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EVALUATION OF MICROSCOPIC PARAMETERS FOR ETU PROCESS IN DIODE-PUMPED Nd : YLF

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The visible up-conversion fluorescences from the $^4G_{7/2}$ level in Nd-doped $LiYF_4$ crystals have been studied at 300 K under c.w. diode laser pumping at 797 nm. These emissions originate from either two-step excitations, involving an excited-state absorption from the $^4F_{3/2}$ metastable level or from energy transfer up-conversion (ETU) processes. Assuming a dipole–dipole interaction between two Nd^{3+} ions in the excited state separated by the distance R, one can obtain the microscopic ETU rate given by $K_{D-A}(R) = (C_{D-A}/R^6)$, where C_{D-A} is the transference constant (cm^6/s). A model to estimate the probability of the ETU process based on the average of microscopic probability rate derived from Förster–Dexter method, was proposed.

Keywords: Diode-pumped lasers; Laser spectroscopy; Energy transfer

1 INTRODUCTION

All solid state Nd laser systems pumped by high power diode lasers have high efficiencies and are now standard tools for many applications [1,2]. In particular the Nd-doped $YLiF_4$ (YLF) crystal under diode pumping has been intensely investigated. On the other hand, high power – high brightness diode lasers are efficient sources to promote non-linear effects in laser media, as the up-conversion fluorescence. Most of the

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processes rely either on excited-state absorption (ESA) or energy-transfer up-conversion (ETU). Recently Chuang *et al.* [3] observed yellow and blue fluorescences induced by laser excitations in the range of 785–811 nm in Nd : YLF.

In this paper, we report the determination of ETU parameters of Nd-doped YLF crystals excited with high-power diode laser at 797 nm. These processes constitute an important source of loss mechanisms in Nd laser systems, which is not well characterized in the literature.

2 EXPERIMENTAL RESULTS

The crystals were grown by Czochralski technique. The studied Nd concentration was 0.6 mol% ($8.50 \times 10^{19}\,cm^{-3}$) for the YLF crystal. The infrared pumping was performed with a GaAlAs laser diode, model SDL-2382-P1, operating at 797 nm. The luminescence signal was processed using a PAR lock-in amplifier. The lifetimes of excited Nd^{3+} ions were measured using a pulsed laser excitation (10 ns) from a nitrogen laser pumped dye laser tuned to 413 nm. The time-dependence signal was detected by a fast S-20 extended-type photomultiplier detector and analyzed using a signal-processing Box-Car averager (PAR 4402).

Under pumping with 797 nm, the Nd ions are excited to $^4F_{5/2}$ manifold, from which they rapidly relax to the metastable $^4F_{3/2}$ level, that has a fluorescence lifetime of 538 μs for YLF. A second photon of 797 nm can excite few Nd ions from $^4F_{3/2}$ level to the phonon sideband of the $^2P_{1/2}$ manifold, than relaxing mainly to the $^4G_{7/2}$ level. Another excitation can be induced by a 797 nm photon, exciting the $^2P_{3/2}$ level, being one of the possibilities is due to the $^4F_{5/2} \rightarrow {}^2P_{3/2}$ transition (Fig. 1). There are also the ETU processes involving two excited Nd^{3+} ions, leaving one ion in either the $^4G_{7/2}$ manifold or the $^2G(1)_{9/2}$ multiplet and the other ion in the states $^4I_{13/2}$ or $^4I_{11/2}$, respectively.

