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

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## 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  $\mu\text{m}$  supporting vibronic laser operation.

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

## 1. INTRODUCTION

Cubic (C-type) rare-earth sesquioxides such as Lutetia ( $\text{Lu}_2\text{O}_3$ ) are attractive laser host media for doping with rare-earth ions ( $\text{RE}^{3+}$ ) 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  $\text{RE}^{3+}$  doping and strong crystal-fields for the dopant ions resulting in broad emission bands. Thulium ions ( $\text{Tm}^{3+}$ ) in  $\text{Lu}_2\text{O}_3$  crystals emit slightly above 2  $\mu\text{m}$  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- $\mu\text{m}$  laser sources are of interest for molecular spectroscopy and fiber gas photonics. In this work, we have systematically studied long-wave emissions of  $\text{Tm}^{3+}$  ions in  $\text{Lu}_2\text{O}_3$  and demonstrated efficient vibronic laser operation of this crystal at wavelengths beyond 2.2  $\mu\text{m}$ .

## 2. PHONON SIDEBANDS OF $\text{Tm}^{3+}$ IONS

The experimental crystal-field splitting of the ground-state ( ${}^3\text{H}_6$ ) and the first excited state ( ${}^3\text{F}_4$ ) of  $\text{Tm}^{3+}$  ions in  $C_2$  symmetry sites in  $\text{Lu}_2\text{O}_3$  is shown in Fig. 1(a). Given the total Stark

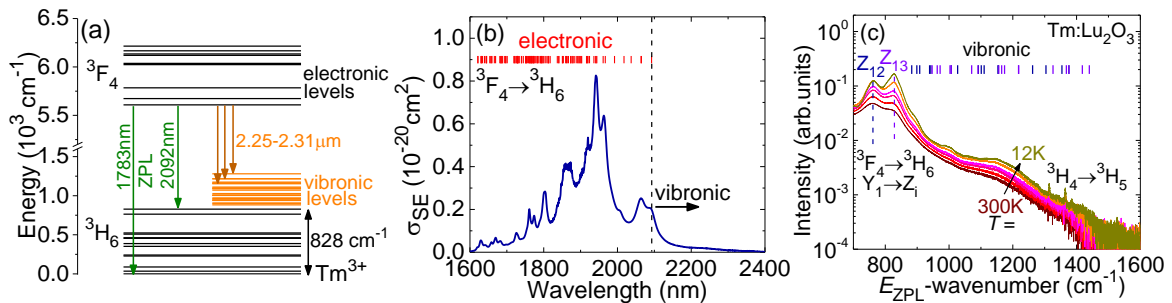


Figure 1: Spectroscopy of  $\text{Tm}^{3+}$  ions in  $\text{Lu}_2\text{O}_3$ : (a) experimental crystal-field splitting for the  ${}^3\text{H}_6$  and  ${}^3\text{F}_4$  multiplets (black) and vibronic energy levels (orange) responsible for long-wave emissions; (b) stimulated-emission cross-sections,  $\sigma_{\text{SE}}$ , for  $\text{Tm}^{3+}$  ions around 2  $\mu\text{m}$ , dashes – electronic transitions, (c) vibronic sideband of the emission spectrum at various temperatures, dashes – vibronic transitions.

splitting of the ground-state of  $828\text{ cm}^{-1}$ , the longest wavelength of the purely electronic  ${}^3\text{F}_4 \rightarrow {}^3\text{H}_6$  transition is  $2092\text{ nm}$ . As depicted in Fig. 1(b), the stimulated-emission cross-section,  $\sigma_{\text{SE}}$ , for the corresponding emission peak amounts to  $0.21 \times 10^{-20}\text{ cm}^2$ . Furthermore,  $\text{Tm}^{3+}$  ions in  $\text{Lu}_2\text{O}_3$  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  $12\text{ K}$ , 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  $5\text{ mm}$  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  $T_{\text{OC}}$  of  $0.65\%$  or  $0.8\%$  at  $2.3\text{ }\mu\text{m}$ . A Ti:sapphire laser tuned to  $796\text{ nm}$  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  $654\text{ mW}$  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  $0.70\text{ W}$ , 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).

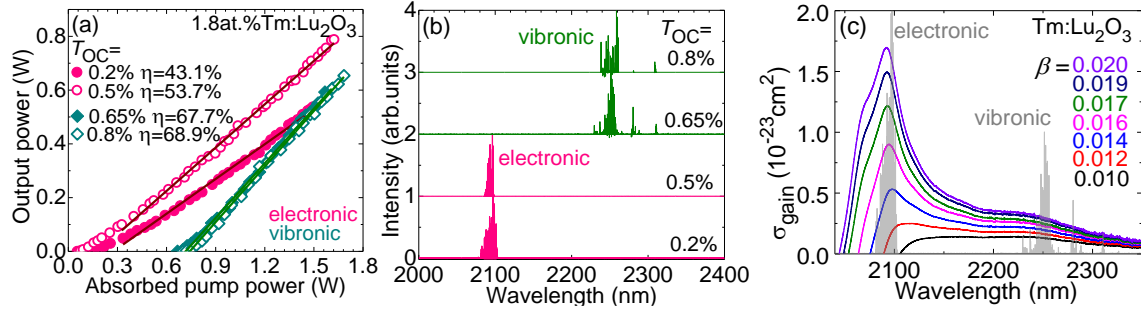


Figure 2: Vibronic  $1.8\text{ at.}\%$   $\text{Tm}:\text{Lu}_2\text{O}_3$  laser: (a) input-output dependences,  $\eta$  – slope efficiency; (b) typical spectra of laser emission; (c) gain profiles for  $\text{Tm}^{3+}$  ions above  $2\text{ }\mu\text{m}$ ,  $\beta$  – inversion ratio.

### 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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