Spectral and Luminescence Study of Er³⁺ Doped Phosphate Glasses for the Development of 1.5 µm Broadband Amplifier

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ABSTRACT

Ytterbium zinc lithium lead alumino antimony borophosphate glasses containing Er^{3+} in $(25-x)P_2O_5$: 10ZnO: 10Li₂O: 10PbO: 10Al₂O₃: 10Sb₂O₃: 10Y₂O₃: 15B₂O₃: Er₂O₃ (where x=1, 1.5,2 mol %) have been prepared by melt-quenching method. The amorphous nature of the glasses was confirmed by x-ray diffraction studies. Optical absorption, excitation Spectrum and fluorescence spectra were recorded at room temperature for all glass samples. Judd-Ofelt intensity parameters Ω_{λ} ($\lambda=2$, 4, 6) are evaluated from the intensities of various absorption bands of optical absorption spectra. Using these intensity parameters various radiative properties like spontaneous emission probability, branching ratio, radiative life time and stimulated emission cross– section of various emission lines have been evaluated.

Keywords: YZLLAABP Glasses, Optical Properties, Judd-Ofelt Theory, Rare earth ions.

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I. Introduction

Rare earth doped ceramic glasses are widely used mainly for solid state lasers, white light-emitting diodes (WLEDs), up-conversion lasers, optical amplifiers, optical communications and electro-chromic display devices [1-4].Phosphate glasses have attracted a great attention due to their several properties, such as good thermal and mechanical stabilities, low refractive index, low melting temperature, high transparency, high refractive index and high density [5-10]. Phosphate based glasses have a wide range of potential applications in optoelectronic devices, transmission, solid state lighting and laser technologies [11-13]. Glasses containing heavy metal oxides exhibits good non-linear optical properties and good chemical durability [14,15]. Boric oxide (B_2O_3) acts as a good glass former and flux material [16]. PbO is heavy metal oxide because it improves the chemical durability, thermal stability and decrease the melting temperature. Lead oxide acts as both modifier and former [17]. The low glass melting temperature .low phonon energy, good chemical durability and the optical fiber development compatibility makes the phosphate glasses suitable candidates for photonic applications [18,19]. Among different rare-earth ions, the Er^{3+} ions have been identified as the most efficient ion for obtaining the lasing action, frequency up-conversion and optical amplification [20-24].

In this work, the spectroscopic properties of Er^{3+} -doped (25-x)P₂O₅: 10ZnO: 10Li₂O: 10PbO: 10Al₂O₃: 10Sb₂O₃: 10Y₂O₃: 15B₂O₃:xEr₂O₃ (where x=1, 1.5,2 mol %) glasses were investigated for operation at the 1.55 µm wavelength. The optical absorption, excitation and fluorescence spectra of Er^{3+} of the glasses were investigated. The intensities of the transitions for the rare earth ions have been estimated successfully using the Judd-Ofelt theory, The laser parameters such as radiative probabilities(A),branching ratio (β), radiative life time(τ_R) and stimulated emission cross section(σ_p) are evaluated using J.O.intensity parameters(Ω_{λ} , λ =2,4 and 6).

Preparation of glasses

II. Experimental Techniques

The following Er^{3+} doped ytterbium zinc lithium lead alumino antimony borophosphate glass samples (25-x)P₂O₅: 10ZnO: 10Li₂O: 10PbO: 10Al₂O₃: 10Sb₂O₃: 10Y₂O₃: 15B₂O₃: xEr₂O₃ (where x=1, 1.5.2) have been prepared by melt-quenching method. Analytical reagent grade chemical used in the present study consist of P₂O₅,ZnO,Li₂O,PbO,Al₂O₃,Sb₂O₃,Y₂O₃, B₂O₃and Er₂O₃. All weighed chemicals were powdered by using an Agate pestle mortar and mixed thoroughly before each batch (10g) was melted in alumina crucibles in silicon carbide based an electrical furnace.

