



Open Access

Tm³⁺:LiGdF₄ Laser, Passively Q-Switched With a Cr²⁺:ZnSe Saturable Absorber

IEEE Photonics Journal

An IEEE Photonics Society Publication

Volume 7, Number 3, June 2015

Bozhidar Oreshkov Stefano Veronesi Mauro Tonelli Alberto di Lieto Valentin Petrov Uwe Griebner Xavier Mateos Ivan Buchvarov



DOI: 10.1109/JPHOT.2015.2427737 1943-0655 © 2015 IEEE





Tm³⁺:LiGdF₄ Laser, Passively Q-Switched With a Cr²⁺:ZnSe Saturable Absorber

Bozhidar Oreshkov,^{1,4} Stefano Veronesi,² Mauro Tonelli,^{2,3} Alberto di Lieto,^{2,3} Valentin Petrov,¹ Uwe Griebner,¹ Xavier Mateos,⁵ and Ivan Buchvarov^{4,6}

¹Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, 12489 Berlin, Germany ²NEST Istituto Nanoscienze—CNR, 56127 Pisa, Italy ³Dipartimento di Fisica dell'Università di Pisa, 56127 Pisa, Italy ⁴Faculty of Physics, Sofia University, 1164 Sofia, Bulgaria ⁵Física i Cristallografia de Materials i Nanomaterials (FiCMA-FiCNA), Universitat Rovira i Virgili (URV), 43007 Tarragona, Spain ⁶Laser Engineering and Biomedical Optics Department, ITMO University, 197101 St. Petersburg, Russia

DOI: 10.1109/JPHOT.2015.2427737

1943-0655 © 2015 IEEE. Translations and content mining are permitted for academic research only. Personal use is also permitted, but republication/redistribution requires IEEE permission. See http://www.ieee.org/publications_standards/publications/rights/index.html for more information.

Manuscript received March 25, 2015; revised April 23, 2015; accepted April 24, 2015. Date of current version May 22, 2015. This work was supported in part by the Bilateral Exchange Programme between Germany and Bulgaria under DAAD Project-ID 54391860; by the EC's Seventh Framework Programme (LASERLAB-EUROPE) under Grant 284464; and by the Bulgarian Ministry of Education, Science, and Youth under Grant DO/02/134/2009. Corresponding author: B. Oreshkov (e-mail: b.oreshkov@phys.uni-sofia.bg).

Abstract: We demonstrate efficient passively Q-switched laser operation, based on Tm^{3+} :LiGdF₄ and Cr²⁺:ZnSe, as active medium and saturable absorber, respectively. The polarized emission was centered at 1876 nm, with pulse parameters of 13 ns (duration), 467 μ J (energy), and 37 kW (peak power) at a repetition rate of 350 Hz.

Index Terms: Diode-pumped, solid state, Q-switched, infrared laser.

1. Introduction

Lasers based on the Tm³⁺-ion (Tm) emission near 1.9 μ m are interesting for many medical applications [1], remote sensing (LIDAR) and pumping Ho³⁺ (Ho)- [2], [3] and Cr²⁺ (Cr)-lasers [4], as well as optical parametric oscillators (OPOs) for frequency down-conversion into the mid-IR. Although, very recently, continuous-wave (CW) operation of orientation-patterned GaAs (OPGaAs) based OPO was also demonstrated, such devices normally require short pulses from Q-switched Tm- or Ho-lasers to reach the threshold without optical damage and achieve sufficient conversion efficiency. High pulse energy and peak power are advantageous for these applications since they can be obtained at relatively low average power if compared with the CW regime of operation. Furthermore, the most promising nonlinear materials, ZnGeP₂ and OPGaAs exhibit residual or two-photon absorption at shorter wavelengths. While energies of the order of 10 mJ have been demonstrated by active Q-switching and pulsed pumping of bulk Tm-lasers [5], passive Q-switching (PQS) is much simpler and cheaper for realization and more importantly, shorter pulses are generated with higher peak power, which is advantageous for OPO pumping.

In recent years, PQS of diode-pumped solid-state (DPSS) lasers by the use of intracavity saturable absorber (SA) was successfully demonstrated in several CW-pumped, bulk Tm-doped materials. Those include, yttrium aluminium garnet YAG [6], yttrium aluminium perovskite YAP [7],



Fig. 1. Set-up of the PQS Tm:GLF laser. $M_1:$ dichroic pump mirror, $M_2:$ HR-laser, HT-pump mirror, $M_3:$ output coupler.

monoclinic double tungstates — $KY(WO_4)_2$ (KYW) [8], $KLu(WO_4)_2$ (KLuW) [9], and fluorides — LiYF₄ (YLF) [10], LiLuF₄ (LLF) [11], and BaY₂F₈ (BYF) [12], in combination with different SAs, such as Cr:ZnSe [6] and Cr:ZnS [9]–[12] crystals/ceramics, PbS quantum dots [8], and InGaAs/GaAs semiconductor based SAs [7].

