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SPECTROSCOPIC AND STIMULATED EMISSION PROPERTIES OF Er-DOPED CRYSTALS FOR THREE-MICRON LASERS

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Abstract - Based on spectroscopic data the various processes (energy transfers, excited state absorption) that influence the population dynamics of the excited levels in concentrated Er^{3+} doped laser crystals are discussed and their role in determining the population inversion between the $^4\text{I}_{11/2}$ and $^4\text{I}_{13/2}$ involved in three-micron laser emission is estimated by solving the laser rate equations for stationary regime under selective or non-selective pumping. The role of co-doping the crystals with ions that modify the mechanism of population of the initial laser level (such as Cr^{3+}) or of ions that de-excite the terminal level (such as Nd^{3+} , Tm^{3+} or Ho^{3+}) is also discussed. It is thus shown that by a proper choice of the active medium and by correlation of the spectroscopic properties with the pump conditions a good control of the emission (wavelength, regime of emission, efficiency and so on) could be obtained.

1. Introduction

Despite of a lifetime of the laser terminal level much longer than that of the emitting level, efficient room temperature three-micron laser emission has been obtained between the levels $^4\text{I}_{11/2}$ and $^4\text{I}_{13/2}$ of Er^{3+} in highly concentrated crystals [1]. Preliminary studies [2 - 4] on the population dynamics of excited levels in YAG:Er have shown that this is determined by three energy transfer processes by cross-relaxation inside the system of Er^{3+} ions: (i) a process of rate W_4 , bi-ionic for low- and three-ionic for high- Er concentrations [5], that de-activates the level $^4\text{S}_{3/2}$ leading to an equal population of levels $^4\text{I}_{11/2}$ and $^4\text{I}_{13/2}$; however a small population between definite pairs of crystal field components x_1 and y_j of these levels may exist and this could be sufficient to start the laser emission; a process (w_1) of up-conversion from level $^4\text{I}_{13/2}$: ($^4\text{I}_{13/2} \rightarrow ^4\text{I}_{15/2}$) + ($^4\text{I}_{13/2} \rightarrow ^4\text{I}_{9/2}$), followed by a very fast multiphonon relaxation on $^4\text{I}_{11/2}$; (ii) an up-conversion process (w_2) from $^4\text{I}_{11/2}$: ($^4\text{I}_{11/2} \rightarrow ^4\text{I}_{15/2}$) + ($^4\text{I}_{11/2} \rightarrow ^4\text{F}_{7/2}$), followed by a rapid relaxation on $^4\text{S}_{3/2}$. Of these processes w_2 is essential for three-micron laser emission since it depopulates the terminal level and re-pumps the emitting level thus precluding the self-saturation of the emission. A practical problem that has to be answered to is: why the three-micron laser emission of Er^{3+} is restricted to only a few of the transitions observed in the $^4\text{I}_{11/2} \rightarrow ^4\text{I}_{13/2}$ luminescence spectra (figure 1 shows

these spectra for Er^{3+} in YAG and YSGG) and what can be done in order to extend the laser emission wavelength range.

Fig. 1. Luminescence spectra of $^4I_{11/2} \rightarrow ^4I_{13/2}$ transition of Er^{3+} in YAG (a) and YSGG (b).

Table 1 lists the main luminescence lines of this spectrum for Er^{3+} in YAG and YSGG together with the identification of crystal field components involved in transition and with a figure of merit p that will be defined later.

Table 1. The main lines in the $^4I_{11/2} \rightarrow ^4I_{13/2}$ luminescence spectrum of Er^{3+} in YAG and YSGG.

Crystal	Luminescence lines cm^{-1}	μm	Assignment $x_i \rightarrow y_j$	Figure of merit p
YAG:Er	3858	2.598	$6 \rightarrow 1$	3.982
	3807.6	2.626	$5 \rightarrow 3$	2.98
	3766	2.655	$(4 \rightarrow 3) + (3 \rightarrow 3)$	2.43
	3707	2.698	$1 \rightarrow 1$	1.848
	3682	2.716	$2 \rightarrow 3$	1.624
	3654	2.737	$1 \rightarrow 3$	1.412
	3575.8	2.797	$3 \rightarrow 4$	0.994
	3567	2.807	$4 \rightarrow 5$	0.940
	3530.3	2.833	$5 \rightarrow 7$	0.780
	3502	2.855	$2 \rightarrow 4$	0.690
	3481	2.873	$3 \rightarrow 5$	0.613
	3454.7	2.877	$1 \rightarrow 5$	0.541
	3403	2.938	$2 \rightarrow 7$	0.426
YSGG:Er	3790.7	2.638	$(4 \rightarrow 2) + (5 \rightarrow 3) +$ $(3 \rightarrow 1) + (6 \rightarrow 3)$	3.20
	3701	2.702	$(1 \rightarrow 1) + (2 \rightarrow 2)$	1.917
	3579	2.794	$(3 \rightarrow 4) + (4 \rightarrow 5)$	1.09.

