Growth, Spectroscopy and Laser Operation of Tm3+,Li + -Codoped Ca3Ta1.5Ga3.5O12 -Type Disordered Garnet Crystal

A. Alles, Z. Pan, J. Serres, P. Loiko, K. Tang, S. Yingming, Y. Wang, Y. Zhao, E. Dunina, A. Kornienko, et al.

Growth, Spectroscopy and Laser Operation of Tm^{3+}, Li^{+}-Codoped $Ca_3Ta_{1.5}Ga_{3.5}O_{12}$-Type Disordered Garnet Crystal

A. Alles[1,2,*], Z. Pan[3,4], J. M. Serres[1,2], P. Loiko[5], K. Tang[3], S. Yingming[3], Y. Wang[4], Y. Zhao[4,6], E. Dunina[7], A. Kornienko[7], P. Camy[5], W. Chen[4,8], L. Wang[4], U. Griebner[4], V. Petrov[4], R. M. Solé[1], M. Aguiló[1], F. Díaz[1], and X. Mateos[1,9]

*1. Universitat Rovira i Virgili (URV), Marcel·lí Domingo 1, 43007 Tarragona, Spain. *E-mail: adrian.alles@eurecat.org*
2. Eurecat, Centre Tecnològic de Catalunya, Advanced Manufacturing Systems Unit (AMS), Marcel·lí Domingo 2, 43007 Tarragona, Spain
3. Institute of Chemical Materials, China Academy of Engineering Physics, 621900 Mianyang, China
4. Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Str. 2a, 12489 Berlin, Germany
5. CIMAP, UMR 6252 CEA-CNRS-ENSICAEN, Université de Caen Normandie, 6 Boulevard du Maréchal Juin, 14050 Caen, France
6. Jiangsu Key Laboratory of Advanced Laser Materials and Devices, Jiangsu Normal University, 221116 Xuzhou, China
7. Vitebsk State Technological University, 72 Moskovskaya Ave., 210035 Vitebsk, Belarus
8. Key Laboratory of Optoelectronic Materials Chemistry and Physics, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou, 350002 Fujian, China
9. Serra Húnter Fellow

Calcium niobium gallium garnet (CNGG) crystals doped with thulium (Tm^{3+}) ions possess disordered structure leading to inhomogeneously broadened emission bands which makes them attractive for generation of ultrashort pulses at ~2 μm [1]. The actual composition of CNGG deviates from the stoichiometry and cationic vacancies are present to ensure charge compensation. They can be eliminated by codoping with univalent alkali cations (Li^+, Na^+). The related calcium tantalum gallium garnet (CTGG) shows better thermal properties than CNGG [2]. Here, we report on the growth, spectroscopy and first laser action in a Tm^{3+},Li^+-codoped CTGG (Tm:CLTGG) crystal.

The Tm:CLTGG single crystal was grown by the Czochralski method using an [111]-oriented seed and argon atmosphere in an iridium crucible. The crystal was transparent with slight green coloration, inset in Fig. 1(a). The Tm^{3+} concentration was determined to be 3.17 at.%. Tm:CLTGG belongs to the cubic class (sp. gr. O^{10}_h, a = 12.5158(0) Å). Its structure was refined by the Rietveld method, Fig. 1(a). The Raman spectroscopy revealed a maximum phonon energy of 842 cm^{-1}. The absorption cross-section, σ_{abs}, for the $^3H_6 \rightarrow {}^3H_4$ Tm^{3+} transition is 3.1×10^{-21} cm^2 at 795.0 nm, Fig. 1(b). The maximum SE cross-section for the $^3F_4 \rightarrow {}^3H_6$ Tm^{3+} transition σ_{SE} is 3.7×10^{-21} cm^2 at 1865 nm with smooth and broad spectrum extending well above 2 μm, Fig. 1(c). The luminescence lifetime of the $^3F_4$ state τ_{lum} is 5.26 ms, Fig. 1(d). The transition probabilities for the Tm^{3+} ion were calculated using the modified Judd-Ofelt (mJ-O) theory yielding a radiative lifetime $\tau_{rad}(^3F_4)$ = 5.33 ms.

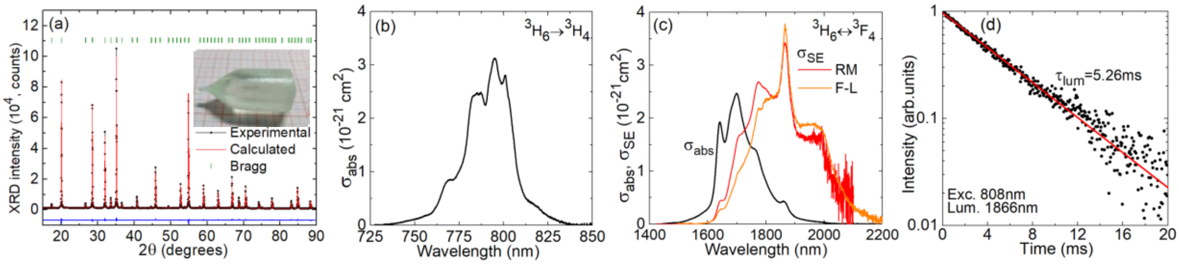

Fig. 1 Tm:CLTGG crystal: (a) X-ray powder diffraction (XRD) pattern showing the Rietveld refinement, *inset* – photograph of the as-grown crystal; (b) σ_{abs} spectrum for the $^3H_6 \rightarrow {}^3H_4$ transition; (c) σ_{abs} and σ_{SE} spectra for the $^3F_4 \leftrightarrow {}^3H_6$ transition (RM: reciprocity method, F-L: Fuchtbauer–Ladenburg equation); (d) luminescence decay curve, λ_{exc} = 808 nm, λ_{lum} = 1866 nm.

CW laser operation was achieved with a compact plane-parallel cavity and a fiber-coupled 793 nm AlGaAs laser diode. The uncoated crystal was 8.19 mm-thick. The Tm:CLTGG laser (unpolarized) generated a maximum output power of 1.08 W at 1995 and 2003 nm with a slope efficiency of 23.8% and a laser threshold of 0.91 W (for T_{OC} = 5%), Fig. 2. Tm:CLTGG is promising for broadly tunable and mode-locked lasers emitting above 2 μm.

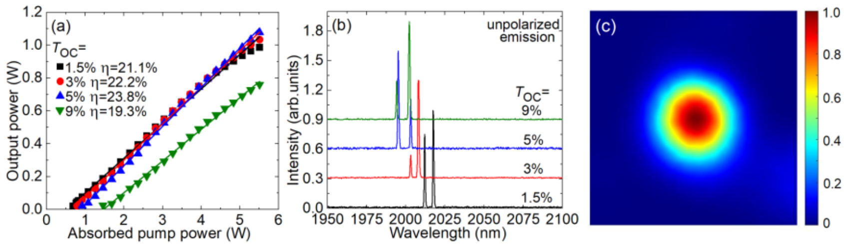

Fig. 2 Diode-pumped Tm:CLTGG laser: (a) input-output dependences, T_{OC}: output coupler transmission, η – slope efficiency; (b) typical spectra of laser emission; (c) typical spatial profile of the laser mode in the far-field.

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] C. Ma, Y. Wang, X. Cheng, M. Xue, C. Zuo, C. Gao, S. Guo, and J. He, "Spectroscopic, thermal, and laser properties of disordered garnet Nd:CLTGG crystal," J. Cryst. Growth **504**, 44-50 (2018).