



**HAL**  
open science

## Dual-wavelength-pumping of mid-infrared Tm:YLF laser at 2.3 $\mu\text{m}$ : demonstration of pump seeding and recycling processes

Hippolyte Dupont, Lauren Guillemot, Pavel Loiko, Alain Braud, Jean-Louis Doualan, Patrice Camy, Patrick Georges, Frédéric Druon

### ► To cite this version:

Hippolyte Dupont, Lauren Guillemot, Pavel Loiko, Alain Braud, Jean-Louis Doualan, et al.. Dual-wavelength-pumping of mid-infrared Tm:YLF laser at 2.3  $\mu\text{m}$ : demonstration of pump seeding and recycling processes. *Optics Express*, 2022, 30 (18), pp.32141. 10.1364/OE.468695 . hal-03798899

**HAL Id: hal-03798899**

**<https://hal-iogs.archives-ouvertes.fr/hal-03798899v1>**

Submitted on 5 Oct 2022

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

# Dual-wavelength-pumping of mid-infrared Tm:YLF laser at 2.3 $\mu\text{m}$ : demonstration of pump seeding and recycling processes

HIPPOLYTE DUPONT,<sup>1</sup> LAUREN GUILLEMOT,<sup>2</sup> PAVEL LOIKO,<sup>2</sup> ALAIN BRAUD,<sup>2</sup> JEAN-LOUIS DOULAN,<sup>2</sup> PATRICE CAMY,<sup>2</sup> PATRICK GEORGES,<sup>1</sup> AND FRÉDÉRIC DRUON<sup>1,\*</sup>

<sup>1</sup>Université Paris-Saclay, Institut d'Optique Graduate School, CNRS, Laboratoire Charles Fabry, 91127 Palaiseau, France

<sup>2</sup>Centre de Recherche sur les Ions, les Matériaux et la Photonique (CIMAP), UMR 6252 CEA-CNRS-ENSICAEN, Université de Caen, 6 Boulevard Maréchal Juin, 14050 Caen Cedex 4, France

\*frederic.druon@institutoptique.fr

**Abstract:** Upconversion pumping of thulium lasers emitting around 2.3  $\mu\text{m}$  (the  $^3\text{H}_4 \rightarrow ^3\text{H}_5$  transition) has recently attracted a lot of attention as it is compatible with the mature Yb-laser technology. To explore this possibility, we built a mid-infrared Tm:LiYF<sub>4</sub> laser pumped by an Yb:CaF<sub>2</sub> laser at 1.05  $\mu\text{m}$  delivering an output power of 110 mW at 2.31  $\mu\text{m}$  for a maximum incident pump power of 2.0 W. A strong absorption issue appeared in the Tm laser: the slope efficiency vs. the incident pump power was 7.6% while that vs. the absorbed pump power reached 29%. To overcome this issue, a dual-wavelength pumping at 0.78  $\mu\text{m}$  and 1.05  $\mu\text{m}$  was explored (combining both the direct and upconversion pumping schemes). The reciprocal interplay between the two pumps was studied to evaluate their benefits in terms of the pump absorption and laser efficiency. We observed an interesting decrease of the laser threshold for upconversion pumping when adding a small fraction of the direct pump revealing a seeding effect for the excited-state absorption from the metastable  $^3\text{F}_4$  level. A recycling process of this manifold by excited-state absorption in the  $^3\text{F}_4 \rightarrow ^3\text{F}_{2,3}$  loop was also observed. The pump absorption seeding is a viable route for the development of low-threshold upconversion pumped thulium lasers.

© 2022 Optical Society of America under the terms of the [OSA Open Access Publishing Agreement](#)

## 1. Introduction

Mid-infrared (MIR) lasers emitting at the wavelengths around 2.3  $\mu\text{m}$  (falling into the 2.0–2.4  $\mu\text{m}$  atmospheric window, the K band) are of practical importance for spectroscopy of various atmospheric and biological species such as HF, CO, CH<sub>4</sub>, H<sub>2</sub>CO and C<sub>6</sub>H<sub>12</sub>O<sub>6</sub> leading to applications in the atmosphere gas sensing and pollutant detection, combustion studies and non-invasive glucose blood measurements [1,2]. Such laser sources are also interesting for pumping of mid-infrared optical parametric oscillators (OPOs) [3].

There exist several approaches to address this spectral range, namely by using lasers based on zinc chalcogenide crystals (ZnS, ZnSe) doped with Cr<sup>2+</sup> ions [4]; semiconductor (GaInAs / InP or GaInAsSb / GaSb) Vertical-Cavity Surface-Emitting Lasers (VCSELs) [5]; Raman-shifted pulsed Tm or Ho lasers with a fundamental emission at 2  $\mu\text{m}$  [6] or (iv) OPOs [3]. Finally, the thulium ions (Tm<sup>3+</sup>) themselves can provide a direct generation of the 2.3  $\mu\text{m}$  radiation [7,8] according to the  $^3\text{H}_4 \rightarrow ^3\text{H}_5$  electronic transition, Fig. 1(a). Note that Tm<sup>3+</sup> ions can be efficiently pumped at 0.78-0.8  $\mu\text{m}$  (directly to the  $^3\text{H}_4$  state) using commercial high-power AlGaAs laser diodes [9]. The main advantages of 2.3  $\mu\text{m}$  bulk Tm lasers are the developed growth technology of high optical quality Tm<sup>3+</sup>-doped fluoride and oxide crystals, the availability of cheap ; simple and high-power pump sources (e.g., fiber-coupled AlGaAs diode lasers and Yb fiber lasers, see below) ; direct laser emission at 2.2-2.4  $\mu\text{m}$  avoiding any

46 frequency conversion steps ; the possibility to operate both in the continuous-wave and pulsed  
47 regimes and high quality of the laser beam. In this way, Tm lasers operating on the  ${}^3\text{H}_4 \rightarrow {}^3\text{H}_5$   
48 transition may represent a simple and cheap alternative to  $\text{Cr}^{2+}$ -ion-based lasers at the expense  
49 of much narrower gain bandwidth. The emission range of such Tm lasers is also filling the gap  
50 between other direct emission of rare-earth-ions in the near-mid-IR, namely  $\text{Ho}^{3+}$  ( ${}^5\text{I}_7 \rightarrow {}^5\text{I}_8$ ,  
51  $\sim 2.1 \mu\text{m}$ ) and  $\text{Er}^{3+}$  ( ${}^4\text{I}_{11/2} \rightarrow {}^4\text{I}_{13/2}$ ,  $\sim 2.8 \mu\text{m}$ ).