Silicon Carbide Muffle furnace was heated to working temperature of 1060°C, for preparation of Ytterbium zinc lithium lead alumino antimony borophosphate glasses, for two hours to ensure the melt to be

free from gases. The melt was stirred several times to ensure homogeneity. For quenching, the melt was quickly poured on the steel plate & was immediately inserted in the muffle furnace for annealing. The steel plate was preheated to 100° C. While pouring; the temperature of crucible was also maintained to prevent crystallization. And annealed at temperature of 350° C for 2h to remove thermal strains and stresses. Every time fine powder of cerium oxide was used for polishing the samples. The glass samples so prepared were of good optical quality and were transparent. The chemical compositions of the glasses with the name of samples are summarized in Table 1

Table 1 Chemical composition of the glasses

Sample	Glass composition (mol %)
YZLLAABP (UD)	25P ₂ O ₅ : 10ZnO: 10Li ₂ O: 10PbO: 10Al ₂ O ₃ : 10Sb ₂ O ₃ : 10Y ₂ O ₃ : 15B ₂ O ₃
YZLLAABP (ER 1)	24P ₂ O ₅ : 10ZnO: 10Li ₂ O: 10PbO: 10Al ₂ O ₃ : 10Sb ₂ O ₃ : 10Y ₂ O ₃ : 15B ₂ O ₃ : 1 Er ₂ O ₃
YZLLAABP (ER 1.5)	23.5P ₂ O ₅ : 10ZnO: 10Li ₂ O: 10PbO: 10Al ₂ O ₃ : 10Sb ₂ O ₃ : 10Y ₂ O ₃ : 15B ₂ O ₃ 1.5 Er ₂ O ₃
YZLLAABP (ER 2)	23P ₂ O ₅ : 10ZnO: 10Li ₂ O: 10PbO: 10Al ₂ O ₃ : 10Sb ₂ O ₃ : 10Y ₂ O ₃ : 15B ₂ O ₃ : 2 Er ₂ O ₃

YZLLAABP (UD)—Represents undoped Ytterbium zinc lithium lead alumino antimony borophosphate glass specimens

YZLLAABP (ER) -Represents Er³⁺ doped Ytterbium zinc lithium lead alumino antimony borophosphate glass specimens

III. THEORY

3.1 Oscillator Strength

The intensity of spectral lines are expressed in terms of oscillator strengths using the relation [25]. $f_{\text{expt.}} = 4.318 \times 10^{-9} [\epsilon (v) \text{ d } v$ (1)

where, $\varepsilon(v)$ is molar absorption coefficient at a given energy v (cm⁻¹), to be evaluated from Beer–Lambert law. Under Gaussian Approximation, using Beer–Lambert law, the observed oscillator strengths of the absorption bands have been experimentally calculated, using the modified relation [26].

$$P_{\rm m}=4.6\times10^{-9}\times\frac{1}{cl}\log\frac{I_0}{I}\times\Delta\upsilon_{1/2}$$
(2)

where c is the molar concentration of the absorbing ion per unit volume, I is the optical path length, $logI_0/I$ is absorbtivity or optical density and $\Delta v_{1/2}$ is half band width.

3.2. Judd-Ofelt Intensity Parameters

According to Judd [27] and Ofelt [28] theory, independently derived expression for the oscillator strength of the induced forced electric dipole transitions between an initial J manifold $|4f^N(S, L) J\rangle$ level and the terminal J' manifold $|4f^N(S'L') J'\rangle$ is given by:

$$\frac{8\Pi^2 m c \bar{\upsilon}}{3h(2J+1)} \frac{1}{n} \left[\frac{\left(n^2+2\right)^2}{9} \right] \times S(J, J^{-})$$
(3)

where, the line strength S(J, J') is given by the equation

 $S (J, J') = e^{2} \sum_{\lambda < 4} \Omega_{\lambda} < 4f^{N}(S, L) J \| U^{(\lambda)} \| 4f^{N}(S', L') J' > 2 \qquad (4)$ $\lambda = 2, 4, 6$

In the above equation m is the mass of an electron, c is the velocity of light, v is the wave number of the transition, h is Planck's constant, n is the refractive index, J and J' are the total angular momentum of the initial and final level respectively, Ω_{λ} ($\lambda = 2, 4$ and 6) are known as Judd-Ofelt intensity parameters. **3.3. Radiative Properties**

The Ω_{λ} parameters obtained using the absorption spectral results have been used to predict radiative properties such as spontaneous emission probability (A) and radiative life time (τ_R), and laser parameters like fluorescence branching ratio (β_R) and stimulated emission cross section (σ_p).

