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## Spectral and Thermal Properties of Tm<sup>3+</sup> Doped in Zinc Lithium Lead AluminoTungsten Borophosphate Glasses

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**ABSTRACT:** Glass sample of Zinc Lithium LeadAluminoTungsten Borophosphate: (35-x) P<sub>2</sub>O<sub>5</sub>:10ZnO:10Li<sub>2</sub>O:10PbO:10Al<sub>2</sub>O<sub>3</sub>:10WO<sub>3</sub>:15B<sub>2</sub>O<sub>3</sub>:xTm<sub>2</sub>O<sub>3</sub>. (where x=1,1.5 and 2 mol%) have been prepared by melt-quenching technique. The amorphous nature of the prepared glasssamples was confirmed by X-ray diffraction. Optical absorption and fluorescence spectra were recorded at roomtemperature for all glass samples.Judd-Ofeltintensity parameters  $\Omega_{\lambda}$  ( $\lambda$ =2, 4 and6) are evaluated from theintensities of various absorption bands of opticalabsorption spectra. Using these intensity parameters variousradiative properties like spontaneous emission probability, branching ratio, radiative life time and stimulatedemission cross-section of various emission lines have been evaluated

KEYWORDS: ZLLATBP Glasses, Optical Properties, Judd-Ofelt Theory, Rare earth ions.

#### I. INTRODUCTION

Transparent glass–ceramic as host materials for active optical ions have attracted great interest recently due to their potential application in optical devices such as frequency-conversion materials, waveguide laser and optical fiber amplifiers [1-6].Rare- earth doped glasses are technologically important materials as they find applications in lasers, white light emitting diodes and light converting devices[7-10].Glasses based on heavy metal oxide have received increased attention due to their manifold possible applications in the field of glass ceramics, layers for optoelectronics devices, thermal and mechanical sensors [11-15].The host glass materials should have high refractive index with good chemical and thermal stability. Phosphate glasses are promising laser hosts because they are able to accommodate higher content of rare earth ions and still remain amorphous in comparison with other glass systems. The up-conversion of phosphate glasses is also compressed because of their relatively large phonon energy. Phosphate glasses can be a good matrix for fiber lasers because they exhibit good mechanical property [16-25].

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 absorption and emission properties of  $\text{Tm}^{3+}$ doped zinc lithium lead aluminotungsten borophosphateglasses. 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  $\text{Tm}^{3+}$ doped borophosphateglass samples (35-x) P<sub>2</sub>O<sub>5</sub>:10ZnO:10Li<sub>2</sub>O:10 PbO:10Al<sub>2</sub>O<sub>3</sub>:10WO<sub>3</sub>:15B<sub>2</sub>O<sub>3</sub>:xTm<sub>2</sub>O<sub>3</sub> (where x=1,1.5 and 2 mol%) have been prepared by melt-quenching method. Analytical reagent grade chemical used in the present study consist of P<sub>2</sub>O<sub>5</sub>,ZnO,Li<sub>2</sub>O,PbO,Al<sub>2</sub>O<sub>3</sub>, WO<sub>3</sub>,B<sub>2</sub>O<sub>3</sub> and Tm<sub>2</sub>O<sub>3</sub>. They were thoroughly mixed by using an agate pestle mortar. then melted at 965<sup>o</sup>C by an electrical muffle furnace for 2h., After complete melting, the melts were quickly poured in to a preheated stainless steel mould and annealed at temperature of 250<sup>o</sup>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**. International Journal of Innovative Research in Science, Engineering and Technology (IJIRSET)

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Table 1.

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ZLLATBP (UD) -Represents undopedzinc lithium lead aluminotungsten borophosphate glass specimen. ZLLATBP (TM) -Represents Tm<sup>3+</sup>dopedzinc lithium lead aluminotungsten borophosphate glass specimens.

#### **III. THEORY**

#### **3.1Oscillator Strength**

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

$$f_{\text{expt.}} = 4.318 \times 10^{-9} \mathrm{fe}(v) \,\mathrm{dv}$$
 (1)

where,  $\varepsilon(v)$  is molar absorption coefficient at a given energy v (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 [27], using the modified relation:

$$P_{m}=4.6 \times 10^{-9} \times \frac{1}{cl} \log \frac{I_{0}}{I} \times \Delta v_{1/2}(2)$$

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

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

strength S (J, J') is given by the equation S (S', L') = $e^2 \sum \Omega_{\lambda} \leq 4f^N(S, L) J \| U^{(\lambda)} \| 4f^N(S', L')J' > 2$  (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,  $\Omega_{\lambda}$  ( $\lambda$ =2,4and 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  $|4f^{N}(S', L') J'>$  to a final manifold  $|4f^{N}(S,L) J>|$  is given by:

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

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

Where, the line

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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

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

SLJ

where, the sum is over all terminal manifolds.

