



HAL
open science

Excited-state absorption of Tm^{3+} ions in cubic sesquioxides

Kirill Eremeev, Pavel Loiko, Christian Kränkel, Patrice Camy, Alain Braud

► **To cite this version:**

Kirill Eremeev, Pavel Loiko, Christian Kränkel, Patrice Camy, Alain Braud. Excited-state absorption of Tm^{3+} ions in cubic sesquioxides. OPTIQUE Normandie 2024, La Société Française d'Optique, Jul 2024, Rouen, France. hal-04654928

HAL Id: hal-04654928

<https://hal.science/hal-04654928>

Submitted on 20 Jul 2024

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Excited-state absorption of Tm^{3+} ions in cubic sesquioxides

Kirill Ereemeev^{1,*}, Pavel Loiko¹, Christian Kränkel², Patrice Camy¹ and Alain Braud¹

¹Centre de Recherche sur les Ions, les Matériaux et la Photonique (CIMAP), UMR 6252 CEA-CNRS-ENSICAEN, Université de Caen Normandie, 6 Boulevard Maréchal Juin, 14050 Caen, France

²Leibniz-Institut für Kristallzüchtung (IKZ), Max-Born-Str. 2, 12489 Berlin, Germany

*kirill.eremeev@ensicaen.fr

RÉSUMÉ

Excited-state absorption of Tm^{3+} ions in R_2O_3 ($\text{R} = \text{Y}, \text{Lu}, \text{Sc}$) cubic sesquioxides was studied by the pump-probe method focusing on the ${}^3\text{F}_4 \rightarrow {}^3\text{F}_{2,3}$ (at $\sim 1 \mu\text{m}$) and ${}^3\text{F}_4 \rightarrow {}^3\text{H}_4$ (at $\sim 1.45 \mu\text{m}$) transitions. The energy-transfer upconversion is also quantified in these materials. The performed studies are essential for the design of $2.3 \mu\text{m}$ Tm lasers.

MOTS-CLEFS: *sesquioxides; thulium ions; optical spectroscopy; excited-state absorption.*

1. INTRODUCTION

Thulium ions (Tm^{3+}) enable laser operation at several transitions in the short-wave infrared, at $\sim 2 \mu\text{m}$ (${}^3\text{F}_4 \rightarrow {}^3\text{H}_6$) and $\sim 2.3 \mu\text{m}$ (${}^3\text{H}_4 \rightarrow {}^3\text{H}_5$). $2.3\text{-}\mu\text{m}$ laser sources find applications in remote sensing of atmospheric species, metrology of combustion processes, non-invasive glucose measurements and as pump sources for mid-infrared gas fiber photonics. Recently, an upconversion pumping scheme relying on a resonant excited-state absorption (ESA) ${}^3\text{F}_4 \rightarrow {}^3\text{F}_{2,3}$ at $\sim 1 \mu\text{m}$ was proposed for $2.3\text{-}\mu\text{m}$ Tm lasers [1]. It benefits from the commercialized technology of high power and high brightness Yb-fiber lasers. Cubic rare-earth sesquioxides R_2O_3 ($\text{R} = \text{Y}, \text{Lu}, \text{or Sc}$) are well-known for doping with rare-earth ions including Tm^{3+} ones [2]. They belong to the cubic class (sp. gr. $Ia\bar{3}$) and combine good thermal properties, low phonon energies and strong crystal-fields for the dopant ions. The latter underlines broadband emission properties of these materials. In the present work, we have studied excited-state absorption and energy-transfer upconversion (ETU) for Tm^{3+} ions in three cubic sesquioxides with the goal of developing novel materials for $2.3 \mu\text{m}$ lasers.

