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ScienceDirect
Journal of Luminescence 122–123 (2007) 474–477
JOURNAL OF LUMINESCENCE
www.elsevier.com/locate/jlumin

Study of optical properties of YLF:Nd:Yb:Tm crystals

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Available online 24 March 2006

Abstract

In this paper we present a comparison between YLF:Yb:Tm and YLF:Yb:Tm:Nd systems identifying the most important processes that lead to an enhancement of thulium blue up conversion emission, under excitation around 792 nm, for the double sensitized system. Analysis of the 483 nm and 1030 nm emissions for the samples with different concentrations of Nd^{3+} ions showed that energy transfer between Nd^{3+} and Yb^{3+} is the main mechanism and is responsible for the enhancement in up-conversion.

Keywords: YLF; Thulium; Blue; Laser; Up-conversion; Energy transfer

1. Introduction

Nowadays laser research is focused in the development of visible compact sources. Particularly, blue laser are important in the compact disc industry, optical storage systems, color displays, in new medical and dentistry applications [1,2] and in atmospheric and physics research. Solid state lasers are attractive for most applications because they are rugged, relatively simple and easy to use.

Thulium-doped materials generate blue laser radiation through the nonlinear conversion of radiation from the infrared into visible range. The dynamics of up-conversion [3] is explained by taking into account various cross-relaxation (CR) and excited-state absorption (ESA) processes. The emissions at 480 and 450 nm can be observed after pumping thulium ions with two or three red or infrared photons. $YLiF_4$ (YLF) crystals doped with thulium and also co-doped with ytterbium [4,5] are well-known as active media that generate stimulated radiation on a number of lines over a wide spectral range from 450 to 2350 nm, upon selective laser, laser diode and flash lamp pumping.

In YLF, neodymium upper laser level present a very long storage time, a natural birefringence and a relatively weak thermal lensing. These characteristics make the YLF:Nd crystal a very important laser medium [6]. Recently, single-doped YLF fibers were produced showing a very interesting new way to develop compact low-loss and low-cost laser systems [7].

We present here the spectroscopic characterization of the new composition YLF:Yb:Tm:Nd crystal, under pumping at 792 nm. This pump, in Nd absorption band, results in a blue up-conversion emission, which can be used to make an YLF fiber laser.

2. Materials and methods

In this work we used YLF crystals grown by Czhochraski method in different compositions: YLF:1%Tm; 10%Yb;1%Tm; and 20%Yb,0.5%Tm,*x*%Nd. Three samples were cut from this last crystal from the beginning (#2), half (#5) and end (#8) that have a little Nd concentration change due to segregation coefficient. The samples were cut and polished with 2 mm thickness.

For the absorption measurements a spectrometer Cary 17D-Olis was used. For emission measurements, the samples were excited by a SDL diode laser at 792 nm and observed by a 0.5 m Spex monochromator, Stanford

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doi:10.1016/j.jlumin.2006.01.210

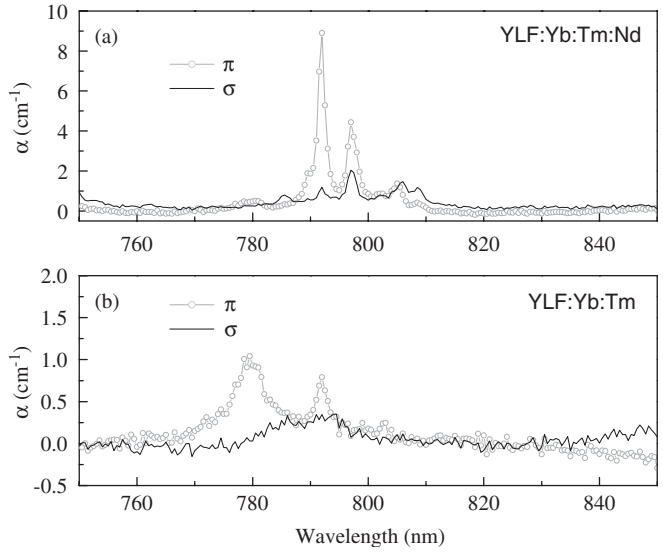

Fig. 1. Polarized absorption spectra of YLF:Yb:Tm:Nd (a) and YLF:Yb:Tm (b) samples.

