Fluorescence Saturation and cross relaxation in Tb^{3+} doped LSCAS glasses

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Due to their novel electronic and optical properties resulting from the 4f and 4f5d electrons, rare earth materials have attracted more research interest in the last decades. Tb^{3+} doped materials have been studied in many systems (crystalline and amorphous) due to their very interesting spectroscopic properties[1,2]. The strong emission in the green and weak emission in the UV blue levels usually is attributed to the cross relaxation (**CR**) process. In this paper, the luminescence properties of Tb^{3+}-doped low silica calcium aluminosilicate glasses are analyzed for UV (λ_{exc}=325nm) and visible (λ_{exc}=488 nm) excitations. The intensity of green luminescence increases proportionally to the concentration while the blue luminescence is strongly reduced with the increase of Tb^{3+} concentration from 0.5 – 15.0 wt%. This behavior can be related to the blue fluorescence quenching due to CR between $^5D_4$ and $^5D_3$ states. In the case of 488 nm excitation, a saturation behavior of the green emission is observed at intensities two orders of magnitude smaller than expected for bleaching of the ground state population (see Fig.1.a). Using a rate equation model[1] we show that, this behavior can be explained by an excited state absorption cross section ($\boldsymbol{\sigma_{esa}}$) two orders of magnitude larger than the ground state absorption ($\boldsymbol{\sigma_g}$), which is a spin forbidden transition. A strong concentration quenching of emission from $^5D_3$ state was observed in the decay times and integrated emission blue intensity ($\mathbf{I_B}$). Fig. 1.b shows this behavior. We then concluded that CR is not the only loss mechanism responsible for the $^5D_3$ depopulation and others mechanisms involving upper lying states (4f5d, charge transfer, host matrix, defects, etc) should play a significant role[1].

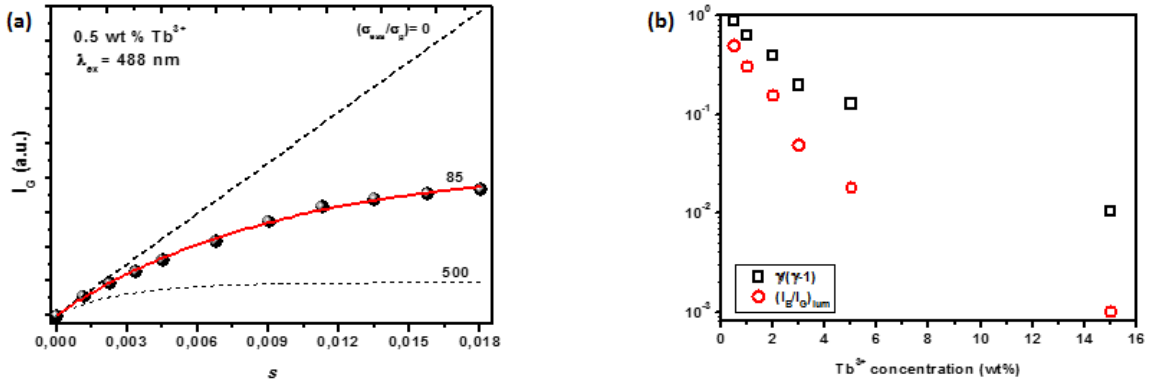

Figure 1. (a) Experimental data of the green emission (from $^5D_4$ level) dependence on pump power and pump saturation parameter (*s*), under 488 nm excitation, for the 0.5 wt% of Tb^{3+} sample. (b) The comparison between the integrated luminescence ratio, (I_B/I_G), with (γ/γ-1), as a function of Tb^{3+} concentration (0.5 – 15.0 wt%), where γ is the $^5D_4$ quantum efficiency. The ratio of the integrated emission intensities of the $^5D_3 \rightarrow {}^7F_j$ transitions to the $^5D_4 \rightarrow {}^7F_j$ transitions (where J = 3-6), referred as the blue-to-green luminescence intensity ratio (I_B/I_G), for different Tb^{3+} concentrations.

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[2] A.D. Sontakke and K. Annapurna, *Spectrochim. Acta A.* **2012**, 94, 180