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Spectral, Transmittance and Radiative Properties of Pr³⁺ doped Borotellurite Glasses

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ABSTRACT: Yttrium zinc lithium tungsten vanadium borotellurite glasses containing Pr³⁺ in (30-x):TeO₂:10ZnO:10Li₂O:10WO₃:10V₂O₅:10Y₂O₃:20B₂O₃:xPr₂O₃ (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. Transmittance, Optical absorption, Excitation spectra 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: YZLTVBT Glasses, Optical Properties, Transmittance properties, Judd-Ofelt Theory, Radiative properties.

I. INTRODUCTION

Transparent ceramic glasses have attracted great interest due to their potential application in optical devices such as optical fiber amplifiers, frequency-conversion materials, lasers and solar cells [1–5]. Tellurite glasses have large transparency window, low phonon energy, high refractive index, good mechanical, high density, good thermal and chemical durability [6-8]. Tellurite doped glasses are very important because of the possibility of their application in optoelectronic, optic device fields, such as lasers, fiber optic large bandwidth, high emission and absorption cross-section [9,12]. The addition of network modifier (NWF) Li₂O is to improve optical, electrical and mechanical properties of such glasses. Recently, rare earth doped tellurite glass has attracted much interest because of high rare earth ion solubility and optical data transmission. [13-15].

In this work, the spectroscopic properties of Pr³⁺ -doped (30-x):TeO₂:10ZnO:10Li₂O:10WO₃:10V₂O₅:10Y₂O₃:20B₂O₃:xPr₂O₃ (where x=1, 1.5,2 mol %) glasses were investigated. absorption, Excitation spectra and fluorescence spectra, The transmittance, absorption, Excitation and fluorescence spectra of Pr³⁺ of the glasses were investigated. The J-O intensity parameters render significant information regarding local structure and bonding in the vicinity of rare- earth ions.

II. EXPERIMENTAL TECHNIQUES

Preparation of glasses

The following Pr³⁺ doped yttrium zinc lithium tungsten vanadium borotellurite glass samples (30-x):TeO₂: 10ZnO: 10Li₂O: 10WO₃: 10V₂O₅: 10Y₂O₃:20B₂O₃:xPr₂O₃ (where x=1, 1.5,2) have been prepared by melt-quenching method. Analytical reagent grade chemical used in the present study consist of TeO₂,ZnO,Li₂O,WO₃,V₂O₅,Y₂O₃,B₂O₃and Pr₂O₃. 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 1055^oC, for preparation of Yttrium zinc lithium tungsten vanadium 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 to100^oC.While pouring; the temperature of crucible was also maintained to prevent crystallization. And annealed at temperature of 350^oC 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 %)
YZLTVBT (UD)	30TeO ₂ :10ZnO:10Li ₂ O:10WO ₃ :10V ₂ O ₅ :10Y ₂ O ₃ :20B ₂ O ₃
YZLTVBT PR (1.0)	29TeO ₂ :10ZnO:10Li ₂ O:10WO ₃ :10V ₂ O ₅ :10Y ₂ O ₃ :20B ₂ O ₃ :1 Pr ₂ O ₃
YZLTVBT PR (1.5)	28.5TeO ₂ :10ZnO:10Li ₂ O:10WO ₃ :10V ₂ O ₅ :10Y ₂ O ₃ :20B ₂ O ₃ : 1.5 Pr ₂ O ₃
YZLTVBT PR (2.0)	28TeO ₂ :10ZnO:10Li ₂ O:10WO ₃ :10V ₂ O ₅ :10Y ₂ O ₃ :20B ₂ O ₃ : 2 Pr ₂ O ₃

YZLTVBT (UD)—Represents undoped Yttrium zinc lithium tungsten vanadium borotellurite glass specimen.
YZLTVBT (PR) -Represents Pr³⁺ Yttrium zinc lithium tungsten vanadium borotellurite glass specimens.

III. THEORY

3.1 Oscillator Strength

The intensity of spectral lines are expressed in terms of oscillator strengths using the relation [16].

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

where, $\epsilon(\nu)$ is molar absorption coefficient at a given energy ν (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 [17].

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

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 [18] and Ofelt [19] 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:

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

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

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

In the above equation m is the mass of an electron, c is the velocity of light, $\bar{\nu}$ 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, 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:

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

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

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

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

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

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

3.4 Nephelauxetic Ratio (β') and Bonding Parameter ($b^{1/2}$)

The nature of the R-O bond is known by the Nephelauxetic Ratio (β') and Bonding Parameters ($b^{1/2}$), which are computed by using following formulae [20, 21]. The Nephelauxetic Ratio is given by

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

where, ν_a and ν_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.