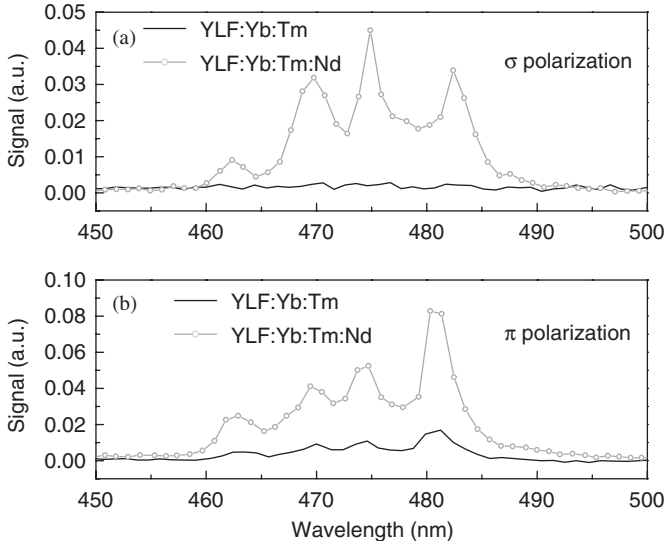

Fig. 2. Polarized emission spectra of YLF:Yb:Tm:Nd (a) and YLF:Yb:Tm (b) samples excited at 792 nm.

chopper, PAR-EG&G lock-in, Hammamatsu S-20 PMT and Germanium detector.

3. Results

Fig. 1 shows the absorption spectra of these the YLF:Yb:Tm and YLF:Yb:Tm:Nd samples in both polarizations σ and π. When YLF samples containing Tm^{3+} co-doped with Yb^{3+} or Yb^{3+} and Nd^{3+}, are excited at 792 nm, a strong blue emission is observed, Fig. 2. It is important to mention that the same excitation of YLF:1%Tm does not result in any detectable blue emission.

The blue emission observed in these spectra can be understood referring to the energy levels diagram shown in the Fig. 3. When the co-doped Yb/Tm sample is excited at 792 nm the following processes *a*′, *c*, *d* and *f* occur and populate the Tm^{3+} [$^1G_4$] level that emits blue. The same processes are complemented by the processes *a*, *b*, *e*, *g*, *h* and *i* with the addition of Nd as co-dopant in YLF:Yb:Tm.

Table 1 shows the energy transfer parameters obtained by overlap-integral between sensitizators-emissions and receptors-absorptions cross sections bands [8]. The energy transfer processes *f*, *c* and *g* are more important than the

Table 1
Energy transfer parameters for YLF:Yb:Tm:Nd crystal

Parameters	Values
σ_{abs} (Nd) (792 nm)	1.5×10^{-19} cm^2
σ_{em} (Nd) (1047 nm)	6.0×10^{-19} cm^2
σ_{em} (Nd) (960 nm)	2.1×10^{-21} cm^2
σ_{abs} (Yb) (960 nm)	2.1×10^{-21} cm^2
σ_{abs} (Tm) (792 nm)	7.3×10^{-21} cm^2
σ_{em} (Tm) (475 nm)	3.5×10^{-21} cm^2
$\tau(^2F_{7/2})$	2.0 ms
$\tau(^3F_4)$	15.0 ms
$\tau(^3H_5)$	1.0 ms
$\tau(^3H_4)$	2.1 ms
$\tau(^1G_4)$	75.0 μs
$\tau(^4F_{3/2})$	57.0 μs
f	($C_{YbTm} = 13.7 \times 10^{-40}$ cm^6/s; Rc = 12.0 Å)
c	($C_{TmYb} = 9.1 \times 10^{-40}$ cm^6/s; Rc = 11.2 Å)
b	($C_{NdYb} = 5.5 \times 10^{-40}$ cm^6/s; Rc = 9.8 Å)

