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Excited-state luminescence properties and laser performance of the Tm$^{3+}$ doped perovskite YAlO$_3$

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1. Introduction

Polarized absorption and emission spectra have been recorded in a broad spectral range to determine the best pumping and laser emission conditions of the crystals in the infrared domain and to show their potentialities in the blue/green and red regions. These data are completed by lifetime measurements made as a function of the dopant concentration and a discussion of the radiative versus the non-radiative relaxations between the various excited-states is developed on the basis of the Judd-Ofelt formalism. Pointed excited-state excitation/absorption measurements are also performed to derive cross section values in a spectral region which has been found [1] interesting in this material for photon-avalanche up-conversion.

2. Experimental results

Figure 1a and 1b show the room temperature polarized absorption spectra of a YAP crystal doped by about 5% Tm$^{3+}$ ions. According to Weber [2] these spectra lead to the Judd-Ofelt parameters:

\[ \Omega_2 = 0.67 \times 10^{-20} \text{ cm}^2, \quad \Omega_4 = 2.3 \times 10^{-20} \text{ cm}^2, \quad \Omega_6 = 0.74 \times 10^{-20} \text{ cm}^2 \]

which give in turn the radiative lifetimes and branching ratios given in Table 1.

Figures 2 and 3 show the room temperature visible emission spectra of a 1% Tm$^{3+}$ doped powder sample obtained after $^1D_2$ and $^3G_4$ excitations at 355 and 470 nm respectively. These spectra show the spectral extents and give an idea of the respective intensities of the transitions originating from the $^1D_2$ and $^1G_4$ and terminating on the $^3H_6$ and $^3F_4$ levels.

Figure 4 gives the room temperature polarized emission spectra of a 5% Tm$^{3+}$ doped crystal as a function of polarization, the spectra being calibrated between each other.

Table 2 gives the room temperature 1/e fluorescence time constants of the emissions originating from the various energy levels as a function of the dopant concentration.

In the end we give in Figures 5a and 5b room temperature polarized excited state excitation (ESE) spectra which were recorded with the aid of a two beam experiment, one laser beam being used to bring the ions in
their $^3H_4$ excited state, after pumping of the $^3F_2$-$^3F_3$ levels and multiphonon relaxation, and the other, which could be triggered at various time delays after the former, being used to probe the fluorescences coming from the $^1D_2$ and $^1G_4$ levels. The ESE spectra were recorded around 650 nm, in a wavelength domain where a strong ESA line responsible for photon avalanche in this material should appear. By comparing the positions of the lines appearing in these spectra with the ones expected, from the energy level scale in this material, for $^3H_4$-$^1D_2$ (large bars in the figures) and $^3F_4$-$^1G_4$ (small bars in the figures) excited state absorption (ESA) transitions, it seems clear that the former dominates. This was confirmed further by delaying the probe beam by a few $\mu$s with respect to the pump beam since in this case the signal disappeared gradually and immediately. Indeed if the lines around 650 nm were assigned to $^3F_4$-$^1G_4$ ESA transitions, then, because of the 210 $\mu$s feeding time of the $^3F_4$ metastable state after $^3H_4$ excitation [3] the ESE signal would increase with the time delay between the two laser pulses. This result is very surprising since the model proposed by the authors of Ref. 1 to account for their photon avalanche process only included the possibility of a $^3F_4$-$^1G_4$ ESA transition. We have already planned to work on crystals doped by different amounts of Tm$^{3+}$ ions and by pumping them in different excited states to clarify this question.

Acknowledgments: Thanks are expressed to C. Borel and Ch. Wyon (LETI-CEA Grenoble) and N. Garnier (Labo. TSI, St Etienne) for their assistance in this research.

Reference:
2. M.J. Weber

![Figure 1: Room temperature absorption spectra of YAP:Tm$^{3+}$ for light polarized parallel (||) and perpendicular (⊥) to the c axis of the crystal]
<table>
<thead>
<tr>
<th>TRANSITION</th>
<th>Average wavelength (nm)</th>
<th>$A_{cm}^{(1)}$</th>
<th>$A_{cm}^{(2)}$</th>
<th>$\beta$</th>
<th>$t_\tau$</th>
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<tr>
<td>$3H_6 \rightarrow 3H_4$</td>
<td>215.1</td>
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<td>4.44</td>
<td>1</td>
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<tr>
<td>$3H_5 \rightarrow 3H_4$</td>
<td>209.7</td>
<td>0.3</td>
<td>2.48</td>
<td>0.99</td>
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<tr>
<td>$3H_4 \rightarrow 3H_3$</td>
<td>206.8</td>
<td>0</td>
<td>0.92</td>
<td>0.92</td>
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<tr>
<td>$3H_4 \rightarrow 3H_3$</td>
<td>196.8</td>
<td>0</td>
<td>0.92</td>
<td>0.92</td>
<td></td>
</tr>
<tr>
<td>$3H_3 \rightarrow 3H_2$</td>
<td>182.3</td>
<td>0</td>
<td>0.30</td>
<td>0.30</td>
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<td>$3H_2 \rightarrow 3H_1$</td>
<td>165.5</td>
<td>0</td>
<td>0.74</td>
<td>0.74</td>
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<td>$3H_1 \rightarrow 3H_2$</td>
<td>150.4</td>
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<td>0.74</td>
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<td>$3H_3 \rightarrow 3H_1$</td>
<td>144.9</td>
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<td>0.74</td>
<td>0.74</td>
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<tr>
<td>$3H_4 \rightarrow 3H_1$</td>
<td>135.8</td>
<td>0</td>
<td>0.74</td>
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<tr>
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<td>0.74</td>
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<tr>
<td>$3H_2 \rightarrow 3H_1$</td>
<td>111.4</td>
<td>0</td>
<td>0.74</td>
<td>0.74</td>
<td></td>
</tr>
</tbody>
</table>

Table 1: Transitions, average wavelengths, electric and magnetic dipole transition probabilities, radiative lifetimes and branching ratios in YAP:Tm

![Polarized Emission](image)

**YAP: Tm**

**Polarized Emission**

**Figure 4.** Room temperature calibrated and polarized infrared emission spectra of a 5% Tm doped crystal
Figure 2: Room temperature visible emission spectra of a 1% Tm$^{3+}$ doped YAP powder sample obtained after 355 nm excitation.

Figure 3: Room temperature visible emission spectra of a 1% Tm$^{3+}$ doped YAP powder sample obtained after 470 nm excitation.

Figure 5: Room temperature ESE spectra recorded with a 5% Tm$^{3+}$ doped YAP sample and comparison with the expected positions of the lines corresponding to the $^3H_4 \rightarrow D_2$ (large bars) and $^3F_4 \rightarrow ^1G_4$ ESA transitions. (a) $\pi$ polarization, (b) $\sigma$ polarization.