52 During the last years, a great progress in developing continuous-wave (CW) [10-14] and  
53 mode-locked (ML) [15,16] Tm lasers operating on the  ${}^3\text{H}_4 \rightarrow {}^3\text{H}_5$  transition has been  
54 demonstrated using various (both fluoride and oxide) laser crystals. Guillemot *et al.* reported  
55 on a CW Tm:KY<sub>3</sub>F<sub>10</sub> laser directly pumped at 0.78  $\mu\text{m}$  and generating 0.84 W at 2.34  $\mu\text{m}$  [12].  
56 Loiko *et al.* showed that the slope efficiency of such a laser may exceed the Stokes limit owing  
57 to the efficient energy-transfer upconversion (ETU) process for adjacent Tm<sup>3+</sup> ions,  ${}^3\text{F}_4 + {}^3\text{F}_4$   
58  $\rightarrow {}^3\text{H}_6 + {}^3\text{H}_4$ , Fig. 1(a), refilling the upper laser manifold [10]. Soulard *et al.* reported on a  
59 SESAM ML Tm:LiYF<sub>4</sub> laser delivering 94 ps pulses at 2.31  $\mu\text{m}$  [15]. Canbaz *et al.* achieved  
60 femtosecond pulses (514 fs) from a similar laser using the Kerr-lens mode-locking technique  
61 [16]. Note that both ML lasers were pumped by a Ti:Sapphire laser at 0.78  $\mu\text{m}$ .

62 Later, alternative upconversion (UC) pumping schemes at 1  $\mu\text{m}$  and 1.5  $\mu\text{m}$  were proposed  
63 for such lasers [17,18]. Among them, the first one is more attractive as it is compatible with the  
64 mature Yb-bulk and Yb-fiber laser technologies. It is based on a weak (non-resonant) ground-  
65 state absorption (GSA) corresponding to the short-wave vibronic sideband of the  ${}^3\text{H}_6 \rightarrow {}^3\text{H}_5$   
66 Tm<sup>3+</sup> absorption band and a strong (resonant) excited-state absorption (ESA) from the  
67 metastable Tm<sup>3+</sup> state,  ${}^3\text{F}_4 \rightarrow {}^3\text{F}_{2,3}$ , Fig. 1(a). The population of the intermediate level  ${}^3\text{F}_4$  is  
68 mainly provided by the photon avalanche effect [19] relying on both the above-mentioned  
69 resonant ESA channel and the well-known cross-relaxation (CR) process for Tm<sup>3+</sup> ions,  ${}^3\text{H}_4 +$   
70  ${}^3\text{H}_6 \rightarrow {}^3\text{F}_4 + {}^3\text{F}_4$ . Note that due to the non-resonant nature of the GSA transition, the population  
71 of the  ${}^3\text{F}_4$  state via the non-radiative path from the short-living  ${}^3\text{H}_5$  state is almost negligible.  
72 The photon avalanche process recycles the populations of Tm<sup>3+</sup> multiplets in favor of the  ${}^3\text{H}_4$ ,  
73 and  ${}^3\text{F}_4$  ones acting as the upper laser level and the “effective” ground-state, respectively.  
74 Tyazhev *et al.* reported on a watt-level Tm fluoride fiber laser generating 1.24 W at 2.28  $\mu\text{m}$   
75 with a slope efficiency of 37% based on a single-wavelength UC pumping at 1.05  $\mu\text{m}$  using an  
76 Yb-fiber laser [20].

77 So far, the main disadvantages of the UC pumping scheme for bulk MIR Tm lasers are the  
78 limited pump absorption and relatively high laser threshold. This is explained by the need of  
79 populating the intermediate  ${}^3\text{F}_4$  Tm<sup>3+</sup> state via the photon avalanche mechanism which becomes  
80 efficient only at high pump intensities. It would be attractive to combine both pump schemes  
81 (direct and UC pumping) to improve these performances. Wang *et al.* studied dual-wavelength  
82 pumping at 0.79 and 1.47  $\mu\text{m}$ : a diode-pumped Tm:LiYF<sub>4</sub> laser delivered 1.8 W at 2.31  $\mu\text{m}$  for  
83 a total incident pump power of 70 W [21]. Besides the low optical-to-optical efficiency of only  
84 2.5%, the output power of the Tm laser saturated while increasing the pump power at 1.47  $\mu\text{m}$   
85 and no lasing under single UC pumping was achieved. Thus, the potential of dual-wavelength  
86 pumping was not clearly revealed.

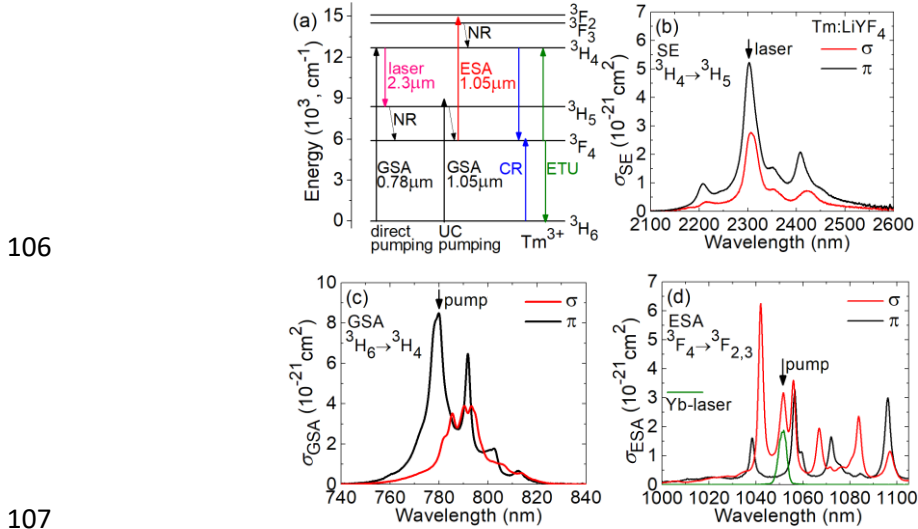
87 In the present work, we report on a mid-infrared thulium laser operating on the  ${}^3\text{H}_4 \rightarrow {}^3\text{H}_5$   
88 transition with a dual-wavelength (direct and upconversion) pumping at 0.78 and 1.05  $\mu\text{m}$  (i.e.,  
89 relying on the Yb-laser technology). As a gain material, we have selected Tm<sup>3+</sup>-doped lithium  
90 yttrium fluoride crystal (LiYF<sub>4</sub>) which currently appears as a state-of-the-art material for this  
91 type of lasers.

