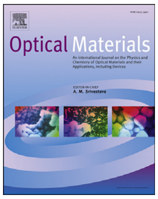
Optical Materials 72 (2017) 578–582

Contents lists available at ScienceDirect

Optical Materials

journal homepage: www.elsevier.com/locate/optmat



1.87 μm laser operation by 980 nm pumping in Yb,Tm:CaF_2

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ARTICLE INFO

Article history:
Received 20 April 2017
Received in revised form
19 June 2017
Accepted 25 June 2017

Keywords:
Laser materials
Rare earth
Energy transfers
Infrared lasers

ABSTRACT

We report the first laser operation of an Yb:Tm:CaF_2 crystal around 1.9 μm after pumping at 980 nm. A maximum power of 140 mW was obtained corresponding to a slope efficiency of 14.5% with respect to the incident pump power. The spectroscopic study of this laser medium allows the optimization of the Yb/Tm concentration ratio to achieve laser operation and tune the energy transfers efficiency between Yb and Tm ions.

1. Introduction

The need for diode-pumped solid state lasers emitting around 2 μm is a growing field due to an increasing number of applications [1]. Because of the water absorption in this spectral region, laser sources can be used for medical purposes as well as for monitoring molecular species in the atmosphere with the advantage to fit the eye-safe spectral window. The 2 μm lasers are also suitable for non-linear crystals pumping in Mid-Infrared Optical Parametric Oscillators [2]. The main reported works in the spectral region around 2 μm are based on Tm and Ho doped materials. Tungstate, fluoride bulk crystals and glass fibers are the most studied materials to achieve CW (continuous-wave) and, more frequently, Q-switched laser operation. Mode-locking of Tm-Ho systems, to reach pico-seconds or femtoseconds regimes has been also widely considered for the past couple of years to satisfy the requirement for broad bands to obtain ultrashort pulse laser operation [3].

The 2 μm emission of Tm-doped materials occurs between the first excited level and the ground state of the Tm^{3+} active ions, and the optical pumping of such quasi-three levels laser systems is either centered around 1.7 μm, for most fiber lasers, or around 800 nm for Tm-doped crystals. This latter pumping scheme leads in principle to an efficient "2 for 1" cross relaxation process, but usually requires highly rare-earth doped materials (several %) or hosts with ion clusters such as Tm:CaF_2 [4].

Another pumping scheme can be achieved with a co-doped Yb, Tm system by using energy transfers between Yb^{3+} and Tm^{3+} ions [5], in a similar way as Yb, Er systems which involve Yb^{3+} to Er^{3+} energy transfers to obtain the 1.5 μm laser emission. In fact, Yb, Tm co-doped materials can be used to achieve both visible and IR laser emissions from different levels [6–9]. Most importantly for the purpose of this paper, this also means that Tm^{3+} ions can be efficiently excited by pumping Yb^{3+} ions around 980 nm, with high power and cost-effective laser diodes.

Energy transfers are most efficient when the distance between rare earth ions is short. This condition is usually fulfilled in highly Yb-doped materials, with typical Yb doping levels of about 20%in the case Yb, Er fibers [10] in which ions are randomly distributed. Distances are drastically reduced in crystals having like CaF_2 where ion-clustering occurs because of charge compensation during the fabrication process. These clusters may involve several ions depending on the rare earth ion size. It was shown for instance that Yb:CaF2 should be considered as a multisite system whose luminescent and lasing properties are dominated by a series of Yb^{3+} clusters ranging from dimers to tetramers [11,12]. Consequently, ions are very close to each other within clusters and

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http://dx.doi.org/10.1016/j.optmat.2017.06.050
0925-3467/

energy transfers are therefore particularly efficient in CaF_2. For these reasons, cross relaxation is observed in Tm:CaF_2, even at relatively low dopant concentration (0.1%at.), and efficient tunable CW laser operation around 2 μm was previously demonstrated in this material [4] by using a dopant level of only about 1.9%. The pump wavelength, however, was centered at 767 nm which is not as standard as 792 nm, a wavelength for which commercial laser diodes are available. Moreover, by directly pumping Tm^{3+} ions at 767 nm, the absorption was relatively small, leading then to a relatively low overall laser efficiency. Therefore, in order to maximize the laser performance of this specific crystal, it makes sense to co-dope the crystals with Yb^{3+} ions, to pump them around 980 nm and take advantage of Yb, Tm energy transfers. The impact of this codoping on the laser performance is at the core of this paper where we report the results of a detailed spectroscopic analysis of co-doped Yb,Tm:CaF_2 single crystals, and we demonstrate CW laser operation around 1.87 μm by pumping Yb^{3+}ions around 980 nm.

2. Experimental conditions

CaF_2 laser crystals were grown in our laboratory using the Bridgman technique in a homemade furnace, operating with a high purity carbon crucible. Starting materials were CaF_2, YbF_3 and TmF_3 powders. After growth, the crystals were cut and polished with parallel end faces. Several crystals, with different rare-earth concentrations (Yb-Tm = 8%–0.2%, 8%–0.8%, and 5%–2%) were synthetized to realize the spectroscopic study as well as the laser tests. Absorption spectra were obtained with the aid of a PE 1050 Spectrophotometer. The lifetime measurements were acquired by exciting the samples with an OPO laser (Continuum Horizon) and analyzing the fluorescence signals with an ORIEL monochromator and a photomultiplier or an InGaAs photodiode depending on the investigated spectral domain. Emission spectra were obtained by exciting the samples with a CW Ti:Sapphire laser and recording the sample emission first in the visible and the near-infrared (between about 300 nm and 1.1 μm) with a calibrated Optical Spectrum Analyzer (OSA). The near- and mid-infrared emission spectra (between about 900 nm and 2.1 μm) were recorded with a Jobin Y von HRS2 monochromator, an InGaAs detector and a lock-in amplifier, the whole setup being calibrated with a blackbody. Laser experiments were achieved by using a two-mirror plano-concave laser cavity with an output coupler having a transmission of 1.8% at 1.9 μm and a radius of curvature of 100 mm and by pumping the laser element with a high brightness ($M^2 = 1.1$) laser source (Azur Light Systems) emitting up to 15 W at 980 nm.

