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## CW laser operation around 2- $\mu\text{m}$ in (Tm,Yb):KLu(WO<sub>4</sub>)<sub>2</sub>

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### Abstract

Laser generation in continuous wave (CW) regime at 1.94- $\mu\text{m}$  from (Tm,Yb) codoped system has been investigated in two different hosts: KLu(WO<sub>4</sub>)<sub>2</sub> and KY(WO<sub>4</sub>)<sub>2</sub>. The high quality crystals were grown by the Top-Seeded Solution Growth Slow Cooling (TSSG-SC) method with doping levels of 2.5 at. %Tm and 5 at. %Yb. The active media were pumped with a diode laser at 980 nm. We demonstrated the superior performance of KLu(WO<sub>4</sub>)<sub>2</sub> compared to that of KY(WO<sub>4</sub>)<sub>2</sub> and improved the results already obtained in the literature. The maximum laser output power reached was 157 mW for (Tm,Yb):KLu(WO<sub>4</sub>)<sub>2</sub> and 123 mW for (Tm,Yb):KY(WO<sub>4</sub>)<sub>2</sub>.

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### 1. Introduction

The laser emission around 2- $\mu\text{m}$  based on the  $^3\text{F}_4 \rightarrow ^3\text{H}_6$  transition of thulium (Tm) has become interesting for medical applications and atmospheric sensing, mainly due to the strong absorption of water around this wavelength, as well as for pumping OPO's for conversion in the mid-IR [1]. The 2- $\mu\text{m}$  laser emission can easily be achieved by pumping directly around 800 nm with AlGaAs diode lasers in the case of single doped Tm laser crystals or glasses, or by pumping around 980 nm with InGaAs diode lasers when using a sensitizer ion such as Yb<sup>3+</sup>. The laser

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operation with single doped  $\text{Tm}^{3+}$  has been successfully demonstrated in a wide variety of hosts such as YAG ( $\text{Y}_3\text{Al}_5\text{O}_{12}$ ) [2],  $\text{YLiF}_4$  [3],  $\text{YVO}_4$  [4] and the double tungstates  $\text{KRE}(\text{WO}_4)_2$  (Hereafter, KREW; RE=Y, Gd, Lu) [5,6]. On the other hand, there are many works focused on the  $\text{Yb}^{3+}$  as sensitizer ion for  $\text{Tm}^{3+}$  particularly due to the high absorption cross section of  $\text{Yb}^{3+}$  and the effective energy transfer from  $\text{Yb}^{3+}$  to  $\text{Tm}^{3+}$  [7, 8, 9].

The high absorption and emission cross sections for the rare earth ions and also the possibility of high concentration doping levels make the double tungstates very attractive materials to be used as laser host. For instance the maximum values of the absorption cross section of the Yb doped KYW and KLuW single crystals are  $11.7 \times 10^{-20} \text{ cm}^2$  [10] and  $11.8 \times 10^{-20} \text{ cm}^2$  [11] respectively. These values are considerably large compared to other commonly used hosts such as YAG,  $\text{YAlO}_3$ , YLF [12] or YVO [13]. Regarding the emission cross section of thulium in KREW's, it has been found that  $1.15 \times 10^{-20} \text{ cm}^2$  at 1910 nm for  $E//N_m$ , and  $1.20 \times 10^{-20} \text{ cm}^2$  at 1950 nm for  $E//N_m$  are the maximum values for KYW [14] and KLuW [11], respectively. The laser action of single doped Tm and Yb ions has been successfully achieved in both hosts with high efficiencies. The laser emission at 1910 nm of (Tm,Yb) codoped system has been achieved in KYW in [9], but to the best of our knowledge, lasing of (Tm,Yb) codoped KLuW single crystals has not yet been demonstrated.

The  $\text{Yb}^{3+}$  ion used as sensitizer is of great advantage since it has only two manifolds, the ground state  $^2F_{7/2}$  and the excited state  $^2F_{5/2}$ . Once the ion is excited it only can decay to the ground state or transfer part of its energy to another ion if the material is codoped and if the levels are reasonably resonant in energy.

