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Optical gain by upconversion in Tm–Yb oxyfluoride glass ceramic

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Abstract Evidence of positive optical gain is observed in Tm^{3+}–Yb^{3+}-codoped oxyfluoride glass ceramic in an upconversion pump and probe experiment. The $^1G_4$ level of the Tm^{3+} ions is populated by an upconversion mechanism under excitation of the Yb^{3+} ions at 975 nm with a high-power pulsed laser and give rise to an intense emission from the $^1G_4$ to the $^3F_4$ levels. The $^1G_4 \rightarrow {}^3F_4$ electronic transition is stimulated with a low signal at 650 nm as a probe.

Under this condition, we reach the population inversion necessary between the Tm^{3+} levels of the transition $^1G_4 \rightarrow {}^3F_4$ and we observed an increase of the emission intensity at the signal wavelength due to the stimulated emission. A positive optical gain of around 4 cm^{-1} (∼17 dB/cm) has been measured in Tm–Yb-codoped oxyfluoride glass ceramic.

1 Introduction

Rare earth (RE) doped low phonon optical materials are important for infrared solid lasers, optical amplifiers, upconversion lasers and visible display devices [1–4]. With the rapid increment of demand in high transmission capacity, Tm^{3+}-doped materials have attracted significant interest in 1.4 μm broadband application. Meanwhile, Tm^{3+}-doped materials also catch much attention in the area of visible upconversion processes because it does not require any nonlinear medium for harmonic generation to obtain a blue laser. Because Tm^{3+}-doped materials do not have any absorption band near 980 nm, commercial 980 nm diode lasers cannot be used as a powerful pumping source. To overcome the shortcoming, co-doping with Yb^{3+} is adopted to achieve the strong energy absorption at 980 nm [5].

In this sense, oxyfluoride glass ceramics have shown to be an interesting matrix for RE ions due to the lower phonon energy in the fluoride environment, which reduces the nonradiative decay rates and shortening of distances between the RE ions, which favors the energy transfer processes [6–8]. Recently, upconversion mechanisms have been reported in Tm^{3+} single-doped oxyfluoride glass ceramics and in Tm^{3+}–Yb^{3+}-codoped samples [7, 9]. These emissions are drastically increased in the codoped samples.

In this context we are investigating the optical amplification properties of trivalent lanthanide ions doped into the oxyfluoride glass ceramics, with the purpose of extending the knowledge and the application perspective of this material [10–14]. In this paper we present the results of a series of pump and probe experiments carried out on oxyfluoride glass ceramics activated with Tm^{3+}–Yb^{3+}.

2 Experimental

The samples of this study have been reported in a previous work [7, 9]. Measurements of optical amplification were carried out in a pump and probe experimental setup. The pump radiation was provided by an optical parametric oscillator (OPO) tuned at 975 nm with high-energy pulses between 50 and 115 mJ/cm^2 of about 5 ns of duration. The probe

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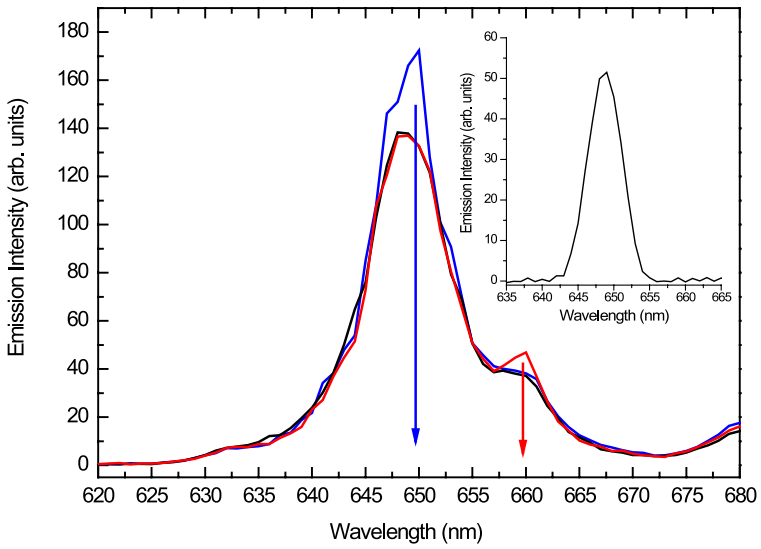

Fig. 1 Emission spectrum for Tm^{3+}–Yb^{3+}-codoped oxyfluoride glass ceramic associated to the $^1G_4 \rightarrow {}^3F_4$ electronic transition. *The black line* gives the emission of the sample under pump excitation, *the blue line* shows the emission under pump and probe excitation at 650 nm (subtracting the signal of the probe shown *in the inset*) and *the red line* shows the emission under pump and probe excitation at 660 nm (subtracting the signal of the probe). The probe wavelengths are indicated with *arrows*

beam was obtained by a continuous 1000-W lamp together with a monochromator, giving a signal power density of 195 μW/cm^2 at 650 nm with a spectral width of 5 nm.