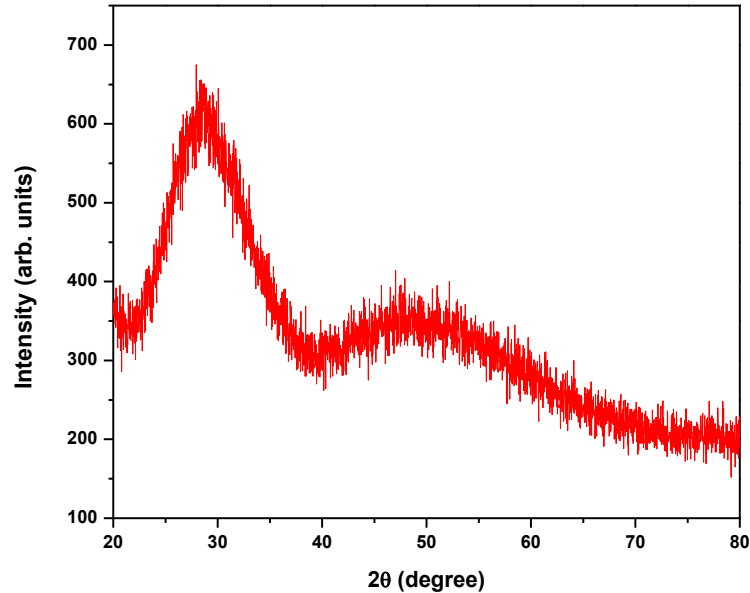


Fig.1: X-ray diffraction pattern of YZLTVBT PR(1.0) glass.

4.2 Transmittance Spectra

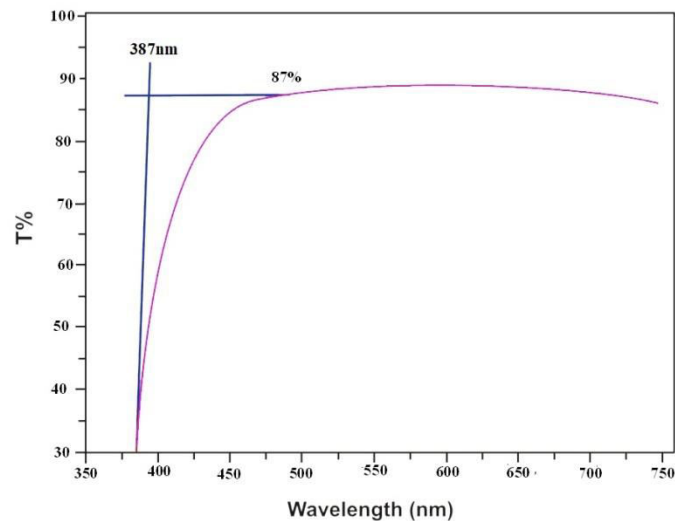


Fig.2: Transmittance Spectra of YZLTVBT PR (1.0) glass.

4.3. Absorption spectra

The absorption spectra of YZLTVBT PR(1.0) glass, 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 YZLTVBT PR(1.0) glass.

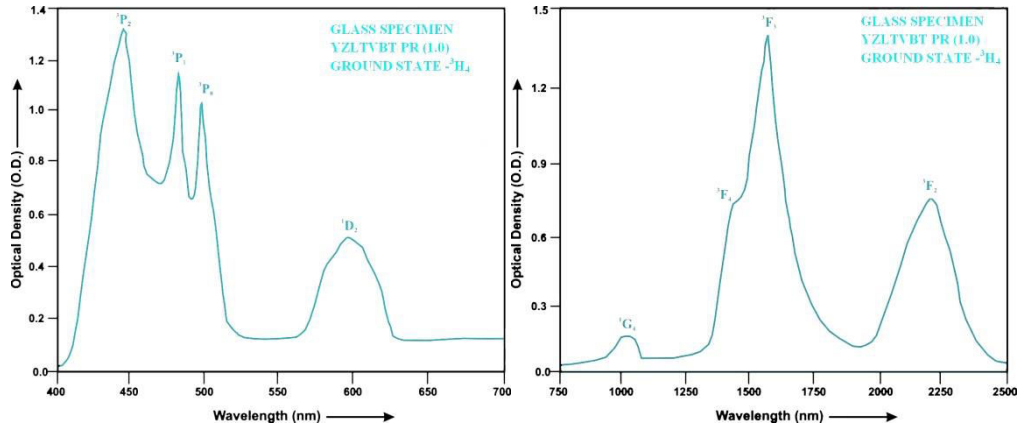


Fig.3: Absorption spectra of YZLTVBT PR (1.0) glass.

