 2.7 kHz, with peak power of 6 kW. In fact this energy corresponded to saturation at incident pump powers ~4 W, with further increase of the average output power only due to the increasing repetition rate (Fig. 3). The quality of the beam, determined by the knife-edge method, was $M_x^2 = M_y^2 = 1.2$.

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4. Theoretical analysis of the passively *Q*-switched Tm:KLuW laser performance

The optimal PQS parameters can be calculated following an analytical approach [15]. They can be expressed in terms of a single variable $z = 2g_0 l/\delta$, where $2g_0 l$ is the small-signal gain and δ is the round-trip loss due to diffraction, scattering and absorption. To determine these two, it is possible to apply the Findlay-Clay method [15] assuming good thermal management and minimum reabsorption losses so that Tm:KLuW can be considered as quasi-three level laser. This method uses the relationship between loss and threshold gain, so that the pump power at the laser threshold is expressed in terms of the reflectance of output coupler $R = 1-T_{oc}$:

$$-\ln R = \frac{2\eta_{exc}}{A_p I_s} P_p - \delta = 2g_0 l - \delta.$$
⁽¹⁾

where η_{exc} is the excitation efficiency, A_p is the area of the pump beam in the crystal and I_s is the saturation intensity. Using mirrors with $T_{\text{oc}} = 1.5$, 3, 5, 9 and 15% in CW operation the loss per round trip obtained is 9%, and the small-signal gain is 0.204 P_p , in this way, the parameter z is determined and depends only on P_p . The output energy is then given by

$$E_p = \frac{Ah\nu\delta}{2\sigma_0\gamma}(z-1-\ln z).$$
⁽²⁾

where A is the area of the laser beam (70 μ m radius), hv is the photon energy and γ is one for four-level lasers or two for three-level lasers. Finally, the pulse duration is also determined by

$$\Delta t = \frac{2L_c}{c\delta} \left(\frac{\ln z}{z[1 - a(1 - \ln a)]} \right)$$
(3)

with $a = (z-1)/(z \ln z)$. The calculated optimum parameters for the Tm:KLuW laser are shown in Fig. 5 together with the experimental results obtained with $T_{oc} = 10\%$ and $T_0 = 92\%$, 85% and 78%. There is a good agreement in the pulse energy for incident power in the 2.2 – 3.4 W range ($T_0 = 92\%$), at higher powers saturation is experimentally observed, not taken into account in the model. The theory predicts well also the pulse durations for $T_0 = 85\%$ and 78%.



Fig. 5. Experimental results compared with a) calculated pulse energy and b) pulse duration, depending on the incident power for the Tm:KLuW crystal and $T_{oc} = 10\%$.

5. Conclusion

We have achieved passive *Q*-switching operation of a diode-pumped Tm:KLuW laser emitting near 1920 nm using polycrystalline Cr^{2+} :ZnS samples as saturable absorbers. The best results without optical damage were obtained with $T_{oc} = 10\%$ and $T_0 = 92\%$ for 3 at.% Tm doping in terms of maximum energy of 145 µJ, pulse durations in the 24 – 30 ns range,

#156170 - \$15.00 USD (C) 2012 OSA repetition rate around 2.7 kHz and 0.39 W of average output power. In comparison with [3], based on similar laser crystal and SA, the improvement achieved is by a factor >3 in the average power, >20 in the pulse energy and >50 in the peak power. In fact, with the present setup, average output powers of 0.6 W and pulse energies of 200 μ J could be reached at incident pump power of 7 W for $T_0 = 92\%$ and $T_{oc} = 10\%$ at 3 kHz. However, at such high intracavity fluence we observed SA damage within minutes, even in positions close to the output coupler. Further improvement of the present results can be expected for SAs with $T_0>92\%$. We conclude that polycrystalline Cr²⁺:ZnS is superior as SA at wavelengths around 2 μ m in comparison with PbS quantum dots-doped glass [4] or semiconductor SAs [5] for PQS of diode-pumped Tm-lasers.

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