Best results in PQS of DPSS lasers around 1.9 μ m, in terms of pulse energy and peak power have been achieved with Cr:ZnS and Cr:ZnSe SAs. Such SAs were successfully employed in Tm:YAG, Tm:KYW, and Tm:KLuW, as well as in co-doped Yb,Tm:KYW [13] lasers. While monoclinic double tungstates are known for their high cross sections, they exhibit one of the shortest lifetimes. Such properties are more adequate for mode-locking, however, in PQS, long upper state lifetimes of the active medium ensure high energy storage capability and consequently pulses of higher energy can be generated. It is well known that fluoride hosts, due to their low phonon energies, exhibit the longest lifetimes for rare earth dopants [14]. Indeed, using Tm:YLF [10] and Tm:LLF [11], combined with Cr:ZnS SAs, we achieved the highest so far pulse energies — on the millijoule level, the shortest pulse durations -<10 ns, and the highest peak powers ->100 kW, for any DPSS PQS Tm laser. Such peak powers already exceed the best values reported for flashlamp pumped PQS systems, like Tm:YAG, in combination with Cr:ZnSe SA [15].

In the present work, we investigate the PQS performance of Tm-doped LiGdF₄ (GLF). This is a relatively new laser crystal isomorphous to the well-known YLF and LLF, in which the dopant substitutes Gd³⁺ host ions. Although, CW [16], [17] and mode-locked [18] operation of Tm:GLF were previously studied, to the best of our knowledge, this is the first report of Q-switching of this laser medium. The values for the maximum emission cross section $(3.1 \times 10^{-21} \text{ cm}^2 \text{ at}$ 1877 nm for π polarization) and fluorescence decay time of the ³F₄ manifold (16.0 ms) derived in [17] are comparable and even slightly higher than those found for other fluorides [19], [20]. This makes Tm:GLF, combined with the simplicity of PQS, another very attractive material for generation of short laser pulses and high peak powers in the 1.9 μ m wavelength range.

2. Experimental Set-Up

For our experiments, an L-shape hemispherical resonator as shown in Fig. 1 was employed. This scheme was chosen for the non-absorbed pump power not to reach the SA, an effect causing instabilities in simple two-mirror cavities. The pump beam is delivered through the plane mirror (M₁), antireflection (AR) coated for the pump wavelength and high reflection (HR), > 99.9%, coated for 1800–2090 nm. As output coupler (OC, M₃) we used curved mirrors with radius of curvature of -75 mm and transmission $T_{OC} = 5\%$, 10%, and 20% (at 1900 nm). The folding mirror (M₂) was plane, highly transmitting (HT) for the pump and HR for the laser radiation at 45°. The pump source was a fiber-coupled (NA = 0.15, 105 μ m core diameter) AlGaAs diode laser delivering > 10 W unpolarized beam. The pump power incident on the Tm:GLF crystal was limited to 1.9 W to avoid any risk of optical damage. The wavelength of the pump laser was temperature-tuned to 792.5 nm, which is the second highest absorption peak of Tm:GLF for π



Fig. 2. (a) Absorption of the Tm:GLF crystal versus incident pump power measured with $T_{OC} = 10\%$. (b) CW output power versus absorbed pump power (symbols) and linear fits (lines) for calculation of the slope efficiency η for the three different OCs.

polarization — $\sigma_{abs} = 5.0 \times 10^{-21} \text{ cm}^2$, with a $\Delta\lambda(\text{FWHM}) = 4.5 \text{ nm}$ [16]. The focal spot achieved in the position of the crystal with an f = 60 mm lens assembly had a diameter of 230 μ m.