The population of the  $^3F_4$  level of  $\text{Tm}^{3+}$  via energy transfer from  $\text{Yb}^{3+}$  can be described using the energy levels scheme in figure 1. The 980 nm pump light is absorbed by  $\text{Yb}^{3+}$  ions, a part of this energy is transferred ( $T_1$ ) from the  $^2F_{5/2}$  level of  $\text{Yb}^{3+}$  to the  $^3H_5$  level of  $\text{Tm}^{3+}$  that decays via non-radiative process to the  $^3F_4$  level. Part of these electrons in  $^3F_4$  level could decay to the ground level, another could absorb energy ( $T_2$ ) and get up to  $^3F_{2,3}$  levels, decaying to  $^3H_4$  level via non-radiative process. Finally, the  $^3F_4$  level can be populated via cross relaxation process such as  $R_1$  and  $R_2$  in figure 1. Another processes in the (Tm,Yb) system, such as  $T_3$  process, give rise to the population of the  $^1G_4$  level that decays emitting at 480 nm ( $^1G_4 \rightarrow ^3H_6$ ) and 650 nm ( $^1G_4 \rightarrow ^3F_4$ ).

In this work, we report on the laser oscillation of (Tm,Yb) codoped KLuW and KYW single crystals grown by the Top Seeded Solution Growth Slow Cooling (TSSG-SC) method. The obtained crystals (2.5 at. % Tm and 5 at. % Yb) were cut according to the principal optical axes for better laser performance and placed in a hemispherical resonator for the laser experiments.

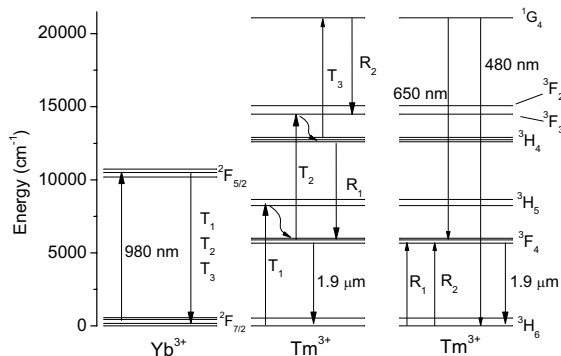


Figure 1. Energy levels scheme indicating the energy transfer processes in the (Tm,Yb) system.

## 2. Active media

For laser experiments, we used two different hosts from monoclinic double tungstates family:  $\text{KY}(\text{WO}_4)_2$  (KYW) and  $\text{KLu}(\text{WO}_4)_2$  (KLuW) doped with Tm and Yb. The cell parameters of KYW, in the  $C2/c$  space group, are  $a = 10.631 \text{ \AA}$ ,  $b = 10.345 \text{ \AA}$ ,  $c = 7.555 \text{ \AA}$ ,  $\beta = 130.75^\circ$  and  $Z = 4$  [15], similarly, the cell parameters for KLuW are  $a = 10.576 \text{ \AA}$ ,  $b = 10.214 \text{ \AA}$ ,  $c = 7.487 \text{ \AA}$ ,  $\beta = 130.68^\circ$  and  $Z = 4$  [16]. For both hosts, the principal optical axis

$N_p$  is parallel to  $b$  crystallographic direction, while  $N_m$  and  $N_g$  lie in the  $a$ - $c$  crystallographic plane. The principal optical axis  $N_g$  is at  $18.5^\circ$  clockwise with respect to  $c$  crystallographic direction.

The (Tm,Yb)-doped crystals were grown by the Top-Seeded Solution Growth Slow-Cooling (TSSG-SC) method described in detail in [17]. The crystals grew from a KLuW (or KYW) seed oriented along  $b$  crystallographic direction. The doping levels were 2.5 at. % Tm and 5 at. % Yb. After growth, the chemical composition of the crystals was  $\text{KLu}_{0.919}\text{Tm}_{0.030}\text{Yb}_{0.051}(\text{WO}_4)_2$  and  $\text{KY}_{0.923}\text{Tm}_{0.026}\text{Yb}_{0.051}(\text{WO}_4)_2$  measured by Electron-Probe Micro-Analysis (EPMA).

For the laser experiments, we constructed a hemispherical resonator consisting of a planar mirror with antireflecting coating (AR) for the pump wavelength (770–1050 nm) and highly reflecting (HR) coating for the laser wavelength (1800–2075 nm). As output coupler we tested different mirrors with several transmissions at the laser wavelength (1820–2050 nm)  $T_{oc} = 1.5\%$ ,  $3\%$ ,  $5\%$  and  $9\%$  and different radius of curvature  $R_{oc} = 25$ ,  $50$  and  $75$  mm. The pump source was a fiber-coupled (NA = 0.22, core diameter =  $200 \mu\text{m}$ ) high power InGaAs diode laser delivering up to  $50$  W emitting in the  $976$ – $980$  nm range depending on the current level. The active media were cut for propagation along the  $N_g$  direction with dimensions  $3 \times 3 \times 1.6 \text{ mm}^3$  along  $N_p \times N_m \times N_g$ . The uncoated high optical quality polished samples were mounted in a water cooled copper holder for heat dissipation. The normal incident pump beam was focused to a  $200 \mu\text{m}$  spot diameter onto the crystal with a lens of  $20$  mm focal length.

