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Influence of looping mechanisms in up-conversion processes in Yb-Tm-Ho doped $\text{Gd}_3\text{Ga}_5\text{O}_{12}$

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Abstract: Two looping mechanisms are studied in Tm and Tm-Ho-Yb doped $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ (GGG). The first one is obtained under pulsed laser excitation, the second one under continuous laser excitation. The gain, losses and condition of stability of the loop are given.

I. Introduction

Up-conversion by excited-state absorption can be either beneficial or unfavorable. The first case is the case of up-conversion lasers, the second one is the case of energy losses in infrared emitting lasers. Because of cross-relaxation processes, the up-converted energy can be returned towards the lower excited states of the ions, then can be used again for up-conversion and so the energy describes loops. These loops are general mechanisms whose we exhibit two examples. The first one was studied in Tm-doped $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ under high repetition rate pulsed laser excitation, the second one was studied in Yb-Tm-Ho-doped $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ under continuous laser excitation.

II. Looping mechanism in pulsed laser excitation regime

Blue emission from the $^1\text{G}_4$ (Tm) level was obtained in Tm-doped $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ under pulsed excitation from a excimer pumped dye laser at 240 Hz repetition rate. The first pulse, weakly absorbed by the ground state towards the $^3\text{F}_2$ level sideband at 638.6 nm, feeds the long-living $^3\text{F}_4$ manifold (fig. 1), which is still partly populated when the second laser pulse comes (fig. 2). Because the wavelength of the laser is resonant with the $^3\text{F}_4 \rightarrow ^1\text{G}_4$ transition an excited state absorption of the second pulse (and of the following ones) occurs and feeds the $^1\text{G}_4$ level. Then two cross-relaxation processes from the $^1\text{G}_4$ level feed back the initial $^3\text{F}_4$ manifold.

Fig. 1 Looping mechanism in GGG:Tm. Fig. 2 Fluorescences of 3F_4 (a), 1G_4 (b).

We can say that the energy accumulated in the levels describes the following loop : $^3\text{F}_4 \rightarrow ^1\text{G}_4 \rightarrow ^3\text{F}_4$ and so on. If we start from one ion in the $^3\text{F}_4$ level, the number of ions in this level at the end of a loop can be called the gain G of the loop. It can be expressed as:

$$(1) \quad G = (\eta + (1-\eta) e^{-\phi\sigma/S}) e^{-1/F\tau}$$

where η is the quantum yield of the cross-relaxation processes, ϕ is the number of photons in each laser excitation pulse, σ is the excited state absorption cross-section, S the section of the laser beam, F the repetition rate of the laser excitation, τ is the lifetime of the $^3F_4$ level. If we neglect any time evolution of G due to the ground state depletion (linear approximation) the $^1G_4$ population immediately after the laser pulse number i (fig. 2) is proportional to :

$$(2) \quad N_i = N_1 (1 - G^i) / (1 - G)$$

In our experiment we found $G=0.87$. From expression (2) we see that if the gain G is larger than one then an instability occurs in the linear approximation and the $^1G_4$ population should grow without limit. In this case the ground state depletion occurring after a certain number of pulses prevents the population to become infinite. A more detailed description of the dynamics will be published in the future.

III. Looping mechanism in continuous laser excitation regime

In a previous work [1] we obtained green emission from $^5S_2-^5F_4$ Ho level under continuous laser excitation in Yb-Tm-Ho doped $Gd_3Ga_5O_{12}$. The $^4F_4$ wavelength of the laser was resonant with the $^5I_7 \rightarrow ^5S_2-^5F_5$ (Ho) transition, the $^5I_7$ level being first fed by absorption from the ground state of the Tm ions (fig. 3) (The Yb ions have no role at 77 K).

3 Looping mechanism in
GGG:Tm:Ho at 77 K.

Then the energy was returned from the $^5S_2-^5F_4$ levels towards the $^5I_7$ one by cross-relaxation processes involving the Tm ions, closing the $^5I_7$ (Ho) $\rightarrow$ $^5S_2-^5F_5$ (Ho) $\rightarrow$ $^3H_4$ (Tm) $\rightarrow$ $^3F_4$ (Tm) $\rightarrow$ $^5I_7$ (Ho) loop. We give here a new model based on the function transfer method [2] in order to predict the threshold of stability of the loop, valid in the case where rate equations of population cannot be used due to the fact that the efficiency of the cross-relaxation processes depends on the environment of the donor ion (non exponential decay after δ excitation in the donor level). For simplicity the model concerns one chemical specie of ions with three energy levels (fig. 4) and was shown to be equivalent to the more complex real one in fig. 3 [1].

Let us write $f(s)$ the Laplace transform of a function $f(t)$, s being the Laplace variable and t the time. Then the time evolution $N_A(t)$ of the population of an acceptor level A (level 2 or 3 in fig. 4) due to an excitation $e(t)$ starting at time 0 is such that :

$$(3) \quad N_A(s) = K_A(s) e(s)$$

where $K_A(t)$ is the time evolution of the population due to a δ excitation, called response function of the acceptor. Let us also recall that the rate $e(t)$ of an energy transfer (for example the cross-relaxation process in fig.

