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# Spectroscopic Properties of Pr<sup>3+</sup> Doped Zinc Lithium Alumino Lead Borotellurite Glasses

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**ABSTRACT:** Zinc lithium alumino lead borotellurite glasses containing Pr<sup>3+</sup> in (35-x):TeO<sub>2</sub>:10ZnO:10Li<sub>2</sub>O:10Al<sub>2</sub>O<sub>3</sub>:10PbO:25B<sub>2</sub>O<sub>3</sub>:xPr<sub>6</sub>O<sub>11</sub> (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 and fluorescence spectra were recorded at room temperature for all glass samples. Judd-Ofelt intensity parameters  $\Omega_{\lambda}$  ( $\lambda=2, 4, 6$ ) 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:** ZLALBT Glasses, Optical Properties, Judd-Ofelt Theory, Rare earth ions.

## I. INTRODUCTION

Among different glass matrices, scientific interest on tellurite glasses is attributed to their favorable properties such as, high refractive index, high density, large third order nonlinear optical susceptibility [1-6]. Tellurite glass is an extremely promising material for laser, mechanical sensors, reflecting windows and nonlinear applications in optics due to some of its essential characteristic features, such as low phonon maxima, low melting temperature and excellent transparency in the far infrared region [7-12]. They have high thermal stability, high transparency, a low melting point, a high gain density and low dispersion rates [13-15]. Zinc oxide is added in the glass matrix to increase glass forming ability and to ensure low rates of crystallization in the glass system [16]. Tellurite Glasses are both scientifically and technologically important materials because they generally offer some unique physical properties better than other Glasses [17-19]. Among active rare-earth ions Pr<sup>3+</sup> exhibits high solubility in ceramic glasses, which also possess excellent optical and physical properties [20-25].

The present work reports on the preparation and characterization of rare earth doped heavy metal oxide (HMO) glass systems for lasing materials. I have studied on the Optical absorption and fluorescence properties of Pr<sup>3+</sup> doped zinc lithium alumino lead borotellurite glasses. 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 ( $\beta$ ), radiative life time ( $\tau_R$ ) and stimulated emission cross section ( $\sigma_p$ ) are evaluated using J.O. intensity parameters ( $\Omega_{\lambda}$ ,  $\lambda=2, 4$  and 6).

## II. EXPERIMENTAL TECHNIQUES

### Preparation of glasses

The following Pr<sup>3+</sup> doped zinc lithium alumino lead borotellurite glass samples (35-x):TeO<sub>2</sub>:10ZnO:10Li<sub>2</sub>O:10Al<sub>2</sub>O<sub>3</sub>:10PbO:25B<sub>2</sub>O<sub>3</sub>: xPr<sub>6</sub>O<sub>11</sub> (where x=1, 1.5, 2) have been prepared by melt-quenching method. Analytical reagent grade chemical used in the present study consist of TeO<sub>2</sub>, ZnO, Li<sub>2</sub>O, Al<sub>2</sub>O<sub>3</sub>, PbO, B<sub>2</sub>O<sub>3</sub> and Pr<sub>6</sub>O<sub>11</sub>. 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 1075<sup>0</sup>C, for preparation of zinc lithium alumino lead borotellurite 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 %)
ZLALBT (UD)	35TeO <sub>2</sub> :10ZnO:10Li <sub>2</sub> O:10Al <sub>2</sub> O <sub>3</sub> :10PbO:25B <sub>2</sub> O <sub>3</sub>
ZLALBT (PR1)	34TeO <sub>2</sub> :10ZnO:10Li <sub>2</sub> O:10Al <sub>2</sub> O <sub>3</sub> :10PbO:25B <sub>2</sub> O <sub>3</sub> :1 Pr <sub>6</sub> O <sub>11</sub>
ZLALBT(PR 1.5)	33.5TeO <sub>2</sub> :10ZnO:10Li <sub>2</sub> O:10Al <sub>2</sub> O <sub>3</sub> :10PbO:25B <sub>2</sub> O <sub>3</sub> : 1.5 Pr <sub>6</sub> O <sub>11</sub>
ZLALBT (PR2)	33TeO <sub>2</sub> :10ZnO:10Li <sub>2</sub> O:10Al <sub>2</sub> O <sub>3</sub> :10PbO:25B <sub>2</sub> O <sub>3</sub> : 2 Pr <sub>6</sub> O <sub>11</sub>

ZLALBT (UD)—Represents undoped Zinc Lithium Alumino Lead Borotellurite glass specimen.