The Tm:GLF sample we used was 2.7 mm thick with a-cut and aperture of $3.40(a) \times$ 3.11(c) mm², doped with 8 at.% of Tm ($N = 1.07 \times 10^{21}$ at/cm³). It was uncoated and mounted with two lateral surfaces in contact with a Cu holder, cooled by circulating water at 18°C for heat dissipation. For PQS, it is required that the absorption cross section of the SA is higher than the emission cross section of the gain material or in more general treatment these cross sections shall be normalized to the corresponding intracavity beam cross sections. Cr:ZnS absorption maximum is at 1680 nm, with peak absorption cross-section $\sigma_{asa} = 10.0 \times 10^{-19} \text{ cm}^2$, whilst Cr:ZnSe absorption maximum is shifted by 100 nm to longer wavelengths at 1780 nm, with $\sigma_{gsa} = 11.0 \times 10^{-19} \text{ cm}^2$ [21]. Moving to longer wavelengths, the difference in the ground state absorption becomes greater, and in the spectral region around 1900 nm the ground state absorption of Cr:ZnSe is nearly twice higher than that of Cr:ZnS. Thus, polycrystalline Cr²⁺:ZnSe was chosen in the present work as SA. The sample available (IPG Photonics) was specified with low signal transmission (corrected for Fresnel reflections) of $T_0(1910 \text{ nm}) = 85\%$ which is equivalent to $T_0(1880 \text{ nm}) = 82\%$. It was AR-coated which reduced the Fresnel reflection to < 0.5% per surface at 1910 nm. The SA element was 1.81 mm thick with lateral dimensions of 3.6×7.2 mm². The separations L₁ and L₂ on Fig. 1 were 50 mm and 21 mm respectively, in both CW and PQS regimes. The Gaussian diameter of the TEM₀₀ mode was calculated to be 232 μ m in the Tm:GLF crystal and 715 μ m in the SA, which ensured operation in the fundamental transversal mode. With the peak Tm emission cross section at 1877 nm for π -polarization [17], this gives a ratio of \sim 30 for the normalized cross sections of the SA and Tm:GLF.

3. Results and Discussion

The performance of the Tm:GLF laser was first studied in CW regime of operation and after that in PQS. CW operation was realized with the three different OCs up to absorbed pump power of ~1.0 W. The crystal absorption was measured to be on average 57.0% in lasing conditions, slightly decreasing with the pump power, from 59.0% at threshold, to 55.9% at 1.0 W. The absorption results shown on Fig. 2(a) are with the $T_{OC} = 10\%$, but the absorption was the same with the other two OCs. Maximum output power of 382 mW and slope efficiency of 52.4% were achieved with $T_{OC} = 10\%$ [see Fig. 2(b)]. The slope efficiencies and output powers were similar, but slightly lower with the other two OCs. The laser output was always π -polarized ($E \parallel c$). The central wavelength shifted slightly when increasing the transmittance of the OC, from 1902 nm for $T_{OC} = 5\%$ to 1880 nm for $T_{OC} = 10\%$, and 1878 nm for $T_{OC} = 20\%$, as expected when higher inversion is required in the three-level Tm-laser system.

With the SA inside the cavity for PQS operation, the best results, in terms of pulse energy, duration, and peak power, were achieved with $T_{OC} = 20\%$. We have chosen to start with the



Fig. 3. (a) Output power in CW and PQS regimes versus absorbed pump power (symbols) and linear fits (lines) for calculation of the slope efficiency η . (b) Pulse characteristics in the PQS regime versus absorbed pump power.



Fig. 4. Q-switched pulse shape recorded at maximum pump power. The inset shows the pulse train.

highest output coupler transmission, in order to prevent damage in the active element. The maximum average output power reached 152 mW, at an absorbed pump power of 1.0 W, corresponding to slope efficiency of 29.8% [see Fig. 3(a)] and 42.5% of Q-switching to CW extraction efficiency at maximum power. The output pulses were monitored with a high speed InGaAs photodiode (> 12.5 GHz at 2 μ m) and a 1 GHz oscilloscope. The results in terms of repetition rate, pulse duration, single pulse energy and peak power are summarized in Fig. 3(b). The repetition rate increases from 30 to 350 Hz with the pump power. The pulse duration was almost constant — 13 ns. The maximum pulse energy reached 467 μ J, and the maximum peak power reached 37 kW. These values were measured close to the PQS threshold corresponding to absorbed pump power of 0.59 W, average output power of 14 mW and pulse repetition rate of 30 Hz. With the increase of the pump power the pulse energy increased up to a certain point, and then started to decrease. Thus, if the measurements near the threshold where the overall performance was not very stable are excluded, maximum pulse energy of 441 μ J and peak power of 35 kW (of the same order of magnitude), corresponding to much higher average output power of 128 mW at a repetition rate of 290 Hz, were obtained for absorbed pump power of 0.96 W.

The typical shape of the Q-switched pulses and the pulse train at maximum pump power are shown in Fig. 4. As in the CW regime, the output in PQS was π -polarized. The central wavelength was slightly shorter, 1876 nm, due to the increased overall cavity losses for the three-level Tm-laser system.

