Journal of Alloys and Compounds 509 (2011) 5084–5089

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

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

Optical properties of Yb^{3+}-doped phosphate laser glasses

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ARTICLE INFO

Article history:
Received 12 October 2010
Received in revised form 8 January 2011
Accepted 21 January 2011
Available online 2 February 2011

PACS:
70
78.20–e
42.70–a
78.30–j

Keywords:
Ytterbium ion or Yb^{3+} ion
Phosphate glass
Optical properties
Laser parameters
Energy transfer

ABSTRACT

Ytterbium-doped phosphate glasses have been prepared and studied their spectroscopic properties through absorption, emission and Fourier transform infrared (FTIR) spectral studies and time-resolved luminescence decay curves. The absorption cross-section has been found to vary with the variation of Yb_2O_3 concentration. The results of the FTIR spectra show that the OH^- content is increasing with increase of the Yb_2O_3 concentration in these glasses. The decay curves of the $^2F_{5/2}$ level of Yb^{3+} ions exhibit a single exponential nature for all the concentrations. The lifetimes of the $^2F_{5/2}$ level of Yb^{3+} ions decreases from 1.04 to 0.27 ms when the Yb_2O_3 concentration is increased from 0.1 to 6.0 mol%. The quenching of lifetimes has been found to vary directly with the inter-ionic distance between the Yb^{3+} ions. The concentration quenching of the lifetime has been analyzed using different energy transfer processes and no evidence of cooperative luminescence of Yb^{3+} ions has been found in these glasses, which reveals that the present glasses are useful for photonic device applications. The laser performance properties have also been evaluated for these glasses and compared with those of other reported Yb^{3+}-doped glass systems.

1. Introduction

Research on trivalent lanthanide (Ln^{3+})-doped transparent glasses are of great interest as these materials are used as solid state lasers, optical amplifiers, upconverters and phosphors [1,2]. The gain of these devices is limited by fast energy transfer and non-radiative de-excitation probabilities which take place inside Ln^{3+} clusters [3,4], which can be determined in glasses with the measurement of the cooperative luminescence of the Yb^{3+} ions [5,6]. In order to preserve good performances, it is necessary to limit the clustering of Ln^{3+} ions. In this regard, the selection of the chemical composition and the concentration of the optically active ions play vital role in the formation of clusters in glasses [7].

The investigation on spectroscopic properties of Yb^{3+}-doped glasses have been widely fascinated due to the special features of Yb^{3+} ions: (i) a simple energy level scheme with the $^2F_{7/2}$ ground and the $^2F_{5/2}$ excited states, for which excited state absorption and up-conversion can be ignored, (ii) the absence of cross-relaxation mechanisms that can enhance the effective laser cross-section, (iii) a higher Ln^{3+} ion solubility because of a weak concentration quenching, (iv) an excellent sensitizer for other Ln^{3+} (Er^{3+}, Tm^{3+}, Ho^{3+}, etc.) ions [8,9] through energy transfer and back-transfer efficient processes, (v) the Yb^{3+} ion is used as a local probe for evaluating the Ln^{3+} ion clustering in solids from its cooperative luminescence [10], and finally, (vi) the broad emission band of Yb^{3+} ions allows the generation of ultra-short pulses in the femtosecond range [11].

The development of optical materials for practical applications requires a systematic understanding of bonding environment of Ln^{3+} ions and their spectroscopic properties. The properties, such as the absorption and emission cross-sections, the lifetimes of excited states, and the radiative quantum efficiencies, etc., are becoming important and useful in order to understand and improve the optical device performances. In this direction, up to now, considerable work has been carried out on the spectroscopic and the laser characteristics of the Yb^{3+} ions in different glassy matrices, including

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0925-8388/$ – see front matter
doi:10.1016/j.jallcom.2011.01.148

lead silicate [12,13], lead germinate [14], heavy metal fluoride [15], zinc telluride [16], phosphate [17], fluorophosphate [18,19] and chalcogenide [20] glasses.

The selection of the chemical composition of the host for the Ln^{3+} ions is one of the more fundamental issues, since it plays a key role in the performance of practical devices. Among oxide based glasses, metaphosphate glasses are suitable hosts for Ln^{3+} ions due to their potentiality as laser hosts, good Ln^{3+} ion solubility, high transparency, low dispersion, good thermo-optical performance, low rupture strength and good chemical durability. In addition, these glasses exhibit high stimulated emission cross-sections, weak up-conversion luminescence and low probability of energy back transfer [21–24] which are crucial in design of good laser devices.