### 3. Laser experiments results

The input-output characteristics of (Tm,Yb):KLuW and (Tm,Yb):KYW for several  $T_{oc}$  and  $R_{oc}$  are summarized in table 1, and the results for  $R_{oc} = 50$  mm are shown in fig. 2 as representative of the whole results obtained in the present work.

**Table 1. Maximum output powers and their corresponding incident powers and laser wavelengths of the 2.5 at.% Tm, 5 at.% Yb:KLuW (left) and KYW (right) depending on the  $T_{oc}$  and  $R_{oc}$ .**

$T_{oc}(\%)$	(TmYb):KLuW			(TmYb):KYW		
	$R_{oc}(\text{mm})$			$R_{oc}(\text{mm})$		
	25	50	75	25	50	75
1.5	$P_{out}=153$ mW	$P_{out}=157$ mW	$P_{out}=140$ mW	$P_{out}=122$ mW	$P_{out}=123$ mW	$P_{out}=126$ mW
	$P_{in}=5.2$ W	$P_{in}=5.2$ W	$P_{in}=5.2$ W	$P_{in}=4.9$ W	$P_{in}=4.7$ W	$P_{in}=5.2$ W
	$\eta=3.6\%$	$\eta=3.8\%$	$\eta=3.4\%$	$\eta=3.0\%$	$\eta=3.5\%$	$\eta=3.1\%$
3	$P_{out}=143$ mW	$P_{out}=148$ mW	$P_{out}=121$ mW	$P_{out}=103$ mW	$P_{out}=94$ mW	$P_{out}=107$ mW
	$P_{in}=4.4$ W	$P_{in}=4.9$ W	$P_{in}=4.9$ W	$P_{in}=4.9$ W	$P_{in}=4.4$ W	$P_{in}=4.9$ W
	$\eta=4.3\%$	$\eta=4.0\%$	$\eta=3.3\%$	$\eta=2.6\%$	$\eta=3.1\%$	$\eta=3.2\%$
5	$P_{out}=118$ mW	$P_{out}=121$ mW	$P_{out}=109$ mW	$P_{out}=87.1$ W	$P_{out}=80$ mW	$P_{out}=83$ mW
	$P_{in}=4.9$ W	$P_{in}=4.7$ W	$P_{in}=4.9$ W	$P_{in}=4.7$ W	$P_{in}=4.4$ W	$P_{in}=4.9$ W
	$\eta=3.4\%$	$\eta=3.7\%$	$\eta=3.2\%$	$\eta=2.8\%$	$\eta=3.0\%$	$\eta=2.8\%$
9	$P_{out}=73.5$ mW	$P_{out}=75.3$ W	$P_{out}=74.8$ W			
	$P_{in}=4.7$ W	$P_{in}=4.9$ W	$P_{in}=4.9$ W			
	$\eta=2.7\%$	$\eta=2.6\%$	$\eta=2.5\%$			

The results, in terms of wavelength and slope efficiency, are very similar for different  $R_{oc}$  in KLuW and KYW. However, the best efficiency for KLuW was achieved with  $T_{oc} = 3\%$  and  $R_{oc} = 25$  mm while for KYW the highest efficiency was obtained with  $T_{oc} = 1.5\%$  and  $R_{oc} = 50$  mm. In general, saturation of the output power was observed for pump powers higher than 5 W in the case of (Tm,Yb):KLuW and 4 W for (Tm,Yb):KYW. This effect is mainly due to thermal load because when a chopper is used (duty cycle of 50%) the linear dependence is maintained for higher powers. In any case, no cracking of the crystals was observed. The laser wavelengths were, in general, longer for (Tm,Yb):KLuW than for (Tm,Yb):KYW in agreement with the maximum values of emission cross section for Tm in these hosts, though there was a broader emission in the laser wavelength for  $T_{oc} = 1.5\%$  and  $R_{oc} = 25$  mm for (Tm,Yb):KLuW, and  $T_{oc} = 1.5\%$ ,  $R_{oc} = 50$  mm for (Tm,Yb):KYW.