The ETU process, in which an excited ion transfers non-radiatively its energy to an already excited neighboring ion, is one of the most efficient up-conversion mechanisms. This process is probably weak in the considered system since the sample was doped by 0.6 mol% Nd^{3+} ions. In this paper we quantify the ETU rate. Assuming a dipole–dipole interaction between two Nd^{3+} ions in the excited state separated by the distance R, the microscopic ETU rate is given by $K_{D-A}(R) = (C_{D-A}/R^6)$,

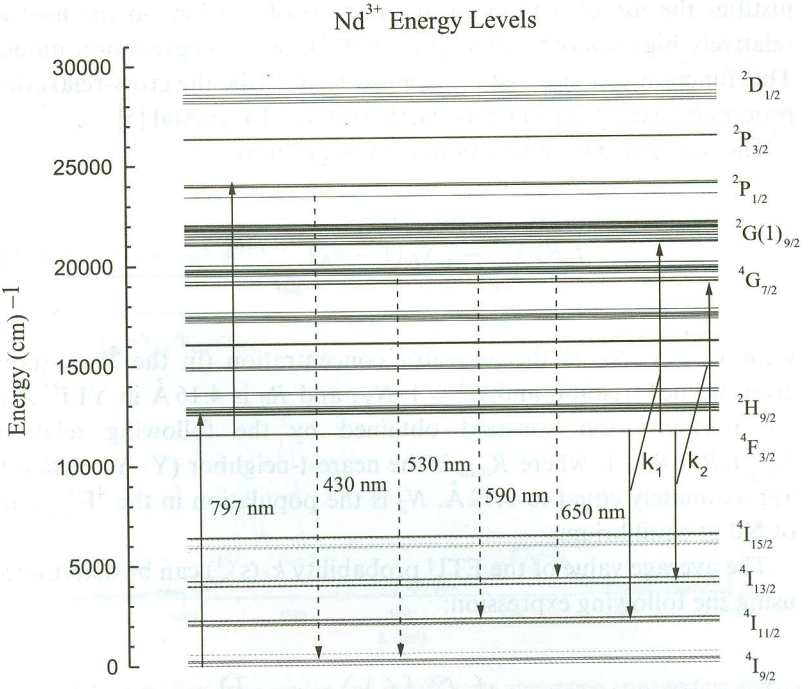

FIGURE 1 Energy level diagram of Nd, showing the up-conversion processes under diode laser pumping at 797 nm and anti-Stokes emissions observed.

where C_{D-A} is the transference constant (cm^6/s) [4] for two ETU processes:

$$(k_1) \quad {}^4F_{3/2} \rightarrow {}^4I_{13/2} : {}^4F_{3/2} \rightarrow {}^2G(1)_{9/2}$$
$$(k_2) \quad {}^4F_{3/2} \rightarrow {}^4I_{11/2} : {}^4F_{3/2} \rightarrow {}^4G_{7/2}$$

Considering a random ion distribution in the crystal, it is possible to define the function which expresses the fraction of finding Nd pairs as the closest neighbor inside of a shell between the distances R and $R + dR$. Since the $4f^N$ orbital of the (3+) rare-earth ions is always shielded by the most external fulfilled orbital ($5p^6$), one expects a negligible interaction between them in the ground state which, in turn, produces no tendency for either preference or rejection of closer pair configurations away from statistical distribution. This argument

justifies the use of a random distribution of Nd ions in the host at relatively high concentration ($x_A > 0.005$), in the deexcitation model. This function was successfully applied to describe the cross-relaxation process between trivalent rare-earth ions in YLF crystal [5].

The ion distribution function, $f(R)$, is given by:

$$f(R) = \frac{4\pi R^2}{AR_0^3} x_A (1 - x_A)^{4\pi R^3/3R_0^3 - 2} \tag{1}$$

were $x_A = N_2/N_Y$ is the activator concentration (in the $^4F_{3/2}$ state) given in mol fraction and $R_0^3 = 1/N_Y$, and R_0 is 4.16 Å in YLF. A is the normalization constant obtained by the following relation: $\int_{R_{min}}^{\infty} f(R)\,dR = 1$, where R_{min} is the nearest-neighbor (Y–Y) distances approximately equal to 3.72 Å. N_2 is the population in the $^4F_{3/2}$ state of Nd at equilibrium.

