Investigation of energy transfer between lanthanides for Nd:Er and Nd:Ho systems in $LiYF_4$.

Fábio Henrique Jagosich*, Luiz Vicente Gomes Tarelho#,
Gregorio Perez Peiro, Laércio Gomes e Izilda Márcia Ranieiri.

Divisão de Materiais Optoeletrônicos - MEO
Instituto de Pesquisas Energéticas e Nucleares - IPEN
Travessa R, 400 - Cid. Universitária "Armando Salles de Oliveira"
CX. Postal 11049 - CEP: 05422-970 - phone: 55-11-816-9317
lgomes@net.ipen.br

Abstract

Solid-state laser systems available on 3 μm are very useful to several medical and environmental applications. Although the applicational aspect, these systems have academic interest in order to study the deactivation mechanism present on both systems.

This study enables us to characterize the energy transfer required to improve laser action in this region. The energy transfer probability and the optimal dopant concentrations could be estimated using the microparameters evaluated from the extended Förster-Dexter method.

Introduction

The characterization of new laser materials is generally focused on the three main properties : emission wavelength and tunability, emission efficiency and the energy transfer mechanisms[1]. These properties are necessary to indicate how easy laser action is achieved . Besides, these properties are ruled by two basic interactions. The dopant-host interaction determines the local structural environment and defines, for instance, the lengths and angles of the chemical bonds and the crystal field acting on the dopant ion. The second interaction, the dopant-dopant one, can be more useful to the energy transfer study because it is responsible for the mechanism control and effectiveness of the process. This interaction is a sensitive function of the mean distance between dopant ions[2].

Among these dopant-dopant interactions we can emphasize the interactions between two ions whose role at the process is very clear. The dopant ions can work as activator, sensitizer or deactivator ions.

The sensitization is named it the dopant ion receives the input laser energy and supply (donor) it to the activator which accepts it. The deactivation mechanism occurs when the activator ion acts as a donor and transfer its excitation energy to the dopant.

These interactions can be studied mainly based on the donor-donor and donor-acceptor interactions. The acceptor-donor ones are called back-transfer mechanism elsewhere[3].

Experimental Procedure

The absorption and emission spectra needed as input data to Förster-Dexter[4,5] method were obtained from absorption technique. The optical absorption of the samples were measured using a Olis computer interfaced Cary 17D. The emission data were obtained from the absorption ones in order to avoid phonon coupled transitions . The absorption spectra contain thermal broaded zero phonons transitions and the emission spectra evaluated from them have only zero phonon contributions[6].

The experimental parameter critical radius (R_C) is a measurement of the required distance to allow interactions between ions.

DR - FAPESP , * PIBIC/CNPq

Förster-Dexter Method

Förster and Dexter[4,5] method could be applied to rare earth impurities in fluoride crystals as soon as Kushida[7] had proved so. This method is very consistent to its assumptions and the dipole-dipole interactions are usually used to explain several phenomena in the energy transfer investigations.

There are two main parameters which allow us to understand the characteristics of the energy transfer processes. The first one is the energy transfer constant between donors and donors-acceptors. This constant C_{X-Y} is given by the following expression[6]:

$$C_{x\cdots y} = \frac{6c}{(2\pi)^4 n^2} \frac{g_a^{\cdots}}{g_a^{\cdots}} \sum_{N=0}^{\infty} \int \sigma_{\cdots}^{a}(\lambda_x^{\cdots}) \sigma_{\cdots}^{a}(\lambda) d\lambda \left(\sum_{k=0}^{N} P_{(N-k)}^{\cdots} P_z^{\cdots} P_z^{\cdots} \right) \quad (1)$$

$$W_{X-Y} = \frac{C_{X-Y}}{R^6} \quad (2)$$

where W_{X-Y} represents the microscopic energy transfer rate and R the distance between the two ions. The X,Y subscripts are the notation for the Donor (D) and the acceptor (A), respectively. The migration between donors can de denoted by D-D and the energy transfer between donor -acceptor is easily denoted by D-A. The back transfer mechanism can be denoted by A-D.

The second parameter is the critical radius (R_C) which gives us a measurement of the required distance to allow interaction between ions. The R_C determines the distance between these ions when the energy transfer has the same probability as the radiative decay . It can be evaluated by the expression 3.

$$C_{X-Y} = \frac{R_C^6}{\tau} \quad (3)$$

where τ is radiative decay time.