The radiative life time is given by

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

SLJ

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

 $4f^{N}(S,L)J > |$  is expressed as

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

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

#### **IV. RESULT AND DISCUSSION**

#### 4.1XRD Measurement

Figure 1 presents the XRD pattern of the sample contain -  $P_2O_5$  which is show no sharp Bragg's peak, but only a broad diffuse hump around low angle region. This is the clear indication of amorphous nature within the resolution limit of XRD instrument

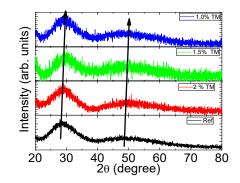


Fig. 1 X-ray diffraction pattern of P2O5:ZnO:Li2O:PbO:Al2O3:WO3:B2O3:Tm2O3.

#### 4.2 Thermal Property

Figure 2, shows the thermal properties of ZLLATBP glass from  $300^{\circ}$ C to  $1000^{\circ}$ C. From the DSC curve of present glasses system, I can find out that no crystallization peak is apparent and the glass transition temperature T<sub>g</sub> are 355,450 and 585 respectively. The T<sub>g</sub> increase with the contents of Tm<sub>2</sub>O<sub>3</sub> increase. I could conclude that thermal properties of the ZLLATBP glass are good for fiber drawing from the analysis of DSC curve.

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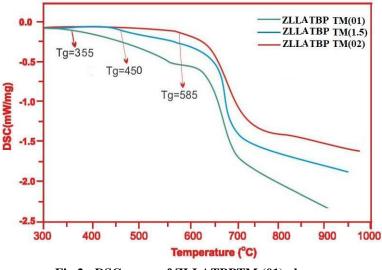


Fig.2: DSC curve of ZLLATBPTM (01) glasses.

#### 4.3 Absorption Spectrum

The absorption spectra of  $\text{Tm}^{3+}$  doped ZLLATBPglass specimens have been presented in Figure 3 in terms of optical density versus wavelength. Five absorption bands have been observed from the ground state  ${}^{3}\text{H}_{6}$  to excited states  ${}^{3}\text{F}_{4}$ ,  ${}^{3}\text{H}_{5}$ ,  ${}^{3}\text{H}_{4}$ ,  ${}^{3}\text{F}_{3}$  and  ${}^{1}\text{G}_{4}$  for Tm<sup>3+</sup> doped ZLLATBPglasses.

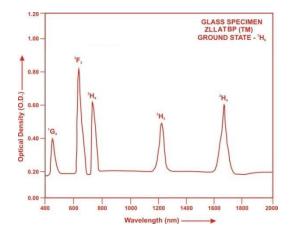


Fig. (3) Absorption spectrum of ZLLATBP TM (01) glass.

The experimental and calculated oscillator strength for Tm<sup>3+</sup>ions in ZLLATBP glasses are given in **Table 2**.

**Table 2:** Measured and calculated oscillator strength ( $P_m \times 10^{+6}$ ) of Tm<sup>3+</sup>ions in ZLLATBP glasses.

Energy level from	Glass		Glass		Glass		
<sup>3</sup> H <sub>6</sub>	ZLLATBP	(1M01)	ZLLATB	<u>(1M1.5)</u>	ZLLATBP(TM02)		
	P <sub>exp</sub> .	P <sub>cal</sub> .	P <sub>exp</sub> .	P <sub>cal</sub> .	P <sub>exp</sub> .	P <sub>cal</sub> .	
${}^{3}F_{4}$	1.80	1.84	1.78	1.83	1.80	1.75	
$^{3}\text{H}_{5}$	1.35	1.44	1.32	1.43	1.41	1.28	
${}^{3}\text{H}_{4}$	1.97	2.04	1.95	2.03	2.01	1.92	
${}^{3}F_{3}$	2.95	3.02	2.91	2.99	2.96	2.87	
${}^{1}G_{4}$	0.75	0.89	0.72	0.88	0.87	0.69	
r.m.s. deviation	±0.08894		±0.1037		±0.1180		

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\*Low r.m.s.deviationvalues clearly indicate the accuracy of fitting.

In theZinc Lithium Lead AluminoTungsten Borophosphateglasses (ZLLATBP) $\Omega_2$ ,  $\Omega_4$  and  $\Omega_6$ parameters decrease with the increase of x from 1 to 2 mol%. The order of magnitude of Judd-Ofelt intensity parameters is  $\Omega_4 > \Omega_2 > \Omega_6$  for all the glass specimens. The spectroscopic quality factor ( $\Omega_4/\Omega_6$ ) related with the rigidity of the glass system has been found to lie between 1.439 and 1.459 in the present glasses.

The values of Judd-Ofelt intensity parameters are given in Table 3.