ZLALBT(PR) -Represents Pr<sup>3+</sup> doped Zinc Lithium Alumino Lead Borotellurite glass specimens.

### III. THEORY

#### 3.1 Oscillator Strength

The intensity of spectral lines are expressed in terms of oscillator strengths using the relation [26].

$$f_{\text{expt.}} = 4.318 \times 10^{-9} \int \epsilon(\nu) d\nu \quad (1)$$

where,  $\epsilon(\nu)$  is molar absorption coefficient at a given energy  $\nu$  (cm<sup>-1</sup>), 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 [27].

$$P_m = 4.6 \times 10^{-9} \times \frac{1}{cl} \log \frac{I_0}{I} \times \Delta\nu_{1/2} \quad (2)$$

where  $c$  is the molar concentration of the absorbing ion per unit volume,  $l$  is the optical path length,  $\log I_0/I$  is absorbivity or optical density and  $\Delta\nu_{1/2}$  is half band width.

#### 3.2. Judd-Ofelt Intensity Parameters

According to Judd [28] and Ofelt [29] 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:

$$\frac{8\pi^2 mc \bar{\nu}}{3h(2J+1)n} \left[ \frac{(n^2+2)^2}{9} \right] \times S(J, J') \quad (3) \quad \text{where, the line}$$

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

$$S(J, J') = e^2 \sum \Omega_\lambda \langle 4f^N(S, L) J || U^{(\lambda)} || 4f^N(S', L') J' \rangle^2 \quad (4)$$



$\lambda = 2, 4, 6$

In the above equation  $m$  is the mass of an electron,  $c$  is the velocity of light,  $\nu$  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,  $\Omega_\lambda$  ( $\lambda = 2, 4$  and  $6$ ) are known as Judd-Ofelt intensity parameters.

### 3.3. Radiative Properties

The  $\Omega_\lambda$  parameters obtained using the absorption spectral results have been used to predict radiative properties such as spontaneous emission probability ( $A$ ) and radiative life time ( $\tau_R$ ), and laser parameters like fluorescence branching ratio ( $\beta_R$ ) and stimulated emission cross section ( $\sigma_p$ ).

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:

$$A[(S', L') J'; (S, L) J] = \frac{64 \pi^2 \nu^3}{3h(2J'+1)} \left[ \frac{n(n^2+2)^2}{9} \right] \times S(J', \bar{J}) \quad (5)$$

$$\text{Where, } S(J', J) = e^2 [\Omega_2 \|U^{(2)}\|^2 + \Omega_4 \|U^{(4)}\|^2 + \Omega_6 \|U^{(6)}\|^2]$$

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

$$\beta[(S', L') J'; (S, L) J] = \sum \frac{A[(S', L') J'; (S, L) J]}{A[(S', L') J'; (S, L) J]} \quad (6)$$

where, the sum is over all terminal manifolds.

The radiative life time is given by

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

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

$$\sigma_p(\lambda_p) = \left[ \frac{\lambda_p^4}{8\pi c n^2 \Delta\lambda_{eff}} \right] \times A[(S', L') J'; (S, L) J] \quad (8)$$

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

### 3.4 Nephelauxetic Ratio ( $\beta$ ) and Bonding Parameter ( $b^{1/2}$ )

The nature of the R-O bond is known by the Nephelauxetic Ratio ( $\beta$ ) and Bonding Parameters ( $b^{1/2}$ ), which are computed by using following formulae [30, 31]. The Nephelauxetic Ratio is given by

$$\beta' = \frac{\nu_g}{\nu_a} \quad (9)$$

where,  $\nu_a$  and  $\nu_g$  refer to the energies of the corresponding transition in the glass and free ion, respectively. The values of bonding parameter  $b^{1/2}$  are given by



$$b^{1/2} = \left[ \frac{1-\beta'}{2} \right]^{1/2} \quad (10)$$

#### IV. RESULT AND DISCUSSION

##### 4.1. XRD Measurement

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.