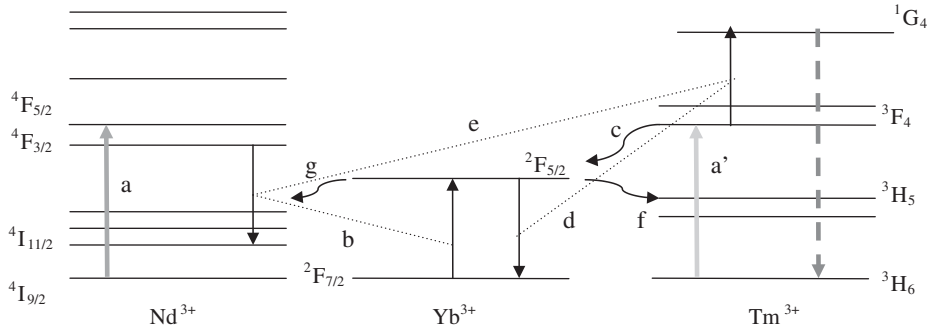

Fig. 3. Energy levels scheme and the energy transfer mechanism of Yb/Tm/Nd system.

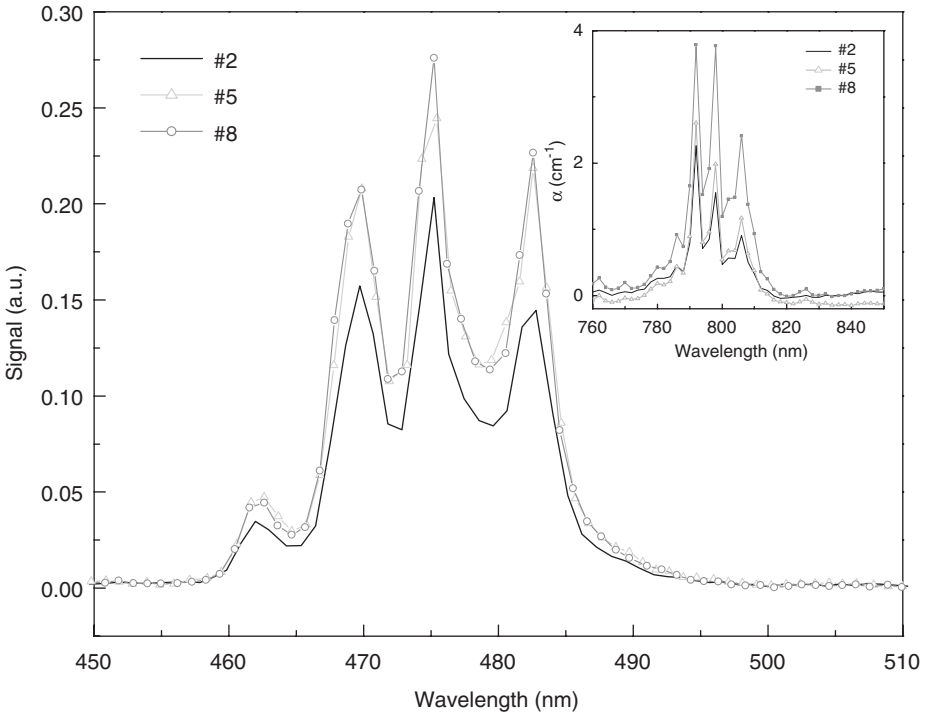

Fig. 4. Variation of Tm blue emission intensity in the three samples YLF:Yb:Tm:Nd containing different Nd concentrations. The inside figure shows the absorption coefficient of the three different samples of YLF:Yb:Tm:Nd crystal.

back transfer mechanism *e* since this last one involves absorption of phonons least probable that creation of phonons.