This paper shows that a good understanding of the three-micron laser processes in concentrated Er systems could be obtained by analysing the analytical solutions that can be obtained by solving the system of

laser rate equations for the stationary emission regime, important for long-pulse or cw Erbium lasers.

2. Laser rate equations

The three-micron laser processes for Er^{3+} can be described by the system of rate equations [2, 3, 6] :

$$\begin{aligned}\frac{dN_4}{dt} &= -\frac{N_4}{T_4} - W_4 N_0^{\kappa} N_4 + W_2 N_2^2 + R_{p4} N_0 \\ \frac{dN_3}{dt} &= -\frac{N_3}{T_3} + \frac{N_4}{T_4} + R_{p3} N_0 \\ \frac{dN_5}{dt} &= -\frac{N_5}{T_5} + \frac{N_3}{T_3} + W_1 N_1^2 + W_4 N_0^{\kappa} N_4 + R_{p5} N_0 \\ \frac{dN_2}{dt} &= -\frac{N_2}{T_2} - 2W_2 N_2^2 + \frac{N_5}{T_5} - \sigma\phi(\alpha N_2 - \beta N_1) + R_{p2} N_0 \\ \frac{dN_1}{dt} &= -\frac{N_1}{T_1} + \frac{N_2}{T_2} - 2W_1 N_1^2 + W_4 N_0^{\kappa} N_4 + \sigma\phi(\alpha N_2 - \beta N_1) + R_{p1} \\ \frac{d\phi}{dt} &= \phi v [\sigma(\alpha N_2 - \beta N_1) - \rho]\end{aligned}\quad (1)$$

where σ is the laser emission cross-section, ϕ the photon flux density, ρ the total losses at the wavelength of emission, v the speed of light in the active medium, $R_{pi} N_0$ the pump rate in level i and α and β are the Boltzmann thermal population coefficients of crystal field components $x_i(^4I_{11/2})$ and $y_i(^4I_{13/2})$ involved in the laser emission. N_1 to N_5 are the populations of the levels $^4I_{13/2}$, $^4I_{11/2}$, $^4F_{9/2}$, $^4S_{3/2}$ and $^4I_{9/2}$ respectively, T_1 to T_5 being their lifetimes. The exponent κ of N_0 in equations for N_4 , N_5 and N_1 takes the value of 1 for two-ion processes W_4 and 2 for three-ion processes.

For stationary regime ($\frac{dN_i}{dt} = 0$, $\frac{d\phi}{dt} = 0$ and for medium or high Er concentrations ($W_4 N_0 \gg 1/T_4$) the system (1) gives an exact solution for population N_1 of $^4I_{13/2}$:

$$N_1 = \varphi \left[\frac{(R_1 + R_2) N_0}{W_1} \right]^{1/2} = [(\mu + \varepsilon^2)^{1/2} - \varepsilon] \left[\frac{(R_1 + R_2) N_0}{W_1} \right]^{1/2} \quad (2)$$

with $\mu = 1$.

$$\varepsilon = \frac{1}{2T_1} [(R_1 + R_2) N_0 W_1]^{-1/2} \quad (3)$$

and

$$R_1 = R_{p1} + R_{p4}$$

$$R_2 = R_{p2} + R_{p3} + R_{p4} + R_{p5}$$

Below the laser threshold ($\phi \sim 0$)

$$N_2 = \delta \left[\frac{(R_1 + 2R_2)N_0}{W_1} \right]^{1/2} = [(v + \gamma^2)^{1/2} - \gamma] \left[\frac{(R_1 + 2R_2)N_0}{W_2} \right]^{1/2} \quad (4)$$