Among various phosphate glass compositions, the P_2O_5–K_2O–MO–Al_2O_3 (MO = BaO, MgO, SrO, CaO) composition is found to be an excellent host for laser media [25]. In phosphate glasses, Al_2O_3 is often used to modify the glass structure which improves chemical stability and physical properties [22]. The Al^{3+} ions not only affect thermo–mechanical properties but also laser properties [26]. The low field strength cations such as K^+ are added to improve mechanical properties which are prerequisite for a good laser glass [26]. It is desirable that the laser glass has a large emission cross-section, long fluorescence lifetime and narrow emission bandwidth. The reported data show that K_2O is perhaps the best modifier to achieve the above properties [26]. As use of K_2O as the only modifier is not practical, generally a group II modifier such as BaO/MgO/SrO/CaO is added. The design of the base composition of the present glass is based on the ranges of compositions of the two most widely used commercial metaphosphate laser glasses, LG-750 and LHG-8 [27], except that the modifier BaO has been replaced by the SrO.

The present work reports the effect of the Yb_2O_3 concentration on the spectroscopic properties of the Yb^{3+} ions in a novel metaphosphate glasses. The absorption and emission cross-sections have been calculated from the absorption spectra in order to assess their capabilities of being used as laser active medium. The decay curves for the $^2F_{5/2}$ level have been measured as a function of the Yb_2O_3 concentration in order to understand the excited state dynamics of this level. The Fourier transform infrared (FTIR) transmission spectra have been measured for evaluating the OH^- concentration and its effects. Finally, the spectroscopic and the laser performance properties have been compared with those of some reported laser glasses.

2. Experimental details

2.1. Preparation of glasses

The precursor glasses doped with Yb^{3+} ions were prepared by a high-temperature melting followed by sudden quenching method using high-purity chemicals of $Al(PO_3)_3$, KH_2PO_4, $Sr(PO_3)_2$ and Yb_2O_3. The molar compositions of these phosphate glasses, labeled as PKSAYb, are $(60 - x/2)\,P_2O_5 + 15\,K_2O + (15 - x/2)\,SrO + 10\,Al_2O_3 + x\,Yb_2O_3$, where x = 0.1, 0.5, 1.0, 2.0, 3.0, 4.0 and 6.0 mol% (see Table 1). The precursors of about 25 g were thoroughly mixed/grinded in an agate mortar, taken in a platinum crucible and kept in an electric furnace at 1075 °C for 1 h and then the melt was poured onto a preheated (350 °C) brass plate. The obtained glasses were annealed at 350 °C for 10 h to remove thermal strains. Finally, the samples were cut in a rectangular shape and polished to get smooth, transparent and uniform surface for measuring the optical properties. The density of the polished samples has been determined by Archimede's method with water as an immersion liquid [28]. The refractive indices of the samples were determined with an Abbe refractometer using monobromo naphthalene as an adhesive coating.

2.2. Measurement of spectroscopic properties

The optical absorption spectra of the polished samples were measured on a spectrophotometer (Perkin Elmer Lambda – 950) in the wavelength range of 875–1050 nm at room temperature. The emission spectra were measured by exciting glass sample with the 854 nm radiation of a Ti:sapphire pumped by a multiline 10 W Ar^+ laser (2060-10 Beamlock Spectra Physics). The emissions were focused with a convergent lens onto a 0.18 m single-grating monochromator (Jobin Yvon Triax180) and then detected with a photomultiplier tube (Hamamatsu R406). The decay curves were measured by exciting the glass samples with the 950 nm radiation of optical parametric oscillator (EKSPLA/NT342/3/UVE) by monitoring the 980 nm emission using a photomultiplier tube (Hamamatsu R406). The signal was acquired by a digital oscilloscope (LeCroy 200 MHz Oscilloscope). The FTIR spectra were recorded with a Nicolet Magna (FTIR 550) for evaluating the OH^- concentration in the studied glasses.

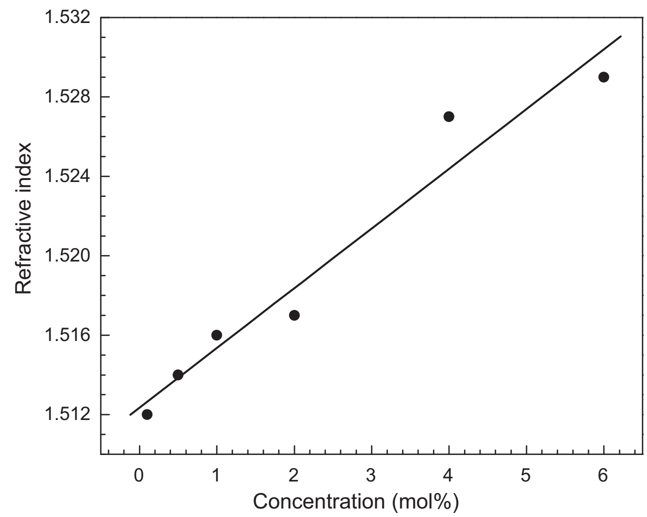

Fig. 1. The variation of refractive index for PKSAYb glasses as a function of Yb_2O_3 concentration.

3. Results and discussion

The absorption and emission spectra and decay rates of the title glasses have been analyzed by following the procedure and methodologies described elsewhere [2,14,28–40].