The spontaneous emission probability from initial manifold $|4f^N(S', L') J\rangle$ to a final manifold $|4f^N(S, L) J\rangle$ is given by:

$$A[(S', L') J'; (S, L) J] = \frac{64 \pi^2 \nu^3}{3h(2j'+1)} \left| \frac{n(n^2+2)^2}{9} \right| \times S(J', \bar{J})$$
(5)
Where, S (J', J) = e² [\Omega_2 || U⁽²⁾ ||² + \Omega_4 || U⁽⁴⁾ ||² + \Omega_6 || U⁽⁶⁾ ||²]

The fluorescence branching ratio for the transitions originating from a specific initial manifold $|4f^{N}(S', L') J^{>}$ to a final many fold $|4f^{N}(S, L) J^{>}$ is given by

$$\beta [(S', L') J'; (S, L) J] = \sum_{k=1}^{\infty} \frac{A[(S' L)]}{A[(S' L) J'(S L)]}$$
(6)
S L J

where, the sum is over all terminal manifolds.

The radiative life time is given by

$$\tau_{rad} = \sum A[(S', L') J'; (S,L)] = A_{Total}^{-1}$$
(7)
$$S L J$$

where, the sum is over all possible terminal manifolds. The stimulated emission cross -section for a transition from an initial manifold $|4f^{N}(S', L') J^{2}\rangle$ to a final manifold $|4f^{N}(S, L) J^{2}\rangle$ is expressed as

$$\sigma_{\rm p}(\lambda_{\rm p}) = \left[\frac{\lambda_{\rm p}^4}{_{8\pi{\rm cn}^2\Delta\lambda_{\rm eff}}}\right] \times A[(S',L') J';(\bar{S},\bar{L})\bar{J}]$$
(8)

where, λ_p the peak fluorescence wavelength of the emission band and $\Delta \lambda_{eff}$ is the effective fluorescence line width.

4.1. XRD Measurement

IV. Result and Discussion

Figure 1 presents the XRD pattern of the samples containing show no sharp Bragg's peak, but only a broad diffuse hump around low angle region. This is the clear indication of amorphous nature with in the resolution limit of XRD instrument.



4.2. Absorption spectra

The absorption spectra of YZLLAABP (ER) glasses, consists of absorption bands corresponding to the absorptions from the ground state ${}^{4}I_{15/2}$ of Er³⁺ ions. Nine absorption bands have been observed from the ground state ${}^{4}I_{15/2}$ to excited states ${}^{4}I_{11/2}$, ${}^{4}F_{9/2}$, ${}^{4}S_{3/2}$, ${}^{2}H_{11/2}$, ${}^{4}F_{7/2}$, ${}^{4}F_{3/2}$, ${}^{2}H_{9/2}$ and ${}^{4}G_{11/2}$ for Er³⁺ doped YZLLAABP (ER) glasses.



Fig.2: Vis-NIR absorption spectra of YZLLAABP ER(01) glass.

The experimental and calculated oscillator strengths for Er^{3+} ions in ytterbium zinc lithium lead alumino antimony borophosphate glasses are given in **Table 2**

			8			9
Energy level	Glass YZLLAABP		Glass YZLLAABP		Glass YZLLAABP	
${}^{4}I_{15/2}$	(ER01)		(ER1.5)		(ER02)	
	Pexp.	P _{cal.}	Pexp.	P _{cal.}	Pexp.	P _{cal.}
${}^{4}I_{11/2}$	0.86	0.67	0.84	0.67	082	0.67
${}^{4}I_{9/2}$	0.43	0.14	0.40	0.14	0.38	0.14
${}^{4}F_{9/2}$	2.46	1.45	2.43	1.44	0.33	1.43
${}^{4}S_{3/2}$	038	0.60	0.36	0.61	0.31	0.61
${}^{2}\mathrm{H}_{11/2}$	6.50	2.38	6.47	2.39	6.44	2.39
${}^{4}F_{7/2}$	5.28	2.10	5.25	2.11	5.22	2.10
${}^{4}F_{5/2}$	0.68	0.77	0.66	0.77	0.64	0.77
${}^{4}F_{3/2}$	0.38	0.47	0.36	0.47	0.33	0.47
${}^{2}H_{9/2}$	1.72	0.90	1.69	0.90	1.65	0.90
${}^{4}G_{11/2}$	486	6.77	4.83	6.77	4.79	6.77
R.m.s.deviation	1.8044		1.7916		1.7813	

