Energy transfer from Fe^{3+} to rare-earth ions in YAG and the problem of sensitization of laser emission

V. Lupei, A. Lupei, S. Bonea, M. Nanau, M. Bolog

► To cite this version:

V. Lupei, A. Lupei, S. Bonea, M. Nanau, M. Bolog. Energy transfer from Fe^{3+} to rare-earth ions in YAG and the problem of sensitization of laser emission. Journal de Physique IV Proceedings, 1994, 04 (C4), pp.C4-505-C4-508. 10.1051/jp4:19944121 . jpa-00252572

HAL Id: jpa-00252572

<https://hal.science/jpa-00252572>

Submitted on 1 Jan 1994

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Energy transfer from Fe^{3+} to rare-earth ions in YAG and the problem of sensitization of laser emission

V. LUPEI, A. LUPEI, S. BONEA, M.P. NANAU and M. BOLOG

Institute of Atomic Physics, Bucharest 76900, Romania

Abstract

The static and dynamic measurements of Tm^{3+} spectra in YAG:Fe:Tm show that the $^3\text{H}_4$ level is efficiently populated by transfer from Fe^{3+} in tetrahedral sites. The features of the transfer processes, by investigation of Tm^{3+} acceptor luminescence characteristics, are presented.

The two micron Tm^{3+} laser emission is hampered by the low absorption of the radiation from the existing sources. To overcome this difficulty, the co-doping with transition metal ions has been used, Cr^{3+} ion being the most used sensitizer.

The possibility of sensitization of Tm^{3+} $^3\text{H}_4$ luminescence in YAG by co-doping with Fe^{3+} was already demonstrated [1, 2]. The energy transfer involves Fe^{3+} in d-tetrahedral sites, whose emission from $^4\text{T}_1$ (^4G) level has a good superposition with $^3\text{H}_4$ Tm^{3+} multiplet absorption in this matrix, especially at elevated temperatures.

Our previous studies concern mainly the static and dynamics spectral characteristics of the donor, Fe^{3+} (d), in the YAG:Fe:Tm samples. The data show clearly two features: (i) a complex behaviour of the global Fe^{3+} (d) luminescence decay under non selective pumping. It depends on Tm^{3+} content and at long times after the laser pulse could be described by a Forster law, indicating a dipole-dipole energy transfer mechanism with a temperature increasing C_{DA} micro-parameter. This interaction could not describe the beginning of the decay suggesting stronger interactions between near neighbours Fe-Tm pairs; (ii) the presence of new satellite lines in Fe^{3+} (d) absorption spectra (near the sharp line corresponding to $^6\text{A}_1 \rightarrow ^4\text{T}_2$ (^4D) transition) and a complete quenching of the donor Fe^{3+} (d) emission at pumping in these absorption peaks. These satellites could be assigned to Fe^{3+} (d)-Tm (c-dodecahedral) near neighbour pairs. The transfer within these pairs is very efficient. The YAG structure allows several types of close Tm^{3+} (c) - Fe^{3+} (d) pairs. Thus, near an Y^{3+} c-site there are two Al^{3+} d-sites at 3 Å, four at 3.67 Å, eight at 5.16 Å etc. The presence of pairs and the almost 100% transfer efficiency within them might be taken into account in the analysis of Fe^{3+} sensitizations of Tm^{3+} emission.

Since Tm^{3+} acceptor luminescence lines are much sharper, the satellite structure induced by Fe^{3+} co-doping is more complex and can provide additional information on the energy transfer processes. The aim of this paper is to present new data on Tm^{3+} spectra in YAG:Fe:Tm samples. Single crystals of YAG:Fe, YAG:Tm, and YAG:Fe:Tm, with various concentrations up to 5% have been comparatively investigated.

The absorption spectra (10K) of co-doped YAG:Fe:Tm samples present a complex satellite structure around Tm^{3+} main lines. Besides the lines observed in YAG: Tm^{3+} samples (line N corresponding to unperturbed Tm^{3+} centers and P-lines from centers perturbed by non-stoichiometric defects) [3], up to three new satellites F_i have been detected in various Tm^{3+} transitions, as illustrated in Fig. 1a for the ${}^3\text{H}_6(Z_1) \rightarrow {}^3\text{H}_4(W_1)$ transition of a YAG:Fe(5%):Tm(5%) sample. The intensity of F_i lines increases with dopants content, but due to the overlapping with P-lines and to their relative low intensity, a concentration dependence is difficult to measure.