Results

Several systems can be studied using the Förster-Dexter method including rare-earth doped systems. The systems studied were Nd:Er and Nd:Ho in YLF and the results could be compared to evaluate which one is the most promising systemto obtain laser action by pumping them with laser diodes in the 800 nm region.

Figures 1 and 2 show the schematic diagrams of the interacting systems .

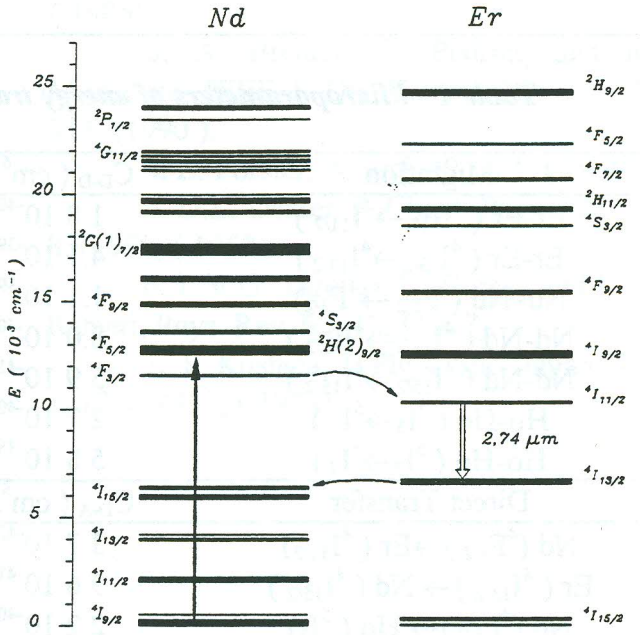

Figure 1 - Schematic diagram of Nd:Er system

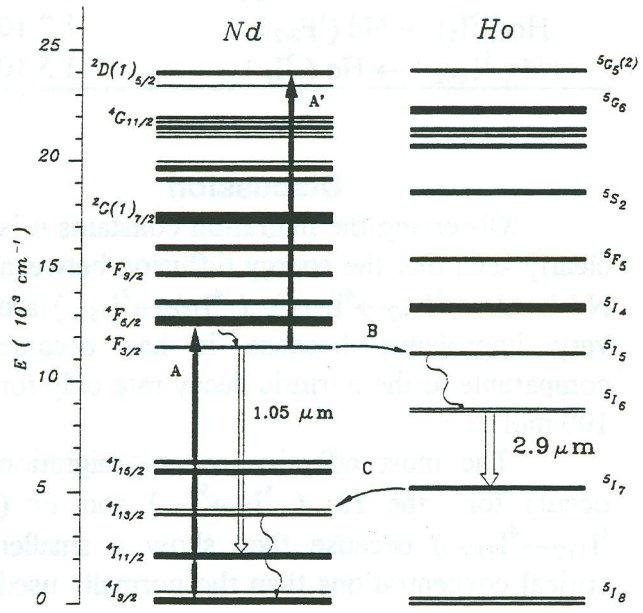

Figure 2 - Schematic Diagram of Nd:Ho system.

Both systems have an important deactivation mechanism that enhances the laser population inversion.

The main results of this study are the microparameters listed in table 1.

The critical concentration denotes the required concentration to achieve effectiveness of the energy transfer process.

Table 1 - Microparameters of energy transfer between some levels of Nd, Er and Ho.