2. EXCITED-STATE ABSORPTION

The ESA studies were conducted using a pump-probe set-up depicted in Fig. 1(a). The double lock-in detection technique was used. Two ESA transitions were considered, ${}^3\text{F}_4 \rightarrow {}^3\text{F}_{2,3}$ (at

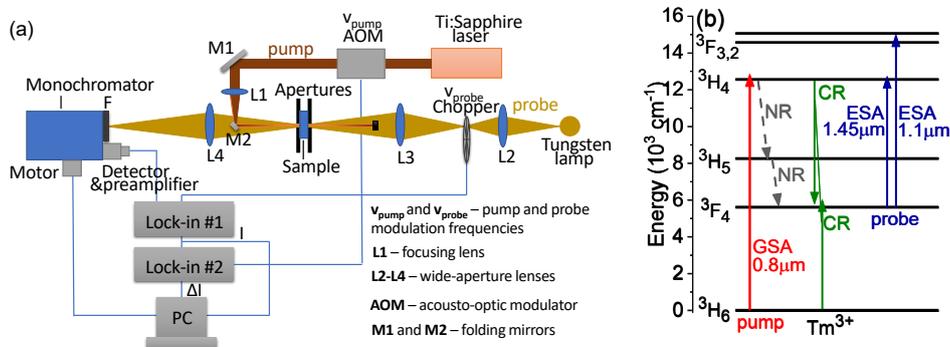


Figure 1: (a) Scheme of the pump-probe setup used for the ESA measurements; (b) Energy-level scheme of Tm^{3+} ion illustrating the principle of the pump-probe experiment, CR – cross-relaxation.

$\sim 1 \mu\text{m}$) and ${}^3\text{F}_4 \rightarrow {}^3\text{H}_4$ (at $\sim 1.45 \mu\text{m}$). The pump source was a 1 W Ti:Sapphire laser tuned to $0.79 \mu\text{m}$. The source of probe radiation was a stabilized 90 W tungsten lamp. The spectra were measured using a 0.5 m monochromator with a cooled InGaAs detector, which has a resolution better than 0.5 nm. The energy-level scheme of Tm^{3+} ions, illustrating the principle of the pump-probe method, is shown in Fig. 1(b).

The excited-state absorption cross-section, σ_{ESA} , spectra for $\text{Tm}:\text{Y}_2\text{O}_3$ are shown in Fig. 2(a). The maximum σ_{ESA} amounts to $1.40 \times 10^{-20} \text{ cm}^2$ at 1024.4 nm (for the ${}^3\text{F}_4 \rightarrow {}^3\text{F}_{2,3}$ transition) and $0.63 \times 10^{-20} \text{ cm}^2$ at 1423.7 nm (for the ${}^3\text{F}_4 \rightarrow {}^3\text{H}_4$ one). In Fig. 2(b), we compare the ESA cross-section spectra for Tm^{3+} ions in all three studied cubic sesquioxides in the spectral range slightly above $1 \mu\text{m}$ covered by the emission of Yb-fiber lasers. Both $\text{Tm}:\text{Y}_2\text{O}_3$ and $\text{Tm}:\text{Lu}_2\text{O}_3$ show a great promise for realization of the upconversion pumping scheme at $\sim 1 \mu\text{m}$.

Energy-transfer upconversion from the metastable Tm^{3+} state, ${}^3\text{F}_4 + {}^3\text{F}_4 \rightarrow {}^3\text{H}_4 + {}^3\text{H}_6$, is an important effect influencing the performance of $2.3 \mu\text{m}$ lasers. In the present work, the concentration-independent ETU parameter C_{ETU} of Tm^{3+} ions in cubic sesquioxides was calculated using the Burnstein model. The ground-state absorption (GSA), stimulated-emission (SE) and ESA cross-section spectra for the relevant Tm^{3+} transitions in Y_2O_3 used for this calculation are plotted in Fig. 2(c). This analysis considers the exponential Stokes (S) and anti-Stokes (AS) phonon sidebands, as the ETU process is non-resonant and phonon assisted. For $\text{Tm}:\text{Y}_2\text{O}_3$, C_{ETU} amounts to $4.40 \times 10^{-40} \text{ cm}^6 \text{ s}^{-1}$. The determined C_{ETU} value is two orders of magnitude smaller than that for the cross-relaxation (CR) process, ${}^3\text{H}_6 + {}^3\text{H}_4 \rightarrow {}^3\text{F}_4 + {}^3\text{F}_4$, $C_{\text{CR}} = 0.5 \times 10^{-37} \text{ cm}^6 \text{ s}^{-1}$ for the same $\text{Tm}:\text{Y}_2\text{O}_3$ crystal [2].

