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Study of the most suitable new glass laser to incorporate ytterbium: alkali niobium tellurite, lead fluorborate or heavy metal oxide

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Abstract

Diode-pumped Yb-doped glass lasers have received considerable attention for applications such as high-power beam production or femtosecond pulses generation. In this paper, we evaluate the laser potential of three different glass families doped with Yb^{3+}: alkali lead fluorborate ($PbO–PbF_2–B_2O_3$), heavy metal oxide ($Bi_2O_3–PbO–Ga_2O_3$) and niobium tellurite ($TeO_2–Nb_2O_5–K_2O–Li_2O$). Spectroscopic properties were studied for the samples and calculations of the minimum laser pump intensity (I_{min}), saturation fluence (U_{sat}) and the theoretical limit of peak power (P_{max}) are also presented. A comparison of laser properties of these three different glasses and their importance is shown and analyzed.
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1. Introduction

Developments in high-field lasers for next generation nuclear fusion have indicated that Yb-doped materials, particularly glasses, are the best host materials to efficient energy storage in the excited state [1]. Since Yb^{3+} has the [Xe] $4f^{13}$ electron configuration, it offers the advantage of a small number of 4f states. The small quantum defect reduces the heat load in the Yb:glass. Materials doped with Yb^{3+} ions have efficient emission when pumped by diode lasers without the possibility of excited state absorption [2]. The Yb^{3+} ions are of interest also as a sensitizer of energy transfer for infrared to visible up conversion and infrared lasers [2].

Glasses are materials capable of high peak power generation due to their high saturation fluence, broad emission bandwidth and long upper-state lifetime. Laser glasses are usually evaluated by means of these spectroscopic properties. Emission cross section and fluorescence

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lifetime are calculated using intensity parameters based on the Judd–Ofelt theory [3,4]. Since there is only the $^2F_{5/2} \rightarrow {}^2F_{7/2}$ transition for Yb^{3+}, it is impossible to calculate directly the Judd–Ofelt parameters for this ion. For this reason, the compositional dependence of the spectroscopic properties of Yb^{3+}-doped glasses is not well established. Up to now, there are only a few papers involving the effect of composition on the emission cross section of Yb^{3+} in simple systems as borate, phosphate, silicate and tellurite glasses [5–8].

It was shown recently that Yb-doped tellurite glasses have more advantages over some other excellent laser glasses, being comparable to laser crystals like Yb:YAG and Yb:YAP, exhibiting high emission cross sections and long luminescence lifetimes [6,9]. In this paper, we present the study and characterization of three different families: lead fluorborate ($PbO–PbF_2–B_2O_3$), heavy metal oxide ($Bi_2O_3–PbO–Ga_2O_3$) and alkali niobium tellurite ($TeO_2–Nb_2O_5–K_2O–Li_2O$), prepared containing Yb^{3+}, stable against crystallization.

A comparison of laser properties of these three different glasses and their importance is shown and analyzed in the following pages.

2. Experimental procedure

The lead fluorborate glasses used in this work were prepared with 99.99% pure elements (all products from Fluka and Aldrich). Concentration of 0.5 mol% of Yb_2O_3 was added to a glass matrix composed of: (mol%) $43.5H_3BO_3–22.5PbCO_3–34.0PbF_2$. These materials were melted in air at 1000°C for approximately 1 h and a half, using alumina crucibles. The melt was poured into preheated brass molds for a quick solidification and then annealed at 400°C (below the glass transition temperature) for 12 h. Yellow-colored, transparent and homogeneous samples were obtained.

The heavy metal oxide glasses presented in this work, with 0.5 mol% of Yb_2O_3 were prepared, with (mol%)$25.0Bi_2O_3–57.0PbO–18.0Ga_2O_3$. After melting the starting powders in platinum crucibles at 1000°C for 1 h and a half, they were poured onto a preheated mold and annealed for 3 h at 400°C, and then cooled inside the furnace.

To obtain the niobium tellurite glasses, appropriate mixtures of reagent-grade TeO_2, Nb_2O_5, Li_2CO_3 and K_2CO_3 were melted in gold crucibles for 30 min at 830°C, in air. The liquids were quenched at room temperature in steel molds and, annealing treatments were performed at a temperature near Tg (glass transition temperature) for 30 min. Samples with compositions of: $80TeO_2–10Nb_2O_5–5K_2O–5Li_2O$, stable against crystallization, were also prepared containing ytterbium ions. Table 1 shows the concentrations, densities and refractive index of studied glasses.