The incidence of pump and probe beams were normal to the surface of the sample, which was situated after a 1 mm diameter pinhole. A dichroic mirror was employed to align both laser beams. In order to cover only the whole area of the pinhole, the pump beam was focused a 20 cm focal length lens.

The detection chain was formed by a TRIAX-180 monochromator with 1 nm resolution and the output of the photomultiplier tube was registered by a digital oscilloscope TEKTRONIX-2430A for temporal analysis of the decay curves.

To determinate the optical gain, two kinds of emission spectra were measured. In the first one, the pump and probe beams were present simultaneously, while the probe was blocked for the second one. The two spectra were compared after subtracting the continuous background due to the probe.

3 Result and discussion

As mentioned in the experimental section, we have recorded two upconversion emission spectra: first with pump and probe present and then when the probe is blocked [10, 14]. This result is given in Fig. 1.

An increase of the detected intensity at the signal wavelength, 650 nm (indicated by an arrow in the figure), can be clearly appreciated. This increment is due to the stimulated emission associated with the $^1G_4 \rightarrow {}^3F_4$ transition that occurs at the probe wavelength and it is the physical basis of signal amplification. Moreover, to corroborate that this enhancement of the emission is due to the probe beam, the wavelength of the probe beam is shifted from 650 nm to the sides of the spectrum. It is found that the intensity enhancement shifted accordingly to the signal wavelength as can be seen in Fig. 1. Under this pumping scheme population inversion for $^1G_4 \rightarrow {}^3F_4$ transition at 650 nm is expected at $t = 0$ s because $^3F_4$ is not initially populated.

The Tm^{3+} :$^3H_6 \rightarrow {}^1G_4$ ground state absorption (GSA) is centred about 460 nm. In the pump and probe experiments, the high-power pump pulses at 975 nm induce a nearly resonant GSA strongly populating the $^1G_4$ excited level by upconversion mechanism. In these conditions, a probe beam tuned at 650 nm can induce a relaxation process involving the stimulated emission of a photon at the same frequency. The energy levels diagram shows the proposed mechanisms to find optical amplification in this setup (see Fig. 2).

The net optical gain resulting from the stimulated emission process can be determined by evaluating the signal enhancement (SE) when the probe beam passes through the crystal, defined as follows [14, 15]:

$$\mathrm{SE} = \frac{I_{\mathrm{pp}} - I_{\mathrm{p}}}{I_{\mathrm{probe}}} \tag{1}$$

where I_{pp} is the intensity detected at 650 nm in the direction of the probe beam coming out from the sample when it is irradiated simultaneously with the pump and the probe beams, I_{p} is the spontaneous emission intensity at the same wavelength when the probe is blocked before the sample, and I_{probe} is the intensity of the probe beam. When a probe beam passes through a material, its intensity decreases according to the exponential law:

$$I_{\mathrm{probe}} = I_0 e^{-\alpha L} \tag{2}$$

where I_0 is the probe intensity at the entrance of the medium, α the absorption coefficient of the sample at the probe wavelength, and L the length of the sample.

With both pump and probe beams incident onto the sample, I_{pp} can be written as

$$I_{pp} = I_p + I_0 e^{(g_{int}-\alpha)L} \tag{3}$$

where g_{int} is the internal gain coefficient due to stimulated emission of the sample at the probe wavelength. The GSA at 650 nm ($^3H_6 \rightarrow {}^3F_2$) is 1.5 cm^{-1}, then a net optical gain coefficient g_{net} can be expressed as

$$g_{net} = g_{int} - \alpha \tag{4}$$

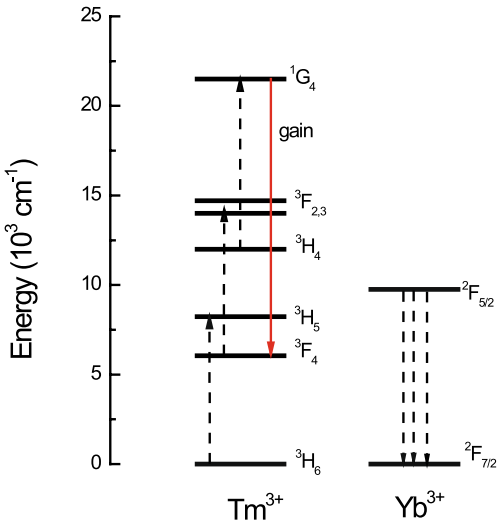

Fig. 2 Energy level scheme of Tm^{3+}–Yb^{3+}-codoped oxyfluoride glass ceramic. *The dashed lines* show the upconversion process to populate $^1G_4$ level of Tm^{3+} ions by energy transfer from Yb^{3+} ions, which are produced by using OPO laser at 975 nm. *The solid line* shows the optical gain transition

By introducing (2), (3) and (4) into (1) the following expression for the net gain coefficient g_{net} is obtained:

$$SE = \exp(g_{net} L) \tag{5}$$

The intensities I_p, I_{pp} and I_{probe} can be experimentally measured (see Fig. 3a). As we use a pulsed excitation source, I_p and I_{pp} are experimental curves that decay after the excitation pulse. Therefore, the values of SE and g_{net} are calculated by using (1) and (5) from these experimental curves (see Fig. 3b and 3c). The maximum of the detected intensity just after the pump pulse was used in order to obtain the g_{net} value.

