ELSEVIER

Journal of Luminescence 58 (1994) 284–286

JOURNAL OF LUMINESCENCE

Looping mechanism in Yb^{3+}–Tm^{3+}–Ho^{3+} doped $Gd_3Ga_5O_{12}$ garnet

A. Brenier*, L.C. Courrol, C. Pédrini, C. Madej, G. Boulon

Laboratoire de Physico-Chimie des Matériaux Luminescents, Unité de recherche associée au CNRS no. 442, Université Claude Bernard Lyon I, 43 Bd 11 Novembre 1918, 69622 Villeurbanne Cedex, France

Abstract

Conversion of near infrared laser light into green emission has been obtained in $Gd_3Ga_5O_{12}$:Yb:Tm:Ho by a two step process: absorption from the ground states of the ions followed by a mechanism we have called the looping mechanism. The latter is composed of an absorption from the $^5I_7$(Ho) excited state followed by a positive feedback process via the Tm ions. A two ion (Tm–Ho) model is given to describe the dynamics and the gain and losses of the loop are evaluated.

1. Introduction

In a previous paper [1] we have presented a study devoted to the sensitization of the Ho^{3+} 2 μm laser emission by the Yb^{3+}–Tm^{3+} ions in $Gd_3Ga_5O_{12}$ garnet. Such system is attractive for laser diode pumping. We had measured with an integrating cavity the up-conversion energy losses quantum yield after infrared pumping into the Yb ion and we had shown that in the triply doped crystal it is weaker than in the Yb–Ho doubly doped one. The reason for this could be the existence of efficient down-conversion mechanisms that bring back the up-converted energy in the $^5S_2$–$^5F_4$(Ho) levels to the 2 μm emitting level in the $^5I_7$(Ho) manifold. If this is true it means that a positive feedback increases the population of the $^5I_7$ manifold and the purpose of this work is to point out this process. The excitation source chosen is a CW Ti:sapphire laser operating at 775 nm. This wavelength corresponds to the $^5I_7 \rightarrow {}^5S_2$–$^5F_4$(Ho) excited state absorption in such a way that the so-called looping mechanism occurs. The result is the conversion of the infrared excitation into a green emission from the $^5S_2$–$^5F_4$(Ho) levels as shown in Fig. 1. All results given in this paper are for the triply Yb(5%)–Tm(5%)–Ho(0.5%) doped sample at 77 K. We also want to mention that only Tm and Ho ions are involved and we assume that at 77 K the Yb ions do not play a role.

2. Excited state absorption

The laser excitation spectrum of the Ho green emission (near 540 nm, transition $^5S_2$–$^5F_4 \rightarrow {}^5I_8$) is shown in Fig. 2 (lower curve) at 77 K. Because in this range of wavelengths there is no *peak* of absorption from the Yb–Tm–Ho fundamental levels, there is no difficulty in attributing the absorption of the laser beam to the $^5I_7 \rightarrow {}^5S_2$–$^5F_4$(Ho) excited state transition, the $^5I_7$ level being fed first by a structureless vibronic absorption from the Tm ground state to the $^3H_4$(Tm), followed by a

* Corresponding author.

SSDI 0022-2313(93)E0137-M

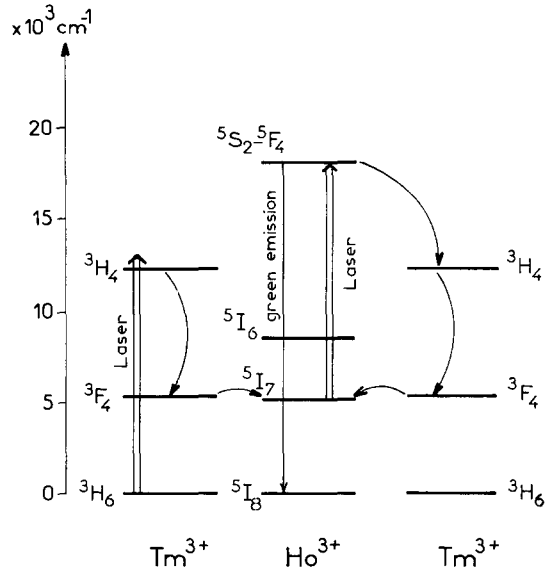

Fig. 1. Scheme of energy levels involved in the model of looping mechanism. The arrows represent the channels of energy mainly by cross-relaxation processes. The first step after non-resonant absorption in Tm ions feeds the $^5I_7$(Ho) level. The second step corresponds to resonant excited-state absorption of the Ho ions by the laser pump and the so-called loop can be seen between $^5S_2$–$^5F_4$, $^3H_4$, $^3F_4$, $^5I_7$ and again $^5S_2$–$^5F_4$ levels of both Tm and Ho ions.

