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Up-conversion and excited state absorption studies of Nd³⁺ doped Y₃Al₅O₁₂, YLiF₄ and LaMgAl₁₁O₁₉

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1. Introduction

Excited-state absorption (E.S.A.) in the rare earth ion doped crystals is a determining parameter for their laser potentialities. It leads to optical transitions which can reduce the laser efficiencies and increase the oscillation thresholds very significantly when they occur in the ground state absorption and/or the stimulated emission spectral domains. In some cases, it can preclude laser emission completely. It can also induce supplementary multiphonon relaxations and increase the temperature of the laser materials [1].

On the other hand, ESA can play a positive role, allowing laser action in the visible domain after pumping in the infrared for example, via so-called up-conversion and photon-avalanche processes; and it is precisely for that purpose that we have registered ESA spectra and measured ESA cross sections in the well known Nd³⁺ doped Y₃Al₅O₁₂, YLiF₄ and LaMgAl₁₁O₁₉ laser crystals around 470 and 600 nm. This has been done by using an experimental set-up in which the sample to be studied is a laser rod placed in a flash lamp pump chamber [2], inserted itself in a two-mirror resonator to measure E.S.A. in the presence or not of laser action. A particular attention has been addressed to YLiF₄:Nd³⁺ by registering ESA spectra at low as well as at room temperatures, because it is the laser system which is the more adequate for up-conversion laser operation [3], and by analysing the effect of up-conversion on its energy storage capacity via lifetime measurements.

In each case, the positions of the observed lines are compared with that deduced experimentally from the energy levels of the Nd³⁺ ion in these materials, and their cross sections are compared with that deduced theoretically within the framework of the JUDD-OFELT model.

2. Experimental results

The figures 1 to 3 show the ESA results obtained in the region around 600 nm. I_p and I_u stand for the transmitted intensities of the probe beam when the system is and is not pumped respectively. They are related to the ground state absorption (GSA) and ESA at a particular wavelength by the usual expression:

$$\ln(I_u/I_p) = (\sigma_{\text{esa}} - \sigma_{\text{gsa}}) N_1 d \quad (1)$$

in which N_1 is the excited state population density and d the length of the active medium. N_1 is determined precisely at a wavelength where $\sigma_{\text{esa}} = 0$ and $\sigma_{\text{gsa}} \neq 0$.

In the case of YAG:Nd³⁺ there are two sets of lines around 595 and 620 nm. The positions of the energy levels indicate that they can be assigned to ${}^4F_{3/2} \rightarrow {}^4D_{3/2}$ and ${}^4F_{3/2} \rightarrow {}^4D_{3/2}$ ESA transitions respectively. Every observed peak correspond to a well defined inter-Stark level optical transition, the one is indicated on the figure by a cross or a bar.

Similar results are obtained in the case of LMA:Nd and YLF:Nd, but for a probe light polarized either parallel (E//c) or perpendicular (E⊥c) to the optical axis of the crystal. The lines are substantially broader than in the case of YAG. They are assigned, without being so well separated than in the case of YAG, to the same types of inter-Stark level ESA transitions.

These spectra have been calibrated experimentally in units of cross sections and the values compared with those obtained theoretically by using the Judd-Ofelt theory. This is shown in the following table for the most important ESA peaks. The results are quite good in the case of YLF and LMA. They differ, however, by a factor 5 in the case of YAG. This discrepancy is attributed to an uncertainty in the calculated value of the Judd-Ofelt parameter Ω_2 , knowing that the intensity of the ${}^4F_{3/2} \rightarrow {}^4D_{3/2}$ ESA transition depends exclusively on this parameter.