3.1. Spectroscopic properties: absorption and emission cross-sections

The physical properties such as density (d), concentration (N), refractive index (n) of the PKSAYb glasses are shown in Table 1 along with their glass compositions. It is observed that the density, concentration and refractive index of the present glasses are increasing with the increase of the Yb_2O_3 concentration. The variation of the refractive index of the PKSAYb glasses as a function of the Yb_2O_3 concentration is shown in Fig. 1. The refractive index of the glass mainly depends on the individual polarizabilities of the cations and their concentration and also increases with increase of cation size [29]. But in the present glass host materials, all cations are fixed and refractive index is to be mainly determined by the concentration of the Yb^{3+} ions per unit volume. Therefore, the increase in refractive index with Yb^{3+} ion concentration can be attributed to the increase of Yb^{3+} ionic polarizability besides increase of the glass density (see Table 1).

The spectroscopic properties such as absorption (σ_{ab}) and emission (σ_{em}) cross-sections, experimental (τ_{exp}) and radiative (τ_R) lifetimes of PKSAYb glasses have been evaluated and are shown in Table 2 along with those of some reported Yb^{3+}-doped systems [2,14,30–39]. The concentration dependent absorption and emission cross-sections spectra of the PKSAYb glasses are presented in Fig. 2 in the wavelength region of 900–1050 nm. The broad absorption and emission of Yb^{3+} ions are due to electronic transitions involving the Stark sublevels of the $^2F_{7/2}$ and the $^2F_{5/2}$ levels. The peak at 975 nm in absorption and emission spectra are due to $4f^{13}$-$4f^{13}$ transitions between lower Stark sublevels of the $^2F_{7/2}$ and $^2F_{5/2}$ levels. The line shapes are similar to other Yb^{3+}-doped glasses [30–39], exhibiting the same asymmetries despite differ-

Table 1
The labels, compositions and physical properties (density (*d*), concentration (*N*) and refractive index (*n*)) of Yb^{3+}-doped phosphate glasses.

Label	Glass composition	d (g/cm^3)	N (ions/cm^3)	n
PKSAYb01	59.95 P_2O_5 + 15 K_2O + 14.95 SrO + 10 Al_2O_3 + 0.1 Yb_2O_3	2.54	0.23	1.512
PKSAYb05	59.75 P_2O_5 + 15 K_2O + 14.75 SrO + 10 Al_2O_3 + 0.5 Yb_2O_3	2.67	1.22	1.514
PKSAYb10	59.5 P_2O_5 + 15 K_2O + 14.5 SrO + 10 Al_2O_3 + 1.0 Yb_2O_3	2.74	2.48	1.516
PKSAYb20	59 P_2O_5 + 15 K_2O + 14 SrO + 10 Al_2O_3 + 2.0 Yb_2O_3	2.69	4.77	1.517
PKSAYb40	58 P_2O_5 + 15 K_2O + 13 SrO + 10 Al_2O_3 + 4.0 Yb_2O_3	2.90	9.90	1.527
PKSAYb60	57 P_2O_5 + 15 K_2O + 12 SrO + 10 Al_2O_3 + 6.0 Yb_2O_3	2.85	14.06	1.529

Table 2
Spectroscopic and laser performance parameters of Yb^{3+}: glasses.

Glass	σ_{ab} (10^{-20} cm^2)	σ_{em} (10^{-20} cm^2)	τ_R (ms)	τ_{exp} (ms)	β_{min}	I_{sat} (kW/cm^2)	I_{min} (kW/cm^2)	$\sigma_{em} \times \tau_{exp}$ (10^{-20} cm^{2}ms)	SFL
PKSAYb01	1.57	2.11	1.03	1.04	0.151	12.99	1.96	2.19	1.117
PKSAYb05	2.19	2.92	1.07	0.80	0.171	11.73	2.00	2.34	1.170
PKSAYb10	1.85	2.47	1.10	0.74	0.157	16.40	2.58	1.83	0.709
PKSAYb20	1.46	1.91	1.14	0.38	0.161	36.94	5.97	0.73	0.122
PKSAYb40	0.98	1.30	1.33	0.28	0.150	84.03	12.68	0.36	0.028
PKSAYb60	1.29	1.74	1.26	0.27	0.147	67.66	9.95	0.47	0.047
PN-20 [2]	–	1.36	–	1.09	0.058	8.89	0.51	1.48	2.902
GP [14]	1.20	0.60	–	0.80	–	–	2.80	0.48	0.171
30PT1Yb [30]	1.26	1.70	1.19	1.26	–	–	1.79	2.14	1.200
30PT3Yb [30]	1.16	1.55	2.11	1.15	–	–	2.14	1.78	0.832
30PT5Yb [30]	1.12	1.50	1.28	0.94	–	–	2.70	1.41	0.522
QX-Kigre [31]	0.50	0.70	–	2.00	0.171	10.79	1.82	1.40	0.769
PNK [32]	–	1.08	–	2.00	–	–	1.29	2.16	1.670
YTG [33]	–	2.35	–	0.90	–	–	0.81	2.12	2.620
GPB1 [34]	1.50	0.80	–	0.54	–	–	2.00	0.43	0.215
PFB [35]	2.56	1.07	–	0.81	–	–	–	0.87	–
HMO [35]	2.20	0.75	–	0.40	–	–	–	0.30	–
Niobium tellurites [35]	4.09	1.10	–	0.59	–	–	–	0.65	–
BSBMYb1 [36]	1.77	1.39	0.71	0.66	0.210	17.30	3.70	0.92	0.249
BSBMYb2 [36]	1.46	0.97	1.05	0.50	0.210	27.00	6.00	0.49	0.082
BSBMYb3 [36]	1.39	0.72	1.20	0.41	0.210	31.00	6.90	0.29	0.042
TWZ4 [37]	–	1.32	–	0.93	–	–	0.92	1.23	1.330
SPb [38]	–	0.49	–	2.00	0.093	16.85	1.56	0.98	0.628
SAl [38]	–	0.32	–	1.42	0.165	22.78	3.76	0.45	0.120
LSY-8 [39]	–	0.56	–	1.04	0.078	22.23	1.75	0.58	0.331