Migration	C_{D-D} (cm^6/s)	R_C (Å)	$Conc_{crit}$ (mol %)
Er-Er ($^4I_{11/2} \rightarrow {}^4I_{11/2}$)	$1.8\ 10^{-40}$	10.4	6.3
Er-Er ($^4I_{13/2} \rightarrow {}^4I_{13/2}$)	$4.1\ 10^{-39}$	18.9	1.1
Nd-Nd ($^4F_{3/2} \rightarrow {}^4F_{3/2}$)	$1.1\ 10^{-39}$	9.2	9.1
Nd-Nd ($^4I_{13/2} \rightarrow {}^4I_{13/2}$)	$8.0\ 10^{-40}$	0.7	100
Nd-Nd ($^4I_{15/2} \rightarrow {}^4I_{15/2}$)	$5.9\ 10^{-41}$	2.9	100
Ho-Ho ($^5I_5 \rightarrow {}^5I_5$)	$2.7\ 10^{-40}$	5.3	47.9
Ho-Ho ($^5I_7 \rightarrow {}^5I_7$)	$5.8\ 10^{-38}$	31.0	0.2
Direct Transfer	C_{D-A} (cm^6/s)	R_C (Å)	$Conc_{crit}$ (mol %)
Nd ($^4F_{3/2}$) $\rightarrow$ Er ($^4I_{11/2}$)	$3.7\ 10^{-42}$	3.6	100
Er ($^4I_{13/2}$) $\rightarrow$ Nd ($^4I_{15/2}$)	$9.6\ 10^{-41}$	10.6	6.0
Nd ($^4F_{3/2}$) $\rightarrow$ Ho ($^5I_5$)	$2.2\ 10^{-40}$	7.0	20.8
Ho ($^5I_7$) $\rightarrow$ Nd ($^4I_{13/2}$)	$8.3\ 10^{-41}$	10.4	6.4
Back transfer	C_{A-D} (cm^6/s)	R_C (Å)	$Conc_{crit}$ (mol %)
Er ($^4I_{11/2}$) $\rightarrow$ Nd ($^4F_{3/2}$)	$2.8\ 10^{-44}$	1.0	100
Nd ($^4I_{15/2}$) $\rightarrow$ Er ($^4I_{13/2}$)	$6.3\ 10^{-41}$	8.7	10.8
Ho ($^5I_5$) $\rightarrow$ Nd ($^4F_{3/2}$)	$1.2\ 10^{-39}$	3.1	100
Nd ($^4I_{13/2}$) $\rightarrow$ Ho ($^5I_7$)	$1.5\ 10^{-43}$	0.2	100

Discussion

Observing the migration constants it is clearly seen that the energy diffusion between Nd levels ($^4I_{13/2} \rightarrow {}^4I_{13/2}$), ($^4I_{15/2} \rightarrow {}^4I_{15/2}$) are very improbable because it has became comparable to the intrinsic decay rate only for 100 mol %.

The most effective energy migration occurs for the Ho ($^5I_7 \rightarrow {}^5I_7$) and Er ($^4I_{13/2} \rightarrow {}^4I_{13/2}$) because they show a smaller critical concentrations than the normally used as dopant.

As a matter of fact, the direct transfer mechanisms studied in this paper allowed us to predict that the deactivation processes of Er ($^4I_{13/2}$) by Nd ($^4I_{15/2}$) and Ho ($^5I_7$) by Nd ($^4I_{13/2}$) are very similar and having the same probability to occur. Although this similarity, the activation mechanism which would allow the population inversion is more effective in Ho than in Er system. The energy transfer Nd ($^4F_{3/2}$) $\rightarrow$ Ho ($^5I_5$) is more probable than Nd ($^4F_{3/2}$) $\rightarrow$ Er ($^4I_{11/2}$).

The back transfer mechanisms are very weak ones. The only back transfer that cannot be neglected is the Nd ($^4I_{15/2}$) $\rightarrow$ Er ($^4I_{13/2}$).

Conclusions

The method seemed to be appropriate to characterize these systems and to predict the most promising one.

The Nd:Ho systems is more promising than the Nd:Er due to the activation process be more efficient in Nd:Ho.

The deactivation mechanism in Nd:Er is similar to the Nd:Ho. So both systems are effective deactivators.

Acknowledgments

The authors thank FAPESP and CNPq for their financial support.

References

1. P.R. Wamsley, K.L. Bray , J. of Lumin., **59**, 11-17 (1994).

2. R.B. Barthem, R. Buisson, J.C. Vial and H. Harmand, J. of Lumin., **34**, 295-305, (1986).

3. A. Brenier, C. Pedrini, and R, Moncorgé, IEEE J. Qu. Electr., **26**, 6, 967-971, (1990).

4. Z. Förster, Naturf, **49**, 321, (1949).

5. D.L. Dexter, J. Chem. Phys. , **21**, 5, 836-850, (1952).

6. L.V.G. Tarelho, L. Gomes and I.M. Ranieri, Phys. Rev, **52**, 12, (1997).

7. T. Kushida, J. Phys. Soc. Japan, **34**, 5, 1318-1337, (1973).