Table3: Judd-Ofelt intensity parameters for Tm<sup>3+</sup> doped ZLLATBPglass specimens.

Glass Specimen	$\Omega_2(pm^2)$	$\Omega_4(pm^2)$	$\Omega_6(pm^2)$	$\Omega_4/\Omega_6$
ZLLATBP (TM 01)	6.633	8.299	5.688	1.459
ZLLATBP B(TM 1.5)	6.671	8.163	5.645	1.446
ZLLATBP(TM 02)	6.592	8.037	5.586	1.439

#### 4.4. Fluorescence Spectrum

The fluorescence spectrum of  $\text{Tm}^{3+}$ doped in zinc lithium lead aluminotungsten borophosphateglass is shown in Figure 4. There are seven broad bands observed in the Fluorescence spectrum of  $\text{Tm}^{3+}$ dopedzinc lithium lead aluminotungsten borophosphateglass. The wavelengths of these bands along with their assignments are given in Table 5. The peak with maximum emission intensity appears at 798nm and corresponds to the  $({}^{3}\text{H}_{4} \rightarrow {}^{3}\text{H}_{6})$  transition.

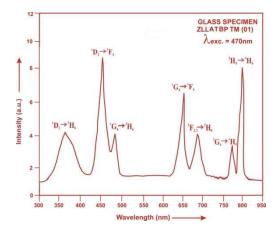


Fig. (4). Fluorescence spectrum of ZLLATBP TM (01) glass.

Table4: Emission peak wave lengths  $(\lambda_p)$ , radiative transition probability  $(A_{rad})$ , branching ratio  $(\beta)$ , stimulated emission cross-section  $(\sigma_p)$  and radiative life time  $(\tau_R)$  for various transitions in Tm<sup>3+</sup> doped ZLLATBP glasses

Transition		ZLLATBP(TM01)			ZLLATBP(TM1.5)			ZLLATBP(TM 02)					
	$\lambda_{max}$	A <sub>rad</sub> (s <sup>-1</sup> )	β	σp	τ <sub>R</sub> (μs)	A <sub>rad</sub> (s <sup>-1</sup> )	β	σ <sub>p</sub>		$A_{rad}(s^{-1})$	β	σ <sub>p</sub>	τ <sub>R</sub> (10 <sup>-20</sup>
	(nm)			$(10^{-20}  \text{cm}^2)$				$(10^{-20} \mathrm{cm}^2)$	τ <sub>R</sub> (μs)			$(10^{-20}  \text{cm}^2)$	cm <sup>2</sup> )
$^{1}D_{2}\rightarrow ^{3}H_{\delta}$	365	54552.2	0.6370	1.469		53871.10	0.6342	1.509		53273.10	0.6341	1.547	
$^{1}D_{2}\rightarrow ^{3}F_{4}$	455	17578.6	0.2053	2.230		17653.80	0.2078	2.419		17457.90	0.2078	2.586	
$^1G_4 {\rightarrow} ^3H_6$	480	2242.64	0.0262	0.535		2227.31	0.0262	0.581		2200.65	0.0262	0.633	
$^{1}G_{4}\rightarrow ^{3}F_{4}$	651	736.44	0.0086	1.344	11.6775	730.11	0.0086	1.655	11.7728	723.21	0.0086	2.163	11.9031
<sup>3</sup> F <sub>2,3</sub> → <sup>3</sup> H <sub>6</sub>	689	5572.44	0.0651	2.274		5523.98	0.0650	2.352		5467.46	0.0651	2.444	
$^1G_4 \rightarrow ^3H_5$	785	2133.59	0.0249	2.397		2126.11	0.0250	2.581		2106.74	0.0251	2.764	
$^{3}\mathrm{H}_{4}\mathrm{\rightarrow}^{3}\mathrm{H}_{6}$	798	2818.93	0.0329	4.206		2809.40	0.0331	4.506		2782.36	0.0331	4.962	

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#### V. CONCLUSION

In the present study, the glass samples of composition (35-x)  $P_2O_5$ :10ZnO:10Li<sub>2</sub>O:10PbO:10Al<sub>2</sub>O<sub>3</sub>:10WO<sub>3</sub>:15B<sub>2</sub>O<sub>3</sub>:xTm<sub>2</sub>O<sub>3</sub>(where x =1, 1.5and 2mol %) have been prepared by melt-quenching method. The value of stimulated emission cross-section ( $\sigma_p$ ) is found to be maximum for the transition ( ${}^{3}H_{4} \rightarrow {}^{3}H_{6}$ ) for glass ZLLATBP(TM 01), suggesting that glass ZLLATBP(TM 01) is better compared to the other two glass systems ZLLATBP(TM1.5) and ZLLATBP(TM02).

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