The output spectra with $T_{OC} = 20\%$ are shown on Fig. 5(a) for CW and in the inset for PQS mode of operation. The central wavelengths correspond to the main emission peak for this polarization identified at 1878 nm in [17]. The computed gain coefficient $N\sigma_{gain}$ of Tm:GLF is



Fig. 5. (a) Output spectra of the Tm:GLF laser in CW and PQS (inset) regimes with $T_{OC} = 20\%$. (b) Tm:GLF π -polarized gain coefficient for four values of the inversion parameter β .



Fig. 6. Tm:GLF crystal (a) before and (b) after the PQS experiment with the $T_0(1910 \text{ nm}) = 76\% \text{ SA}$.

shown in Fig. 5(b) for four inversion rates, i.e., $\beta = 0.15$, 0.2, 0.3, and 0.4, with the gain cross section defined by $\sigma_{gain} = \beta \sigma_{emi} - (1 - \beta) \sigma_{abs}$, where σ_{emi} and σ_{abs} are the emission and absorption cross sections at room temperature. Compared to Tm:YLF and Tm:LLF the fluorescence peaks for both π and σ polarizations are shifted to slightly shorter wavelengths and this is seen in the emission spectra of the short pulse lasers, too [10], [11]. However, the more significant distinct feature compared to these isomorphs is that in Tm:GLF the σ polarization almost completely overlaps with the stronger π polarization, and as evident from the CW performance there is no polarization switching, when wavelength exceeds 1900 nm in contrast to the other two hosts [10], [11].

In order to increase the output energy and shorten the pulse duration we inserted another SA with lower initial transmission—Cr:ZnSe with $T_0(1910 \text{ nm}) = 76\%$ corresponding to $T_0(1880 \text{ nm}) = 71\%$. PQS operation was achieved very briefly, after which the output power dropped to zero, and after few attempts we found evidence for laser induced damage, closer to the entrance surface (for the pump beam) in the Tm:GLF crystal, as can be seen in Fig. 6. The cavity mode size is slightly smaller at this surface.

The occurrence of damage spots could be attributed to the poor quality of the laser crystal. As shown in Fig. 6(a), many small micro-defects inside the crystal were present even before starting the laser experiments. They are related to the extreme difficulty in the growth process of Tm:GLF. In fact GLF is strongly incongruent and it is not possible to grow more than about 30% of the raw material loaded in the crucible. Moreover additional structure stress is introduced by the substitution of Gd by Tm. In any family of rare earth hosts, that is the most difficult, due to the largest difference in the ionic radii compared to Y and Lu. For fluoride hosts, which is our case, the radii of these elements are as follows: Y—97.7 pm (in YLF), Lu—101.9 pm (in LLF), Gd—105.3 pm (in GLF), and for Tm—99.4 pm [22]. These numbers are taken from [22, Ref. 10] for a coordination number of 8. All these occurrences can result in the formation of micro-defects inside the crystal, as is evident in Fig 6.

4. Conclusion

In conclusion, we demonstrated PQS of a DPSS Tm:GLF laser, for the first time. Pulse duration as short as 13 ns, with moderate pulse energy and high peak power, i.e., 467 μ J and 37 kW, were achieved using Cr:ZnSe as a SA. These initial results are better compared to different Tm hosts such as garnets and double tungstates. However, they are still inferior compared to the other two fluoride isomorphs (Tm:YLF and Tm:LLF). We attribute this to the inferior quality of the available Tm:GLF crystal sample. We believe that with better optical quality samples, higher energies and shorter pulse durations could be achieved with this promising new material. The advantage in comparison to the other fluoride materials is the stable intrinsic polarization selection which also supports a different (somewhat shorter) oscillation wavelength.