The experimental and calculated oscillator strengths for Pr^{3+} ions in Yttrium Zinc lithium tungsten vanadium borotellurite glasses are given in Table 2

Table 2 Measured and calculated oscillator strength ($P^m \times 10^{+6}$) of Pr^{3+} ions in YZLTVBT glasses.

Energy level 3H_4	Glass YZLTVBT PR(1.0)		Glass YZLTVBT PR(1.5)		Glass YZLTVBT PR(2.0)	
	$P_{exp.}$	$P_{cal.}$	$P_{exp.}$	$P_{cal.}$	$P_{exp.}$	$P_{cal.}$
3F_2	4.54	3.82	3.65	2.98	2.55	2.09
3F_3	6.88	5.99	5.46	4.64	4.74	3.95
3F_4	4.25	3.72	3.45	2.97	2.21	2.40
1G_4	0.35	0.31	0.28	0.25	0.23	0.20
1D_2	2.37	1.06	1.88	0.83	1.39	0.70
3P_0	4.57	1.52	3.64	0.93	2.58	1.20
3P_1	4.78	2.64	3.86	1.75	2.73	1.98
3P_2	12.64	3.53	11.53	2.77	10.64	2.32
R.m.s.deviation	3.5412		3.3695		3.0213	

Table3. Computed values of Slater-Condon, Lande', Racah, nephelauxetic ratio and bonding parameter for Pr^{3+} doped YZLTVBT glass specimens

Parameter	Free ion	YZLTVBT PR (1.0)	YZLTVBT PR (1.5)	YZLTVBT PR (2.0)
$F_2(\text{cm}^{-1})$	322.09	300.01	300.02	300.01
$F_4(\text{cm}^{-1})$	44.46	44.27	44.28	44.27
$F_6(\text{cm}^{-1})$	4.867	4.412	4.413	4.412
$\xi_{4f}(\text{cm}^{-1})$	741.00	858.41	858.26	858.42
$E^1(\text{cm}^{-1})$	4728.92	4450.98	4451.48	4450.97
$E^2(\text{cm}^{-1})$	24.75	22.01	22.01	22.01
$E^3(\text{cm}^{-1})$	478.10	454.74	454.73	454.73
F_4/F_2	0.13804	0.14755	0.14758	0.14755
F_6/F_2	0.01511	0.01470	0.01471	0.01470
E^1/E^3	9.8911	9.7881	9.7893	9.7881
E^2/E^3	0.0518	0.0484	0.0484	0.484
β'		0.8887	0.8888	0.8887
$b^{1/2}$		0.2359	0.2358	0.2359

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

Table 4 Judd-Ofelt intensity parameters for Pr³⁺ doped YZLTVBT glass specimens.

Glass Specimen	$\Omega_2(\text{pm}^2)$	$\Omega_4(\text{pm}^2)$	$\Omega_6(\text{pm}^2)$	Ω_4/Ω_6	Ref.
YZLTVBT (PR01)	2.760	2.140	5.272	0.4059	P.W.
YZLTVBT (PR1.5)	2.310	1.312	4.244	0.3091	P.W.
YZLTVBT (PR02)	1.166	1.698	3.388	0.5012	P.W.
TWPL (PR)	2.087	1.591	3.780	0.421	[22].

4.4 Excitation Spectrum

Excitation spectra of YZLTVBT PR (1.0) glass recorded at the emission wavelength 395 nm is depicted as figure 4. The excitation spectra consists of three peaks corresponding to the transitions from the ground state 3H_4 to the various excited states 3P_2 , 3P_1 and 3P_0 at the wavelengths of 448, 465 and 486 nm respectively. Among these, a prominent excitation band at 448 nm has been selected for the measurement of emission spectrum of YZLTVBT PR (1.0) glass.