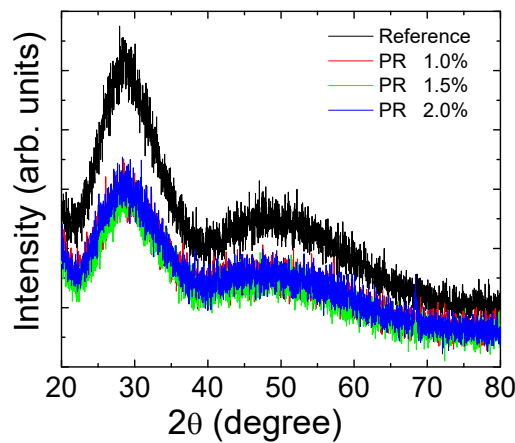


Fig.1:X-ray diffraction pattern of ZLALBT (PR) glass.

##### 4.2. Absorption spectra

The absorption spectra of ZLALBT (PR) glasses, consists of absorption bands corresponding to the absorptions from the ground state  $^3H_4$  of  $Pr^{3+}$  ions. Eight absorption bands have been observed from the ground state  $^3H_4$  to excited states  $^3F_2$ ,  $^3F_3$ ,  $^3F_4$ ,  $^1G_4$ ,  $^1D_2$ ,  $^3P_0$ ,  $^3P_1$  and  $^3P_2$  for  $Pr^{3+}$  doped ZLALBT (PR) glasses.

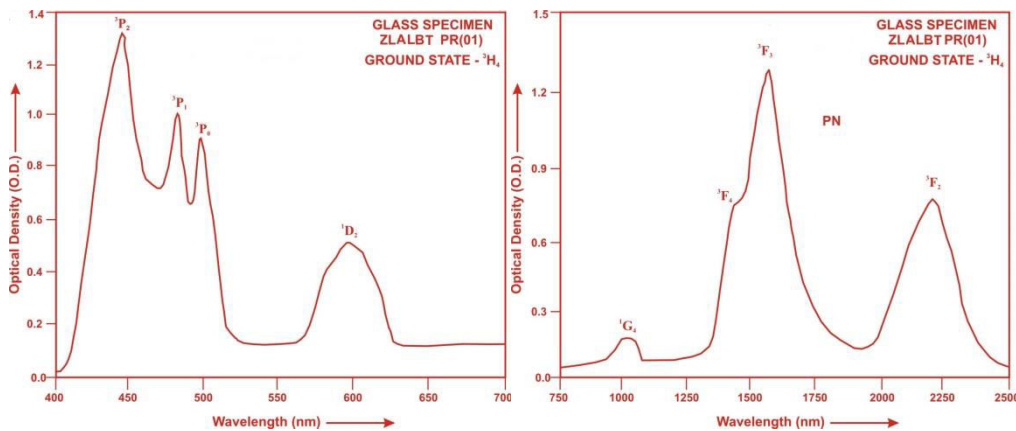


Fig.2: Absorption spectra of ZLALBT (PR01) glass.





The experimental and calculated oscillator strengths for Pr<sup>3+</sup> ions in zinc lithium alumino lead borotellurite glasses are given in Table 2

**Table 2. Measured and calculated oscillator strength ( $P^m \times 10^{+6}$ ) of Pr<sup>3+</sup> ions in ZLALBT glasses.**

Energy level <sup>3</sup> H <sub>4</sub>	Glass ZLALBT (PR01)		Glass ZLALBT (PR1.5)		Glass ZLALBT (PR02)	
	P <sub>exp.</sub>	P <sub>cal.</sub>	P <sub>exp.</sub>	P <sub>cal.</sub>	P <sub>exp.</sub>	P <sub>cal.</sub>
<sup>3</sup> F <sub>2</sub>	4.54	3.94	3.85	3.34	2.74	2.44
<sup>3</sup> F <sub>3</sub>	7.85	6.91	6.74	5.88	5.85	5.15
<sup>3</sup> F <sub>4</sub>	4.35	4.17	3.55	3.54	2.55	3.02
<sup>1</sup> G <sub>4</sub>	0.49	0.36	0.35	0.30	0.29	0.26
<sup>1</sup> D <sub>2</sub>	3.36	1.22	2.95	1.03	1.79	0.90
<sup>3</sup> P <sub>0</sub>	4.58	2.23	3.88	1.91	2.79	1.97
<sup>3</sup> P <sub>1</sub>	4.99	3.64	3.99	3.12	2.87	3.08
<sup>3</sup> P <sub>2</sub>	11.37	4.03	10.29	3.43	9.34	2.98
R.m.s.deviation	2.8940		2.6537		2.3116	