Table 2. Measured and calculated oscillator strength (1 \times 10) of Er 1005 in 1 EEEAAD1 glasses

Computed values of Slater-Condon, Lande, Racah, nephelauexetic ratio and bonding parameter for Er^{3+} doped YZLLAABP glass specimens are given in **Table 3**.

Table3. Computed values of Slater-Condon, Lande', Racah, nephelauexetic ratio and bonding parameter
for Er ³⁺ doped YZLLAABP glass specimens.

	1		
Free ion	YZLLAABP ER01	YZLLAABP	YZLLAABP
		ER1.5	ER02
441.680	433.93	433.99	433.98
68.327	67.049	67.0452	67.047
7.490	7.046	7.049	4.048
2369.400	2414.625	2414.523	2414.537
6855.300	6663.203	6664.296	6664.117
32.126	31.345	31.356	31.353
645.570	643.606	643.602	643.596
0.15470	0.15451	0.15448	0.15449
0.01696	0.01624	0.016242	0.016241
10.61899	10.35292	10.35469	10.35451
0.049764	0.048702	0.0487189	0.0487156
	0.99580	0.99598	0.995954
	0.04583	0.04483	0.04498
	Free ion 441.680 68.327 7.490 2369.400 6855.300 32.126 645.570 0.15470 0.01696 10.61899 0.049764	Free ion YZLLAABP ER01 441.680 433.93 68.327 67.049 7.490 7.046 2369.400 2414.625 6855.300 6663.203 32.126 31.345 645.570 643.606 0.15470 0.15451 0.01696 0.01624 10.61899 10.35292 0.049764 0.048702 0.99580 0.04583	Free ion YZLLAABP ER01 YZLLAABP ER1.5 441.680 433.93 433.99 68.327 67.049 67.0452 7.490 7.046 7.049 2369.400 2414.625 2414.523 6855.300 6663.203 6664.296 32.126 31.345 31.356 645.570 643.606 643.602 0.15470 0.15451 0.15448 0.01696 0.01624 0.016242 10.61899 10.35292 10.35469 0.049764 0.048702 0.0487189 0.99580 0.99598 0.04483

The values of Judd-Ofelt intensity parameters are given in Table 4.

Table 4. Judd-O	felt intensity p	arameters for	· Er ³⁺ doped	YZLLAABP glas	ss specimer	IS

Glass Specimen	$\Omega_2(pm^2)$	$\Omega_4(pm^2)$	$\Omega_6(pm^2)$	Ω_4/Ω_6	Ref.
YZLLAABP (ER01)	0.8579	0.3155	0.9595	0.3288	P.W.
YZLLAABP (ER1.5)	0.8650	0.3041	0.9636	0.3156	P.W.
YZLLAABP (ER02)	0.8673	0.2994	0.9623	0.3111	P.W.
ZLMVBB (PR)	2.059	1.683	4.493	0.3746	[29]
SYB (PR)	10.57	2.94	17.85	0.1647	[30]

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4.3. Excitation Spectrum

The Excitation spectra of Er^{3+} doped YZLLAABP glass specimens have been presented in Figure 3 in terms of Excitation Intensity versus wavelength. The excitation spectrum was recorded in the spectral region 300–600 nm fluorescence at 550nm having different excitation band centered at 350,365, 381, 425, 450, 470and 515 nm are attributed to the ${}^{2}K_{15/2}$, ${}^{4}G_{9/2}$, ${}^{4}G_{11/2}$, ${}^{2}G_{9/2}$, ${}^{4}F_{5/2}$ and ${}^{2}H_{11/2}$ transitions, respectively. The highest absorption level is ${}^{4}G_{11/2}$ and is at 381nm.So this is to be chosen for excitation wavelength.