The average value of the ETU probability k_i (s^{-1}) can be determined using the following expression:

$$k_i = C_6 \left\{ \int_{R_{min}}^{\infty} \left[\left(\frac{1}{R^6} \right) f(R)\,dR \right] \right\} \tag{2}$$

$C_{D-A} = C_6 = R_c^6/\tau_D$, where τ_D is the donor lifetime of $^4F_{3/2}$ level. R_c is the critical distance for which excitation transfer and intracenter decay of the sensitizer have equal probability. R_c can be written as

$$R_c^6 = \tau_D \left(\frac{1}{2\pi} \right)^4 \left(\frac{6c}{n^2} \right) \frac{g_l}{g_u} \int \sigma^*(\lambda) \sigma_e(\lambda)\,d\lambda \tag{3}$$

where σ^* (λ) [6] and $\sigma_e(\lambda)$ are the excited state absorption cross section and the emission cross-section normalized line shape functions, respectively, (Fig. 2, Table I), n the refractive index and g_l and g_u are the level degeneracies. By substituting $f(R)$ in Eq. (2), one obtains k_i and subsequently $K_{ETU} = \Sigma k_i$, and the points were fitted by a linear function of the inverted population (N_2), given by: $K_{ETU} = -25.28 \times 10^{-15} N_2$ (for Nd : YLF). K_{ETU} is given in s^{-1}.

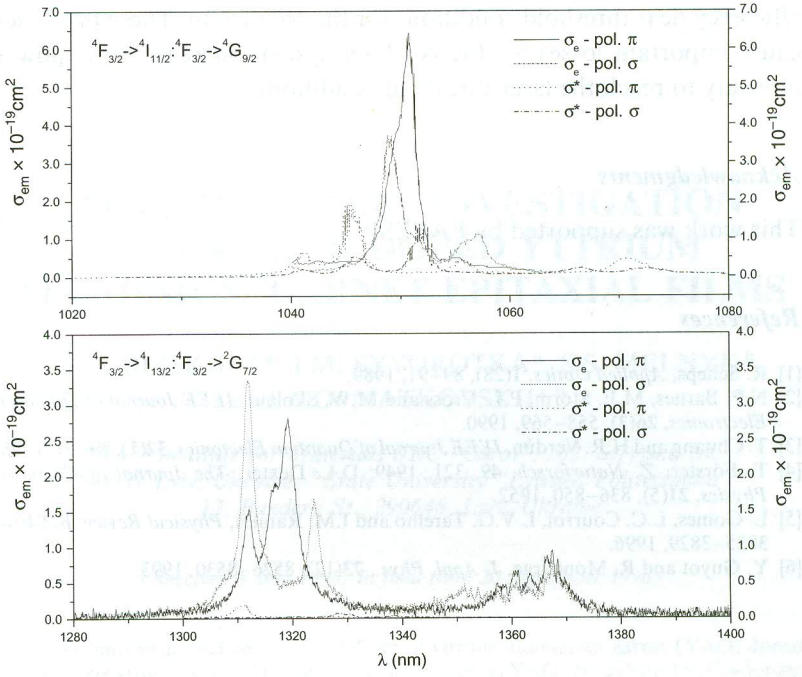

FIGURE 2 Emission cross-section and excited state absorption cross-section spectra in the range of 1050 and 1300 nm for the YLF : Nd.

TABLE I Relevant spectroscopic parameters for upper laser level population calculation, accounting for the up-conversion losses, in Nd : YLF

Nd–Nd interaction	C_{D-A} (cm^6/s)	R_c (Å)
($^4F_{3/2}$/$^4I_{11/2}$) – 1.05 μm	1.36×10^{-37}	20.4
($^4F_{3/2}$/$^4I_{13/2}$) – 1.3 μm	1.83×10^{-37}	21.4

3 CONCLUSION

A model to estimate the probability of the ETU process based on the average of microscopic probability rate derived from Förster–Dexter method was proposed and the ETU parameters were calculated. With these parameters and solving the rate equations for the system under continuous pumping, it was possible to estimate the up-conversion

efficiency near threshold conditions for the Nd system. These processes cause important losses for the Nd laser system, increasing the power intensity to reach the laser threshold conditions.

Acknowledgments

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