## 92 2. Experimental

### 93 2.1 Spectroscopy of Tm:LiYF<sub>4</sub>: An overview

94 Prior to the laser experiments, let us briefly describe the relevant spectroscopic parameters of  
95 Tm<sup>3+</sup> ions in the LiYF<sub>4</sub> crystal. The stimulated-emission cross-section for the  ${}^3\text{H}_4 \rightarrow {}^3\text{H}_5$

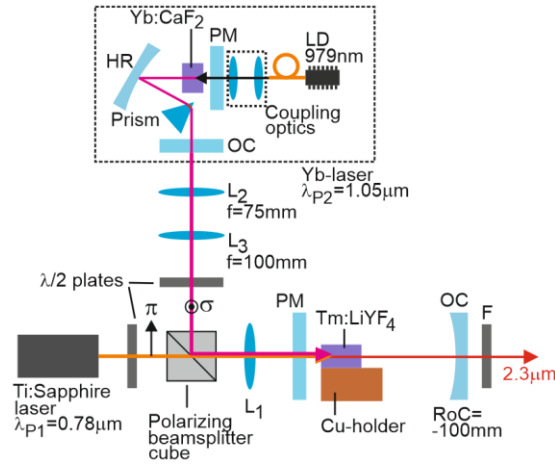
96 transition reaches  $\sigma_{SE} = 0.52 \times 10^{-20} \text{ cm}^2$  at 2303 nm for  $\pi$ -polarization, Fig. 1(b). The intrinsic  
 97 lifetime of the upper laser level  ${}^3\text{H}_4$   $\tau_{lum,0} = 2.3 \text{ ms}$  and for 3.5 at.% Tm doping, the self-  
 98 quenching via CR leads to a reduced value of  $\tau_{lum} \sim 155 \mu\text{s}$  [10]. For the  ${}^3\text{H}_6 \rightarrow {}^3\text{H}_4$  GSA  
 99 transition, the peak cross-section  $\sigma_{GSA} = 0.85 \times 10^{-20} \text{ cm}^2$  at 779.9 nm for  $\pi$ -polarization  
 100 (bandwidth at FWHM: 7.6 nm), see Fig. 1(c). For the  ${}^3\text{F}_4 \rightarrow {}^3\text{F}_{2,3}$  ESA transition, the peak  
 101 cross-section  $\sigma_{ESA} = 0.63 \times 10^{-20} \text{ cm}^2$  at 1042.0 nm and at longer wavelengths, another less  
 102 intense peak appears with  $\sigma_{ESA} = 0.32 \times 10^{-20} \text{ cm}^2$  at 1051.6 nm (for  $\sigma$ -polarization), Fig. 1(d).  
 103 The FWHMs of these peaks are 2.5 nm and 4.3 nm, respectively. The  ${}^3\text{H}_6 \rightarrow {}^3\text{H}_5$  GSA is weak  
 104 between 1000 nm and 1100 nm ( $\sigma_{GSA} \approx 0.01 \times 10^{-20} \text{ cm}^2$  in this region) since it is located in the  
 105 tail of the peak at 1.20  $\mu\text{m}$  (for  $\pi$ -polarization) that has a value of  $\sigma_{ESA} = 0.62 \times 10^{-20} \text{ cm}^2$ .



108 **Fig. 1.** (a) The scheme of energy levels of thulium ions relevant for MIR laser operation: GSA and ESA – ground- and excited-state absorption, respectively, NR – non-radiative relaxation,  
 109 CR – cross-relaxation, ETU – energy-transfer upconversion; (b-d) Spectroscopy of  $\text{Tm}^{3+}$  ions in  $\text{LiYF}_4$ : (b) simulated-emission (SE) cross-sections,  $\sigma_{SE}$ , the  ${}^3\text{H}_4 \rightarrow {}^3\text{H}_5$  transition; (c) GSA cross-  
 110 sections,  $\sigma_{GSA}$ , the  ${}^3\text{H}_6 \rightarrow {}^3\text{H}_4$  transition; (d) ESA cross-sections,  $\sigma_{ESA}$ , the  ${}^3\text{F}_4 \rightarrow {}^3\text{F}_{2,3}$  transition.  
 111 The light polarizations:  $\pi$  and  $\sigma$ . In (d), the spectrum of the Yb:CaF<sub>2</sub> laser is shown for  
 112 comparison.  
 113  
 114

## 115 2.2 Laser set-up

116 The scheme of the dual-wavelength-pumped MIR thulium laser is shown in Fig. 2. As a gain  
 117 medium, we use an  $\alpha$ -cut 3.5 at.% Tm:LiYF<sub>4</sub> crystal ( $\Phi=8.5 \text{ mm}$ , thickness=8.14 mm). Its faces  
 118 are polished to laser-quality with good parallelism and left uncoated. The crystal is glued to a  
 119 Cu-holder and it is passively cooled. A hemispherical laser cavity is composed of a flat pump  
 120 mirror (PM) coated for high transmission (HT) at the pump wavelengths ( $T = 95\%$  at 1.05  $\mu\text{m}$   
 121 and 0.78  $\mu\text{m}$ ) and high reflection (HR,  $R > 99.9\%$ ) at 2.3  $\mu\text{m}$ , and a concave (radius of curvature:  
 122 RoC = -100 mm) output coupler (OC) having a transmission at the laser wavelength  $T_{OC} = 2\%$ .  
 123 The OC also provides HT at 1.9  $\mu\text{m}$  to suppress the laser on the  ${}^3\text{F}_4 \rightarrow {}^3\text{H}_6$  transition. The  
 124 geometrical cavity length is 99 mm.



**Fig. 2.** Set-up of the dual-wavelength-pumped MIR thulium laser: PM – pump mirror, OC – output coupler, HR – highly-reflective mirror, L<sub>1</sub>, L<sub>2</sub>, L<sub>3</sub> – aspherical lenses, λ/2 – half-wave plates, LD – laser diode, F – band-pass filter.

125  
126  
127  
128

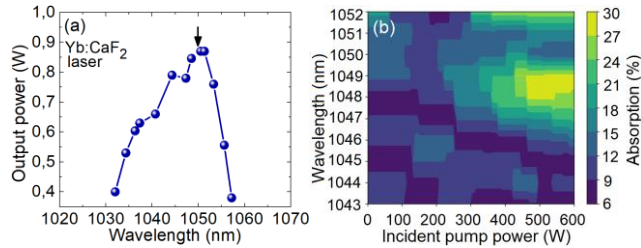
129 Two pump sources are used. The first one addresses the  ${}^3\text{H}_6 \rightarrow {}^3\text{H}_4$  GSA transition (the  
130 direct pumping). It is a Ti:Sapphire laser (Spectra Physics, model 3900J) delivering up to 3.0  
131 W at  $\lambda_{\text{p,GSA}} = 780$  nm (linewidth at FWHM: 0.1 nm) with a beam quality factor  $M^2 \approx 1$  and a  
132 linear polarization aligned to correspond to  $\pi$  in the Tm-crystal. Its output is focused into the  
133 laser crystal using a lens (L<sub>1</sub>). Several L<sub>1</sub> focusing lenses with a focal length in the range of 50  
134 – 150 mm are tested. For  $f = 150$  mm (the optimum one for single wavelength pumping at  $\lambda_{\text{p,GSA}}$   
135 = 0.78  $\mu\text{m}$ ), the measured pump beam waist  $2w_{\text{p}}$  is  $61 \pm 5$   $\mu\text{m}$ .