Fig.3: Excitation Spectrum of YZLLAABP ER (01) glass.

4.4. Fluorescence Spectrum

The fluorescence spectrum of Er^{3+} doped in ytterbium zinc lithium lead alumino antimony borophosphate glass is shown in Figure 4. There are seven broad bands (${}^{4}\text{F}_{7/2} \rightarrow {}^{4}\text{I}_{15/2}$), (${}^{2}\text{H}_{11/2} \rightarrow {}^{4}\text{I}_{15/2}$), (${}^{4}\text{S}_{3/2} \rightarrow {}^{4}\text{I}_{15/2}$), (${}^{4}\text{F}_{9/2} \rightarrow {}^{4}\text{I}_{15/2}$), (${}^{4}\text{F}_{9/2} \rightarrow {}^{4}\text{I}_{15/2}$), (${}^{4}\text{I}_{13/2} \rightarrow {}^{4}\text{I}_{1$



Fig.4: Excitation Spectrum of YZLLAABP ER (01) glass.

Table 5. Emission peak wave lengths (λ_p) , radiative transition probability (A_{rad}) , branching ratio (β_R) , stimulated emission crosssection (σ_p) , and radiative life time (τ) for various transitions in Er^{3+} doped YZLLAABP glasses.

Transition		YZLLAABP ER 01				YZLLAABP ER 1.5				YZLLAABP ER 02			
	λ_{max}	$A_{rad}(s^{-1})$	β	σ_p	$\tau_R(\mu s)$	$A_{rad}(s^{-1})$	β	σ_p	$\tau_R (\mu s)$	$A_{rad}(s^{-1})$	β	σ_p	$\tau_{\rm R}$
	(nm)			(10-20				(10-20				(10-20	(10-20
				cm ²)				cm ²)				cm ²)	cm ²)
${}^{4}F_{7/2} \rightarrow {}^{4}I_{15/2}$	485	1925.20	0.4065	0.529		1931.22	0.4067	0.520		1927.98	0.4064	0.492	
$^{2}H_{11/2} \rightarrow ^{4}I_{15/2}$	530	1191.51	0.2516	0.382		1195.16	0.2517	0.368		1196.37	0.2522	0.357	
${}^{4}S_{3/2} \rightarrow {}^{4}I_{15/2}$	550	850.73	0.1796	0.259		855.53	0.1802	0.246		855.94	0.1804	0.240	
${}^{4}F_{9/2} \rightarrow {}^{4}I_{15/2}$	657	576.93	0.1218	0.312	211.17	573.82	0.1209	0.298	210.62	571.59	0.1205	0.284	210.78
${}^{4}I_{11/2} \rightarrow {}^{4}I_{15/2}$	990	98.31	0.0208	0.348		98.94	0.0208	0.342		98.96	0.0208	0.334	
${}^{4}I_{13/2} \rightarrow {}^{4}I_{15/2}$	1538	79.48	0.0168	1.210		79.88	0.0168	1.189		79.85	0.0168	1.156	
${}^{4}I_{11/2} \rightarrow {}^{4}I_{13/2}$	2711	13.34	0.0028	0.791		13.40	0.0028	0.777		13.39	0.0028	0.760	

V. Conclusion

In the present study, the glass samples of composition (25-x)P₂O₅: 10ZnO: 10Li₂O: 10PbO: 10Al₂O₃: 10Sb₂O₃: 15B₂O₃:xEr₂O₃ (where x =1, 1.5, 2 mol %) have been prepared by melt-quenching method. The value of stimulated emission cross-section (σ_p) is found to be maximum for the transition (${}^{4}I_{13/2} \rightarrow {}^{4}I_{15/2}$) for glass YZLLAABP (ER 01), suggesting that glass YZLLAABP (ER 01) is better compared to the other two glass systems. The transition (${}^{4}I_{13/2} \rightarrow {}^{4}I_{15/2}$) is useful for the development of 1.5 µm broadband Amplifier

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