Growth, Spectroscopy and Laser Operation of $\text{Tm}^{3+}, \text{Li}^{+}$ -Codoped $\text{Ca}_3\text{Ta}_{1.5}\text{Ga}_{3.5}\text{O}_{12}$ -Type Disordered Garnet Crystal

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A. Alles, Z. Pan, J. Serres, P. Loiko, K. Tang, et al.. Growth, Spectroscopy and Laser Operation of $\text{Tm}^{3+}, \text{Li}^{+}$ -Codoped $\text{Ca}_3\text{Ta}_{1.5}\text{Ga}_{3.5}\text{O}_{12}$ -Type Disordered Garnet Crystal. 2021 Conference on Lasers and Electro-Optics Europe & European Quantum Electronics Conference (CLEO/Europe-EQEC), Jun 2021, Munich, Germany. pp.1-1, 10.1109/CLEO/Europe-EQEC52157.2021.9541945 . hal-03368745

HAL Id: hal-03368745

<https://hal.science/hal-03368745>

Submitted on 7 Oct 2021

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Growth, Spectroscopy and Laser Operation of Tm^{3+} , Li^+ -Codoped $\text{Ca}_3\text{Ta}_{1.5}\text{Ga}_{3.5}\text{O}_{12}$ -Type Disordered Garnet Crystal

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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 $\sim 2 \mu\text{m}$ [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 , $a = 12.5158(0) \text{ \AA}$). 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 $^3\text{H}_6 \rightarrow ^3\text{H}_4$ Tm^{3+} transition is $3.1 \times 10^{-21} \text{ cm}^2$ at 795.0 nm , Fig. 1(b). The maximum SE cross-section for the $^3\text{F}_4 \rightarrow ^3\text{H}_6$ Tm^{3+} transition σ_{SE} is $3.7 \times 10^{-21} \text{ cm}^2$ at 1865 nm with smooth and broad spectrum extending well above $2 \mu\text{m}$, Fig. 1(c). The luminescence lifetime of the $^3\text{F}_4$ 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 $\tau_{\text{rad}}(^3\text{F}_4) = 5.33 \text{ ms}$.

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 $^3\text{H}_6 \rightarrow ^3\text{H}_4$ transition; (c) σ_{abs} and σ_{SE} spectra for the $^3\text{F}_4 \leftrightarrow ^3\text{H}_6$ transition (RM: reciprocity method, F-L: Fuchtbauer-Ladenburg equation); (d) luminescence decay curve, $\lambda_{\text{exc}} = 808 \text{ nm}$, $\lambda_{\text{lum}} = 1866 \text{ nm}$.

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 $T_{\text{OC}} = 5\%$), Fig. 2. Tm:CLTGG is promising for broadly tunable and mode-locked lasers emitting above $2 \mu\text{m}$.

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.

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