136 The second pump source is designed to address the  ${}^3\text{F}_4 \rightarrow {}^3\text{F}_{2,3}$  ESA transition (the UC  
137 pumping scheme). A home-made tunable Ytterbium-bulk laser is based on a 2 at.% Yb:CaF<sub>2</sub>  
138 crystal pumped by a fiber-coupled InGaAs laser diode at 979 nm. A V-shaped laser cavity is  
139 composed of a flat pump mirror, a concave (RoC = -200 mm) HR mirror and a plane OC ( $T_{\text{OC}}$   
140 = 5%). The wavelength tuning is realized by inserting an SF10 prism close to the OC. The  
141 maximum of the output power of the tunable Yb:CaF<sub>2</sub> laser is observed at  $\sim 1.05$   $\mu\text{m}$ , thus, we  
142 select the thulium ESA peak at 1051.6 nm for UC pumping, cf. Fig. 1(b), even though another  
143 ESA peak at 1042.0 nm is more intense. The tunable Yb-laser delivers up to 1 W at  $\lambda_{\text{p,UC}} =$   
144 1050 nm (linewidth:  $\sim 3$  nm) with a linear polarization corresponding to  $\sigma$  in the Tm-crystal.  
145 Without the prism, in the free-running regime, the Yb-laser can be scaled up to 2 W (after the  
146 polarizer Fig. 2) at a central wavelength of 1050 nm. The size of the pump beam from the Yb-  
147 laser is adjusted using an afocal telescope system composed of two lenses (L<sub>2</sub>:  $f = 75$  mm, L<sub>3</sub>:  
148  $f = 100$  mm) and the polarization state – using a half-wave plate. For this pump source, the  
149 measured pump spot size in the crystal  $2w_{\text{p}}$  is  $30 \pm 5$   $\mu\text{m}$  (for L<sub>1</sub> lens with a focal length of  $f =$   
150 50 mm, being the optimum one for pure UC pumping). The two pump beams are combined  
151 using a polarizing beam-splitter cube (Thorlabs PBS252). The pumping is in double-pass  
152 because of the partial reflectivity of the OC at the pump wavelengths ( $R_{0.78\mu\text{m}} = 64\%$  and  $R_{1.05\mu\text{m}}$   
153 = 28%). The residual (non-absorbed) pump after the OC is filtered out using a band-pass filter  
154 (Thorlabs, FB2250-50).

### 155 3. Results and discussion

156 The wavelength of the Yb:CaF<sub>2</sub> laser is continuously tuned from 1032 to 1054 nm, Fig. 3(a).  
157 To justify the selection of the pump wavelength, we have studied the single-pass pump  
158 absorption as a function of  $\lambda_{\text{p,UC}}$  and the incident pump power, Fig. 3(b). The highest pump  
159 absorption is observed at 1050 nm in agreement with the ESA spectra of Tm<sup>3+</sup> ions in LiYF<sub>4</sub>  
160 for  $\sigma$ -polarization, cf. Fig. 1(d). A certain pump level is needed to reach a kind of plateau for

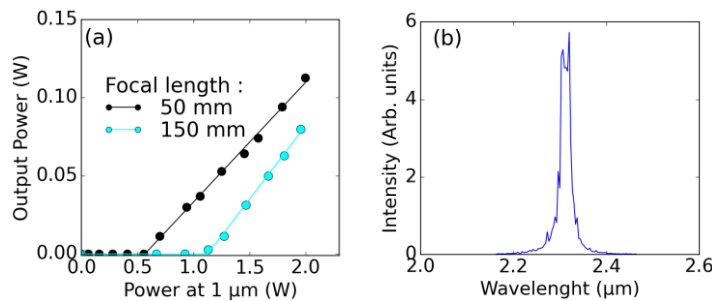
161 pump absorption (~30%). Thus, tuning the pump wavelength to match precisely one of the ESA  
 162 peaks is indeed critical as otherwise we are not able to reach reasonably high pump absorption  
 163 even for high pump levels.



164  
 165 **Fig. 3.** (a) Wavelength tuning curve of the Yb:CaF<sub>2</sub> laser, *arrow* – the selected laser wavelength;  
 166 (b) A 2D plot of single-pass pump absorption in the Tm:LiYF<sub>4</sub> crystal (σ-polarized pump) vs.  
 167 the pump wavelength and the incident pump power.

168 Due to the different physical processes involved in the pump absorption between the cases  
 169 of direct pumping (the GSA  ${}^3\text{H}_6 \rightarrow {}^3\text{H}_4$  transition following a classical Beer-Lambert absorption  
 170 law) and UC pumping (the ESA  ${}^3\text{F}_4 \rightarrow {}^3\text{F}_{2,3}$  transition based on a photon avalanche effect), it  
 171 is found that different focal lengths of the L<sub>1</sub> focusing lens lead to optimum laser performances  
 172 in these two cases. Indeed, shorter focal length ( $f = 50$  mm) is preferable for pure UC pumping  
 173 as it leads to higher incident pump intensity and, thus, stronger photon avalanche effect and  
 174 higher pump absorption. In contrast, for pure direct pumping, an L<sub>1</sub> lens with  $f = 150$  mm  
 175 provides better mode matching with a relatively good absorption efficiency since the absorption  
 176 coefficient is independent of the pump intensity.

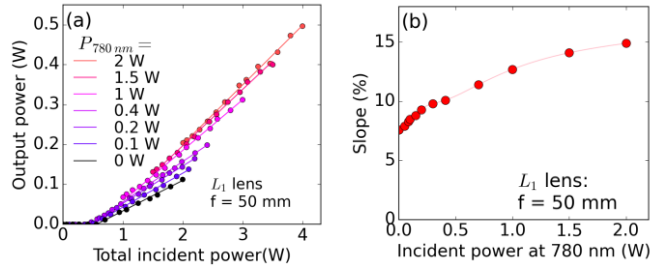
177 First, we study single-wavelength UC pumping. As explained above, an L<sub>1</sub> lens with a focal  
 178 length of  $f = 50$  mm is used, Fig. 4(a). Pumping at  $\lambda_{\text{p,UC}} = 1.05$  μm (the  ${}^3\text{F}_4 \rightarrow {}^3\text{F}_{2,3}$  ESA  
 179 transition), the output power of the MIR Tm-laser reaches 110 mW at 2.31 μm with a slope  
 180 efficiency  $\eta$  of 7.6% (vs. the incident pump power  $P_{1.05\mu\text{m}}$ ) and a laser threshold of 0.51 W.  
 181 However, the slope efficiency vs. the absorbed pump power is relatively high, 29%, which then  
 182 rises the interest of using the Yb-laser technology for pumping Tm<sup>3+</sup>-doped materials emitting  
 183 at 2.3 μm. A typical spectrum of the laser emission is shown in Fig. 4(b). The laser emitted  
 184 linearly polarized radiation ( $\pi$ ) and its polarization state is naturally selected by the anisotropy  
 185 of the gain medium. To illustrate the superiority of the short focal length L<sub>1</sub> lens for pure UC  
 186 pumping, both the input-output dependences of the Tm-laser for  $f = 50$  mm and  $f = 150$  mm  
 187 are given in Fig. 4(a). Using the latter lens, the maximum output power of the MIR Tm-laser  
 188 was only 80 mW corresponding to an increased threshold of 1.2 W.  
 189