(4) is such that :

$$(4) \quad \hat{e}_{DA}(s) = \hat{N}_D(s) [\hat{K}_D(s)^{-1} - (s+1/\tau_D)]$$

where the label D stands for the donor level (level 3 in fig. 4), τ_D being the spontaneous deexcitation lifetime of the level. More the quantum yield H of the energy transfer D $\rightarrow$ A after a δ excitation into level D can be expressed in a very convenient way :

$$(5) \quad H = 1 - (1/\tau_D) \int_0^\infty K_D(t) dt = 1 - \hat{K}_D(0)/\tau_D$$

The expressions (3) or (4) can be represented visually by block diagrams, and doing so for each processes of feeding of the levels 2 and 3 in fig. 4, we can built a diagram representing the whole dynamics of the system by connecting the different blocks :

Fig. 4 Energy level scheme corresponding to the diagram.

The parameters defining the system are τ_2, τ_3 (lifetimes of levels 2, 3), B (branching ratio of the 3 $\rightarrow$ 1 deexcitation), H_{CR} (quantum yield of the cross relaxation). R_1, R_2 are the pumping rates. The diagram can be read as follow :

$$(6) \quad \hat{N}_2 = \hat{K}_2 \{ C \hat{R}_1 - R_1 \hat{N}_2 + [2(\hat{K}_3^{-1} - (s+1/\tau_3)) + (1-B)/\tau_3 - R_1] \hat{K}_3 R_2 \hat{N}_2 \}$$

Or by writing the terms in an other order :

$$(7) \quad \hat{N}_2 = \frac{C \hat{K}_2 \hat{R}_1}{1 - \{ \hat{K}_3 \hat{R}_2 [2(\hat{K}_3^{-1} - (s+1/\tau_3)) + (1-B)/\tau_3 - R_1] - R_1 \} \hat{K}_2}$$

The inverse Laplace transform of expression (7) give the time evolution of the population of level 2 at early times, when we can neglect the influence of the ground state depletion on the efficiency of the cross-relaxation. In this approximation expression (7) predicts an instability if its denominator D(s) vanishes in the right side of the s-plane : $N_2(t)$ should go to infinity at long times if the ground state depletion did not occur. In order to find at which condition the denominator vanishes let us use Nyquist criterion. When the Laplace variable s describes the path drawn in fig. 5-a, we can obtain three kinds of paths in the denominator plane, drawn in fig. 5-b. All of them start at $s=1$ whatever the values of the parameters of the system if the radius of the semi-circle in fig. 5-a is infinite, which is necessary in order to enclose the whole right side of the s-plane. On the contrary the position of D(0) is dependent on the values of the parameters of the system

Fig. 5-a Path in the s-plane.

Fig. 5-b Corresponding path in the denominator plane.

and on the pumping rates and we see that if $D(0) < 0$ then 0 is enclosed by the path, the denominator vanishes in one point of the right side of the s-plane and the system is instable. The condition $D(0) < 0$ has a very simple physical meaning which can be obtained with the use of formula (5) :

$$D(0) < 0 \Leftrightarrow R_2 [2 H_{CR} + \eta - 1] > 1/\tau_2 + R_1 + R_1 R_2 (1 - H_{CR}) \tau_3 \quad (8)$$

where the symbol $\Leftrightarrow$ means "equivalent". The left side of (8) is the net gain of the loop : when an ion is removed (-1) from level 2 by the laser beam (this occurs with a rate R_2) it is returned on this level by the cross-relaxation process with a quantum yield $2H_{CR}$ and by the $3 \rightarrow 2$ spontaneous deexcitation with a quantum yield η . The right side of (8) gives the losses from level 2 : $1/\tau_2$ for the spontaneous deexcitation, R_1 and $R_1 R_2 (1 - H_{CR}) \tau_3$ for the losses of level 2 feeding by the laser pump due to accumulation of population in levels 2 and 3 respectively. So if the net gain of the loop is larger than the losses the loop is instable. Let us consider a last point. If the pumping rate R_1 is zero then $D(0) = 1$: at low R_1 the loop is always stable but an interesting question is to know at which conditions concerning the parameters defining the system it is possible to get instability by increasing the pumping rate R_1 from zero. Actually instability will occur if we succeed to cancel $D(0)$. The equation $D(0) = 0$ is a second order equation whose unknown quantity is only R_1 because $R_2 = R_1 \sigma_1 / \sigma_2$ where σ_1 and σ_2 are absorption cross-sections from the ground state 1_1 and the excited state 2. It has physical solutions (ie R_1 real and positive) only if two conditions are fulfilled : the discriminant must be positive and we must have $2H_{CR} + \eta - 1 - (\sigma_1 / \sigma_2) > 0$. If these conditions are not fulfilled the loop is intrinsically stable whatever the pumping rate.

IV. Conclusion

The role of looping mechanisms in up-conversion has been pointed out. The stability of the loop can be studied at early times when it is still possible to neglect the influence of the ground state depletion on the efficiency of the energy transfers. The condition of instability of the loop has been derived in the case where non exponential decays of the fluorescences are observed after direct δ excitation in the levels. The up-conversion is due to a competition between a two-step absorption and a looping mechanism, their relative efficiency depending if the system works below of above the threshold of instability of the loop.

[1] A. Brenier, L. C. Courrol, C. Madej, C. Pédrini, G. Boulon, Optical Materials, to be published.

[2] Huang Shi-Hua, Lou Liren, Acta Phys. Sin. vol. 38 n°3 (1989) 422.