The ratio of Racah parameters  $E^1/E^3$  and  $E^2/E^3$  are about 9.81 and 0.048 respectively. Computed values of Slater-Condon, Lande', Racah, nephelauxetic ratio and bonding parameter for Pr<sup>3+</sup> doped ZLALBT glass specimens are given in Table 3.

**Table 3. Computed values of Slater-Condon, Lande', Racah, nephelauxetic ratio and bonding parameter for Pr<sup>3+</sup> doped ZLALBT glass specimens.**

Parameter	Free ion	ZLALBT(PR01)	ZLALBT(PR1.5)	ZLALBT(PR02)
F <sub>2</sub> (cm <sup>-1</sup> )	322.09	299.57	299.57	299.56
F <sub>4</sub> (cm <sup>-1</sup> )	44.46	44.35	44.35	44.34
F <sub>6</sub> (cm <sup>-1</sup> )	4.867	4.426	4.426	4.424
ξ <sub>4f</sub> (cm <sup>-1</sup> )	741.00	856.91	856.89	857.08
E <sup>1</sup> (cm <sup>-1</sup> )	4728.92	4452.65	4452.68	4452.09
E <sup>2</sup> (cm <sup>-1</sup> )	24.75	21.95	21.95	21.95
E <sup>3</sup> (cm <sup>-1</sup> )	478.10	453.73	453.74	453.73
F <sub>4</sub> /F <sub>2</sub>	0.13805	0.14803	0.14804	0.14801
F <sub>6</sub> /F <sub>2</sub>	0.01511	0.0147732	0.0147730	0.0147697



$E^1/E^3$	9.8911	9.81341	9.8133	9.812108
$E^2/E^3$	0.0518	0.048366	0.04837	0.048369
$\beta'$		0.88957	0.88957	0.889426
$b^{1/2}$		0.23498	0.23498	0.23513

Judd-Ofelt intensity parameters  $\Omega_\lambda$  ( $\lambda = 2, 4$  and  $6$ ) were calculated by using the fitting approximation of the experimental oscillator strengths to the calculated oscillator strengths with respect to their electric dipole contributions. In the present case the three  $\Omega_\lambda$  parameters follow the trend  $\Omega_2 < \Omega_4 < \Omega_6$ .

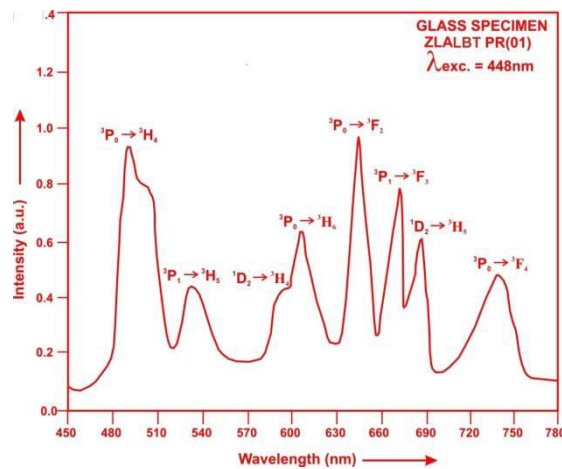
The values of Judd-Ofelt intensity parameters are given in Table 4.

**Table 4. Judd-Ofelt intensity parameters for Pr<sup>3+</sup> doped ZLALBT glass specimens.**

Glass Specimen	$\Omega_2(\text{pm}^2)$	$\Omega_4(\text{pm}^2)$	$\Omega_6(\text{pm}^2)$	$\Omega_4/\Omega_6$
ZLALBT(PR01)	2.493	3.370	6.242	0.5399
ZLALBT(PR1.5)	2.092	2.892	5.295	0.5462
ZLALBT(PR02)	1.005	2.971	4.481	0.6630

### 4.3. Fluorescence Spectrum

The fluorescence spectrum of Pr<sup>3+</sup> doped in zinc lithium aluminoborate glass is shown in Figure 3. There are eight broad bands ( $^3P_0 \rightarrow ^3H_4$ ), ( $^3P_1 \rightarrow ^3H_5$ ), ( $^1D_2 \rightarrow ^3H_4$ ), ( $^3P_0 \rightarrow ^3H_6$ ), ( $^3P_0 \rightarrow ^3F_2$ ), ( $^3P_1 \rightarrow ^3F_3$ ), ( $^1D_2 \rightarrow ^3H_5$ ) and ( $^3P_0 \rightarrow ^3F_4$ ) respectively for glass specimens.