ences in widths and peak positions in individual PKSAYb glasses. The systematic incorporation of Yb^{3+} ions by the vitreous matrix is confirmed by linear increase of the absorption coefficient with Yb_2O_3 concentration as shown in the inset of Fig. 2.

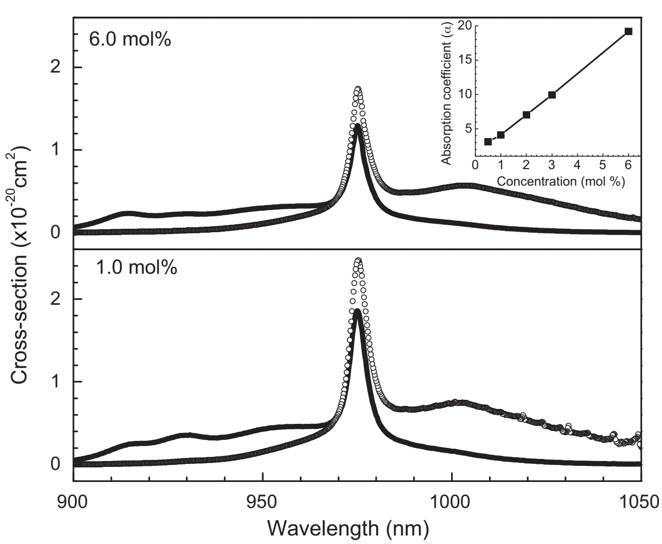

Fig. 2. Absorption (solid line) and emission (open line) cross-sections spectra for the PKSAYb doped glasses for 1.0 and 6.0 mol% Yb_2O_3, the inset shows variation of absorption co-efficient with Yb_2O_3 concentration in PKSAYb glasses.

The emission cross-sections have been calculated by the reciprocity method [40] and are shown in Table 2 and the corresponding Yb_2O_3 concentration dependent spectra of the PKSAYb glasses are shown in Fig. 2. It is well known that the site symmetry and the bonding characteristics of Yb^{3+} ions are strongly affected by the bond strength, polarizability and structure between the network former and oxygen ions. It is observed that the emission cross-section changes from 2.92 to 1.30×10^{-20} cm^2 when the Yb_2O_3 concentration increased from 0.1 to 6.0 mol%. As seen from Table 2, there is no systematic variation in the absorption and emission cross-sections when Yb_2O_3 concentration is increased from 0.1 to 6 mol%. It is found that the PKSAYb05 glass possesses the highest absorption and emission cross-section of around 2.19 and 2.92×10^{-20} cm^2, respectively. Although, the values of absorption and emission cross-sections are obviously different, the line shapes of the emission spectra are sufficiently similar for all the glasses so that the emission peak wavelengths are situated at around 975 nm. This behavior shows that the Yb^{3+} ions are most likely to be occupied at the same sites into which the network modifiers are generally incorporated.

The results obtained in the present study are compared with Yb^{3+}-doped PN-20 [2], lead germanate (GP) [14], tellorophosphate [30], phosphate (QX-Kigre) [31], phosphate (PNK) [32], tellurite (YTG) [33], lead bismuth germinate (GPB1) [34], lead fluoroborate (PFB) [35], heavy metal oxides (HMO) [35], niobium tellurites [35], fluorophosphate: (BSBMYb) [36], zinc tungsten tellurite (TWZ4) [37], lead silicate (SPb) [38], alumino silicate (SAl) [38] and LSY-8 [39] glasses. It is observed from Table 2 that PKSAYb05 glass has relatively higher emission cross-section compared with reported