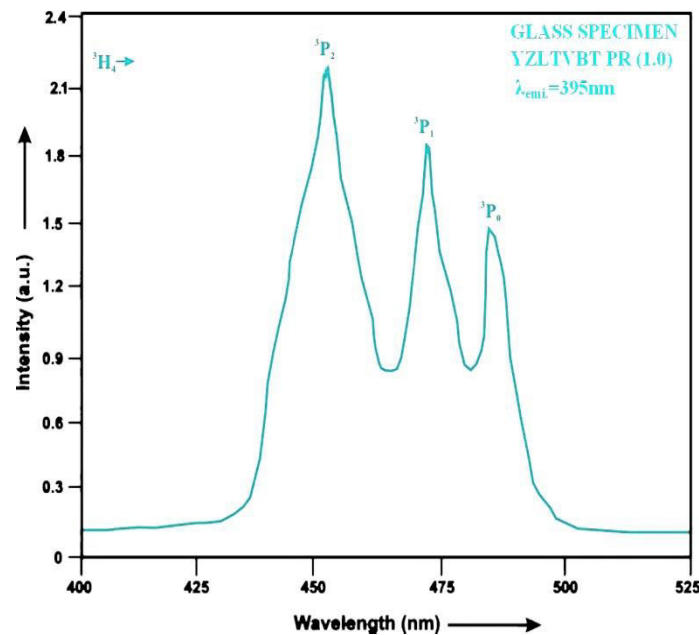


Fig.4: Excitation Spectrum of YZLTVBT PR (1.0) glass.

4.5. Fluorescence Spectrum

The fluorescence spectrum of YZLTVBT PR(1.0) glass is shown in Figure 5. There are eleven broad bands ($^3P_0 \rightarrow ^3H_4$), ($^3P_0 \rightarrow ^3H_5$), ($^1D_2 \rightarrow ^3H_4$) ($^3P_0 \rightarrow ^3H_6$), ($^3P_0 \rightarrow ^3F_2$), ($^3P_1 \rightarrow ^3F_3$), ($^1D_2 \rightarrow ^3H_5$), ($^3P_0 \rightarrow ^3F_4$), ($^1G_4 \rightarrow ^3H_5$) ($^1G_4 \rightarrow ^3H_6$) and ($^1G_4 \rightarrow ^3F_4, ^3F_2$) respectively for YZLTVBT PR(1.0) glass specimen.