**Fig.3: Fluorescence spectrum of ZLALBT (PR01) glass.**



**Table 5.**Emission peak wave lengths ( $\lambda_p$ ), radiative transition probability ( $A_{rad}$ ), branching ratio ( $\beta_R$ ), stimulated emission crosssection ( $\sigma_p$ ), and radiative life time ( $\tau$ ) for various transitions in Pr<sup>3+</sup> doped ZLALBT glasses.

Transition	$\lambda_{max}$ (nm)	ZLALBT(PR 01)				ZLALBT(PR 1.5)				ZLALBT(PR 02)			
		$A_{rad}(s^{-1})$	$\beta$	$\sigma_p$ ( $10^{-20}$ cm <sup>2</sup> )	$\tau_R(\mu s)$	$A_{rad}(s^{-1})$	$\beta$	$\sigma_p$ ( $10^{-20}$ cm <sup>2</sup> )	$\tau_R(\mu s)$	$A_{rad}(s^{-1})$	$\beta$	$\sigma_p$ ( $10^{-20}$ cm <sup>2</sup> )	$\tau_R$ ( $10^{-20}$ cm <sup>2</sup> )
<sup>3</sup> P <sub>0</sub> → <sup>3</sup> H <sub>4</sub>	485	1771.19	0.1627	0.852	91.88	1523.93	0.1645	0.919	107.92	1570.82	0.2103	1.272	133.85
<sup>3</sup> P <sub>1</sub> → <sup>3</sup> H <sub>5</sub>	532	2892.75	0.2658	0.672		2476.90	0.2673	0.604		2390.97	0.3200	0.614	
<sup>1</sup> D <sub>2</sub> → <sup>3</sup> H <sub>4</sub>	599	600.23	0.0552	0.271		511.002	0.0551	0.245		445.82	0.0596	0.228	
<sup>3</sup> P <sub>0</sub> → <sup>3</sup> H <sub>6</sub>	602	503.41	0.0463	0.324		427.89	0.0462	0.314		363.73	0.0486	0.298	
<sup>3</sup> P <sub>0</sub> → <sup>3</sup> F <sub>2</sub>	643	1739.31	0.1598	2.181		146.24	0.1578	2.212		705.03	0.0943	1.343	
<sup>3</sup> P <sub>1</sub> → <sup>3</sup> F <sub>3</sub>	676	3041.35	0.2794	1.911		2575.61	0.02780	1.704		169.78	0.2272	1.202	
<sup>1</sup> D <sub>2</sub> → <sup>3</sup> H <sub>5</sub>	685	7.93	0.00073	0.011		6.80	0.00073	0.011		6.67	0.0008	0.012	
<sup>3</sup> P <sub>0</sub> → <sup>3</sup> F <sub>4</sub>	730	327.38	0.0301	0.287		281.51	0.0304	0.257		290.35	0.0388	0.288	

**V. CONCLUSION**

In the present study, the glass samples of composition (35-x):TeO<sub>2</sub>:10ZnO:10Li<sub>2</sub>O:10Al<sub>2</sub>O<sub>3</sub>:10PbO:25B<sub>2</sub>O<sub>3</sub>:xPr<sub>6</sub>O<sub>11</sub>(where x=1, 1.5, 2 mol %) have been prepared by melt-quenching method. The value of stimulated emission cross-section ( $\sigma_p$ ) is found to be maximum for the transition (<sup>3</sup>P<sub>0</sub>→<sup>3</sup>F<sub>2</sub>) for glass ZLALBT(PR 01), suggesting that glass ZLALBT(PR01) is better compared to the other two glass systems ZLALBT(PR1.5) and ZLALBT(PR02).

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