Table: Comparison between the experimental and theoretical (JO) ESA peak cross sections derived in YAG:Nd³⁺, YLF:Nd³⁺ and LMA:Nd³⁺

	YAG	YLF		LMA	
λ_{esa} (nm)	619	603.6		603.3	599.8
$\sigma_{\text{esa}}^{\text{exp}}$ (10^{-20} cm^2)	9 ± 3	π 1.6 ± 0.6	σ 0.65 ± 0.2	σ ≈ 1.3	π ≈ 0.9
$\sigma_{\text{esa}}^{\text{exp}}$ (10^{-20} cm^2)	1.7 ± 0.3	2 ± 0.5	0.8 ± 0.2	≈ 1.47	≈ 1.06

We also report in Figure 4 ESA spectra recorded in YLF:Nd³⁺ at low and high temperatures around 600 nm (${}^4F_{3/2} \rightarrow {}^4D_{3/2}$ ESA transition). These preliminary data (to be confirmed) show a very curious behaviour and are of particular interest to compare with the data derived from the low temperature photon-avalanche up-conversion experiments recently performed [3]. Our results explain why similar ESA cross sections are found at low and high temperatures at 603.6 nm in the σ polarization and why the previous authors [3] could have triggered their photon avalanche process exclusively in this polarization, since the peak cross section of the ESA line at 603.6 nm seems to be about 30 times larger in the σ than in the π polarization. However we do not yet understand why the oscillator strength of this ESA transition seems to vary so much and why the relative intensity of the main line at 603.6 nm is reversed when going from low to high temperature.

In the end we show in the figures 5 and 6 the results of an experimental and theoretical analysis of the infrared fluorescence decays in YAG:Nd³⁺ and YLF:Nd³⁺ as a function of the excitation pump power assuming up-conversion in the metastable emitting state ${}^4F_{3/2}$.

According to this hypothesis the fluorescence decay shortens and

becomes non-exponential as the excitation pump power increases and it is easy to show that the $1/e$ apparent fluorescence time constant τ_e takes a value τ_e such as:

$$\frac{\tau_e}{\tau} = \ln \left[1 + \frac{e^{-1}}{1 + N_1(0)W\tau} \right] \quad (2)$$

Figure 5 show the experimental data obtained in the case of YLF and the fits of the model for three excitation pump powers labelled FP, GP1 and GP2. Figure 6 shows the expected behaviours in YAG and YLF using fluorescence time constants at low pump powers of 240 and 550 μs respectively. Up-conversion energy transfer rates W of $(2.8 \pm 0.8) 10^{-16} \text{ cm}^3 \text{ s}^{-1}$ and $(1.7 \pm 1) 10^{-16} \text{ cm}^3 \text{ s}^{-1}$ are found in YAG and YLF respectively.

The model fits well our data. It also well accounts for the laser measurements reported by Seelert et al [4] which derived a fluorescence lifetime of 410 μs in the case of a strongly diode pumped YLF crystal.

3. Conclusion

The results given here in this short communication show that ESA might be very important for laser action in case of flash lamp pumping. They also show that ESA can be modeled quite confidently by using the Judd-Ofelt theory. However, the up-conversion losses by energy transfers in the metastable emitting state are proved to be important too since they can be responsible for the limitation of the storage capacity of this level at high excitation pump powers. Complete data and more details on the experimental and theoretical methods will be presented in a subsequent paper [5].

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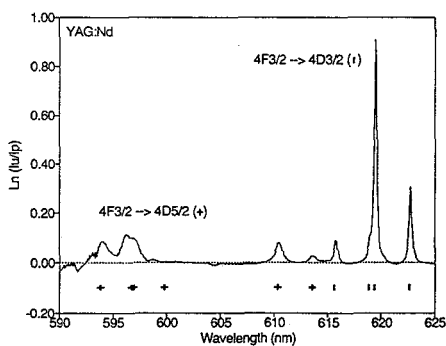