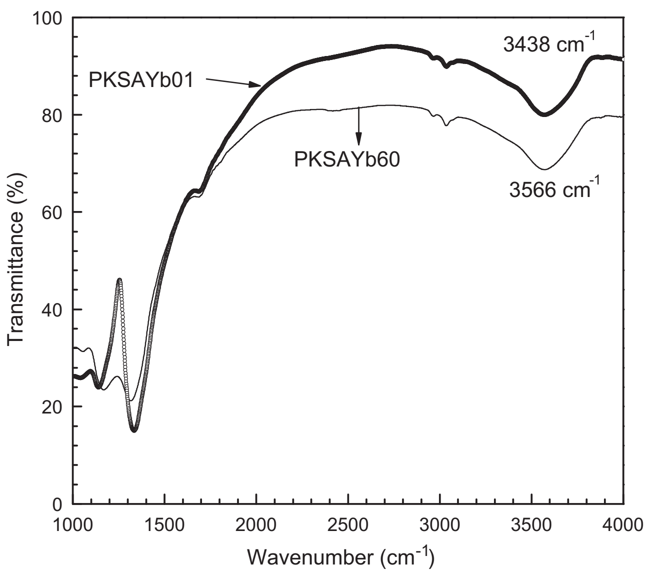

Fig. 3. FTIR transmittance spectra of PKSAYb01 and PKSAYb60 glasses.

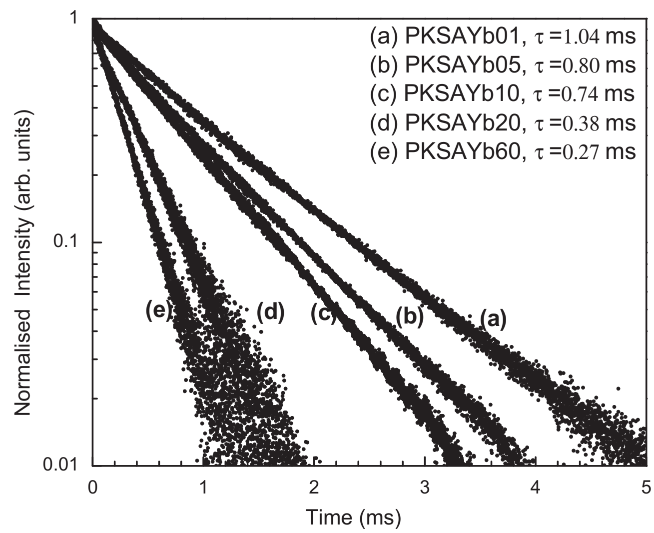

Fig. 4. Normalized decay curves for 0.1, 0.5, 1.0, 2.0, and 6.0 mol% Yb_2O_3 doped PKSAYb glasses.

Yb^{3+}-doped glasses indicating that these glasses can be used for laser applications.

As can be seen in Table 2, there is no considerable variation in radiative lifetimes (τ_R) or transition probabilities of the $^2F_{5/2}$ level of Yb^{3+} ions in PKSAYb glasses with the increase of the Yb_2O_3 concentration. The samples with less covalent character and lower refractive index show the higher values for the radiative lifetime or the lower values for the spontaneous transition probability [39,41]. The concentration dependent luminescence spectra of PKSAYb glasses have been measured and their integrated intensities evaluated as a function of the active ion concentration. It is found that integrated intensity of the $^2F_{5/2} \rightarrow {}^2F_{7/2}$ band increases with the increase of the Yb_2O_3 concentration up to 4.0 mol% and then decreased. Similar behavior was observed by Jiang et al. [32], in Yb^{3+}-doped phosphate glass. The increase of intensity up to 4.0 mol% Yb_2O_3 is due to absence of Ln^{3+} ion clusters in present glass systems and the reduction of intensity after 4.0 mol% Yb_2O_3 is due to energy migration (exchange) between Yb^{3+} ions. The observed broad emission line shape is due to the inhomogeneous broadening that characterizes a glassy host. It is known that the stimulated emission cross-section (σ_{em}) has strong relation with the local environment of the Yb^{3+} site characterized by both the Yb^{3+} site symmetry and its bonding characteristics [18,20,41]. From the laser application point of view, it is good to have higher emission cross-sections, greater gain and longer emission lifetime in order to permit high population inversion density and higher absorption cross-sections at pump wavelength.

The cooperative luminescence of Yb^{3+} ions occurs when there is a simultaneous de-excitation of two excited Yb^{3+} ions from the $^2F_{5/2}$ level resulting in the emission of one visible photon of wavelength around 500 nm. It is worth noting that the cooperative luminescence has not been noticed for all concentrations of Yb_2O_3 -doped PKSAYb glasses. The absence of cooperative luminescence clearly shows that there is no Yb^{3+} ion clustering in the present glasses indicating that these glasses are useful for photonic or laser device applications.

