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Mid-Infrared Laser Emissions of Tm³⁺-doped Garnets: The Case Study of Disordered Tm:CNGG Crystal

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Recently, disordered garnets such as CNGG (calcium niobium gallium garnet) doped with Tm³⁺ and codoped with Tm³⁺ and Ho³⁺ ions have attracted a lot of attention for generation of ultrashort pulses at ~2 μm [1]. Pan *et al.* reported on a Tm oscillator operating on the ³F₄ → ³H₆ transition and delivering 84 fs pulses at 2018 nm (emission bandwidth: >50 nm) and demonstrated continuous wavelength tuning between 1879-2086 nm [1]. It is interesting to extend the emission range of such crystals further into mid-IR. It can be done by exploiting either (i) vibronic emissions relying on electron-phonon-coupling or (ii) less common electronic ³H₄ → ³H₅ transition [2].

A 3.2 at.% Tm:CNGG crystal was grown by the Czochralski method. The stimulated-emission (SE) cross-section, σ_{SE} , spectra for its near- and mid-IR emissions were determined using the Fuchtbauer–Ladenburg equation with the radiative lifetimes τ_{rad} and branching ratios β_{JJ} calculated using the Judd-Ofelt formalism, Fig. 1(a). For the ³F₄ → ³H₆ transition, $\sigma_{SE} = 0.44 \times 10^{-20}$ cm² at 1865 nm and the emission extends until 2.2 μm owing to a phonon sideband. For the ³H₄ → ³H₅ transition, $\sigma_{SE} = 0.28 \times 10^{-20}$ cm² at 2331 nm (emission bandwidth: 132 nm). The gap between these electronic transitions is bridged by vibronic processes. The reabsorption-free luminescence lifetimes $\tau_{lum}(\text{}^3\text{F}_4) = 3.26$ ms and $\langle \tau_{lum}(\text{}^3\text{H}_4) \rangle = 0.181$ ms, Fig. 1(b).

Fig. 1 Spectroscopy of 3.2 at.% Tm:CNGG crystal: (a) Stimulated-emission (SE) cross-sections, σ_{SE} , for (from left to right) the ³F₄ → ³F₄, ³F₄ → ³H₆ and ³H₄ → ³H₅ Tm³⁺ transitions; (b) RT luminescence decay curves from the ³H₄ and ³F₄ states; (c) crystal-field splitting of the ³F₄ and ³H₄ multiplets (revealed at 12 K).

The laser experiments were performed using an [111]-cut uncoated Tm:CNGG sample (thickness: 6.0 mm). A hemispherical cavity was formed by a flat pump mirror coated for HT at ~0.79 μm and for HR at 1.90-2.35 μm and several concave output couplers (OCs) (RoC = -100 mm). We used one “conventional” non-selective OC (#1) and two special “shortpass” OCs (#2 and #3) designed to force the laser oscillations above 2 μm, Fig. 2(a). The pump source was a Ti:Sapphire laser tuned to 785.8 nm. Two types of emission were detected: (i) a purely electronic ³F₄ → ³H₆ one using OC #1 (maximum power: 500 mW at 1.99 μm with a slope efficiency η of 26.1%), and (ii) a mixture of vibronic emission at 2.08 or 2.13 μm and a purely electronic ³H₄ → ³H₅ one at 2.33 or 2.37 μm using OCs #2 and #3 (maximum power: 548 mW with a total $\eta = 58.2\%$), Fig. 2(b,c). The observed laser lines agreed with the crystal-field splitting of the corresponding multiplets determined at 12 K, Fig. 1(c). The weaker contribution of the ³H₄ → ³H₅ emission is assigned to strong self-quenching of the ³H₄ lifetime. Tm:CNGG crystals with optimized doping levels (1 – 3 at.%) are very promising for ultra-broadband mid-IR laser sources.

Fig. 2 Mid-infrared Tm:CNGG laser: (a) transmission spectra of the output couplers (OCs), values specify T_{oc} at 2 μm, 2.13 μm and 2.3 μm; (b) input-output dependences, η – slope efficiency; (c) typical spectra of laser emission.

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