with

$$v = 1 - \frac{N_1}{(R_1 + 2R_2)N_0 T_1} \quad (5)$$

$$\gamma = \frac{1}{2T_2} [(R_1 + 2R_2)N_0 W_1]^{-1/2}$$

Thus the difference of populations of crystal field sublevels of $^4I_{11/2}$ and $^4I_{13/2}$ below laser threshold is:

$$\Delta N = \alpha N_2 - \beta N_1 = \alpha \delta \left[\frac{(R_1 + 2R_2)N_0}{W_2} \right]^{1/2} \left\{ 1 - \frac{\varphi}{\delta} \frac{\beta}{\alpha} \left(\frac{W_2}{W_1} \right)^{1/2} \frac{R_1 + R_2}{(R_1 + 2R_2)} \right\} =$$

$$\alpha \delta \left[\frac{(R_1 + 2R_2)W_0}{W_2} \right]^{1/2} \left\{ 1 - p \frac{\varphi}{\delta} \left(\frac{R_1 + R_2}{R_1 + 2R_2} \right) \right\} \quad (6)$$

The parameter [7]

$$p = \frac{\beta}{\alpha} \left(\frac{W_2}{W_1} \right)^{1/2} \quad (7)$$

in (6) could be considered as a figure of merit for a given transition; it depends on the particular host crystal (via W_1 and W_2) and on the particular laser transition $x_i + y_j$ (via α and β); it also depends on temperature. For a given temperature the transitions originating on the lower crystal field components of $^4I_{11/2}$ and terminating on the upper components of $^4I_{13/2}$ will have small figures of merit and viceversa.

A positive population difference ΔN (i.e. a population inversion) below laser threshold implies that

$$p < \frac{\delta}{\varphi} \left(\frac{R_1 + 2R_2}{R_1 + R_2} \right)^{1/2} ; \quad (8)$$

at laser threshold this condition becomes

$$p < \frac{\delta}{\varphi} \left(\frac{R_1 + 2R_2}{R_1 + R_2} \right)^{1/2} \left(1 - \frac{p}{\sigma \alpha N_2} \right). \quad (9)$$

For the stationary regime the photon flux density above threshold is given by

$$\phi = \frac{N_0}{\rho} [R_2 + (R_1 + R_2)(1 - p^2)] - \frac{1}{\alpha \sigma T_2} - \frac{W_2 \rho}{\alpha^2 \sigma^2} -$$

$$- N_1 \left(\frac{\beta}{\rho \alpha T_2} + \frac{2\beta W_2}{\alpha^2 \sigma} \right) - \frac{1 - p^2}{\rho T_1} N_1 ; \quad (10)$$

a positive flux density implies a condition for figure of merit p identical to (9).

If we define a quantum efficiency as the ratio between the number of quanta generated in the laser cavity and the number of pump quanta, the upper limit of this efficiency for very high pumping in level 1 is given by

$$\eta_{ei} = \frac{R_2 + (R_1 + R_2)(1-p^2)}{R_{p1}} ; \quad (11)$$

thus we have

$$\begin{aligned} \eta_e(^4S_{3/2}) &= 3 - 2p^2 \\ \eta_e(^4F_{9/2}, ^4I_{9/2}, ^4I_{11/2}) &= 2-p^2 \\ \eta_e(^4I_{13/2}) &= 1 - p^2 \end{aligned} \quad (12)$$

All these results show that the cross-relaxation processes W_1, W_2, W_4 can assure laser emission between the levels $^4I_{11/2}$ and $^4I_{13/2}$ of Erbium. The presence of excited state absorption (ESA) of pump radiation from the excited level $^4I_{13/2}$ can have a positive effect on the laser emission, reflected by a positive term $[R_{e1} + (2-p^2)R_{e2}] N_1/p$ in the photon flux density, R_{e1} being the absorption rate of pump radiation of wavelength larger than $0.85 \mu m$ and R_{e2} that corresponding to λ smaller than $0.85 \mu m$. However the effect of ESA depends on the ratio N_1/N_0 that is usually fairly small; thus ESA is not a leading mechanism for three-micron Erbium laser in presence of cross-relaxation W_1 . The expression given above for the populations and photon flux density lead to important conclusions on the role of the various parameters of the system in determining the performances of the three-micron lasers:

(i) The figure of merit p given by (7) plays a crucial role in determining a positive population difference ΔN , a positive photon flux density ϕ and a positive quantum efficiency for a given laser transition. It is thus shown that even in optimal pump conditions, only transitions with $\lambda \geq 2.8 \mu m$ could give stationary laser emission in case of YAG:Er. It also shows that for this crystal stationary emission can be obtained for pump in any of the levels considered above, even in the terminal level $^4I_{13/2}$.