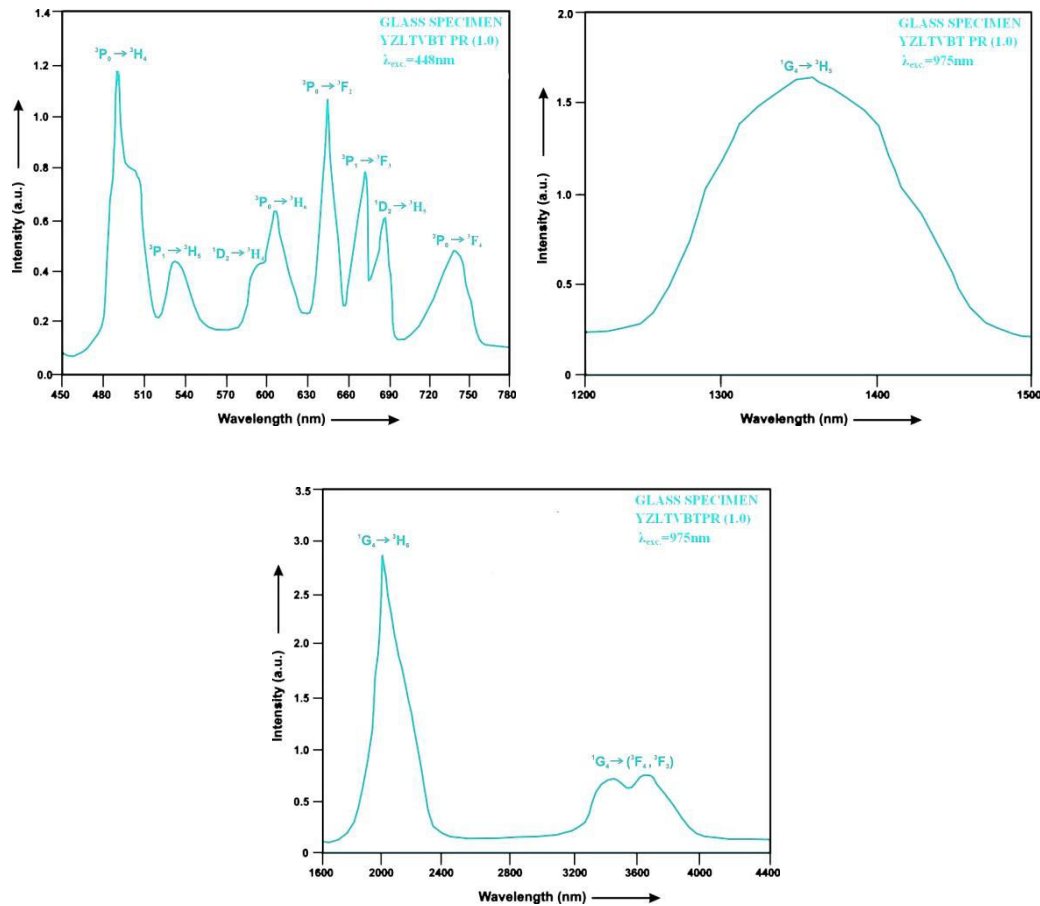


Fig.5: Fluorescence spectrum of YZLTVBT PR (1.0) glass.

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 Pr³⁺ doped YZLTVBT glasses

Transition	YZLTVBT PR (1.0)					YZLTVBT PR (1.5)				YZLTVBT PR (2.0)			
	λ_{max} (nm)	A_{rad} (s ⁻¹)	β	σ_p (10 ⁻²⁰ cm ²)	τ_R (μs)	A_{rad} (s ⁻¹)	β	σ_p (10 ⁻²⁰ cm ²)	τ_R (μs)	A_{rad} (s ⁻¹)	β	σ_p (10 ⁻²⁰ cm ²)	τ_R (10 ⁻²⁰ cm ²)
$^3P_0 \rightarrow ^3H_4$	48	1317	0.11	0.410		809.2	0.09	0.259		1049	0.15	0.349	
	5	.37	80			4	59			.36	76		
$^3P_1 \rightarrow ^3H_5$	52	2410	0.21	0.446		1682.	0.19	0.316		1761	0.26	0.335	
	9	.21	59			00	93			.24	46		
$^1D_2 \rightarrow ^3H_4$	59	573.	0.05	0.215		450.1	0.05	0.173		377.	0.05	0.148	
	9	68	14			1	33			98	68		
$^3P_0 \rightarrow ^3H_6$	60	497.	0.04	0.248		401.6	0.04	0.205		321.	0.04	0.168	
	2	94	46			3	76			25	83		

$^3P_0 \rightarrow ^3F_2$	64 5	2254 .00	0.20 20	1.939	89.6 0	1890. 18	0.22 39	1.684	118. 47	955. 95	0.14 36	0.872	150.2 2
$^3P_1 \rightarrow ^3F_3$	67 6	3408 .19	0.30 54	1.720		2697. 20	0.31 95	1.375		1712 .56	0.25 73	0.887	
$^1D_2 \rightarrow ^3H_5$	68 5	6.44	0.00 06	0.006 4		4.38	0.00 05	0.004 4		4.80	0.00 07	0.005 0	
$^3P_0 \rightarrow ^3F_4$	73 0	243. 35	0.02 18	0.170		149.4 8	0.01 77	0.106		193. 84	0.02 91	0.140	
$^1G_4 \rightarrow ^3H_5$	13 50	272. 77	0.02 44	0.867		216.3 7	0.02 56	0.693		176. 58	0.02 65	0.569	
$^1G_4 \rightarrow ^3H_6$	20 25	156. 90	0.01 41	5.177		124.1 7	0.01 47	4.150		92.6 3	0.01 39	3.136	
$^1G_4 \rightarrow (^3F_4, ^3F_2)$	35 55	20.2 8	0.00 18	3.404		15.92	0.00 19	2.696		10.5 3	0.00 16	1.795	

V. CONCLUSION

In the present study, the glass samples of composition (30-x):TeO₂:10ZnO:10Li₂O:10WO₃:10V₂O₅:10Y₂O₃:20B₂O₃:xPr₂O₃ (where x = 1, 1.5, 2 mol %) have been prepared by melt-quenching method. The stimulated emission cross-section (σ_p) has highest value for the transition ($^1G_4 \rightarrow ^3H_6$) in all the glass specimen doped with Pr³⁺ ion. This shows that ($^1G_4 \rightarrow ^3H_6$) transition is most probable transition and it useful for laser action.

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