3.2. Fourier transform infrared transmittance spectra

The concentration dependent FTIR spectra have been measured in order to know the influence of Yb^{3+} ion concentration on the OH^- content in PKSAYb glasses (see Fig. 3). It is known that the OH^- groups have a broad and intense band at about 3000–3600 cm^{-1} in phosphate glasses. To achieve optimum laser performance, OH^- content should be minimum in the glass. As can be seen from the FTIR spectra, there is one broad absorption band at around 3440 and 3570 cm^{-1} for the PKSAYb01 and the PKSAYb60 glasses, respectively. This band is ascribed to the stretching of the OH^- vibrations [42]. The previous study in sodium silicate glasses [43] showed that the OH^- absorption bands in oxide glasses can be classified into three groups, the free OH^- groups at 3500 cm^{-1}, the strongly hydrogen-bonded OH^- groups at 2650 cm^{-1} and the very strongly hydrogen-bonded OH^- groups at 2300 cm^{-1}. Hence, it is clear that only the free OH^- groups are dominant in Yb^{3+}-doped PKSAYb glasses.

The free OH^- group present in the glasses is associated in the network of the glass former through hydrogen bonding [42]. The free OH^- content and N_{OH} (ions/cm^3) in the Yb^{3+}-doped PKSAYb glasses has been estimated from the measured FTIR spectra [44,45] and is found to be around 9.9×10^{21} ions/cm^3 for the PKSAYb01 and around 12.8×10^{21} ions/cm^3 for the PKSAYb60 glasses, respectively. It is interesting to note that the concentration of OH^- impurities slightly increases with increase in Yb_2O_3 concentration indicating that there exists direct relation between them. This implies that OH^- ions could play a vital role in decreasing the lifetime of the $^2F_{5/2}$ excited state of Yb^{3+} ions in the present glasses.

3.3. Decay curve analysis: concentration dependence of lifetimes

Long fluorescence lifetimes permit high population inversion, which is an important feature for laser action operation. In order to optimize the Yb_2O_3 concentration in these glasses, the decay curves have been measured as a function of Yb_2O_3 concentration. The normalized decay curves for the $^2F_{5/2} \rightarrow {}^2F_{7/2}$ transition of the Yb^{3+} ions in PKSAYb glasses for different Yb_2O_3 concentrations are shown in Fig. 4. The decay curves are found to be single exponential for all the concentrations of Yb^{3+} ions. The experimental lifetime (τ_{exp}) of the $^2F_{5/2}$ level of the Yb^{3+} ions has been evaluated by single exponential fitting (see Fig. 5) and are found to decrease with the increase in Yb_2O_3 concentration (see Table 2). The τ_{exp} of the $^2F_{5/2}$ level is given by [1]

$$\frac{1}{\tau_{exp}} = \left(\frac{1}{\tau_R} + W_{MPR}\right) + W_{ET} = \frac{1}{\tau_0} + W_{ET} \quad (1)$$

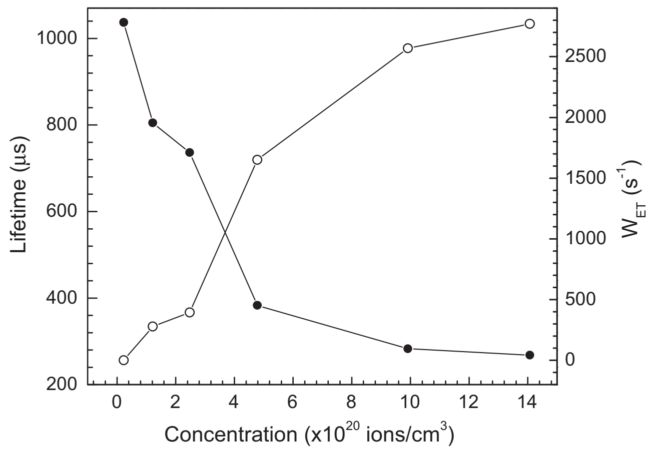

Fig. 5. Variation of experimental lifetime of the $^2F_{5/2}$ level and the energy transfer probability as a function of Yb^{3+} ions concentration (ions/cm^3) in PKSAYb glasses.

where τ_R is the radiative lifetime, W_{MPR} is the multiphonon relaxation probability, τ_0 is intrinsic lifetime and W_{ET} is the energy transfer probability between the Yb^{3+} ions and/or between the Yb^{3+} ions and the OH^- groups.