At the same time, from the main luminescent $^4I_{11/2} \rightarrow ^4I_{13/2}$ Er lines in YSGG listed in Table 1 only the transition at $2.794 \mu m$ could give laser emission and only for pump in levels $^4S_{3/2}, ^4F_{9/2}, ^4I_{9/2}$ and $^4I_{11/2}$, but not in the terminal level $^4I_{13/2}$; even so the quantum efficiency is much lower than in case of Er^{3+} in YAG.

(ii) The lifetime T_2 of $^4I_{11/2}$ should be as large as possible in order to reduce the laser threshold.

(iii) The effect of lifetime T_1 depends on the value of figure of merit p :

- for transitions with $p < 1$ the last term in (10) is negative and reduces the value of photon flux density; in this case a large value of T_1 is desirable and any attempt to reduce T_1 by co-doping with ions such as Nd^{3+} , Ho^{3+} or Tm^{3+} would reduce the laser performances. This explains the negative effect of these ions on the $2.938 \mu m$ emission of Er^{3+} in YAG ($p = 0.426$).
- for $p > 1$ the last term in (10) is positive and a shorter T_1 would increase its contribution to the flux density: for very small values of T_1 this term could become competitive or even dominant in the photon flux density. Thus co-doping with ions that reduce drastically T_1 would make possible laser transitions with $p > 1$ even in crystals where

normally (without co-doping) these transitions are not observed. Co-doping with Nd^{3+} , Ho^{3+} or Tm^{3+} could thus extend the wavelength capabilities of Er^{3+} lasers, even if in some cases the efficiency of the original $p < 1$ transitions in these crystals is lowered because of this co-doping.

(iv) A small W_2 rate is desirable

(v) A large W_1 rate is beneficial.

This analysis shows that the spectroscopic parameters that determine the wavelength range and the efficiency of three-micron laser emission of Er^{3+} laser are the cross-relaxation rates W_1 , W_2 and W_4 , the lifetimes T_1 and T_2 , the Boltzmann population coefficients a and b , the emission cross-section σ and the losses ρ ; at the same time it shows that the laser emission characteristics could be changed by acting on these spectroscopic parameters.

In this respect, this analysis shows that the system $\text{Er}:\text{YSGG}$, despite of a much larger value of T_2 [7] as compared with $\text{Er}:\text{YAG}$ (1.4 msec versus 88 μsec), is expected to have much worse laser parameters than $\text{Er}:\text{YAG}$ because the transitions with large emission cross-section have large figures of merit p . In this case a possibility to overcome the situation would be the modification of the mechanism of population of levels $^4I_{11/2}$ and $^4I_{13/2}$ in a way that would relax the conditions imposed on the figure of merit p in order to have positive pump term in the expression of photon flux. A possible solution in this respect is the co-doping of crystals with Cr^{3+} ; this ion absorbs very effectively the pump radiation due to two intense and broad transitions from the ground state 4A_2 to the levels 4T_2 and 4T_1 . Cr^{3+} enters in the YSGG lattice as a substitutional ion in the large octahedral site of Sc^{3+} : the crystal field effects are fairly weak and the level 2E lies only about 200 cm^{-1} below 4T_2 so that these two levels are thermalized at room temperature and a very effective ($n \sim 1$) energy transfer from Cr^{3+} to the $^4I_{9/2}$ level of Er^{3+} takes place; thus Cr^{3+} contributes to a better utilization of the pump radiation for population of the emitting level $^4I_{11/2}$ of Er^{3+} . At the same time, an effective transfer from $^4S_{3/2}$ Er to 4T_2 Cr could take place. The chain of transfer $^4S_{3/2} \rightarrow ^4T_2\text{Cr} \rightarrow ^4I_{9/2}\text{Er}$, characterized by a global rate W_4 competes with the cross-relaxation process W_4 in de-excitation of $^4S_{3/2}$. However, by difference from W_4 the process W_4 leads only to the population of the initial laser level $^4I_{11/2}$ (and not of both $^4I_{11/2}$ and $^4I_{13/2}$ as in case of W_4) thus improving the building of a population inversion between these two laser levels.

