UPCONVERSION DYNAMICS IN ERBIUM DOPED FLUORIDE GLASSES

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The upconversion dynamics of Er ($^4S_{3/2}$, $^4F_{9/2}$) fluorescences were investigated by time resolved spectroscopy in Er doped fluorozirconate and fluoroindate glasses [1,2] with Er concentration varying from 1 to 20 %. The Er ions were selectively excited by a frequency doubled Nd:YAG pumped OPO pulsed laser tuned in several fundamental absorptions: 1.5 μm, 0.98 μm and 0.78 μm. There are two mechanisms responsible for the green emission: the Excited State Absorption (ESA) exhibiting a fluorescence rise time which follows the short pulse pumping, and the Energy Transfer Upconversion (ETU) process with a much longer rise time due to the non radiative energy transfer between two excited Er ions [3,4]. We also observed that the green emission has a long time decay component greater than radiative time when exciting with a laser intensity higher than 0.2 GW/cm^2. This effect was produced by a partially inhibition of the $^4S_{3/2}$ cross-relaxation process due to a lack of the available acceptor Er ($^4I_{15/2}$) ions in this highly excited volume. In this case, the conditions for energy migration through $^4S_{3/2}$ states are ideal [5], producing a new component with characteristic time decay longer than the expected radiative lifetime.

Introduction

The green upconversion emission of diode pumped Er systems is very interesting due to its applicability to several areas, but specially for the relevant information about the processes which take place in order to produce it. There are several discussions about the roles of ESA (excited state absorption) and ETU (energy transfer upconversion) in the production of this emission.

Some studies shown the possibility of discrimination between the two processes by time resolved sampling. The ESA process can occur only during pumping duration (theoretically instantaneous rise time) but ETU process time can rise slowly [3,4] as represented in Figure 1.

The importance of this kind of determination is related to the maximization of the slope efficiency of the 970 nm diode pumped 3 μm laser. The Stokes limit of $_{st}$=35% can be exceeded due to energy recycling into the upper laser ($^4I_{11/2}$) level by upconversion from the lower laser level ($^4I_{13/2}$)[6].

The Er 3 μm emission is self saturated but efficient laser action denies the occurrence of such effect. In fact, the upconversion processes are responsible for the absence of self saturation in concentrated systems, indicating ETU as the main upconversion effect [7].

12th International Symposium on Non-Oxide Glasses and Advanced Materials
Florianopolis, SC, 2000
Apr. 22-26

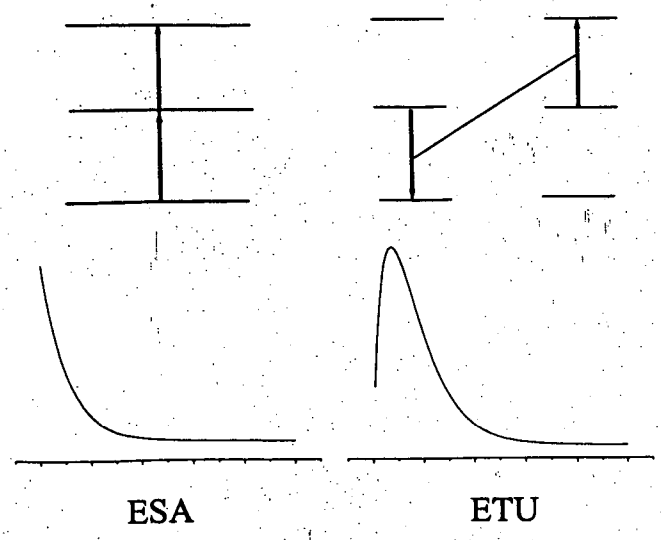

Figure 1 - Diagram of the Er levels involved in the upconversion mechanisms (ESA and ETU) and temporal characteristic of both processes.

Experimental

The samples were prepared with starting composition of (ZBAN) 43 ZrF_3 - 22 BaF_2 - AlF_3 - 12 NaF - x ErF_3 for x varying from 1 to 20, (ZBIGS or ZSBInGa) 15 ZnF_2 - 15 BaF_2 - 25 InF_3 - 15 GaF_3 - 11 SrF_2 - x ErF_3 for x varying from 1 to 20 and (ZBAZYr) 20 ZrF_4 - 25 BaF_2 - 20 AlF_3 - 20 ZnF_2 - 15 YF_3 - x ErF_3 for x varying from 1 to 25.

The excitation system consists of a second harmonic generator pumped by a Quantel Nd:YAG pulsed laser (800 mJ, 4 ns, 10 Hz) that pumps an OPOTEK optical parametric oscilator. The tunability of the OPO allowed excitation wavelengths at the range of 750-850 nm, 960-1000 nm and 1450-1550 nm.

The visible and near infrared emissions were properly injected and dispersed by a 0.25 m SPEX monochromator and detected by a S-20 EMI photomultiplier coupled to a Tektronix digital oscilloscope connected to a computer via GPIB interface. The infrared emissions were focused into a Judson InSb detector whose signal was amplified and connected to the oscilloscope.