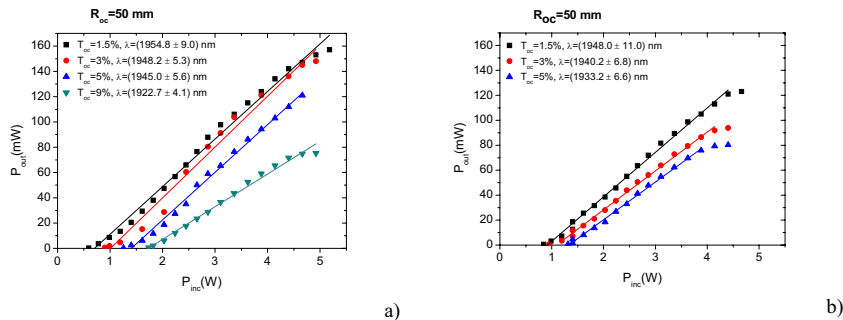


Figure 2. CW laser operation of 2.5% Tm, 5% Yb in a) KLuW, and b) KYW with several output couplers and  $R_{oc} = 50$  mm.

The very low efficiencies were produced from the non-optimized mode matching of the pump and resonator modes. In the work of Batay et al. [9] a beam spot of 80  $\mu\text{m}$  has been used to obtain an efficiency of 19% with respect to the absorbed power. Here we estimate an absorption of 75% and the spot size was 200  $\mu\text{m}$ , so that the bleaching conditions are different and cannot be compared. Moreover, using high power levels the crystal is more likely to suffer thermal effects, as can be seen in figure 2(b) where the thermal effects are characterized by the saturation of the power. This could be avoided with either a reduction of the Yb doping level or an increase of the Tm doping level to optimize the energy transfer from Yb to Tm.

The high laser thresholds are ascribed to the non-perfect resonant energy transfer from Yb to Tm and to the 3-level nature of the Tm ion with partial population of the ground at room temperature. The maximum power for (Tm,Yb):KLuW was 157 mW achieved with  $T_{oc}=1.5\%$  and  $R_{oc}=50$  mm, while for (Tm,Yb):KYW was 123 mW with the same  $T_{oc}$  and  $R_{oc}$ . The latter value is twice than that obtained in [9] also for (Tm,Yb):KYW with an optimum doping level, 6 at. % Tm, 5 at. % Yb, the doping concentration in our case is 2.5 at. % Tm, 5 at. % Yb. In [9], the authors established that low doping levels of Tm, like 3 at. % did not generate laser radiation, probably due to the non-effective cross relaxation mechanism.

#### 4. Conclusions

In summary, we have analysed the laser operation around 2  $\mu\text{m}$  from the  ${}^3F_4 \rightarrow {}^3H_4$  transition, under diode pumping at 980 nm, obtained in 2.5 at. % Tm, 5 at. % Yb codoped system in two similar hosts: KLuW and KYW. We conclude that the doping level of 2.5 at. % Tm is not enough to populate efficiently the  ${}^3F_4$  energy level of Tm via energy transfer of Yb, this would mean that even higher doping levels of Tm in (Tm,Yb):KLuW could increase the results enhancing the cross relaxation process of Tm.