The relative contribution of the two processes (W_4 and W_4') in de-excitation of $^4S_{3/2}$ can be expressed by a parameter [6]:

$$f = \frac{W_4 N_O}{W_4 N_O + W_4' N_{O4}} = \left[1 + \frac{W_4' N_{O4}}{W_4 N_O} \right]^{-1} \quad (13)$$

that ranges between 0 (when only W_4' is active) to 1 (when $W_4' N_{O4}$ is equal to zero). The system of rate equations for this case cannot be solved analytically below-threshold; however, at threshold analytical expression for N_1 and N_2 similar to (2) and (4) could be obtained but with the parameters μ , ϵ , ν and γ dependent on f . At the same time the expression for the flux density is modified to:

$$\phi = \frac{(2-fp^2)R'N_O}{[1+(1-f)p^2]\rho} - \frac{1}{\alpha T_2} - \frac{2-f}{1+(1-f)p^2} \frac{W_2 \rho}{\alpha^2 \sigma} - \frac{1}{\rho} \left[\frac{1}{\alpha T_2} + \frac{2(2-f)}{1+(1-f)p^2} \frac{\beta \rho W_2}{\alpha^2 \sigma} \right] - \frac{1-p^2}{1+(1-f)p^2} \frac{N_1}{\rho T_1} \quad (14)$$

where the pump rate $R'N_0$ takes into account that the pumping is performed in the strong absorption bands of Cr^{3+} . The condition imposed on the figure of merit p in order to have the first term (the pump term) in (14) positive is now

$$p^2 \leq \frac{2}{f} \quad (15)$$

Since f is subunitary and can take quite low values when the ratio $\frac{W_4' N_{Cr}}{W_4 N_0}$ is large the condition (15) is much relaxed and laser transitions with larger p values than for systems doped only with Er^{3+} are now possible; in case when these transitions have $p > 1$, the last term in (14) is positive too, contributing to the photon flux. Thus Cr^{3+} not only increases the efficiency of utilization of pump but, by its effect of short-circuiting the cross-relaxation process W_4 it improves the population inversion and increases the contribution of the positive terms in the photon flux thus leading to the reduction of the laser emission threshold and to the increase of emission efficiency; it also contributes to the extension of the wavelength range of emission. Other possible Er-Cr cross-relaxation processes as, for instance $(^4I_{13/2} \rightarrow ^4I_{15/2}) + (^4T_2 \rightarrow ^4T_1)$ could improve the laser performances of the co-doped crystals. The effect of co-doping Er: YSGG with Cr was the improvement of the long-pulse 2.794 μm emission and the observation of the cw emission at 2.702, a laser transition with $p = 1.917$, that cannot be obtained in absence of Chromium. As expected from eq. (14) the effect of shortening T_1 in case of Cr co-doped crystals is similar to the case of crystals doped only with Er: it can extend the range of emission wavelength and, for transitions with $p > 1$ it reduced the threshold of emission, increases the efficiency and, due to a $(R'N_0)^{1/2}$ dependence on pump via N_1 , could give a non-linear dependence of the output power on pump. These effects have been indeed observed [10] in the 2.702 μm cw emission of Er^{3+} in Cr, Er: YSGG by co-doping with Ho^{3+} that leads to strong reduction of lifetime T_1 .

3. Conclusions

The possibility to obtain analytical expressions for populations of laser levels and photon flux density in case of the stationary regime of three-micron laser emission in Er-doped crystals enables an analysis of the influence of the various spectroscopic parameters of the system and of the pump conditions on this emission. This analysis shows that the three-micron laser emission in concentrated Er systems for which the lifetime of the terminal level is larger than that of the emitting level is essentially determined by the cross-relaxation processes W_1 , W_2 and W_4 . It also shows that the figure of merit p is essential in determining the possibilities to obtain laser emission on a given luminescence transition.

It is also shown that the wavelength range of emission is determined by the conditions imposed on the figure of merit p in order to obtain emission (positive population inversion and positive photon flux) and that these conditions could be modified by acting either on the system (co-doping with ions that modify the mechanism of population of the initial and terminal laser levels or / and with ions that reduce the lifetime of the terminal level for transitions with $p > 1$) or on the parameters that determine the figure of merit p itself. Concerning the effect of co-doping with Chromium, it is now evident that it not only increases the utilization of pump but also, provides a path for overcoming the effect of cross-relaxation W_4 thus, leading to a more efficient population of the initial laser level $^4I_{11/2}$.

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