Novel Calculation for Cross-Relaxation Energy Transfer Parameter Applied on Thulium Highly-Doped Tellurite Glasses

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ABSTRACT

We investigate the cross relaxation parameter of Tm³⁺ ions in tellurite glasses over a wide range of concentrations: from 0.36 mol% up to 10 mol%. We propose a new measurement approach based on monitoring the steady-state emission spectra. The proposed method is very simple and allows to measure even very highly doped samples. The cross-relaxation parameter shows a linear dependence with respect to dopant concentration over the full investigated interval and the measured slope is $1.81 \times 10^{-17} \text{ cm}^3 \text{ s}^{-1} \text{ mol}^{-1}$. The linear dependence suggests a dipole-dipole interaction.

Keywords: Rare-earth, Cross-relaxation, Thulium

1. INTRODUCTION

Among rare-earth doped lasers Thulium laser is an excellent candidate for infrared domain applications thanks to its peculiar and broad emission spectrum at around 1.8 micron [1]. This makes this kind of laser very appealing for several application ranging from precise cut and ablation of biological tissues to LIDAR and sensing applications [2-5]. A further advantage relies on pumping process where cross-relaxation mechanism produce two excited Tm ions out of one pump photon absorbed [6]. For this reason the knowledge of cross-relaxation mechanism is of the outmost importance to properly model and design the laser. So far several methods have been proposed to measure the cross-relaxation parameter, usually based on lifetime measurements [7,8] but also on numerical fitting of fluorescence dynamic versus pump power [9]. In all cases was not possible to define the value of cross-relaxation parameter over a wide range of Tm concentration due to the fact that for very high doping level the pump level is strongly quenched by cross-relaxation and fast detectors are needed. In this paper we investigate the cross-relaxation parameter over a wide range of the doping level up to 10 mol%. To overcome limitation of previous measurement methods we suggests a new method to calculate the cross-relaxation parameters based on steady-state fluorescence measurement. The investigation was done using a set of sample of Tm-doped tellurite glasses. The choice of this glass host was based on the facts they have the lowest phonon energies ($\sim 750 \text{ cm}^{-1}$) of all oxides glasses, which lead to increase in optical efficiency and decrease in probability of non-radiative multiphonon decay as well as an excellent rare earth ions solubility comparing with silicate and germanate glasses. The above advantages explains why Tm-doped tellurite glass fibre lasers are of a great interest [10-12].

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2. MODELLING

Figure 1 energy-level diagram of the Tm^{3+} ion [9] pumped at 790 nm to the $^3\text{H}_4$ level. The $^3\text{F}_4 \rightarrow ^3\text{H}_6$ emission of the neighboring ion is due to the cross-relaxation process: $^3\text{H}_4, ^3\text{H}_6 \rightarrow ^3\text{F}_4, ^3\text{F}_4$ as indicated in Fig. 1

Figure. 1. Simplified energy level diagram of Tm^{3+} ion and scheme of the cross-relaxation

The corresponding rate-equation system, considering empty $^3\text{H}_5$ level, used to analyze our experimental data is:

$$\frac{dN_0}{dt} = -W_{03}N_0 + \frac{N_3}{\tau_{30}} + \frac{N_1}{\tau_1} - C_R N_3 N_0 \quad (1)$$

$$\frac{dN_1}{dt} = \frac{N_3}{\tau_{31}} - \frac{N_1}{\tau_1} + 2C_R N_3 N_0 \quad (2)$$

$$\frac{dN_3}{dt} = W_{03}N_0 - \frac{N_3}{\tau_{30}} - \frac{N_3}{\tau_{31}} - C_R N_3 N_0 \quad (3)$$

where N_0 , N_1 , and N_3 are the populations of Tm^{3+} ions in the $^3\text{H}_6$, $^3\text{F}_4$, and $^3\text{H}_4$ levels respectively, W_{03} is the pump rate (s^{-1}) and C_R the cross relaxation parameter, τ_{xy} is the lifetime of the x to y transition and τ_x is the lifetime of level x . Note that $\tau_3 = (\tau_{31}^{-1} + \tau_{30}^{-1})^{-1}$. The C_R parameter is a function of the Tm doping level. At steady state the time derivatives in the rate equations are equal to zero. Considering the conservation law of Tm^{3+} ions populations, we can also write $N_t = N_0 + N_1 + N_3$. The methods based on fluorescence lifetime measurements [7,8] uses equation 3 and calculate the cross-relaxation parameter from level 3 lifetime quenching. However lifetime values falls below 1 μs for highly-doped samples and measurements of low level signals is difficult. To overcome the limitation of previous methods we propose to investigate the steady-state emission from $^3\text{H}_4$ and the $^3\text{F}_4$ levels. In steady-state condition from Eq. 2 follows:

$$C_R = \frac{1}{2N_0} \left[-\frac{1}{\tau_{31}} + \frac{N_1}{N_3\tau_1} \right] \quad (4)$$