When the Tm^{3+} emission is activated by the transfer from Fe^{3+} , at short times after the laser pulse the ${}^3\text{H}_4$ luminescence is dominated by the three F_i centers emission, as illustrated in Fig. 1b for a YAG:Fe(1%):Tm(1%) sample, excited non selectively with 532nm. This effect is more pregnant at high Tm^{3+} concentrations.

The luminescence decays of various F_i centers are similar, but very different from that of the main line N; a clear rise time being observed in the later case. The rise time is concentration and temperature dependent. The ${}^3\text{H}_4$ luminescence decays for N (Fig. 2a) and F_i (Fig. 2b) centers for a YAG:Fe(1%):Tm(1%) sample at 10K are presented in Fig. 2. The lack of any rise time for F_i emission indicate a very efficient transfer from Fe^{3+} (d) to Tm^{3+} .

The F_i ${}^3\text{H}_4$ emission could be also observed by selective excitation with a dye laser in the corresponding lines of the Tm^{3+} ${}^3\text{F}_3$ multiplet. This way one can obtain a better selectivity and some specific characteristics of F_i spectra could be observed. One feature must be outlined: F_i spectra contain more lines than N spectra. For illustration, in Fig. 3 we present part of the Tm^{3+} ${}^3\text{H}_4 \rightarrow {}^3\text{H}_6$ emission for N and F_i centers at 10K obtained by selective pumping in ${}^3\text{F}_3$ multiplet. If the F_3 emission is similar to that of N, F_1 and F_2 present new intense lines. Their presence can be correlated with structural data. For D_2 group, the local symmetry at the normal c-sites occupied preferentially by Tm^{3+} , the $\Gamma_1 \rightarrow \Gamma_1$ transitions are forbidden. The presence of intense lines such as those corresponding to $W_1(\Gamma_1) \rightarrow Z_2(\Gamma_1)$ for F_1 and F_2 centers indicate a strong perturbation of the crystal field and a lowering of the local symmetry at Tm c-site induced by a near neighbour Fe^{3+} ion. These perturbations could alter all the transition probabilities for these centers.

Thus the static and dynamic luminescence measurements show that the ${}^3\text{H}_4$ level of Tm^{3+} could be very efficiently populated by energy transfer from Fe^{3+} ions in tetrahedral sites. The ${}^3\text{H}_4$ level populates ${}^3\text{F}_4$ laser level the by the known cross-relaxation process (${}^3\text{H}_4, {}^3\text{H}_6 \rightarrow {}^3\text{F}_4, {}^3\text{F}_4$); the efficiency of this process could be reduced to some extent by a possible chain of transfers ${}^3\text{H}_4 \text{ Tm} \rightarrow \text{Fe}^{3+} (\text{a}) \rightarrow {}^3\text{H}_5 \text{ Tm}$, ending again in ${}^3\text{F}_4$. Two possible losses could be introduced here by the presence of iron: excited state absorption from ${}^4\text{T}_1({}^4\text{G})$ to ${}^4\text{T}_2({}^4\text{G})$ of Fe^{3+} and ground state absorption ${}^5\text{E} \rightarrow {}^5\text{T}_2$ in Fe^{2+} . Our measurements show that while ESA in Fe^{3+} peaks at higher energies than the main Tm^{3+} ${}^3\text{F}_4$ emission, GSA of Fe^{2+} is centered on this emission. Therefore, extreme care should be taken in the crystal growth to avoid the formation of Fe^{2+} .

Fig.1.a). Absorption spectrum of a YAG:Fe(5%)Tm(5%) sample at 10K; b). $^3\text{H}_4$ emission for a YAG:Fe(1%)Tm(1%) sample.

Fig.2. $^3\text{H}_4$ luminescence decay for: a). the main line N; b). F_1 satellite.

Fig. 3. Part of the 4H_4 emission of N and F_i centers in a YAG:Fe(1%)Tm(1%) sample at selective excitation in 3F_3 .

References

1. V. Lupei, G. Boulon, A. Brenier, C. Madej, M.J. Elejable, A. Lupei, C. Tiseanu, and S. Georgescu, *Solid State Lasers IV QE/Laser'93*, (Proceed. SPIE).
2. V. Lupei, G. Boulon, M.J. Elejable, A. Brenier, and C. Perdini, *Phys. Rev B*, (to be published).
3. A. Lupei, C. Tiseanu, and V. Lupei, *Phys. Rev. B* 47, 14084 (1993).