Glass samples with two polished faces and 3 mm thickness were used for refractive index, absorption, and emission and lifetime measurements. The sample surfaces were polished flat and parallel. A Carl Zeiss microscope with a $10\times$ objective lens was used to measure the refractive index. The refractive index was determined by means of the "apparent depth method" [10]. This method relates the physical thickness of a transparent specimen to its optical thickness (apparent thickness).

The absorption spectra in the range 920–1120 nm were measured using a Varian Spectrometer Cary 17 D at room temperature. The emission spectra were obtained by exciting the samples, at 968 nm, with a GaAlAs laser diode (Optopower A020). This diode system contains a broad area semiconductor laser with a maximum of 20 W of continuous output power operating at

Table 1
Concentrations, densities and refractive index of studied glasses

Host	Density (g/cm^3)	Refractive index	Yb (mol%) (ions/cm^3)
Lead fluorborate	6.90	2.20	0.5 (1.15×1020)
Heavy metal oxides	7.00	2.52	0.5 (0.64×1020)
Niobium tellurite	5.50	2.09	5 (4.21×1020)

this wavelength. The diode laser beam was treated with a beamshaper [11] and focused by a single $f = 5$ cm lens. Close to the focus, and for a depth of focus of 2 mm, the beam has a square profile, with transverse dimensions of approximately $260 \times 260\,\mu m$. During the emission measurements, the sample was pumped by a 7.5 W and the diode laser beam, modulated at 40 Hz. The emissions of the samples were analyzed with a 0.5 m monochromator (Spex) and a germanium detector. The signal was amplified with an EG&G7220 lock-in and processed by a computer. The lifetimes of excited Yb^{3+} ions were measured using pulsed laser excitation (4 ns) from an OPO pumped by a frequency-doubled Nd:YAG laser (Quantel). The signal was detected by an InSb detector with an appropriated line filter detector and analyzed using a signal processing Box-Car averager (PAR 4402). The densities were measured with the Archimedes method, and the Yb^{3+} concentrations were determined by X-ray fluorescent spectrometry with wavelength dispersion. The relative errors in the emission measurements are estimated to be $<5\%$, errors in the lifetime measurements are $<10\%$.

3. Results

Absorption and emission cross-sections spectra for the three hosts doped with Yb^{3+} are shown in Figs. 1–3. Table 2 presents some of the spectroscopic properties as the fluorescence lifetimes measured (τ_f), fluorescence effective linewidth (Δ_{eff}), as well as the spontaneous emission probabilities (A_R) and the emission cross-sections (σ_{em}) that were calculated using the following equations [5]:

$$A_R = \frac{8\pi c n^2 (2J'+1)}{\lambda_p^4 (2J+1)\rho} \int k(\lambda)\,d\lambda, \quad (1)$$

$$\sigma_{em}(\lambda) = \frac{\lambda^4 g(\lambda) A_R}{8\pi n^2 c}, \quad (2)$$

where c is the velocity of light, n the refractive index of the glass samples, λ_p the absorption peak wavelength (968 nm), ρ the concentration of Yb^{3+} ions, $k(\lambda)$ the absorption coefficient, J' and J the

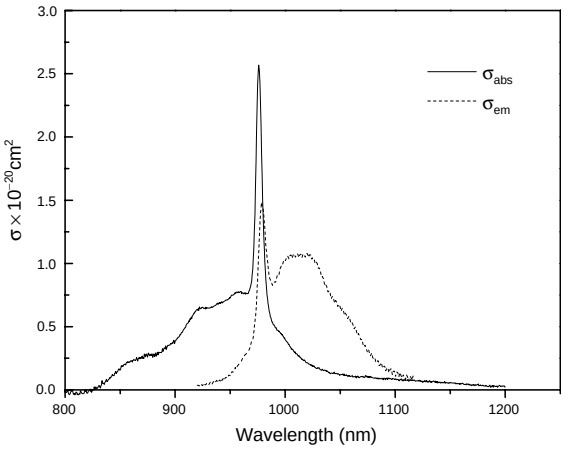

Fig. 1. Absorption and emission cross-sections spectra for the lead fluorborate glass with 0.5 mol% of Yb_2O_3.