The ratio between the population of level 1 and 3 can be rearranged by using the Fuchtbauer-Landenburg rule [13]

$$\sigma_{e,ij}(\lambda_{p,ij}) = H \frac{\Phi_{ij}(\lambda_{p,ij})}{N_i \Delta\nu} \quad (5)$$

and the relationship that provides the amount of the spontaneous emission [14].

$$\sigma_{e,ij}(\lambda_{p,ij}) = K \frac{\lambda_{p,ij}^4}{A_{i,j,norm} n^2(\lambda_{p,ij}) \tau_{ij}} \quad (6)$$

where σ_{ij} , $A_{ij,norm}$, τ_{ij} are the emission cross section, the area of emission spectrum normalized to the maximum and the transition radiative lifetime of the transition from level i to level j ; respectively. The parameters H and K are constants independent from the transition, n is the glass refractive index at the transition wavelength; Φ_{ij} is the number of photons emitted at peak wavelength in the frequency interval $\Delta\nu$, N_i is the excited ion population, and $\lambda_{p,ij}$ is the emission peak wavelength of the transition labeled as 'ij'. By combining the two above written equations we have:

$$\frac{N_i}{\tau_{ij}} \propto \frac{\Phi_{ij}(\lambda_{p,ij})}{\Delta\nu_p \lambda_{p,ij}^4} A_{ij,norm} n^2(\lambda_{p,ij}) \quad (7)$$

After substituting into Eq.4 a final form was found:

$$C_R = \frac{1}{2N_0 \tau_{31}} [kR - 1] \quad (8)$$

Where R is the ratio of the emission spectra from and 'k' contains all other experimental constant. In our case k value was 1.33.

3. EXPERIMENTS

All samples had the same host composition 75TeO₂-20ZnO-5Na₂O (mol%), labeled as TZN, and were doped with Tm³⁺ concentrations ranging from 0.36 mol% to 10 mol% [15,16]. We measured all relevant lifetime parameters by investigation the lowest doping level samples. To calculate the ratio R we excited Tm samples at the wavelength of 785 nm and we observed the emission from the two transition bands centered at 1.47 and 1.8 μ m, which corresponding to ³H₄ → ³F₄ and ³F₄ → ³H₆ respectively. Figure 2 shows the experimental set-up. The collecting lens had a relatively small numerical aperture in order to decrease the effect of self-trapping. Focusing was made on glass edge but we tested that focusing up to 0.6 mm inside the bulk sample did not affect the measured lifetimes.

Figure. 2. Schematic of the set-up layout

The lifetime τ_0 for $^3\text{H}_4$ was 0.37 ms whilst for $^3\text{F}_4$ equal to 2.7 ms. For the samples doped with concentrations higher than 4 mol% the lifetime of $^3\text{H}_4$ level was affected by the response time of our photodiode that is of about few microsecond.

Emission spectra results are shown in Fig. 3 where we normalized all curves to the peak of $^3\text{H}_4 \rightarrow ^3\text{F}_4$ transition at 1.47 μm .

Figure. 3 Emission spectra, inset magnifies the of $^3\text{H}_4 \rightarrow ^3\text{F}_4$ transition at 1.47 μm .

The corresponding ratio to be used in Eq.8 is

Figure. 4 Ratio between the emission from $^3\text{F}_4$ and $^3\text{H}_4$ levels.

Figure 5 shows the cross-relaxation parameter values calculated by using Eq. 8. We can note a quite clear linear increase even for highest doped samples where, however the slope is slightly reduced.

Figure. 5. Cross-relaxation parameter versus Tm concentration.

From the experimental data the fitted CR is $1.81 \times 10^{-17} \text{ cm}^3 \text{ s}^{-1} \text{ \%mol}^{-1}$ is obtained. While this value is below the one previously reported in TZN glass [9] it is consistent with the one used for silica fiber modeling [17]. The observed linear dependence on concentration of the cross-relaxation parameter can be explained with the behavior of the concentration quenching of the lifetimes, as predicted by Auzel in the framework of dipole-dipole interaction processes [18]. We compared our results with values obtained from Eq.3 [7] and up to 4 mol% sample the agreement was within 10%. We believe that the data here reported will be useful to model and design laser in TZN glasses. This measurement method can be easily extended to other type of glasses.

4. CONCLUSIONS

A new and simple approach based on the emission spectra measurements was developed to investigate cross-relaxation parameter in Tm-doped samples with very high doping levels. Thanks to this new method we were able to experimentally calculate the cross relaxation parameter of Tm³⁺ ions in tellurite glasses over a wide range of concentrations. This approach demonstrated to be very sensitive and can be used to investigate other kind of Tm-doped glasses. The obtained values of cross-relaxation show a linear dependence with dopant concentration and value of the linear fit slope was $1.81 \times 10^{-17} \text{ cm}^3 \text{ s}^{-1} \text{ \%mol}^{-1}$. The knowledge of this value allows a proper modeling of Tm-doped tellurite glass laser by properly comparing the impact of different doping level.

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