3. Results and discussion

3.1. Spectroscopic analysis

Absorption spectra of Tm-doped and co-doped Tm, Yb:CaF_2 crystals are reported in Fig. 1. It shows both the absorption peak corresponding to the $^2F_{7/2} \rightarrow {}^2F_{5/2}$transition of Yb^{3+} ions around 980 nm and the absorption peaks associated with the Tm^{3+} ions. Comparison between the Tm singly doped and the Tm, Yb codoped spectra shows that the presence of Yb^{3+} ions does not affect the shape of the Tm^{3+} absorption spectrum. This is the case for all the concentrations studied in this work which means that, like for Yb^{3+} ions [12], at concentration higher than about 1%, there is a dominant center for Tm^{3+} ions with the same absorption spectrum regardless of the codopant concentration.

Judd-Ofelt calculations have been performed using the singly doped Tm:CaF_2crystal absorption spectrum [13]. It allows to estimate the radiative lifetimes as well as the branching ratios of the main transitions coming from the $^3F_4$and $^3H_4$ infrared emitting levels (Table 1).

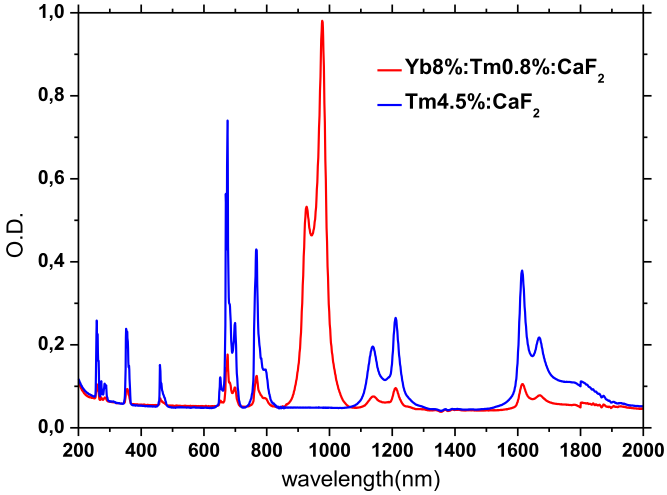

Fig. 1. Absorption spectra in optical density of 4.5%Tm:CaF_2 (blue) and 8%Yb0.8% Tm:CaF_2 (red). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

As indicated by the emission spectra reported in Figs. 2 and 4 and the energy level scheme displayed in Fig. 3 (and discussed in more details later on), exciting the co-doped Yb,Tm:CaF_2 crystal at 980 nm, i.e. the $^2F_{5/2}$ level of the Yb^{3+} ions, leads to the characteristic Yb^{3+} broadband emission around 1000 nm and to a series of emissions coming both from upper lying and lower-lying metastable Tm^{3+} energy levels. Energy transfers thus occur between Yb^{3+} and Tm^{3+} ions but also among Tm^{3+} ions which helps to populate the different emitting levels. According to the energy level scheme of Fig. 3, the observed emission transitions all come from the$^3P_0$, $^1D_2$, $^1G_4$, $^3H_4$ and $^3F_4$ emitting levels down to lower levels, with some transitions overlapping between each other, like the $^3P_0 \rightarrow {}^3H_4$ and $^1D_2 \rightarrow {}^3F_4$ transitions around 470 nm, for instance.

It is worth noting here that when ions are randomly distributed, like in YAG or YLF laser crystals, the transfer efficiency directly depends on the dopant concentration. However, in the case of CaF_2, the situation is much more complicated, and the efficiency of the system depends, in fact, on the distance between Yb^{3+} and Tm^{3+}ions within clusters, and on the real number of emitting clusters since some clusters can be quenched. It means that the relevant parameter is here the ratio between the Yb^{3+} and Tm^{3+}ion concentrations, and the nature (symmetry, mean distance between ions) of the dominant clusters in the co-doped Yb,Tm:CaF_2 system.

Therefore, to understand the emission behavior of co-doped Yb,Tm:CaF_2 crystals, the first approach was to measure fluorescence decays in the same experimental conditions and to compare the results by changing the Yb/Tm concentration ratio. From the decay rates of the emitting levels it was then possible to find the energy transfer efficiency between ions as a function of the Tm^{3+} and Yb^{3+} concentrations.

In a first approximation, the fluorescent lifetime τ_f of a given emitting level can be written as:

Table 1
Tm:CaF_2 lifetimes and branching ratios calculated with JO analysis.

Tm Emission transitions	Lifetime	Branching ratio
$^3F_4 \rightarrow {}^3H_6$	16.8 ms	1
$^3H_4 \rightarrow {}^3H_6$	2.51 ms	0.87
$^3H_4 \rightarrow {}^3F_4$	2.51 ms	0.084
$^1G_4 \rightarrow {}^3H_4$	1.26 ms	0.083