$^3H_4(Tm) + {}^3H_6(Tm) \rightarrow {}^3F_4(Tm) + {}^3F_4(Tm)$ cross-relaxation mechanism, followed by a $^3F_4(Tm) + {}^5I_8(Ho) \rightarrow {}^3H_6(Tm) + {}^5I_7(Ho)$ transfer. We see that the excited state absorption cross-section peak occurs at 753.74 nm and so this wavelength was chosen for the pumping source.

The intensity $I(t)$ of the transmitted beam through the sample shows (Fig. 3) the time evolution of the excited state absorption. At time $t = 0$, when the laser beam is turned on, there is only absorption from the ground state of the Tm ions because the $^5I_7$(Ho) level is empty. Then the $^5I_7$(Ho) level fills and the transmitted intensity decreases. At long times the transmitted intensity $I(\infty)$ becomes constant and its value depends on the incident power.

3. Positive feedback and looping mechanism

We want to examine now what happens to the $^5I_7$(Ho) population when the laser beam is resonantly absorbed by the $^5I_7 \rightarrow {}^5S_2$–$^5F_4$(Ho) transition. The result is given by a laser excitation spectrum of the 2 μm Ho emission because this last one originates from the $^5I_7$ level and its intensity is proportional to the $^5I_7$ population. Such a spectrum is given in Fig. 2 (upper curve). We can see that its peaks correspond exactly to the ones of the green emission laser excitation spectrum (Fig. 2, lower curve). Moreover, we can see that the first

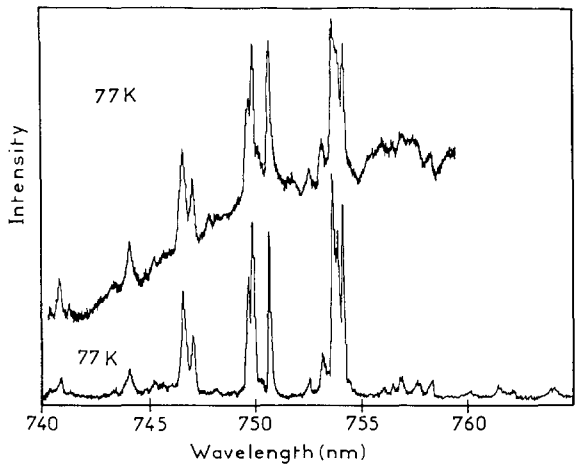

Fig. 2. Correlation between CW Ti:sapphire laser excitation spectra by monitoring either 2 μm emission (upper curve) or green emission (lower curve) of Ho ions.

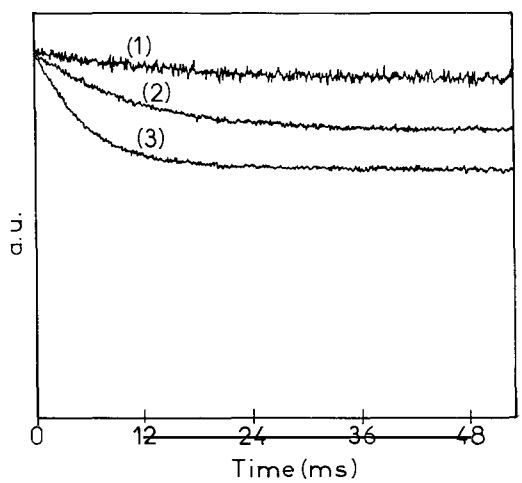

Fig. 3. Time evolution of the intensity $I(t)$ of the transmitted laser beam at different incident power: (1) 0.113 W, (2) 0.580 W, (3) 1.17 W. The pump wavelength is 753.74 nm.