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## References

- [1] A. Godard, *Infrared (2–12  $\mu\text{m}$ ) solid-state laser sources: a review*, C. R. Physique **8**, 1100–1128 (2007).
- [2] E. C. Honea, R. J. Beach, S. B. Sutton, J. A. Speth, S. C. Mitchell, J. A. Skidmore, M. A. Emanuel, and St. A. Payne, *115-W Tm:YAG Diode-Pumped Solid-State Laser*, IEEE J. Quantum Electron. **33**, 1592–1600 (1997).
- [3] X.M. Duan, B.Q. Yao, Y.J. Zhang, C.W. Song, L.L. Zheng, Y.L. Ju, and Y.Z. Wang, *Diode-pumped high efficient Tm:YLF laser output at 1908 nm with near-diffraction limited beam quality*, Laser Phys. Lett. **5**, 347–349 (2008).
- [4] C. Hauglie-Hanssen and N. Djieu, *Further investigations of a 2- $\mu\text{m}$  Tm:YVO<sub>4</sub> laser* IEEE J. Quantum Electron., **30**, 275–279 (1994).
- [5] V. Petrov, F. Güell, J. Massons, J. Gavalda, R. M. Solé, M. Aguiló, F. Díaz, and U. Griebner, *Efficient tunable laser operation of Tm:KGd(WO<sub>4</sub>)<sub>2</sub> in the continuous-wave regime at room temperature*, IEEE J. Quantum Electron., **40**, 1244–1251 (2004).
- [6] X. Mateos, V. Petrov, J. Liu, M.C. Pujol, U. Griebner, M. Aguiló, F. Díaz, M. Galan, and G. Viera, *Efficient 2- $\mu\text{m}$  Continuous wave laser oscillation of Tm<sup>3+</sup>:KLu(WO<sub>4</sub>)<sub>2</sub>*, IEEE J. Quantum Electron., **42**, 1008–1015 (2006).
- [7] Yoh Mita, Takeshi Ide, Masahiro Togashi, and Hajime Yamamoto, *Energy transfer processes in Yb<sup>3+</sup> and Tm<sup>3+</sup> ion-doped fluoride crystals*, J. Appl. Phys., **85**, 4160–4164, (1999).
- [8] P. S. F. de Matos, N. U. Wetter, L. Gomes, I. M. Ranieri and S. L. Baldochi, *A high power 2.3  $\mu\text{m}$  Yb:YLF laser diode-pumped simultaneously at 685 and 960 nm*, J. Opt. A: Pure Appl. Opt. **10** 104009 (2008).
- [9] L.E. Batay, A.A. Demidovich, A.N. Kuzmin, A.N. Titov, M. Mond and S. Kück, *Efficient tunable laser operation of diode-pumped Yb,Tm:KY(WO<sub>4</sub>)<sub>2</sub> around 1.9  $\mu\text{m}$* , Appl. Phys. B, **75**, 457–461 (2002).
- [10] X. Mateos, R. Solé, Jna. Gavalda, M. Aguiló, J. Massons, F. Díaz, *Crystal growth, optical and spectroscopic characterisation of monoclinic KY(WO<sub>4</sub>)<sub>2</sub> co-doped with Er<sup>3+</sup> and Yb<sup>3+</sup>* Opt. Mat., **28**, 423–431 (2006)
- [11] V. Petrov, M. C. Pujol, X. Mateos, O. Silvestre, S. Rivier, M. Aguiló, R. M. Solé, J. Liu, U. Griebner, and F. Díaz, *Growth and properties of KLu(WO<sub>4</sub>)<sub>2</sub>, and novel ytterbium and thulium lasers based on this monoclinic crystalline host* Laser & Photon. Rev., **1**, 179–212 (2007).
- [12] M. Eichhorn, *Quasi-three-level solid-state lasers in the near and mid infrared based on trivalent rare earth ions* Appl. Phys. B, **93**, 269–316 (2008).
- [13] V. E. Kisel, A. E. Troshin, N. A. Tolstik, V. G. Shcherbitsky, N. V. Kuleshov, V. N. Matrosov, T. A. Matrosova, and M. I. Kupchenko, *Spectroscopy and continuous-wave diode-pumped laser action of Yb<sup>3+</sup>:YVO<sub>4</sub>* Opt. Lett., **29**, 2491–2493 (2004).
- [14] A.E. Troshin, V. E. Kisel, A.S. Yasukevich, N.V. Kuleshov, A.A. Pavlyuk, E.B. Dunina and A.A. Kornienko, *Spectroscopy and laser properties of Tm<sup>3+</sup>:KY(WO<sub>4</sub>)<sub>2</sub> crystal*, Appl. Phys. B, **86**, 287–292 (2007).
- [15] S.V. Borisov and R.F. Kletsova, *Crystal structure of KY(WO<sub>4</sub>)<sub>2</sub>*, Sov. Phys. Crystallogr. **13**, 420–421 (1968).
- [16] 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(WO<sub>4</sub>)<sub>2</sub>*, J. Appl. Cryst., **39**, 230–236 (2006).
- [17] R. Sole, V. Nikolov, X. Ruiz, Jna. Gavalda, X. Solans, M. Aguiló, and F. Díaz, *Growth of  $\beta$ -KGd<sub>1-x</sub>Nd<sub>x</sub>(WO<sub>4</sub>)<sub>2</sub> single crystals in K<sub>2</sub>W<sub>2</sub>O<sub>7</sub> solvents*, J. Cryst. Growth, **169**, 600–603 (1996).