190  
 191 **Fig. 4.** Tm:LiYF<sub>4</sub> laser emitting at 2.3 μm with pure UC pumping by an Yb:CaF<sub>2</sub> laser emitting  
 192 at 1050 nm: (a) input-output dependences for different focal lengths of the focusing lens (L<sub>1</sub>):  $f$   
 193 = 50 mm and  $f = 150$  mm; (b) a typical spectrum of laser emission.

194  
 195 The dual-wavelength pumping is studied by fixing the power level of one pump source and

196 varying that of the second one. The laser output powers are plotted versus the total incident  
 197 pump power,  $P_{\Sigma} = P_{0.78\mu\text{m}} + P_{1.05\mu\text{m}}$  and the slope efficiency and the laser threshold are  
 198 determined with respect to  $P_{\Sigma}$  – for fair comparison. To assist UC pumping, the  $P_{0.78\mu\text{m}}$  value is  
 199 fixed at several different levels and  $P_{1.05\mu\text{m}}$  is varied, as shown in Fig. 5(a). With increasing the  
 200 added  $P_{0.78\mu\text{m}}$  power up to 2.0 W, both the maximum output power of the Tm-laser and its slope  
 201 efficiency  $\eta$  gradually increase reaching 500 mW and 15.0%, respectively, Fig. 5(a,b).  
 202



**Fig. 5.** Dual-wavelength-pumped MIR Tm:LiYF<sub>4</sub> laser,  $P_{0.78\mu\text{m}}$  – fixed,  $P_{1.05\mu\text{m}}$  – varied: (a) input-output dependences,  $L_1$  focusing lens:  $f = 50$  mm; (b) laser slope efficiencies vs. the total incident pump power ( $P_{\Sigma}$ ) plotted against the added  $P_{0.78\mu\text{m}}$  pump power

203

204

205

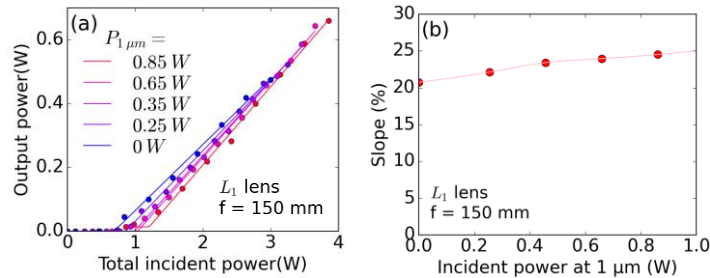
206

207 In all the experiments, the Tm-laser operates solely on the  ${}^3\text{H}_4 \rightarrow {}^3\text{H}_5$  transition and no laser  
 208 at  $1.9\mu\text{m}$  is observed.

209

210 For completeness, we have also studied pure direct pumping using an  $L_1$  lens with a focal  
 211 length  $f = 150$  mm. As the goal of the present work is the observation of the mutual interplay  
 212 between the two pumps, the laser cavity and the pump focusing optics are not separately  
 213 optimized for direct pumping as in the previous studies [10,11]. Pumping at  $\lambda_{\text{P,GSA}} = 0.78\mu\text{m}$   
 214 (the  ${}^3\text{H}_6 \rightarrow {}^3\text{H}_4$  GSA transition), the MIR Tm-laser generates a maximum output power of 480  
 215 mW at  $2.31\mu\text{m}$  with a slope efficiency  $\eta$  of 20.9% (vs. the incident pump power  $P_{0.78\mu\text{m}}$ ) and a  
 216 laser threshold of 0.47 W, Fig. 6(a). Then, we have again implemented dual-wavelength  
 217 pumping: the added  $P_{1.05\mu\text{m}}$  pump power is fixed at different levels and  $P_{0.78\mu\text{m}}$  varies, Fig. 6(a).  
 218 With a maximum added  $P_{1.05\mu\text{m}}$  pump power of 0.86 W, the maximum output power of the Tm-  
 219 laser increases to 664 mW, its slope efficiency  $\eta$  increases to 24.5%, whilst the laser threshold  
 220 also increases up to 0.85 W, Fig. 6(b). In this way, an addition of a small fraction of  
 221 upconversion pump to the direct one has much weaker effect on the laser performance.

221



**Fig. 6.** Dual-wavelength-pumped MIR Tm:LiYF<sub>4</sub> laser:  $P_{0.78\mu\text{m}}$  – varied,  $P_{1.05\mu\text{m}}$  – fixed: (a) input-output dependences; (b) the corresponding laser slope efficiencies plotted against the added  $P_{1.05\mu\text{m}}$  pump power.  $L_1$  lens:  $f = 150$  mm.

222

223

224

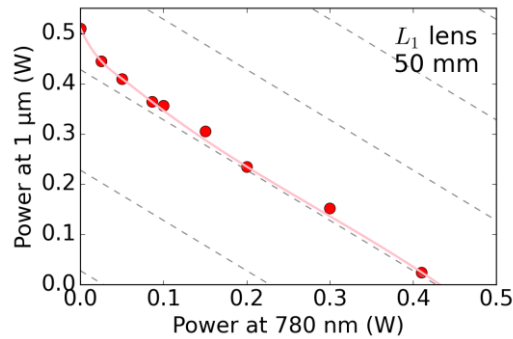
225

226 Let us discuss the observed laser behavior. The pumping at  $0.78\mu\text{m}$  leads to a direct  
 227 excitation of Tm<sup>3+</sup> ions to the upper laser level ( ${}^3\text{F}_4$ ). It can result in an emission of a laser  
 228 photon at  $2.3\mu\text{m}$  followed by a non-radiative (NR) relaxation step to the intermediate