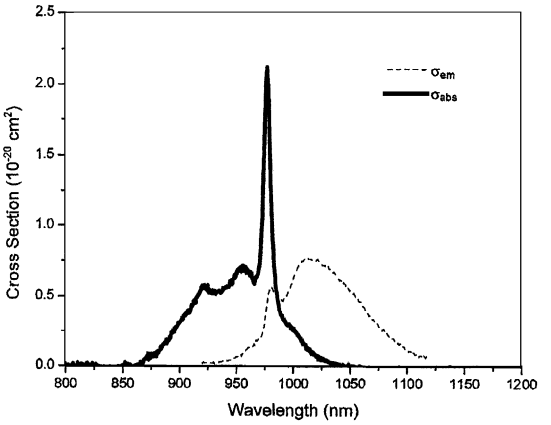

Fig. 2. Absorption and emission cross-sections spectra for the heavy metal oxide glass with 0.5 mol% of Yb_2O_3.

total momentum for the upper and lower levels and $g(\lambda)$ the normalized line shape function of the fluorescence transition of Yb^{3+}. In Table 2 $\sigma_{abs}(\lambda_0)$ and $\sigma_{em}(\lambda_0)$ represent the absorption and the emission cross sections at the extraction wavelength (λ_0) and $\sigma_{abs}(\lambda_p)$ the absorption cross section at the laser pump wavelength (λ_p).

Table 3 presents the minimum pump intensity (I_{min}), which is a measure for the ease of pumping the laser material. I_{min} describes the minimum absorbed pump intensity that is required for transparency to be achieved at the extraction wavelength (λ_0) and is calculated by the following

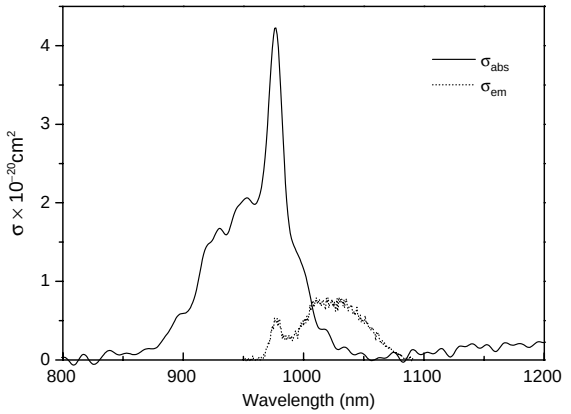

Fig. 3. Absorption and emission cross-sections spectra for the niobium tellurite glass with 5 mol% of Yb_2O_3.

equation [9]:

$$I_{\min} = \beta_{\min} I_{\text{sat}},$$

where

$$\beta_{\min} = \frac{\sigma_{\text{abs}}(\lambda_0)}{\sigma_{\text{em}}(\lambda_0) + \sigma_{\text{abs}}(\lambda_0)}, \tag{3}$$

$$I_{\text{sat}} = \frac{hc}{\lambda_p \tau_f \sigma_{\text{abs}}(\lambda_p)}.$$

In the equations above, I_{sat} is the pump saturation intensity and $\beta_{\min}$ is defined as the minimum fraction of Yb ions that must be excited to balance the gain exactly with the ground-state absorption at λ_0 ($\beta_{\min} = 0.17$ and $I_{\text{sat}} = 9.9\,\text{kW/cm}^2$ for the lead fluorborate glass, $\beta_{\min} = 0.30$, $I_{\text{sat}} = 23.0\,\text{kW/cm}^2$ for the heavy metal oxide one and $\beta_{\min} = 0.20$, $I_{\text{sat}} = 8.0\,\text{kW/cm}^2$ for the niobium tellurite). The $\beta_{\min}$, and I_{sat} in this case, must be small when high intensity lasers must be attained.

When the importance is the efficient storage of optical energy, the laser material must have high doping density, and high fluorescence lifetime (and consequently small emission cross section). This factor determines the rate at which the stored energy is depleted due to spontaneous emission, and the pump power which is needed to create a population inversion in the gain material and is given by

$$U_{\text{sat}} = \frac{h\nu_0}{\sigma_a(\lambda_0) + \sigma_e(\lambda_0)}. \tag{4}$$

Table 2
Spectroscopic properties of Yb^{3+}-doped lead fluorborate, heavy metal oxide and niobium tellurite glasses

