Physical origin of refractive index changes in Tb^{3+} doped glasses

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It is now well known that the refractive index change in laser materials, when the excited state is pumped, due to the polarizability difference ($\Delta\alpha_p$) between ground and excited states. This effect has been studied in many photonic materials (crystals, glasses and fibers) doped with rare earth (RE) and transition metal ions[1,2]. The purpose of this paper is investigate the physical origin of this very high nonlinearity observed in Tb^{3+} doped low calcium aluminosilicate glasses (LSCAS) and its connection with 4f→5d transitions. The time-resolved Z-scan technique were performed using an Ar^+ laser at 488nm. The sample compositions, in wt%, are given by: (47.4-x/2)% CaO + (41.5- x/2)% Al_2O + 4.1% MgO + 7.0% SiO_2 + x% Tb_4O_7, with x = 0.2; 0.5; 1.0; 2.0; 3.0; 5.0 and 15 wt.%.

The refractive index change is given by $\Delta n \sim n_2 I$, where n_2 is proportional to $\Delta\alpha_p$. For Tb^{3+} doped LSCAS glasses $\Delta\alpha_p \sim 10^{-25} cm^3$ was obtained, where $\Delta\alpha_p$ refers to the difference between the excited state ($^5D_4$) and ground state ($^7F_6$) polarizabilities. This value is the highest value ever reported for a RE ion, one order of magnitude larger than observed in Nd^{3+}, Er^{3+} and Yb^{3+} doped materials[2,3]. In principle, the polarizability of a particular state can be calculated by the sum over all transitions contributions. The oscillator strength of 4f→5d transitions are typically 3-4 orders of magnitude higher than 4f→4f transitions. Thus the 4f→5d transitions should give the main contribution to $\Delta\alpha_p$[2,3]. Therefore, the high nonlinearities observed in Tb^{3+} doped glasses are probably due to the lower energy of the 4f→5d transitions of Tb^{3+} (~35.000cm^{-1}). This band was observed in the excitation spectra of Tb^{3+}, as shown in Fig.1a. The linear behavior of $^7F_6 \rightarrow {}^5D_4$ (480 nm) peak area with Tb^{3+} concentration was observed. Normalizing the area of 275nm band by area of the 480 nm peak, the $4f^75d^1$ peak area decrease with Tb^{3+} concentration, as shown in Fig.1b. It is interesting remark this behavior is very similar to the decrease of $\Delta\alpha_p$ with concentration. The strong correlation between $\Delta\alpha_p$ and the area of the $4f^8 \rightarrow 4f^75d^1$ peak in the excitation spectrum is presented for several Tb^{3+} concentrations (in Fig.1.b). This is the first direct experimental evidence of a correlation between $\Delta\alpha_p$ and the $4f^75d^1$ band, in RE doped materials.

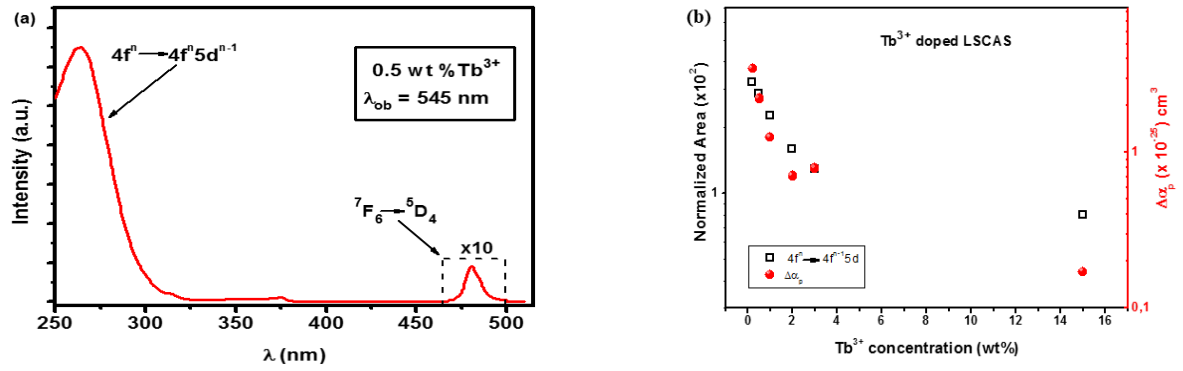

Figure 1. (a) Excitation spectra for λ_{em} = 544nm of Tb^{3+}-doped LCAS (b) The normalized $4f^75d^1$ and the $\Delta\alpha_p$ of Tb^{3+}-doped LSCAS (Tb^{3+}= 0.2, 0.5, 1.0, 2.0, 3.0 5.0 and 15 wt%)

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