The net gain coefficient as a function of the pump energy density is given in Fig. 4. A continuous growth of the net gain coefficient as a function of the pump power density can be observed in the figure. Finally, the maximum value for the net gain has been observed for a pump energy density of 115 mJ/cm^2 corresponding to 4 cm^{-1} (∼17 dB/cm) during the maximum of the detected intensity. However, according to (4), this value is reduced due to the $^3H_6 \rightarrow {}^3F_2$ absorption. Therefore, a value of 5.5 cm^{-1} is obtained for the internal gain of this amplification.

Due to the upconversion excitation mechanism used in this amplification, this process is only efficient during a time of about 0.5 ms (as can be seen in Fig. 3). In this period, an average net gain of 2.1 cm^{-1} (∼9.9 dB) can be calculated.

4 Conclusion

Positive optical gain at 650 nm has been observed in Tm^{3+}–Yb^{3+}-codoped oxyfluoride glass ceramic in an upconversion pump and probe experiment. High-power pump laser pulses at 975 nm were used to induce population inversion

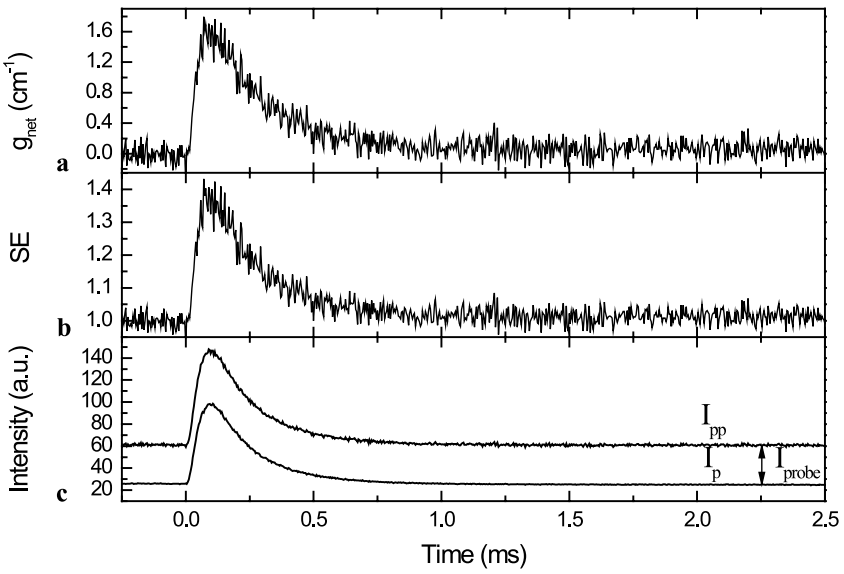

Fig. 3 (**a**) Experimental intensity of the pump (I_p) and pump–probe curve (I_{pp}), where the probe signal is indicated (I_{probe}). (**b**) Signal Enhancement as a function of the time obtained for these experimental data and (**c**) net optical gain as a function of time with pump energy densities of 61.2 mJ/cm^2 with a power probe density of 195 μW/cm^2

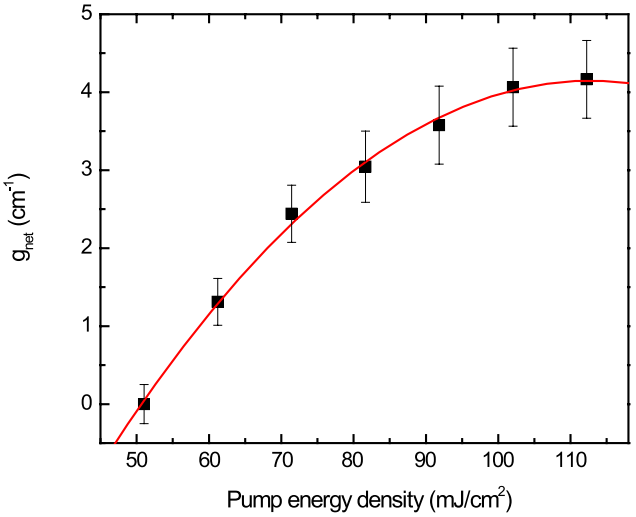

Fig. 4 Net optical gain as a function of the pump energy density from 50 to 115 mJ/cm^2 with a power probe density of 195 μW/cm^2. *The continuous line* is a guide for the eye

between the $^1G_4$ and $^3H_6$ levels, whereas a 650 nm probe beam from a lamp was used to stimulate the $^1G_4 \rightarrow {}^3F_4$ emission transition. A maximum gain coefficient of about 4 cm^{-1} (∼17 dB/cm) was measured for a pump energy density of 115 mJ/cm^2 and the threshold for a positive optical amplification was found for a pump energy density of 65 mJ/cm^2. These new results confirm the remarkable potentialities of in Tm^{3+}–Yb^{3+}-codoped oxyfluoride glass ceramic for applications in the solid-state laser and optical amplifier technologies.

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