Results and discussion

The monitoration of the green emission intensity allows the evaluation of the temporal behavior of the population and depopulation of the $^4S_{3/2}$ and $^2H_{11/2}$ levels, which are responsible for the emission. The population processes of these levels are the ESA and ETU processes while the depopulation mechanisms are radiative decay and cross-relaxation processes. The rise time of the emission has two components as well as the decay time.

The spectral behavior of the rise time can be represented in Figure 2 . The graph is like a zoom on the Figure 1 temporal behavior, where the ESA process is the main mechanism of population for 970 nm pumping and the ETU process is effective for 980 nm pumping. This result can be used in order to evaluate which fraction of the population level is determined by ESA and which one is due to ETU process, constructing an upconversion spectra for each mechanism.

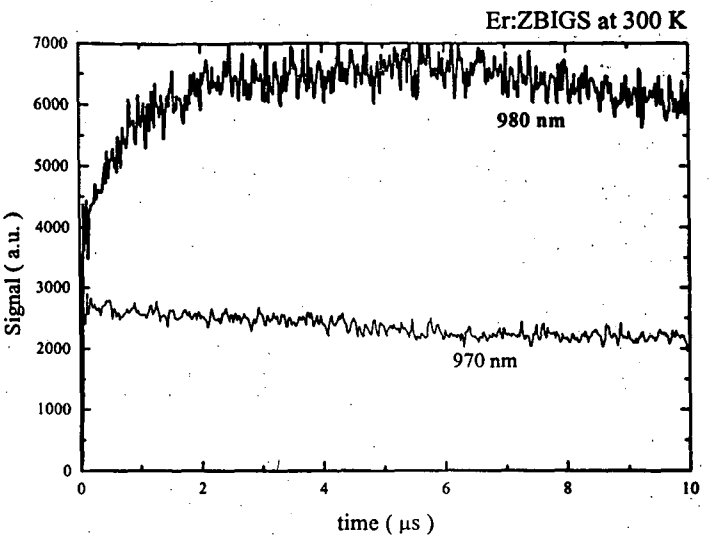

Figure 2 - Comparison between ESA (970 nm) and ETU (980 nm) rise times for the green emission.

The dependence of Er concentration is presented in Figure 3 and demonstrates that the ETU process is more effective for concentrations greater than 5 mol %. The 3 μm laser Stokes efficiency limit can be exceeded for Er concentrations higher than 5 mol %.

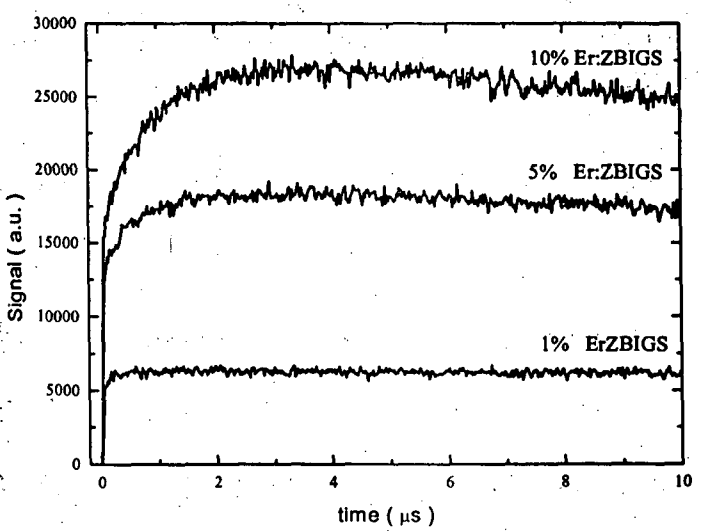

Figure 3 - Comparison between ESA and ETU rise times for the green emission at different Er concentrations. ESA is the main effect for small concentrations and ETU is more relevant for concentrations greater than 5 mol %.

The decay time of the emission has two components, a regular and a greater one. The dependence of these times with the ion concentration is presented in *Table 1*.

Table 1 -($^4S_{3/2}$, $^2H_{11/2}$) level decay times

Er(x%):ZBIGS	Decay time (μs)	
1	200(10)	950(20)
5	58(1)	650(20)
10	13,9(3)	206(2)
15	20,7(8)	154(2)
20	40(4)	175(5)

The radiative lifetime is about 500 μs but there are measurements of decay times greater than this, demonstrating some kind of saturation of non radiative processes. This saturation of the non radiative path obliges the radiative path to be more efficient. The effect is more efficient for concentrations between 5% and 10 % range (the greatest decay time is ten times the smallest decay time). For concentrations greater than 10 % the effect is minimized and for 20 % the ratio between both decay times is similar to the ratio for 1%.

Conclusions

The temporal discrimination between two upconversion mechanisms is very useful for evaluation of positive and negative aspects of the diode pumping scheme. This technique enables us to perform an excited state absorption spectrum without pump-probe technique.

The fractional characterization of both processes is possible due to the time resolved technique linked to wavelength scanning.

The absence of self saturation process in concentrated systems is result of the ETU process; this is the mechanism responsible for slope efficiency greater than Stokes limit

The saturation effect of non radiative processes can be observed and characterized in comparison to the regular decay time. The range of saturation maximization can be determined as well as the upconversion optimization.

Acknowledgements

We would like to thanks FAPESP(95/04166-0), CNPq and CAPES for their financial support.

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