FLUORESCENCE DYNAMICS AND ENERGY TRANSFER PROCESSES IN VARIOUS SOLID-STATE LASER MATERIALS BASED ON Er^{3+} AND Tm^{3+} IONS

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Abstract - A systematic study has been made of the fluorescence dynamics and energy transfer processes in several excited states of Er^{3+} and Tm^{3+} ions in various solid-state laser materials such as $\text{YSO}(\text{Y}_2\text{SiO}_5)$, $\text{YAG}(\text{Y}_3\text{Al}_5\text{O}_{12})$ and $\text{YLF}(\text{YLiF}_4)$. The purpose is to show and compare these properties in the different materials as a function of the dopant concentration and of the excitation conditions.

The importance of the Er^{3+} and Tm^{3+} ions in the development of the diode-pumped solid-state lasers for eye-safe and other applications is now well-known. The Er^{3+} doped materials give rise to laser emissions around $1.55\text{-}1.65\mu\text{m}$ and they can be pumped by InGaAs laser diodes at around $0.98\mu\text{m}$ when they are codoped with Yb^{3+} ions. The Tm^{3+} doped materials usually emit a broadband infrared fluorescence around $1.8\text{-}2.1\mu\text{m}$ and they generally give rise to tunable laser action around $2\mu\text{m}$ when they are pumped with high power AsGa diodes at $0.8\mu\text{m}$. Knowing that the electronic energy levels of the rare earth ions (ex: $^4\text{I}_{13/2}$ in the case Er^{3+} ion) vary slightly from host to host-materials, only one example of absorption spectra is given in figure 1 and 2. The room temperature absorption spectra of YSO doped with Er^{3+} , Yb^{3+} and Tm^{3+} clearly show the regions of diode pumping.

The analysis of the influence of the dopant concentration and of the excitation conditions on the fluorescence dynamics in these materials allow us in particular to determine which of the inter-state relaxation processes, the non-radiative multiphonon relaxation or the cross-relaxation energy transfer processes, are dominant in each system, and depending on the nature of the active ions and their concentration, which are the most appropriate pumping schemes. The results for the Er^{3+} doped systems are reported in table 1 and that for the Tm^{3+}

doped systems in table 2. From these results the following comments can be made.

Exception made for the $^4I_{13/2}$ infrared emission^{<1>}, the fluorescence decays in YSO Er^{3+} doped do not vary much with the Er^{3+} concentration compared to the case of YAG or YLF for which the fluorescence lifetimes are longer and decrease much faster, by a factor 5 to 10 in the case of YAG, and more than a factor 50 in the case of YLF. From these results it can be concluded that the non-radiative multiphonon relaxation predominate in YSO whereas the energy transfers (up-conversion as well as cross-relaxation) predominate in YLF. YAG represents an intermediate case.

In the case of the Tm^{3+} doped systems, the fluorescence decays always vary strongly with the dopant concentrations. In particular the $^3\text{H}_4$ excited state deexcitation is dominated by a cross-relaxation type energy transfer which depends on the square of the dopant concentration. However the risetime of the $^3\text{F}_4$ infrared fluorescence after $^3\text{H}_4$ excitation is systematically equal to half the lifetime of the $^3\text{H}_4$ fluorescence decay; this indicates the occurrence of a very efficient pair process involving the deexcitation of coupled adjacent Tm^{3+} ions both in $^3\text{F}_4$ excited-states.

Fig.1 Room temperature absorption spectrum of a $\text{Y}_2\text{SiO}_5: 10\% \text{Yb}^{3+}, 0.5\% \text{Er}^{3+}$ single crystal (thickness 1,7mm) in the $\langle 010 \rangle$ polarization

Fig.2 Room temperature absorption spectrum of a $\text{Y}_2\text{SiO}_5: 10\% \text{Tm}^{3+}$ single crystal (thickness 1,3 mm)

Table1. Apparent fluorescence lifetimes ($1/e$ values) in various Er^{3+} -doped systems at room temperature, after $^4\text{S}_{3/2}$ excitation

Excited levels	$\text{YSO} : \text{Er}^{3+}$			$\text{YAG} : \text{Er}^{3+}$			$\text{YLF} : \text{Er}^{3+}$	
	1 %	5 %	10 %	1 %	5 %	10 %	1 %	30 %
$^4\text{S}_{3/2}$	$3.7\mu\text{s}$	$2.6\mu\text{s}$	$1.6\mu\text{s}$	$20\mu\text{s}$	$9.2\mu\text{s}$	$3.8\mu\text{s}$	$360\mu\text{s}$	$3.1\mu\text{s}$
$^4\text{F}_{9/2}$	$3.2\mu\text{s}$	$2.6\mu\text{s}$	$1.5\mu\text{s}$	$42\mu\text{s}$	$10.6\mu\text{s}$	$5.6\mu\text{s}$	$350\mu\text{s}$	$3.2\mu\text{s}$
$^4\text{I}_{9/2}$	$3.3\mu\text{s}$	$2.4\mu\text{s}$	$1.5\mu\text{s}$	$30\mu\text{s}$	$10\mu\text{s}$	$3.3\mu\text{s}$	---	$6.3\mu\text{s}$
$^4\text{I}_{11/2}$	$3.5\mu\text{s}$	$2\mu\text{s}$	$1.3\mu\text{s}$	$12\mu\text{s}$	$7.1\mu\text{s}$	$2.7\mu\text{s}$	$200\mu\text{s}$	$5.6\mu\text{s}$
	$18\mu\text{s}$	$18\mu\text{s}$	$18\mu\text{s}$	0.8ms	1ms	1.2ms	4.6ms	5.3ms
$^4\text{I}_{13/2}$	$16\mu\text{s}$	$16\mu\text{s}$	$15\mu\text{s}$	$97\mu\text{s}$	$86\mu\text{s}$	$76\mu\text{s}$	$330\mu\text{s}$	$4\mu\text{s}$
	8ms	7.5ms	4.3ms	6.6ms	6.8ms	6.6ms	17ms	13ms

Table2 . Apparent fluorescence lifetimes ($1/e$ values) in various Tm^{3+} doped systems at room temperature, after 3F_2 , 3F_3 excitation

Excited Levels	YSO : Tm^{3+}			YAG : Tm^{3+}			$YLF:Tm^{3+}$
	1 %	5 %	10 %	1 %	5 %	10 %	1 %
3H_4	80 μ s	7 μ s	1.8 μ s	465 μ s	35 μ s	8.5 μ s	1.45ms
3F_4 $\tau \nearrow$	40 μ s	3.6 μ s	1 μ s	226 μ s	21 μ s	4.5 μ s	735 μ s
3F_4 $\tau \searrow$	1.7ms	1.1ms	0.5ms	11.3ms	8.6ms	4.6ms	22ms

Reference:

/1/ C.LI, C.WYON and R.MONCORGE, special issue of the IEEE J. Quant. Elect. on diode pumped solid-state lasers. Guest ed. F.Welsh and T.Y.Fan (Jan. 1992)

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