spectrum appears in *positive* values. This means that when an excited state absorption occurs from the $^5I_7$ level it does not decrease its population but on the contrary increases it. This fact proves that a positive feedback process occurs, whose origin is a $^5S_2$–$^5F_4$(Ho) + $^3H_6$(Tm) → $^5I_7$(Ho) + $^3H_4$(Tm) transfer, followed by a $^3H_4$(Tm) + $^3H_6$(Tm) → $^3F_4$(Tm) + $^3F_4$(Tm) cross-relaxation mechanism, followed by a $^3F_4$(Tm) + $^5I_8$(Ho) → $^3H_6$(Tm) + $^5I_7$(Ho) transfer as briefly indicated in Fig. 1.

Let us start with one ion in the $^5I_7$(Ho) level. If by excited state absorption the ion is sent into the $^5S_2$–$^5F_4$(Ho) levels, it will be returned back on the $^5I_7$(Ho) level by the feedback process with a certain efficiency and so a loop has occurred. If the number of ions in the $^5I_7$(Ho) level is greater at the end of the loop then gain has occurred. We understand that if the gain is larger than the losses, the dynamics of the system will be qualitatively different than in the case where it is smaller: in the first case we have a photon avalanche. This phenomenon has been studied more than ten years in monodoped crystals [2,3].

Gain and losses of the loop have been studied with the almost equivalent one-ion/three level system represented in Fig. 4, which has been extensively used [3]. We have found that the threshold condition for photon avalanche was:

$$R_2(2H_{CR} + \eta - 1) > 1/\tau_2 + R_1 + R_1R_2\tau_3(1 - H_{CR}), \quad (1)$$

where R_1 and R_2 are the absorption rates from the ground and excited states respectively, H_{CR} and η are the quantum yields of the cross-relaxation and $3 \rightarrow 2$ de-excitation mechanism respectively, τ_2 and τ_3 are the lifetimes of the spontaneous de-excitations of the 2 and 3 levels respectively.

Expression (1) has a very simple physical interpretation. Its left side is the net gain of the loop: when an ion is removed (-1) from the n_2 level by the laser beam (this occurs with a yield R_2), it is returned back by the cross-relaxation process with a quantum yield $2H_{CR}$ and by the $3 \rightarrow 2$ de-excitation with a quantum yield η. The right side of Eq. (1) describes the losses from the n_2 level: $1/\tau_2$ for the spontaneous deexcitation, R_1 and $R_1R_2\tau_3(1 - H_{CR})$ are losses of feeding of level 2 by the laser pump due to accumulation of population

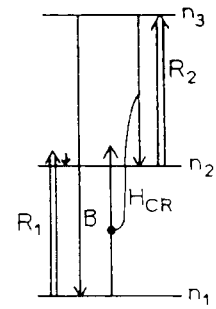

Fig. 4. Scheme of the one-ion/three-level equivalent system.

into the n_2 and n_3 levels respectively. Numerically we have found that our system was working below the threshold because of the high value of R_1, the absorption from the ground state. This is due to the high value of the Tm concentration which is ten times that of the Ho.

4. Conclusion

We have obtained green fluorescence from the $^5S_2$–$^5F_4$(Ho) level under pumping with a CW Ti : sapphire laser at liquid nitrogen temperature in Ho–Tm–Yb doped $Gd_3Ga_5O_{12}$ due to a looping mechanism involving the Ho–Tm pair of ions. It results from an excited state absorption in the Ho ions followed by a positive feed-back process involving the Tm ions. For the first time we have calculated the gain and the losses of the looping mechanism from a theoretical expression leading, for any value of the pump rate R_1 in the ground states of the ions, to the theoretical determination of the threshold for photon avalanche. We show that the threshold is an increasing function of the absorption from the ground state: the weaker it is, the easier it will be to obtain up-conversion via the photon avalanche.

References

[1] A. Brenier, L.C. Courrol, C. Pédrini, C. Madej and G. Boulon, Phys. Rev. B, to be published.
[2] J.S. Chivian, W.E. Case and D.D. Eden, Appl. Phys. Lett. 35 (1979) 124.
[3] U. Oetliker, M.J. Riley, P.S. May and H.U. Gudel, J. Lumin. 53 (1992) 553.