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Phonon sidebands, electron-phonon coupling and vibronic lasing in $\text{Tm}^{3+}:\text{Lu}_2\text{O}_3$ sesquioxide

Pavel Loiko^{1,*}, Kirill Ereemeev¹, Christian Kränkel², Alain Braud¹, and Patrice Camy¹

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

*pavel.loiko@ensicaen.fr

RÉSUMÉ

A vibronic $\text{Tm}:\text{Lu}_2\text{O}_3$ laser generated 654 mW at 2238-2309 nm with a slope efficiency of 68.9% exceeding the Stokes limit. $\text{Tm}:\text{Lu}_2\text{O}_3$ features a strong electron-phonon interaction resulting in a profound long-wave phonon sideband for the ${}^3\text{F}_4 \rightarrow {}^3\text{H}_6$ electronic transition extending up to at least 2.4 μm supporting vibronic laser operation.

MOTS-CLEFS: *sesquioxides; thulium ions; optical spectroscopy; vibronic laser.*

1. INTRODUCTION

Cubic (C-type) rare-earth sesquioxides such as Lutetia (Lu_2O_3) are attractive laser host media for doping with rare-earth ions (RE^{3+}) for the development of lasers emitting in the short-wave infrared spectral range [1]. They benefit from high thermal conductivity, broadband transparency, low phonon energy (among oxides), easy RE^{3+} doping and strong crystal-fields for the dopant ions resulting in broad emission bands. Thulium ions (Tm^{3+}) in Lu_2O_3 crystals emit slightly above 2 μm and $\text{Tm}:\text{Lu}_2\text{O}_3$ crystals find applications in high-power continuous-wave and femtosecond mode-locked lasers. It is interesting to further extend the emission wavelengths of Tm lasers, e.g., by relying on the electron-phonon coupling (vibronic lasers) or the ${}^3\text{H}_4 \rightarrow {}^3\text{H}_5$ electronic transition [2]. Such 2.3- μm laser sources are of interest for molecular spectroscopy and fiber gas photonics. In this work, we have systematically studied long-wave emissions of Tm^{3+} ions in Lu_2O_3 and demonstrated efficient vibronic laser operation of this crystal at wavelengths beyond 2.2 μm .

2. PHONON SIDEBANDS OF Tm^{3+} IONS

The experimental crystal-field splitting of the ground-state (${}^3\text{H}_6$) and the first excited state (${}^3\text{F}_4$) of Tm^{3+} ions in C_2 symmetry sites in Lu_2O_3 is shown in Fig. 1(a). Given the total Stark

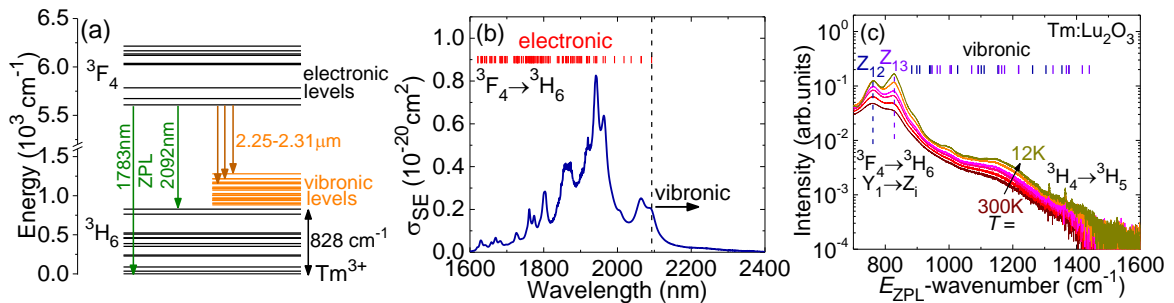


Figure 1: Spectroscopy of Tm^{3+} ions in Lu_2O_3 : (a) experimental crystal-field splitting for the ${}^3\text{H}_6$ and ${}^3\text{F}_4$ multiplets (black) and vibronic energy levels (orange) responsible for long-wave emissions; (b) stimulated-emission cross-sections, σ_{SE} , for Tm^{3+} ions around 2 μm , dashes – electronic transitions, (c) vibronic sideband of the emission spectrum at various temperatures, dashes – vibronic transitions.

splitting of the ground-state of 828 cm^{-1} , the longest wavelength of the purely electronic ${}^3\text{F}_4 \rightarrow {}^3\text{H}_6$ transition is 2092 nm . As depicted in Fig. 1(b), the stimulated-emission cross-section, σ_{SE} , for the corresponding emission peak amounts to $0.21 \times 10^{-20}\text{ cm}^2$. Furthermore, Tm^{3+} ions in Lu_2O_3 exhibit a broad and smooth phonon sideband extending up to at least $2.4\text{ }\mu\text{m}$. By cooling the $\text{Tm}:\text{Lu}_2\text{O}_3$ crystal to 12 K , it is possible to reveal the structure of this long-wave emission and assign it to transitions to vibronic sub-levels coupled to the electronic states of the ${}^3\text{H}_6$ ground manifold, see Fig. 1(c). The same figure clearly presents weak and narrow lines related to the ${}^3\text{H}_4 \rightarrow \text{H}_5$ electronic transition.