References

- [1] K. Scholle, S. Lamrini, P. Koopmann, and P. Fuhrberg, "2 μm laser sources and their possible applications," in Frontiers in Guided Wave Optics and Optoelectronics, B. Pal, Ed. Vukovar, Croatia: Intech, 2010, pp. 471–500.
- [2] P. A. Budni, M. L. Lemons, J. R. Mosto, and E. P. Chicklis, "High-power/high-brightness diode-pumped 1.9-μm thulium and resonantly pumped 2.1-μm holmium lasers," *IEEE J. Sel. Topics Quantum Electron.*, vol. 6, no. 4, pp. 629– 635, Jul./Aug. 2000.
- [3] A. Dergachev, D. Armstrong, A. Smith, T. Drake, and M. Dubois, "3.4-µm ZGP RISTRA nanosecond optical parametric oscillator pumped by a 2.05-µm Ho:YLF MOPA system," Opt. Exp., vol. 15, no. 22, pp. 14 404–14 413, Oct. 2007.
- [4] S. Mirov, V. Fedorov, I. Moskalev, M. Mirov, and D. Martyshkin, "Frontiers of mid-infrared lasers based on transition metal doped II–VI semiconductors," J. Luminescence, vol. 133, pp. 268–275, Jan. 2013.
- [5] M. Eichhorn, "Quasi-three-level solid-state lasers in the near and mid infrared based on trivalent rare earth ions," *Appl. Phys. B*, vol. 93, no. 2/3, pp. 269–316, Nov. 2008.
- [6] M. Mond et al., "Passive Q-switching of a diode-pumped Tm:YAG laser by Cr²⁺:ZnSe," presented at the Proc. Conf. Lasers Electro-Optics Eur., 2003, Paper CA7-5-WED [CD-ROM].
- [7] 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. Exp., vol. 18, no. 13, pp. 13 574–13 579, Jun. 2010.
- [8] M. S. Gaponenko *et al.*, "Diode-pumped Tm : KY(WO₄)₂ laser passively Q-switched with PbS-doped glass," *Appl. Phys. B*, vol. 93, no. 4, pp. 787–791, Dec. 2008.
- [9] M. Segura *et al.*, "Passive Q-switching of the diode pumped Tm³⁺:KLu(WO₄)₂ laser near 2-μm with Cr²⁺:ZnS saturable absorbers," *Opt. Exp.*, vol. 20, no. 4, pp. 3394–3400, Feb. 2012.
- [10] R. Faoro et al., "Passively Q-switched Tm:YLF laser," Opt. Lett., vol. 37, no. 9, pp. 1517–1519, May 2012.
- [11] H. Yu *et al.*, "Compact passively Q-switched diode-pumped Tm:LiLuF₄ laser with 1.26 mJ output energy," *Opt. Lett.*, vol. 37, no. 13, pp. 2544–2546, Jul. 2012.
- [12] H. Yu *et al.*, "Passively Q-switched Tm:BaY₂F₈ lasers," in *Proc. SPIE*, 2013, vol. 8786, p. 878616.
- [13] M. Segura *et al.*, "Passive Q-switching of a diode-pumped (Tm, Yb):KLu(WO₄)₂ laser near 2-μm with a Cr₂₊:ZnS saturable absorber," in *Proc. SPIE*, 2012, vol. 8235, p. 82351U.
- [14] L. H. Guilbert, J. Y. Gesland, A. Bulou, and R. Retoux, "Structure and Raman spectroscopy of Czochralski-grown barium yttrium and barium ytterbium fluorides crystals," *Mater. Res. Bull.*, vol. 28, no. 9, pp. 923–930, Sep. 1993.
- [15] T. Tsai and M. Birnbaum, "Q-switched 2-μm lasers by use of a Cr²⁺:ZnSe saturable absorber," Appl. Opt., vol. 40, no 36, pp. 6633–6637, Dec. 2001.
- [16] F. Cornacchia, A. Di Lieto, and M. Tonelli, "LiGdF₄:Tm³⁺:Spectroscopy and diode-pumped laser experiments," Appl. Phys. B, vol. 96, no. 2/3, pp. 363–368, Aug. 2009.
- [17] N. Coluccelli *et al.*, "High-efficiency diode-pumped Tm:GdLiF₄ laser at 1.9 μm," Opt. Lett., vol. 34, no. 22, pp. 3559– 3561, Nov. 2009.
- [18] J. N. Coluccelli *et al.*, "Passive mode-locking of a diode-pumped Tm:GdLiF₄ laser," *Appl. Phys. B*, vol. 101, no. 1/2, pp. 75–78, Oct. 2010.
- [19] N. Coluccelli *et al.*, "Tm-doped LiLuF₄ crystal for efficient laser action in the wavelength range from 1.82 to 2.06 μm," Opt. Lett., vol. 32, no. 14, pp. 2040–2042, Jul. 2007.
- [20] 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.*, vol. 28, no. 11, pp. 2619–2630, Nov. 1992.
- [21] I. T. Sorokina, "Crystalline mid-infrared lasers," in *Solid-State Mid-IR Laser Sources*, vol. 89, I. T. Sorokina and K. L. Vodopyanov, Eds. Berlin, Germany, Springer-Verlag, 2003, pp. 255–351, ser. Topics in Applied Physics.
- [22] B. M. Walsh, N. P. Barnes, M. Petros, J. Yu, and U. N. Singh, "Spectroscopy and modeling of solid state lanthanide lasers: Application to trivalent Tm³⁺ and Ho³⁺ in YLiF₄ and LuLiF₄," *J. Appl. Phys.*, vol. 95, no. 7, pp. 3255–3271, Apr. 2004.