The energy transfer rate between Tm–Yb in the sample containing 10%Yb:1%Tm is $660\,s^{-1}$ and in the sample containing 20%Yb:1%Tm is $1020\,s^{-1}$ [4]. The energy transfer efficiencies are 47% and 57% in the samples containing 10% Yb and 20% Yb, respectively. It was observed that back transfer Tm–Yb is also an important mechanism, and that Tm–Yb energy transfer grows with Yb concentration.

We observe that a small Nd concentration variation in the YLF:Yb:Tm:Nd samples results in an enhancement of the Tm blue emission as can be seen in the Fig. 4.

The blue emission comes from the Tm^{3+} [$^1G_4$] level and increases with the pumping intensity with a slope of 1.6 in both the Yb:Tm and Yb:Tm:Nd samples, confirming that the upconversion process from the $^3F_4$ state to the $^1G_4$ state is a two-photon process.

4. Conclusions

Comparing the two different YLF compositions Yb/Tm and Yb/Tm/Nd we can conclude that: both compositions are efficient and generate blue emission by a two-photon process mechanism arising from the 792 nm excitation; the YLF:Yb:Tm:Nd sample has a higher absorption coefficient at 792 nm than the YLF:Yb:Tm sample, and consequently a higher absorption cross-section; since Tm ions are directly pumped by 792 nm wavelength, the ground state can be depleted, and therefore, the transition $^1G_4 \rightarrow {}^3H_6$, at ~483 nm can be used to generate stimulated emission using ground state depletion or pump-resonant excitation methods; the cross-relaxation Nd_x Tm leads to a growth in the $^1G_4$ population; a very important energy transfer mechanism between Nd and Yb ions was noticed considering that no Nd emission is observed in infrared region. This fact also impeaches the Tm [$^3F_4$] population which, decreases due to Tm–Nd interaction; the enhancement in blue emission is proportional to the enhancement in Nd absorption.

In conclusion, an enhancement in ~483 nm Tm^{3+} emission of almost 20 times was observed in the samples YLF:Yb:Tm:Nd compared to the YLF:Yb:Tm sample. This system could be interesting for development of compact laser systems using doped YLF fibers with about 5 mm in length and 300 μm in diameter, under diode laser pump.

Acknowledgments

The authors thank the Fundação de Amparo à Pesquisa do Estado de São Paulo, FAPESP, for the support under the grants 00/15135-9 and 05/50437-0.

References

[1] W.P. Risk, T.R. Gosnell, A.V. Nurmikko, Compact Blue-Green Lasers, Cambridge Studies in Modern Optics, Syndicate of the University of Cambridge, Cambridge, UK, 2003.

[2] L. Laser, DPSS 473-nm Microchip Blue Laser, Photonics Mini Magazine, March 2003 Edition.
[3] A. Wnuk, M. Kaczkan, Z. Frukacz, I. Pracka, G. Chadeyron, M. -F. Joubert, M. Malinowski, J. Alloy. Comp. 341 (1/2) (2002) 353.
[4] X.X. Zhang, P. Hong, M. Bass, B.H.T. Chai, Phys. Rev. B 51 (1995) 9298.
[5] N. Rakov, G.S. Maciel, M.L. Sundheimer, L. de S. Menezes, A.S.L. Gomes, Y. Messaddeq, F.C. Cassanjes, G. Poirier, S.J.L. Ribeiro, J. Appl. Phys. 92 (10) (2002) 6337.
[6] E.P. Maldonado, I.M. Ranieri, S.P. Morato, N.D. Vieira Jr., in: C.R. Pollock, W.R. Bosenberg (Eds.), Trends, Optics and Photonics Series, TOPS, Advanced Solid State Lasers, vol. 10, Optical Society of America, Washington DC, 1997, pp. 444–447.
[7] A.M.E. Santo, I.M. Ranieri, G.E.S. Brito, B.M. Epelbaum, S.P. Morato, N.D. Vieira Jr., S.L. Baldochi, J. Cryst. Growth 275 (3–4) (2005) 528.
[8] T. Förster, Ann. Phys. 2 (1948) 55;
D.L. Dexter, J. Chem. Phys. 21 (1953) 836.