Quantum efficiency of $^4F_{3/2}$ level and loss mechanisms in Nd^{3+}-doped phosphate laser glass

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Abstract: We have investigated the quantum efficiency values using a new Thermal Lens approach based on the $^4F_{3/2}$ lifetimes value in Nd-doped phosphate laser glass, Q-98 glass. The losses mechanisms including Auger upconversion are also studies through conventional spectroscopy and energy transfer theories.

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Nd-doped glasses have been extensivelly investigated as host material for 1.06 ?m laser systems. In this way, it is essential to determine of quantum efficiency (?) values and non radiative losses from the laser level ($^3F_{3/2}$) related to the energy transfer process. In the last years, Thermal Lens (TL) spectrometry has been used for ?? determination in rare earth doped host materials by reference sample and multiwalength methods [1]. In this work, we obtained the ? values using a new TL approach based on fluorescent lifetime of $^4F_{3/2}$ level as a function of Nd concentration. This method was applied in Nd-doped phosphate laser glass (Q-98) developed by *Kigre,Inc*. To our knowledge is the first time that ? values of $^4F_{3/2}$ level are determined for Q-98 glass. Moreover, the losses mechanisms were also studies through TL, conventional spectroscopy and energy transfer theories.

The TL measurements were performed in the dual-beam mode-mismatched TL configuration [2]. The samples were excited by a Ti:Al_2O_3 laser. Absorption, near infrared luminescence and lifetime measurements were also performed. The TL method is based on multiphonon decay processes in a doped sample. The transient signal amplitude of the probe laser beam through the TL is proportional the phase shift, ?? = ?.P_{abs}, where P_{abs} is the absorbed pump power. The ? values can be written as a function of $^4F_{3/2}$ lifetime values (?$_{exp}$(N)) as following, ? ? ?[1-(<?$_{em}$>/??$_{exc}$)? ???r(N)], where r(N) = ?$_{exp}$(N)/ ?(0) =?? ?? ??? ?????<?$_{em}$> and ?$_{exc}$ are the average emission and excitation energy, respectively and ?????is the quantum efficiency in a sample with Nd low concentration. Therefore, the linear fit of ? versus r(N) provide the ?????value, and consequently ??? ?. Fig. 1(a) shows that the TL signal decrease linearly with r(N), as expected. In addition, the ??values obtained from TL technique and radiative lifetimes (Judd-Ofelt theory) are depicted in Fig. 1(b). As can be seen, the values achieved using TL technique agrees very well with JO results.

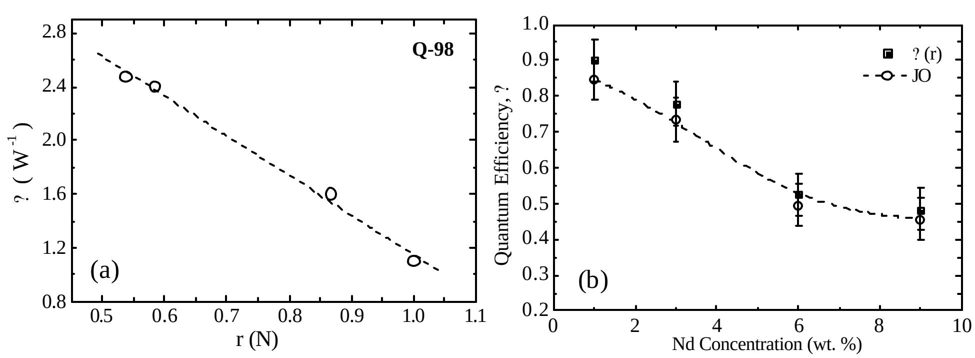

Fig. 1. (a) Plot of the normalized TL amplitude (?) as function of r(N) and (b) Quantum efficiency vs. Nd concentration in Q-98 glasses.

The non radiative energy losses were also studied by Dexter energy transfer theory [3]. The energy transfer micro-parameters for dipole-dipole interaction related to the energy migration (C_{DD}) (EM) and cross relaxation (C_{DA}) (CR) from $^4F_{3/2}$ level were obtained from the absorption and luminescence spectra. According to our calculation the $^4F_{3/2}$,$^4I_{9/2}$? $^4F_{15/2}$,$^4I_{13/2}$ (CR1) process is the most relevant loss mechanism from $^4F_{3/2}$ in contrast with several works that designate the $^4F_{3/2}, ^4I_{9/2} ? \ ^4F_{15/2}, ^4I_{15/2}$ (CR2) mechanism ($C_{DA}^{CR1} / C_{DA}^{CR2} > 100$) . The ? ?dependence on Nd^{3+} ion concentration can be explained by the energy transfer micro-parameters related to both CR1 and EM mechanisms. The ? values were determined by means of the expression given by

$$? \; ? \; \frac{?_0^{?} I(0) e^{?(W_r ? W_{mp})t ? ? \sqrt{t} ? W t} dt}{?_0^{?} I(0) e^{? W_r t} dt}$$

In which W_r and W_{mp} are the radiative and multiphonon rates, ? depends on C_{DA}^{CR1} and W depends on both C_{DA}^{CR1} and C_{DD}^{EM} [4].

Recently, it was demonstrated that the TL technique could be used on upconversion coefficient (??) determination [5]. Therefore, the losses associated to the energy transfer upconversion mechanisms in Q-98 glasses were obtained through TL technique. From pump-probe technique the excited state absorption ($?_{ESA}$) and stimulated-emission ($?_{SE}$) cross section spectra of Q-98 glass were determined. Two upconversion channels clearly observed and assigned as $^4F_{3/2}, ^4F_{3/2} ? \ ^4F_{13/2}, (^4G_{7/2}+^2K_{13/2}+^4G_{9/2})$ and $^4F_{3/2}, ^4F_{3/2} ? \ ^4I_{11/2}, (^4G_{9/2}+^2D_{3/2}+^4G_{11/2}+^2K_{15/2})$. The micro-parameters ($C_{DA}$ and C_{DD}) related to these upconversion processes were also calculated using the spectral overlap between donor emission and acceptor absorption. A good agreement was obtained between the experimental ??values obtained from TL measurements and the calculated by Burshstein migrationally-assisted upconversion rate, given by $W_B ? (C_{DA} C_{DD})^{1/2} N_{ex} N_{Nd}$, where N_{Nd} is total ion and N_{ex} is the excited state population .

In summary, the quantum efficiency values of the laser level were measured by TL method based on $^4F_{3/2}$ lifetime values. It was determined the high quantum efficiency value (~90%) in Q-98 glass with 1 Nd_2O_3 (wt.%). Besides, we verified that CR1 mechanisms is responsible for the quenching concentration instead of CR2 relaxation in Nd-doped host systems since CR2 mechanism involves the annihilation of phonons from host lattice and $^4F_{3/2} ? \ ^4I_{15/2}$ emission has a low emission cross section value. The losses related to the energy transfer upconversion mechanisms were also estimated.

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