Since the τ_0 of the Yb^{3+} donor ions in the absence of acceptors is around 1037 μs, then the decay of the luminescence from any excited level can be described by a single exponential, i.e., $\tau_{exp} = \tau_R$. The multiphonon relaxation probability W_{MPR} can be neglected as it needs 9 phonons to bridge the large energy gap of around 10770 cm^{-1} between the $^2F_{5/2}$ and the $^2F_{7/2}$ levels [46] in phosphate glasses [47]. As there is no intermediate level for the Yb^{3+} ion, the energy transfer between Yb^{3+} ions due to cross-relaxation process do not exist. The Yb^{3+} ion concentration and/or the free OH^- group content are large enough to cause the energy transfer between excited Yb^{3+} ions and free OH^- groups in PKSAYb glasses leading to significant quenching of the $^2F_{5/2}$ level lifetime. Therefore, the OH^- content also influences the Yb^{3+} ions lifetime in the glasses under study. The rate of the energy transfer is increased with increase of the Yb^{3+} ion concentration (see Fig. 5). It is observed that the τ_{exp} of the $^2F_{5/2}$ level of the Yb^{3+} ions shortens from 1.04 to 0.27 ms in PKSAYb glasses when the Yb_2O_3 concentration increases from 0.1 to 6.0 mol%. This quenching of lifetime is clearly due to energy migration between Yb^{3+} ions and/or energy transfer between Yb^{3+} ions and directly coupled OH^- in PKSAYb glasses.

The concentration quenching of the lifetime of the $^2F_{5/2}$ level of the Yb^{3+} ions in PKSAYb glasses has been analyzed by the theory developed by Auzel et al. [48,49], in order to determine whether it is due to a diffusion limited or a fast diffusion regime. In the first case, the order of magnitude for the transfer probability between Yb^{3+} ions and Yb^{3+} ions to host (nonradiative sink) is the same. It generally applies to strong self-quenching materials [48]. The second case, where the quenching step is less probable than the diffusion, usually applies to weak self-quenching materials [49].

According to Auzel's theory, for the diffusion limited case, and assuming an electric dipole-dipole interaction, the self-quenching behavior can be described by [48,49]

$$\tau(N) = \frac{\tau_w}{1 + ((9/2\pi)(N/N_0)^2)} \tag{2}$$

where N is the ion doping concentration, τ_w is the measured lifetime at weak Ln^{3+} ion concentration and N_0 is the critical sensitizer concentration for self-quenching.

On the other hand, self-quenching by fast diffusion is described by [49],

$$\tau(N) = \frac{\tau_w}{1 + 1.45(N/N_{0ss})\exp -(\beta\Delta E/4)} \tag{3}$$

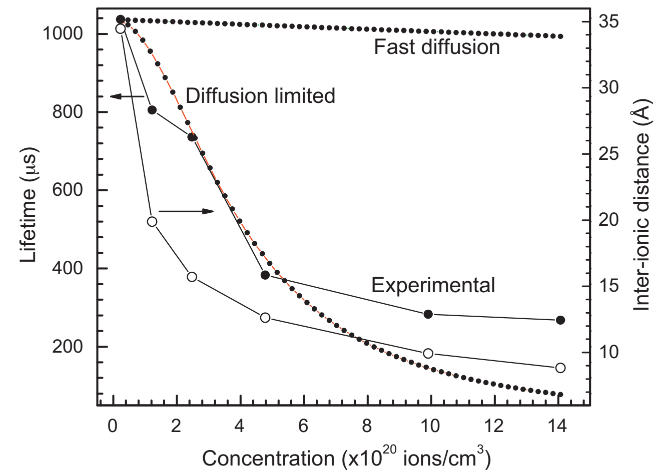

Fig. 6. Quenching of the $^2F_{5/2}$ lifetime (solid circles) and variation of inter-ionic distance (open circles) with Yb^{3+} ions concentration (ions/cm^3) in PKSAYb glasses. The dotted curves are simulations using Eqs. (2) and (3) for diffusion limited (lower) and fast diffusion (upper), respectively, for experimental data (solid circles).

where N_{0ss} is the critical concentration for the diffusion step between active ions, β is the exponential parameter for multiphonon-assisted energy transfer in the considered host and ΔE is the energy of the first excited state of the considered ion.

In the present analysis, N_{0ss} has been taken to be equal to N_0 (4.83×10^{20} ions/cm^3), calculated from Eq. (2) for 1.0 mol% of Yb_2O_3–doped glass by taking $\tau_w = 1037$ μs, as the measured lifetime for the 0.1 mol% Yb_2O_3-doped PKSAYb glass. The β value has been calculated to be 1.7×10^{-3} cm from the equations given in Ref. [50] by taking phonon energy ($\hbar\omega$) of the PKSAYb glass host as 1180 cm^{-1} and an electron–phonon coupling constant (g) as 0.038 found for a similar composition of $58.5P_2O_5$–$17K_2O$–$14.5BaO$–$9Al_2O_3$–$1Eu_2O_3$ glass [51].

Eqs. (2) and (3) have been used to simulate the curves showing the variation of the lifetime of the $^2F_{5/2}$ level for different Yb^{3+} ion concentrations in PKSAYb glasses and the results are shown in Fig. 6. It is clear that the concentration quenching is a purely diffusion limited process at lower concentrations (up to 2.0 mol%), although it deviates towards fast diffusion curve at higher Yb^{3+} ion concentrations. This result would indicate a gradual decrease of diffusion limited process and turns to fast diffusion process at higher Yb^{3+} ion concentrations in the present PKSAYb glasses.