229 metastable level  ${}^3F_4$  or in its population via the CR process for adjacent  $Tm^{3+}$  ions,  ${}^3H_4 + {}^3H_6$   
 230  $\rightarrow {}^3F_4 + {}^3F_4$ . Thus, for direct pumping, the metastable  ${}^3F_4$   $Tm^{3+}$  state accumulates the electronic  
 231 excitations. The UC pumping at  $1.05\ \mu m$  is based on a weak (non-resonant) GSA,  ${}^3H_6 \rightarrow {}^3H_5$ ,  
 232 followed by a NR relaxation step to the  ${}^3F_4$  state, and a resonant ESA,  ${}^3F_4 \rightarrow {}^3F_{2,3}$ , again  
 233 followed by a NR relaxation step terminating at the upper laser level,  ${}^3H_4$ . For low incident  
 234 pump intensities, the ESA process is very weak because of the non-resonant GSA channel and,  
 235 thus, negligible population of the metastable  ${}^3F_4$  state. With increasing the incident pump  
 236 intensity at  $1.05\ \mu m$ , both the  ${}^3H_4$  and  ${}^3F_4$  states accumulate electronic excitations owing to the  
 237 photon avalanche mechanism based on a combination of the ESA and CR.

238 The population of the metastable  ${}^3F_4$  state is crucial for boosting the efficiency of the ESA  
 239 process  ${}^3F_4 \rightarrow {}^3F_{2,3}$  which is a prerequisite for efficient laser operation under pure UC pumping  
 240 and the population of this state could be easily increased by direct pumping representing a  
 241 *seeding* effect on the UC pump absorption. It is indeed priming the UC pumping. Thus, dual-  
 242 wavelength pumping relying mainly on the UC pump with a small addition of the direct pump  
 243 could lead to reduced laser thresholds and boosted laser efficiencies of MIR Tm-lasers as  
 244 compared to the case of pure UC pumping.

245 To illustrate this, we have plotted the measured laser thresholds of the dual-wavelength-  
 246 pumped Tm laser for different combinations of the incident pump powers  $P_{0.78\mu m}$  and  $P_{1.05\mu m}$ ,  
 247 Fig. 7. The  $L_1$  lens with a focal length of 50 mm was chosen as it corresponded to the best  
 248 performance in the case of pure UC pumping. In Fig. 7, each point represents a threshold pump  
 249 power,  $P_{th} = P_{0.78\mu m} + P_{1.05\mu m}$ , then showing a combination of both pumps needed to reach the  
 250 laser threshold. The intersections of this curve with the horizontal and vertical axes represent  
 251 the two extreme cases of single-wavelength pumping: pure direct pumping with  $\lambda_{p,GSA} = 0.78$   
 252  $\mu m$  on the abscise axis, and pure UC pumping with  $\lambda_{p,UC} = 1.05\ \mu m$  on the ordinate axis,  
 253 respectively. Compared to single-wavelength UC-pumping, an addition of even a small  
 254 contribution of the direct pump ( $P_{0.78\mu m} < 0.2\ W$ ) helps to notably reduce the laser threshold in  
 255 terms of the total pump power. This is due to the seeding effect of the direct pump on the  
 256 population of the intermediate  ${}^3F_4$  state as explained above. To highlight this effect, we also  
 257 plotted the isopower lines in order to clearly see that this process is more pronounced for a  
 258 small fraction of the direct pump revealing then a seeding effect.



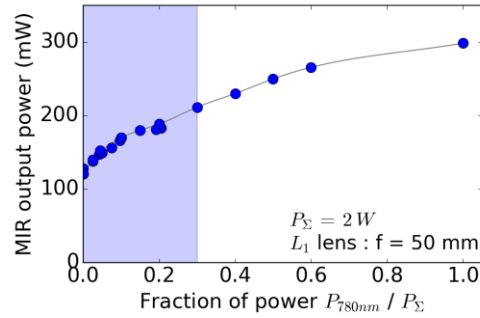
259  
 260  
 261  
 262

**Fig. 7.** Analysis of the threshold behavior of the dual-wavelength pumped MIR Tm:LiYF<sub>4</sub> laser: thresholds for different combinations of the incident pump powers  $P_{0.78\mu m}$  and  $P_{1.05\mu m}$ . Grey lines – isopower lines drawn as guides for eyes.

263 Another way to express the seeding effect of the added direct pump is to monitor the output  
 264 power of the MIR Tm-laser as a function of the ratio between the added  $P_{0.78\mu m}$  power and the  
 265 total incident pump power  $P_{\Sigma}$ , namely  $X = P_{0.78\mu m}/P_{\Sigma}$ . Figure 8 illustrates such analysis for a  
 266 fixed value of  $P_{\Sigma} = 2.0\ W$  and for a focal length for  $L_1$  of  $f = 50\ mm$ . Here, the cases of  $X = 0$   
 267 and  $X = 1$  correspond to pure UC pumping and pure direct pumping, respectively. As can be  
 268 seen from Fig. 8, the main effect of adding the direct pump on the output power is observed for  
 269 small  $X$  values and for  $X > 0.5$ , the dependence saturates, therefore indicating an area of interest



270 towards lower  $X$  values.



271

272

273

274

**Fig. 8.** Output power of the dual-wavelength pumped MIR Tm:LiYF<sub>4</sub> laser as a function of the fraction of the direct pump to the total incident pump power,  $P_{0.78\mu\text{m}}/P_{\Sigma}$ .  $P_{\Sigma} = 2.0$  W,  $L_1$  lens:  $f = 50$  mm. *Blue rectangle:* the zone of interest.

275

276

277

278

279

280

281

282

283

284

285

286

287

288

289

290

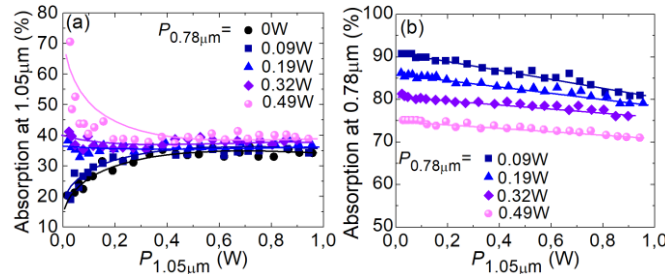
291

292

293

We have studied the total pump absorption in the Tm:LiYF<sub>4</sub> crystal at both wavelengths (0.78  $\mu\text{m}$  and 1.05  $\mu\text{m}$ ) under single- and dual-wavelength pumping and non-lasing conditions, Fig. 9. To assist UC pumping, the added  $P_{0.78\mu\text{m}}$  power is fixed at several different levels and  $P_{1.05\mu\text{m}}$  varies. For this experiment, the focusing lens  $L_1$  with a focal length of  $f = 50$  mm is selected. One can then observe a strong effect of the dual-wavelength pumping on the pump absorption. This effect is also more impacting at lower level of  $\lambda_{\text{P,GSA}} = 0.78$   $\mu\text{m}$