Host	Concentration (10^{20} ion/cm^3)	$\sigma_{em}(\lambda_0)$ (10^{-20} cm^2)	$\sigma_{abs}(\lambda_p)$ (10^{-20} cm^2)	$\sigma_{abs}(\lambda_0)$ (10^{-20} cm^2)	λ_0 (nm)	A_R (s^{-1})	$\Delta\lambda_{eff}$ (nm)	τ_f (ms)	$\sigma_{em}\tau_f$ (10^{20} cm^2 ms)	U_{sat} (J/cm^2)
Lead fluorborate	1.15±0.06	1.07±0.08	2.56±0.18	0.22±0.02	1022	3515.2	60.70	0.81±0.04	0.86	16.20
Heavy metal oxides	0.64±0.03	0.75±0.05	2.20±0.15	0.12±0.01	1012	3000.0	86.00	0.40±0.02	0.10	22.60
Niobium tellurite	4.21±0.21	1.10±0.06	4.09±0.20	0.28±0.01	1028	3200.0	66.1	0.59±0.06	0.65	14.02

Table 3
Spectroscopic properties of some laser glasses and crystals doped with Yb^{3+} [6,7,11]

Materials	σ_{em} (10^{-20} cm^2)	λ_0 (nm)	I_{min} (kW/cm^2)	τ_f (ms)	$\sigma_{em}\tau_f$ (10^{-20} cm^2 ms)
QX	0.70	1018	1.80	2.00	1.40
ADY	1.03	1020	1.12	1.58	1.63
LY	0.80	1028	1.95	1.68	1.35
PN	1.35	1035	0.59	1.36	1.83
PNK	1.08	1016	1.29	2.00	2.16
FP	0.50	1020	0.80	1.20	0.60
YTG	2.35	1024	0.81	0.90	2.12
YAG crystal	2.00	1031	1.53	1.08	2.16
Lead Fluorborate	1.07 ± 0.08	1022	1.69 ± 0.15	0.81 ± 0.04	0.86
Heavy metal	0.75 ± 0.05	1012	3.40 ± 0.31	0.40 ± 0.02	0.10
Niobium Tellurite	1.10 ± 0.06	1028	1.62 ± 0.08	0.59 ± 0.06	0.65

High-intensity lasers produce ultra-intense pulses by concentrating a given amount of optical energy both temporally and spatially. The temporal limit is imposed by the time bandwidth product, $\tau_{min} = 1/\Delta\nu$ where τ_{min} is the minimum pulse duration and $\Delta\nu$ is the fluorescence bandwidth.

The theoretical limit of peak power, P_{max}, which can be generated from a 1-cm^2 area of laser material scales is given by [1]

$$P_{max} \cong \frac{h\nu_0}{\sigma_e \tau_{min}}. \quad (5)$$

The three studied glass families display the bandwidth and the saturation fluence necessary to exceed 1 PW/cm^2 if the potential for energy storage and extraction, and phase control can be met. The obtained values for the studied laser media are shown in Table 2.

4. Discussion and conclusions

Analyzing the optical parameters we conclude that the lead fluorborate and the niobium tellurite glasses doped with Yb^{3+} have more favorable spectroscopic properties than the heavy metal oxide one, which is comparable to some Yb^{3+}-doped laser glasses. This can be seen in Table 3 that shows some phosphates (QX, ADY, LY, PN, PNK), fluorophosphates (FP) and tellurite (YTG) laser glasses, reported in published papers [6,7,11]. The lead fluorborate glass has similar I_{min} as the well-known laser materials YAG and QX, fluorescence lifetime comparable to Yb:YTG (a tellurite laser glass) and emission cross-section comparable to Yb:PNK (a phosphate laser glass).

The niobium tellurite glasses can be doped with a higher ytterbium concentration, than the other ones, without concentration quenching. The high doping concentration allows for thin disk geometries for the gain medium which are advantageous for cooling and reduction of non-linear phase accumulation [1].

We have to remark, for the case of the heavy metal oxide glass having a high fluorescence effective linewidth, of 86 nm, not yet presented in the literature for ytterbium-doped glasses, and an important feature for laser action in short pulse generation under diode pumping.

In conclusion, new glasses of lead fluorborate, of heavy metal oxide and niobium tellurite doped with Yb^{3+} were reported in this paper. High absorption cross-sections (at 968 nm) were measured for all of them, mainly for niobium tellurite. The lead fluorborate glass has more favorable spectroscopic properties than the other ones; it also has very similar properties (0.81 ms for the fluorescence lifetime, emission cross section of 1.07×10^{-20} cm^2 and $I = 1.69$ kW/cm^2) when compared to other known glasses (phosphate and tellurite laser glasses) that were used as active laser media. The saturation fluence $\sim$20 J/cm^2 is smaller than the phosphate glasses (40–80 J/cm^2) but the broad bandwidth compensates this value.

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