Spectroscopy and continuous wave laser operation of Tm$^{3+}$-doped YScO$_3$ mixed sesquioxide crystal

A. Suzuki$^{1,2,3}$, S. Kalusniak$^3$, H. Tanaka$^3$, M. Brützam$^3$, S. Ganschow$^3$, M. Tokurakawa$^{1,2}$, and C. Kränkel$^3$

$^1$Institute for Laser Science, University of Electro-Communications, 1-5-1 Chofugaoka, Chofu Tokyo 182-8585, Japan
$^2$Center for Neuroscience and Biomedical Engineering University of Electro-Communications, 1-5-1 Chofugaoka, Chofu Tokyo 182-8585, Japan
$^3$Leibniz-Institut für Kristallzüchtung (IKZ), Max-Born-Straße 2, 12489 Berlin, Germany

Tm$^{3+}$-doped mixed sesquioxides are attractive gain media for ultrafast 2-$\mu$m lasers due to their superior spectroscopic properties [1]. They exhibit a gain band at the wavelengths exceeding 2000 nm where no water vapor absorption and Tm$^{3+}$ reabsorption is low, and their disordered crystal structure leads to broad and flat gain spectra due to inhomogeneous spectral broadening [2]. These features enable a very broad effective gain bandwidth, which is desirable for ultrashort pulse generation. In addition, high-quality large-size mixed sesquioxide crystals can be grown by the Czochralski method [3]. Conventional pure sesquioxide materials show excellent thermomechanical and spectroscopic properties as a laser medium [4], but they are difficult to fabricate due to their high melting points of more than 2350 ºC. However, our current reinvestigation of the sesquioxide phase diagram revealed the existence of mixed sesquioxide compositions with melting temperatures below 2100 ºC [3]. This enables the Czochralski growth from iridium crucibles, common for other oxide materials. Consequently, mixed sesquioxide crystals are promising candidates as commercially available gain materials for ultrafast lasers. Here, we present spectroscopic investigations and continuous wave laser operation of a Czochralski-grown Tm$^{3+}$-doped YScO$_3$ crystal.

![Fig. 1](image-url) (a) Absorption cross section and (b) Emission cross section at room temperature (RT) and 14 K. (c) Output power characteristics as a function of the absorbed pump power with different OCs.

The laser crystal was prepared by the Czochralski growth from an iridium crucible from a melt composition of (Tm$_{0.02}$Y$_{0.5}$Sc$_{0.5}$)$_2$O$_3$. The actual composition of the grown crystal was determined by inductively-coupled plasma optical emission spectroscopy to be (Tm$_{0.022}$Y$_{0.472}$Sc$_{0.506}$)$_2$O$_3$. Figure 1 (a, b) show the absorption and emission cross sections at room temperature and 14 K. The absorption bands around 0.8 $\mu$m and 1.6 $\mu$m are useful for 2-for-1 pumping as well as in-band pumping for 2 $\mu$m laser operation, respectively. Broad and less-structured absorption and emission spectra are found even at low temperature, which indicates the expected inhomogeneous spectral broadening.

For the laser experiments, we constructed a 48 mm long plane-concave linear cavity. As a gain medium, a 6-mm thick sample was prepared, and a 780 nm single-emitter C-mount laser diode with a maximum output power of 3 W served as a pump source. Using output couplers (OCs) of different transmittance T, we obtained laser characteristics as shown in Fig. 1(c). Using a 4 % OC, the highest slope efficiency of 40% was achieved. The central wavelength was 2100 nm while the Stokes limit determined as the ratio of pump and laser wavelength amounts to only 37%. The highest slope efficiency of 40% was thus only possible by a cross relaxation process between adjacent Tm$^{3+}$ ions in the 2-for-1 pumping scheme.

In conclusion, we investigated the spectroscopic properties and laser operation of a Tm$^{3+}$(2.2 at.%)YScO$_3$ mixed sesquioxide crystal. Its broadband absorption and emission spectra are favorable for ultrafast lasers, and continuous wave laser operation with a slope efficiency of 40% showed that laser-quality crystals can be grown by the Czochralski method.

References

[2] R. Moncorge et al., “Mid-infrared emission properties of the Tm$^{3+}$-doped sesquioxide crystals Y$_2$O$_3$, Lu$_2$O$_3$, Sc$_2$O$_3$ and mixed compounds (Y,Lu,Sc)$_2$O$_3$ around 1.5-, 2- and 2.3-$\mu$m,” J. Lumin. 241, 118537 (2022).