For pure UC pumping, Fig. 9(a), the pump absorption at 1.05  $\mu\text{m}$  increases with the incident pump power and saturates at  $\sim 34\%$  for  $P_{1.05\mu\text{m}} > 0.4$  W. By adding more pump at 0.78  $\mu\text{m}$ , this tendency changes first to almost pump-independent absorption (at  $P_{0.78\mu\text{m}} = 0.19$  W) and then it is even reversed representing an absorption saturation effect. The saturated pump absorption slightly increases with the added  $P_{0.78\mu\text{m}}$  power, from 34% to 40%, as shown in Fig. 9(a), representing a positive effect of the dual-wavelength pumping. These observations are in line with the laser behavior, and they are explained by the increased population of the metastable  $^3\text{F}_4$  state acting as an “effective” ground-state for the  $^3\text{F}_4 \rightarrow ^3\text{F}_{2,3}$  ESA channel. It is for the first time that we observe a *recycling* process of the population of the metastable  $^3\text{F}_4$  state by ESA in the  $^3\text{F}_4 \rightarrow ^3\text{F}_{2,3}$  loop favoring the 2.3  $\mu\text{m}$  laser. Despite the fact that the measurements are performed under non-lasing conditions, as the main effect of the dual-wavelength pumping on the pump absorption is observed below the laser threshold, so that the obtained conclusions held true for the lasing regime as well.



294

295

296

297

**Fig. 9.** Total pump absorption for a dual-wavelength-pumped MIR Tm:LiYF<sub>4</sub> laser under non-lasing conditions measured at: (a)  $\lambda_{\text{P,UC}} = 1.05$   $\mu\text{m}$ , (b)  $\lambda_{\text{P,GSA}} = 0.78$   $\mu\text{m}$ ,  $P_{0.78\mu\text{m}}$  – fixed,  $P_{1.05\mu\text{m}}$  – varied.  $L_1$  lens,  $f = 50$  mm.

298

299

300

301

A very different behavior is observed for the pump absorption at 0.78  $\mu\text{m}$ . With increasing the pump power at 1.05  $\mu\text{m}$ , it decreases marginally due to a slight decrease of the ground-state population since a part of the  $^3\text{F}_4$  population is directly recycled in the upper loop instead of fluorescing back to the ground level. This recycling effect explains the increase of the laser

302 power and slope efficiency, Fig. 6(b), when using dual-pumping despite a lower absorption at  
303 0.78  $\mu\text{m}$ , cf. Fig. 9(b).

304 The observed rapid increase of the pump absorption at 1.05  $\mu\text{m}$  under co-pumping at  
305 0.78  $\mu\text{m}$  (even for relatively low added pump powers) is responsible for the useful decrease of  
306 the laser threshold of the dual-wavelength-pumped Tm:LiYF<sub>4</sub> laser, cf. Fig. 7. It is also partially  
307 responsible of the increase of the laser slope efficiency, as shown in Fig. 5(b). Another possible  
308 reason is the nonlinearity of the input-output dependences of Tm lasers operating on the  $^3\text{H}_4 \rightarrow$   
309  $^3\text{H}_5$  transition [12] due to the enhanced effect of ETU from the metastable  $^3\text{F}_4$  state refilling the  
310 upper laser level.

#### 311 4. Conclusion

312 To conclude, we have studied in detail the dual-wavelength (combining direct and  
313 upconversion) pumping of a Tm:LiYF<sub>4</sub> laser operating on the  $^3\text{H}_4 \rightarrow ^3\text{H}_5$  transition  
314 corresponding to an emission in the MIR, at 2.3  $\mu\text{m}$ . As an UC pump source, we used a Yb:CaF<sub>2</sub>  
315 laser tuned to one of the ESA ( $^3\text{F}_4 \rightarrow ^3\text{F}_{2,3}$ ) peaks of Tm<sup>3+</sup> ions at 1.05  $\mu\text{m}$ . Such fine matching  
316 of the ESA peak was critical to reach a reasonably high pump absorption via the photon  
317 avalanche mechanism. This particular matching between the Yb:CaF<sub>2</sub> pump and Tm:YLF laser  
318 technologies was experimentally demonstrated for the first time, in our best knowledge. For  
319 pure UC pumping, the MIR Tm laser delivered 110 mW at 2.31  $\mu\text{m}$  for 2.0 W of incident pump  
320 power corresponding to a slope efficiencies of 7.6% and 29% versus the incident and absorbed  
321 pump power, respectively. Furthermore, we found that the co-pumping at 0.78  $\mu\text{m}$  helps to  
322 notably reduce the laser threshold due to the seeding effect on the population of the intermediate  
323  $^3\text{F}_4$  Tm<sup>3+</sup> state. We also observed a recycling process of this long-lifetime-manifold population  
324 by resonant ESA that goes in favour of the 2.3  $\mu\text{m}$  laser. Considering the availability of high-  
325 power and high-brightness pump sources at 1  $\mu\text{m}$  based on the Yb-fiber laser technology, and  
326 low- to moderate-power single-mode fiber-coupled 0.8  $\mu\text{m}$  AlGaAs laser diodes, a combination  
327 of these two pump sources could serve as a viable way for the development of MIR Tm lasers,  
328 especially those for which a high-brightness pumping and / or a compatibility with the fiber  
329 laser technology are required, such as fiber and waveguide lasers and bulk femtosecond  
330 oscillators and amplifiers. This is especially relevant for the latter type of laser sources which  
331 are currently pumped by tunable Ti:Sapphire lasers thus strongly limiting their applications.

332 **Funding.** Agence Nationale de la Recherche (ANR-19-CE08-0028, SPLENDID2). “RELANCE” Chair of  
333 Excellence project funded by the Normandy Region.

334 **Disclosures.** The authors declare no conflicts of interest.

335 **Data availability.** Data underlying the results presented in this paper are not publicly available at this time but may  
336 be obtained from the authors upon reasonable request.