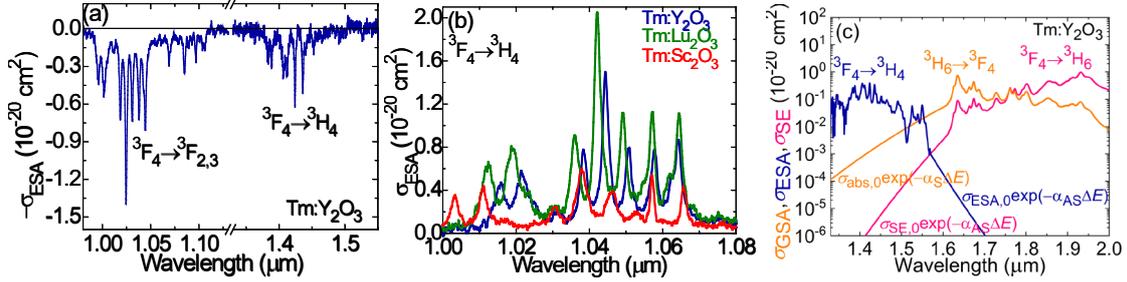


Figure 2: Spectroscopy of Tm^{3+} ions in cubic sesquioxides: (a) ESA cross-sections, σ_{ESA} , for the ${}^3\text{F}_4 \rightarrow {}^3\text{F}_{2,3}$ and ${}^3\text{F}_4 \rightarrow {}^3\text{H}_4$ transitions for $\text{Tm}:\text{Y}_2\text{O}_3$; (b) Comparison of σ_{ESA} spectra around $1 \mu\text{m}$ for the three studied sesquioxides; (c) evaluation of the ETU parameter for $\text{Tm}:\text{Y}_2\text{O}_3$: SE (${}^3\text{F}_4 \rightarrow {}^3\text{H}_6$), GSA (${}^3\text{H}_6 \rightarrow {}^3\text{F}_4$) and ESA (${}^3\text{F}_4 \rightarrow {}^3\text{H}_4$) cross-section spectra plotted in a semi-log scale with the projected phonon sidebands.

3. CONCLUSION

To conclude, excited-state absorption spectra of Tm^{3+} -doped R_2O_3 ($\text{R} = \text{Y}, \text{Lu}, \text{Sc}$) cubic sesquioxides indicate their potential for upconversion pumping using commercially available and power-scalable Yb-fiber lasers to achieve laser emission around $2.3 \mu\text{m}$. The energy-transfer upconversion parameters, which are important for predicting the slope efficiency of such laser sources, were also calculated.

RÉFÉRENCES

- [1] L. Guillemot, P. Loiko, R. Soulard, A. Braud, J. L. Doualan, A. Hideur, R. Moncorgé, and P. Camy, “Thulium laser at $\sim 2.3 \mu\text{m}$ based on upconversion pumping,” *Opt. Lett.* 44(16), 4071–4074 (2019).
- [2] P. Loiko, P. Koopmann, X. Mateos, J. M. Serres, V. Jambunathan, A. Lucianetti, T. Mocek, M. Aguilo, F. Diaz, U. Griebner, and V. Petrov, “Highly efficient, compact $\text{Tm}^{3+}:\text{RE}_2\text{O}_3$ ($\text{RE} = \text{Y}, \text{Lu}, \text{Sc}$) sesquioxide lasers based on thermal guiding,” *IEEE J. Sel. Top. Quantum Electron.* 24(5), 1600713–1–13 (2018).