Figure 1: Room temperature ESA spectra of YAG:Nd^{3+} around 600 nm

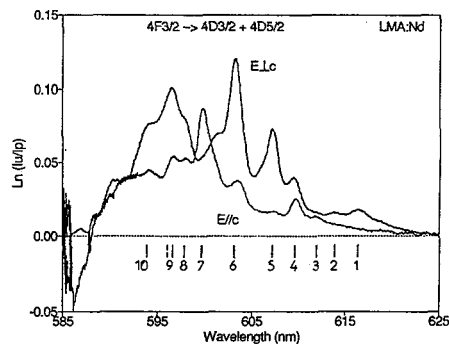


Figure 2: Room temperature polarized ESA spectra of LMA:Nd^{3+} around 600 nm

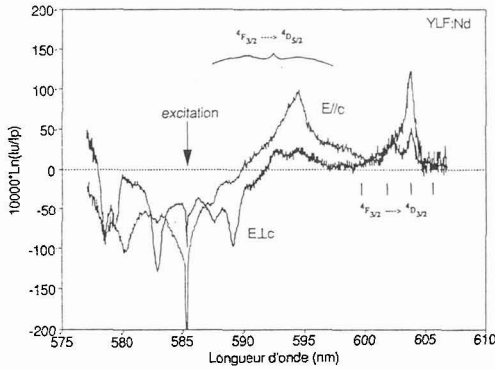


Figure 3: Room temperature polarized ESA spectra of YLF:Nd³⁺ around 600 nm

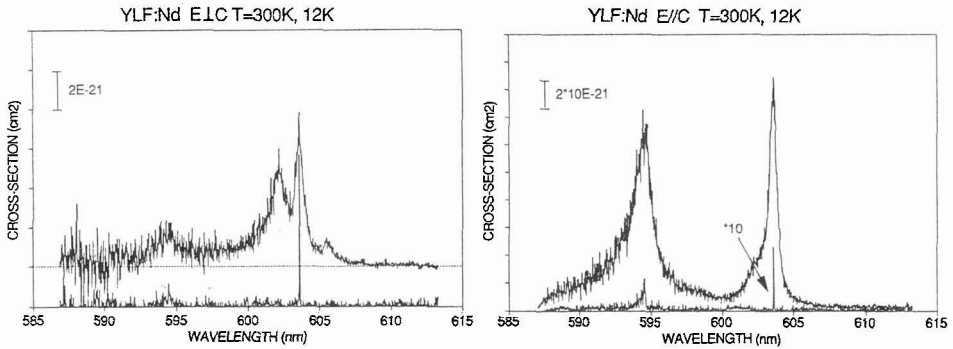


Figure 4: Polarized ESA spectra of YLF:Nd³⁺ recorded at low and high temperatures around 600 nm (preliminary data)

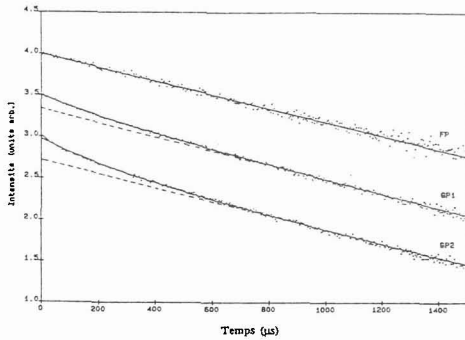


Figure 5: Infrared fluorescence decay of a 1% Nd³⁺ doped YLF crystal for different excitation pump powers FP << GP1 < GP2.

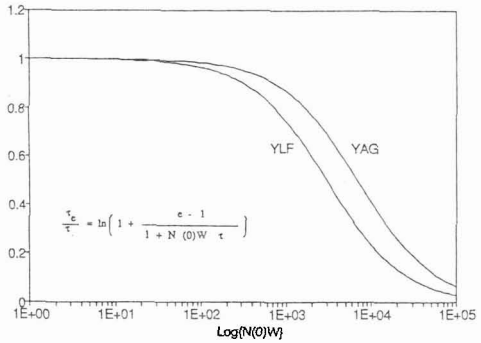


Figure 6: Theoretical variation of the τ_e/τ ratio versus $\log[N_1(0)W]$ in the case of Nd³⁺ doped YLF ($\tau = 550 \mu s$) and YAG ($\tau = 240 \mu s$)