Recent Progress in Laser Crystals and Ceramics for Femtosecond Mode-Locked Lasers at ~2 µm [Invited]

Pavel Loiko,[1,*] Weidong Chen,[2] Xavier Mateos,[3,#] Patrice Camy,[1] Uwe Griebner,[4] and Valentin Petrov[4]

[1]*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*
[2]*Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou, 350002 Fujian, China*
[3]*FiCMA-FiCNA-EMaS, Universitat Rovira i Virgili (URV), Campus Sescelades, E-43007 Tarragona, Spain.* [#]*Serra Húnter Fellow*
[4]*Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Str. 2a, 12489 Berlin, Germany*
pavel.loiko@ensicaen.fr

Lasers emitting femtosecond (fs) pulses in the spectral range of ~2 µm are of interest for development of amplified systems for high harmonic and THz generation, as well as pumping of mid-infrared ultrafast optical parametric oscillators based on non-oxide nonlinear crystals. Such emission can be generated using thulium (Tm^{3+}, $^3F_4 \rightarrow {}^3H_6$) and holmium ($Ho^{3+}$, $^5I_7 \rightarrow {}^5I_8$) ions. From the material point of view, for reaching short (sub-100 fs) pulse durations, there exist two challenges: (i) the need to operate above 2 µm to avoid the structured water vapor absorption in the atmosphere leading to ps pulse durations and (ii) the need of laser-active media with broad, flat and smooth gain profiles supporting the generation of ultrashort pulses. In the present work, we review recent advances in developing singly Tm^{3+} doped and Tm^{3+}, Ho^{3+} codoped laser crystals and transparent ceramics capable of generating sub-100 fs pulses from ~2 µm mode-locked (ML) solid-state lasers.

One solution to address the desired gain properties is the use of structurally disordered host crystals featuring a significant inhomogeneous spectral broadening for the dopant Tm^{3+} and Ho^{3+} ions. Two prominent examples: cubic calcium niobium gallium garnets ($Ca_3Nb_{1.5}Ga_{3.5}O_{12}$-type, CNGG) [1] and tetragonal calcium rare-earth aluminates ($CaGdAlO_4$-type, CALGO), Fig. 1(a) [2]. Even for singly Tm^{3+}-doped crystals, the emission bands are smooth and broad extending above 2 µm. A Tm:CLNGG laser passively ML by a single-walled carbon nanotube (SWCNT) saturable absorber generated 78 fs pulses at 2017 nm at a repetition rate of 86 MHz [1]. Via Tm^{3+}, Ho^{3+} codoping, one can further red-shift the emission wavelength and benefit from the combined gain bandwidths, Fig. 1(b). Using a GaSb-based SESAM, a Tm,Ho:CALGO ML laser delivered 52 fs at 2015 nm with a spectral bandwidth of 82 nm and an average output power of 376 mW [2]. We show that long-wave emissions at wavelengths well exceeding those of purely electronic transitions (the phonon sidebands of Tm^{3+} and Ho^{3+} emission bands) could be responsible for gain up to at least 2.3 µm, Fig. 1(b), thus supporting the generation of shorter pulses from ML lasers. Such emissions of vibronic nature are related to multiphonon-assisted transitions.

Another solution is based on employing materials with strong crystal-fields leading to large Stark splitting of the Tm^{3+} ground-state ($^3H_6$) naturally providing long-wave electronic emissions. One example: cubic sesquioxides A_2O_3 (A = Y, Lu, Sc or their mixture) being very suitable for the transparent ceramic technology. Using the ability of the ceramic sintering technology to fabricate easily compositionally “mixed” solid-solutions such as $(A_{1-x}B_x)_2O_3$, additional spectral broadening can be achieved. A Kerr-lens mode-locked (KLM) Tm:$(Lu,Sc)_2O_3$ ceramic laser generated pulses as short as 58 fs at ~2081 nm with an average output power of 220 mW at a pulse repetition rate of 84.8 MHz (extinction ratio above the noise level: 77 dBc). The emitted spectrum at the long-wavelength wing extended to 2.25 µm due to the vibronic transitions of the Tm^{3+} ions. The latter is found to be essential for generating pulses with durations in the sub-50 fs range.

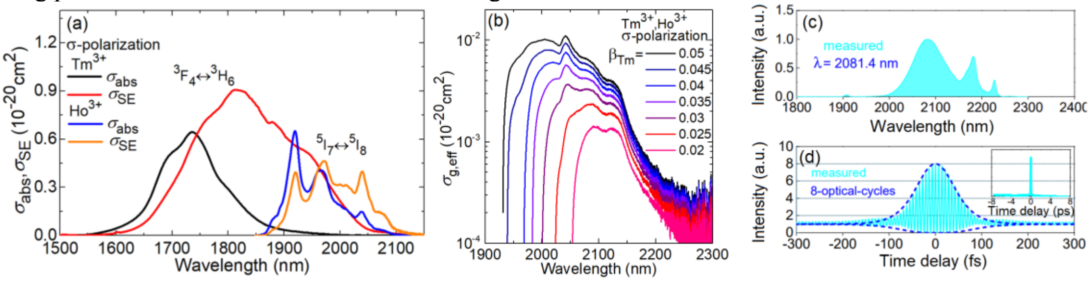

Fig. 1. (a,b) Spectroscopy of Tm^{3+} and Ho^{3+} ions in the disordered CALGO crystal: (a) absorption, σ_{abs}, and stimulated-emission, σ_{SE}, cross-sections for the $^3F_4 \leftrightarrow {}^3H_6$ Tm^{3+} and $^5I_7 \leftrightarrow {}^5I_8$ Ho^{3+} transitions around 2 µm, (b) gain profiles for a Tm^{3+}, Ho^{3+} codoped crystal plotted in a semi-log scale accounting for the long-wave vibronic emission. Light polarization: σ; (c,d) KLM Tm:$(Lu,Sc)_2O_3$ sesquioxide ceramic laser: (c) optical spectrum and (b) interferometric autocorrelation trace.

References

[1] Y. Wang, Y. Zhao, Z. Pan, J. E. Bae, S. Y. Choi, F. Rotermund, P. Loiko, J. M. Serres, X. Mateos, H. Yu, H. Zhang, M. Mero, U. Griebner, and V. Petrov, "78 fs SWCNT-SA mode-locked Tm:CLNGG disordered garnet crystal laser at 2017 nm," Opt. Lett. **43**(17), 4268-4271 (2018).
[2] Y. Wang, P. Loiko, Y. Zhao, Z. Pan, W. Chen, M. Mero, X. Xu, J. Xu, X. Mateos, A. Major, M. Guina, V. Petrov, and U. Griebner, "Polarized spectroscopy and SESAM mode-locking of Tm,Ho:CALGO," Opt. Express **30**(5), 7883-7893 (2022).
[3] Y. Zhao, L. Wang, W. Chen, P. Loiko, Y. Wang, Z. Pan, H. Yang, W. Jing, H. Huang, J. Liu, X. Mateos, Z. Wang, X. Xu, U. Griebner, and V. Petrov, "Kerr-lens mode-locked Tm-doped sesquioxide ceramic laser," Opt. Lett. **46**(14), 3428-3431 (2021).