#### 337 References

- 338 1. F. J. McAleavey, J. O’Gorman, J. F. Donegan, B. D. MacCraith, J. Hegarty and G. Maze, “Narrow linewidth,  
339 tunable Tm<sup>3+</sup>-doped fluoride fiber laser for optical-based hydrocarbon gas sensing,” *IEEE J. Sel. Top. Quantum*  
340 *Electron.* **3**(4), 1103-1111 (1997).
- 341 2. G. G. Taylor, D. Morozov, N. R. Gemmell, K. Erotokritou, S. Miki, H. Terai, and R. H. Hadfield, “Photon  
342 counting LIDAR at 2.3  $\mu\text{m}$  wavelength with superconducting nanowires,” *Opt. Express* **27**(26), 38147-38158  
343 (2019).
- 344 3. V. Petrov, “Frequency down-conversion of solid-state laser sources to the mid-infrared spectral range using  
345 non-oxide nonlinear crystals,” *Progr. Quantum Electron.* **42**, 1–106 (2015).
- 346 4. I. Moskalev, S. Mirov, M. Mirov, S. Vasilyev, V. Smolski, A. Zakrevskiy, and V. Gapontsev, “Ultrafast  
347 middle-IR lasers and amplifiers based on polycrystalline Cr:ZnS and Cr:ZnSe,” *Opt. Mater. Express* **7**(7), 2636-  
348 2650 (2017).
- 349 5. E. Geerlings, M. Rattunde, J. Schmitz, G. Kaufel, H. Zappe, and J. Wagner, “Widely tunable GaSb-based  
350 external cavity diode laser emitting around 2.3  $\mu\text{m}$ ,” *IEEE Photon. Technol. Lett.* **18**(18), 1913-1915 (2006).
- 351 6. U. Sheintop, D. Sebbag, P. Komm, S. Pearl, G. Marcus, and S. Noach, “Two-wavelength Tm:YLF/KGW  
352 external-cavity Raman laser at 2197 nm and 2263 nm,” *Opt. Express* **27**(12), 17112-17121 (2019).

- 353  
354  
355  
356  
357  
358  
359  
360  
361  
362  
363  
364  
365  
366  
367  
368  
369  
370  
371  
372  
373  
374  
375  
376  
377  
378  
379  
380  
381  
382  
383  
384  
385
7. J. Caird, L. DeShazer, and J. Nella, "Characteristics of room-temperature 2.3- $\mu\text{m}$  laser emission from  $\text{Tm}^{3+}$  in YAG and  $\text{YAlO}_3$ ," *IEEE J. Quantum Electron.* **11**(11), 874-881 (1975).
  8. J. F. Pinto, L. Esterowitz, and G. H. Rosenblatt, " $\text{Tm}^{3+}$ :YLF laser continuously tunable between 2.20 and 2.46  $\mu\text{m}$ ," *Opt. Lett.* **19**(12), 883-885 (1994).
  9. E. Kifle, P. Loiko, L. Guillemot, J.-L. Doualan, F. Starecki, A. Braud, T. Georges, J. Rouvillain, and P. Camy, "Watt-level diode-pumped thulium lasers around 2.3  $\mu\text{m}$ ," *Appl. Opt.* **59**(25), 7530-7539 (2020).
  10. P. Loiko, R. Soulard, L. Guillemot, G. Brasse, J.-L. Doualan, A. Braud, A. Tyazhev, A. Hideur, B. Guichardaz, F. Druon, and P. Camy, "Efficient  $\text{Tm}:\text{LiYF}_4$  lasers at  $\sim 2.3$   $\mu\text{m}$ : Effect of energy-transfer upconversion," *IEEE J. Quantum Electron.* **55**(6), 1700212 (2019).
  11. I. Yorulmaz and A. Sennaroglu, "Low-threshold diode-pumped 2.3- $\mu\text{m}$   $\text{Tm}^{3+}$ :YLF lasers," *IEEE J. Sel. Top. Quantum Electron.* **24**(5), 1601007-1-7 (2018).
  12. L. Guillemot, P. Loiko, R. Soulard, A. Braud, J.-L. Doualan, A. Hideur, and P. Camy, "Close look on cubic  $\text{Tm}:\text{KY}_3\text{F}_{10}$  crystal for highly efficient lasing on the  ${}^3\text{H}_4 \rightarrow {}^3\text{H}_5$  transition," *Opt. Express* **28**(3), 3451-3463 (2020).
  13. P. Loiko, E. Kifle, L. Guillemot, J.-L. Doualan, F. Starecki, A. Braud, M. Aguiló, F. Díaz, V. Petrov, X. Mateos, and P. Camy, "Highly efficient 2.3  $\mu\text{m}$  thulium lasers based on a high-phonon-energy crystal: evidence of vibronic-assisted emissions," *J. Opt. Soc. Am. B* **38**(2), 482-495 (2021).
  14. L. Guillemot, P. Loiko, A. Braud, J.-L. Doualan, A. Hideur, M. Koselja, R. Moncorge, and P. Camy, "Continuous-wave  $\text{Tm}:\text{YAlO}_3$  laser at  $\sim 2.3$   $\mu\text{m}$ ," *Opt. Lett.* **44**(20), 5077-5080 (2019).
  15. R. Soulard, A. Tyazhev, J.L. Doualan, A. Braud, A. Hideur, M. Laroche, B. Xu, and P. Camy, "2.3  $\mu\text{m}$   $\text{Tm}^{3+}$ :YLF mode-locked laser," *Opt. Lett.* **42**(18), 3534-3536 (2017).
  16. F. Canbaz, I. Yorulmaz, and A. Sennaroglu, "Kerr-lens mode-locked 2.3- $\mu\text{m}$   $\text{Tm}^{3+}$ :YLF laser as a source of femtosecond pulses in the mid-infrared," *Opt. Lett.* **42**(19), 3964-3967 (2017).
  17. L. Guillemot, P. Loiko, R. Soulard, A. Braud, J.-L. Doualan, A. Hideur, R. Moncorgé, and P. Camy, "Thulium laser at  $\sim 2.3$   $\mu\text{m}$  based on upconversion pumping," *Opt. Lett.* **44**(16), 4071-4074 (2019).
  18. Y. Morova, M. Tonelli, V. Petrov, and A. Sennaroglu, "Upconversion pumping of a 2.3  $\mu\text{m}$   $\text{Tm}^{3+}:\text{KY}_3\text{F}_{10}$  laser with a 1064 nm ytterbium fiber laser," *Opt. Lett.* **45**(4), 931-934 (2020).
  19. M. F. Joubert, S. Guy, and B. Jacquier, "Model of the photon-avalanche effect," *Phys. Rev. B* **48**(14), 10031-10037 (1993).
  20. A. Tyazhev, F. Starecki, S. Cozic, P. Loiko, L. Guillemot, A. Braud, F. Joulain, M. Tang, T. Godin, A. Hideur, and P. Camy, "Watt-level efficient 2.3  $\mu\text{m}$  thulium fluoride fiber laser," *Opt. Lett.* **45**(20), 5788-5791 (2020).
  21. F. Wang, H. Huang, H. Chen, Y. Bao, Z. Li, and D. Shen, "GSA and ESA dual-wavelength pumped 2.3  $\mu\text{m}$   $\text{Tm}:\text{YLF}$  laser on the  ${}^3\text{H}_4 \rightarrow {}^3\text{H}_5$  transition," *Chin. Opt. Lett.* **19**(9), 091405 (2021).