The mean Yb^{3+}-Yb^{3+} inter-ionic distances have been calculated in the present glasses [52] for different Yb^{3+} ion concentrations and are plotted in Fig. 6. As can be seen, it is interesting to note that up to an inter-ionic distance of 12.6 Å, which corresponds to a concentration of 2.0 mol% of Yb^{3+} ions, the lifetime quenching is purely diffusion limited process. After this point, quenching by fast diffusion also becomes important and with further decrease in inter-ionic distance, there is an increase in the fast diffusion component. From the above analysis, it is worth noting that experimental lifetime of the $^2F_{5/2}$ level is directly proportional to the inter-ionic distance between the Yb^{3+} ions and the main channel of lifetime quenching is by energy transfer to the nearby Yb^{3+} ions.

3.4. Laser performance parameters

To evaluate the potentiality of the Yb^{3+}-doped phosphate glasses for laser applications, the laser performance parameters β_{min} (minimum fraction of Yb^{3+} ions that must be excited to balance the gain exactly with the ground state absorption at a laser wavelength), I_{sat} (pump saturation intensity) and I_{min} (minimum absorbed pumping intensity) [53] have been determined and are given in Table 2 along with those of reported Yb^{3+}: glasses [2,31–34,36–39]. As can be seen, their values are in the range of 0.058–0.210 for β_{min}, 8.89–84.03 kW/cm^2 for I_{sat} and 0.51–12.68 kW/cm^2 for I_{min}. These values are comparable with those of the QX-Kiger [31], the PNK [32] and the LSY-8 [39] glasses.

The values of β_{min} do not change very much with the increase of the Yb_2O_3 concentration in the PKSAYb glasses. On the other hand, it is desirable to have an I_{sat} as low as possible to minimize the minimum pumping intensity I_{min} since the emission cross-section is proportional to absorption cross-section [40]. For good laser glasses, figure of merit given by $\sigma_{em} \times \tau_{exp}$ and the energy extraction efficiency are generally desirable to be as large as possible to provide high gain and also I_{min} has to be as small as possible to minimize the pump losses. From Table 2, it is also noticed that PKSAYb05 glass possesses higher value of figure of merit of around 2.3×10^{-20} cm^2 ms when compared to those of reported glasses. Therefore, systematical factor (SFL) [53] of laser properties is very useful to evaluate both pump loses and figure of merit. Laser oscillations are very easy to gain if the SFL is large enough. It is observed from Table 2 that PKSAYb05 glass possess high SFL value of the order of 1.17 compared to those of the commercial QX-Kigre laser glass [31] and some other reported glasses. Further, among the Yb^{3+}:glass systems shown in Table 2, PN-20 [2] glass is having the highest value (2.902) of SFL and the glass systems: 30PT1Yb [30], PNK [32], YTG [33] and TWZ4 [37] are having higher values of SFL compared to the present glass systems. Therefore, higher values of the product of emission cross-section and fluorescence lifetime and/or lower values of I_{min} are desirable for the gain media to be compatible with InGaAs or AlGaAs diode pump sources and to have higher laser efficiency.

4. Conclusions

Absorption and emission spectra and the luminescence decay measurements have been performed for Yb^{3+}-doped phosphate glasses as a function of the Yb_2O_3 concentrations. It is found that PKSAYb05 glasses possess high absorption and emission cross-sections and higher figure of merit. The experimental lifetime of the $^2F_{5/2}$ state shortened from 1.04 to 0.27 ms in PKSAYb glasses when Yb_2O_3 concentration is increased from 0.1 to 6.0 mol% due to energy transfer between Yb^{3+} ions and OH^- ions. Analysis of the concentration quenching of the lifetimes using the Auzel's theory indicates that concentration quenching is purely diffusion limited for lower Yb^{3+} ion concentrations and then gradually deviates to fast diffusion for higher Yb^{3+} ion concentrations. Experimental lifetime of the $^2F_{5/2}$ level is found to be directly proportional to the inter-ionic distance between the Yb^{3+} ions. Out of all the Yb^{3+}-doped glasses studied, the PKSAYb05 glass possess good spectroscopic and laser performance parameters indicating that this glass could be considered for laser applications. The absence of cooperative luminescence also reveals that the title glasses are useful for photonic device applications.

Acknowledgements

Prof. Jayasankar and Dr. Dhareshwar are grateful to the DAE-BRNS, Govt. of India, for the award of the Major Research Project (No. 2007/34/25-BRNS/2415, dt. 18-01-2008) and Dr. Venkatramu, Dr. Babu and Dr. Jayasankar are grateful to Departamento de Física Fundamental y Experimental, Universidad de La Laguna, Spain for their stay as a Guest Scientists. This work has been partly supported by Ministerio de Ciencia e Innovación (MAT2007-65990-C03-02, MALTA CONSOLIDER CSD2007-00045 and International Complementary Action with India PCI2006-A7-0638) and by the EU-FEDER funds.

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