3. VIBRONIC $\text{Tm}:\text{Lu}_2\text{O}_3$ LASER

For the laser experiment, an uncoated $1.8\text{ at.}\%$ $\text{Tm}:\text{Lu}_2\text{O}_3$ crystal having a thickness of 5 mm was used. It was mounted on a passively cooled Cu-holder and placed in a hemispherical cavity formed by a flat pump mirror (HT at $0.80\text{ }\mu\text{m}$ and $2\text{ }\mu\text{m}$, HR at $2.18\text{--}2.35\text{ }\mu\text{m}$) and a concave ($\text{RoC} = -100\text{ mm}$) output coupler with a transmission T_{OC} of 0.65% or 0.8% at $2.3\text{ }\mu\text{m}$. A Ti:sapphire laser tuned to 796 nm was used as pump source. The pump radiation was focused by an achromatic AR-coated lens ($f = 75\text{ mm}$). The continuous-wave $\text{Tm}:\text{Lu}_2\text{O}_3$ laser generated a maximum output power of 654 mW at $2238\text{--}2309\text{ nm}$ (a broad spectrum) with a slope efficiency of 68.9% with respect to the absorbed pump power and a laser threshold of 0.70 W , Fig. 2(a,b). The laser efficiency exceeded the Stokes limit due to a one-for-two pump process enabled by an energy-transfer upconversion, ${}^3\text{F}_4 + {}^3\text{F}_4 \rightarrow {}^3\text{H}_6 + {}^3\text{H}_4$. For comparison, laser operation on the longest electronic transition (at $2081\text{--}2102\text{ nm}$) was studied, Fig. 2(a), featuring a much lower laser threshold. Both laser wavelengths were well explained by the gain spectra considering the phonon sideband, as shown in Fig. 2(c).

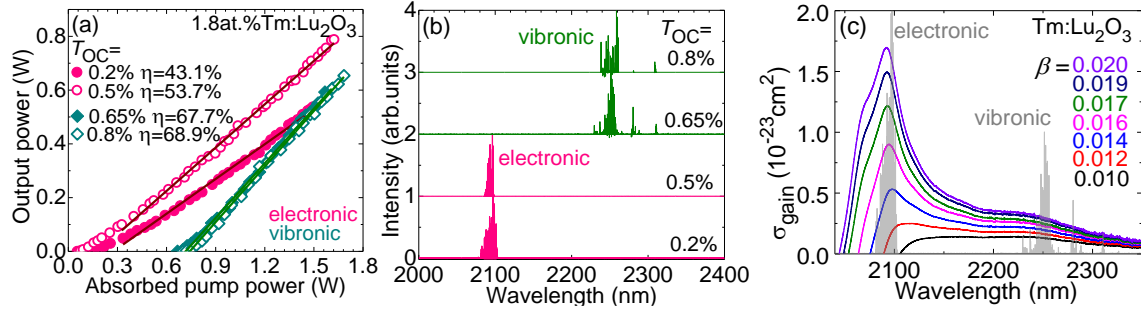


Figure 2: Vibronic $1.8\text{ at.}\%$ $\text{Tm}:\text{Lu}_2\text{O}_3$ laser: (a) input-output dependences, η – slope efficiency; (b) typical spectra of laser emission; (c) gain profiles for Tm^{3+} ions above $2\text{ }\mu\text{m}$, β – inversion ratio.

4. CONCLUSION

To conclude, $\text{Tm}:\text{Lu}_2\text{O}_3$ crystals feature a strong electron-phonon interaction which results in an intense phonon sideband in the emission spectrum extending up to at least $2.4\text{ }\mu\text{m}$ and supporting vibronic laser operation at wavelengths being much longer than those for the ${}^3\text{F}_4 \rightarrow {}^3\text{H}_6$ electronic transition. The obtained results are relevant for the development of broadly tunable and femtosecond mode-locked lasers emitting beyond $2\text{ }\mu\text{m}$.

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