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Spectral and Luminescence Studies of Ho³⁺Doped Phosphate Glasses with Large Stimulated Emission Cross Section

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ABSTRACT: Glasses samples containing Ho³⁺ in Ytterbium Zinc Lithium Tungsten Sodalime Phosphate Glasses (35-x)P₂O₅: 10ZnO: 10Li₂O: 10WO₃: 10Na₂O: 15CaO: 10Y₂O₃:xHo₂O₃ (where x=1, 1.5, 2 mol %) have been prepared by melt-quenching method. The amorphous nature of the prepared glass samples was confirmed by X-ray diffraction. Optical absorption, Excitation and fluorescence spectra were recorded at room temperature for all glass samples. Judd-Ofelt intensity parameters Ω_λ ($\lambda=2, 4$ and 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 (A), branching ratio (β), radiative life time (τ_R) and stimulated emission cross-section (σ_p) of various emission lines have been evaluated.

KEYWORDS: YZLTSLP Glasses, Optical Properties, Luminescence Properties, Radiative Properties.

I. INTRODUCTION

Rare earth glasses have attracted much attention due to they have potential applications in many fields, such as waveguide laser, electro-luminescent devices, memory devices optical fibers, sensors, glass lasers, optical fiber amplifiers, up-conversion lasers and optical fiber amplifiers [1–5]. Among different glasses phosphate glasses have unique properties. They have high refractive index, low dispersion rates, high transparency with good chemical and thermal stability [6-8]. The Luminescence of phosphate glasses is also compressed because of their large phonon energy. Phosphate glasses can be a good matrix for fiber lasers because they exhibit good thermal property. They have some unique physical, optical and spectral properties better than other glasses [9-12]. The addition of network modifier (NWF) Li₂O is to improve both electrical and mechanical properties of such glasses. Zinc oxide is added in the glass matrix to increase glass forming ability and to ensure low rates of crystallization in the glass system [13-15]. Among active rare-earth ions Ho³⁺ exhibits high solubility in ceramic glasses, which also possess excellent Thermal, physical and optical properties [16,17].

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, Excitation and fluorescence spectra of Ho³⁺ doped ytterbium zinc lithium tungsten sodalime phosphate 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 (β), radiative life time (τ_R) and stimulated emission cross section (σ_p) are evaluated using J.O. intensity parameters (Ω_λ , $\lambda=2, 4$ and 6).

II. EXPERIMENTAL TECHNIQUES

Preparation of glasses

The following Ho³⁺doped borate glass samples (35-x)P₂O₅: 10ZnO: 10Li₂O: 10WO₃: 10Na₂O: 15CaO: 10Y₂O₃:xHo₂O₃. (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 P₂O₅, ZnO, Li₂O, WO₃, Na₂O, CaO, Y₂O₃ and Ho₂O₃. They were thoroughly mixed by using an agate pestle mortar. then melted at 1060°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°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**.

Table1. Chemical composition of the glasses

Sample	Glass composition (mol %)
YZLTSLP (UD)	35P ₂ O ₅ : 10ZnO: 10Li ₂ O: 10WO ₃ : 10Na ₂ O: 15CaO: 10Y ₂ O ₃ :15B ₂ O ₃
YZLTSLP HO(1.0)	34P ₂ O ₅ : 10ZnO: 10Li ₂ O: 10WO ₃ : 10Na ₂ O: 15CaO: 10Y ₂ O ₃ :1H ₂ O ₃
YZLTSLP HO(1.5)	33.5P ₂ O ₅ : 10ZnO: 10Li ₂ O: 10WO ₃ : 10Na ₂ O: 15CaO: 10Y ₂ O ₃ :1.5 H ₂ O ₃
YZLTSLP HO(2.0)	33P ₂ O ₅ : 10ZnO: 10Li ₂ O: 10WO ₃ : 10Na ₂ O: 15CaO: 10Y ₂ O ₃ :2 H ₂ O ₃

YZLTSLP (UD) -Represents undoped Ytterbium Zinc Lithium Tungsten Sodalime Phosphate glass specimen.

YZLTSLP (HO)-Represents Ho³⁺ doped Ytterbium Zinc Lithium Tungsten Sodalime Phosphate glass specimens.

III. THEORY

3.1 Oscillator Strength

The intensity of spectral lines are expressed in terms of oscillator strengths using the relation [18].

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

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 [19], using the modified relation:

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

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

3.2. Judd-Ofelt Intensity Parameters

According to Judd [20] and Ofelt [21] 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:

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

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

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

$\lambda = 2, 4, 6$

In the above equation m is the mass of an electron, c is the velocity of light, ν 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.

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:

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

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

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

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

where, the sum is over all terminal manifolds.

The radiative life time is given by

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

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 [22, 23]. 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 value of bonding parameter ($b^{1/2}$) is 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 sample contain – P₂O₅ 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.

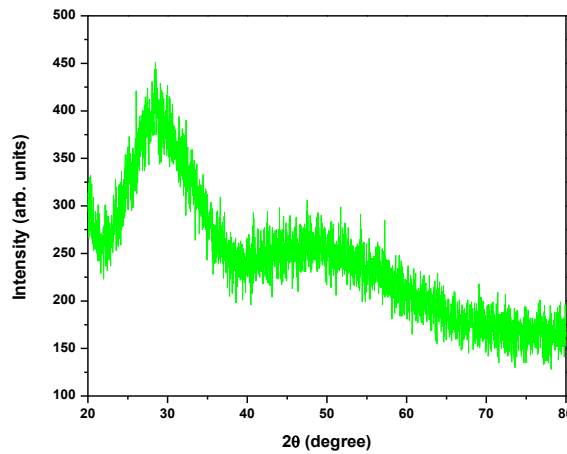


Fig. 1: X-ray diffraction pattern of YZLTSLP HO (1.0) glass.

4.2 Absorption Spectrum

The absorption spectra of Ho³⁺doped YZLTSLP HO(1.0) glass specimen has been presented in Figure 2 in terms of optical density versus wavelength. Twelve absorption bands have been observed from the ground state ⁵I₈ to excited states ⁵I₅, ⁵I₄, ⁵F₅, ⁵F₄, ⁵F₃, ³K₈, ⁵G₆, (⁵G, ³G)₅, ⁵G₄, ⁵G₂, ⁵G₃, and ³F₄ for Ho³⁺ doped YZLTSLP HO(1.0) glass.

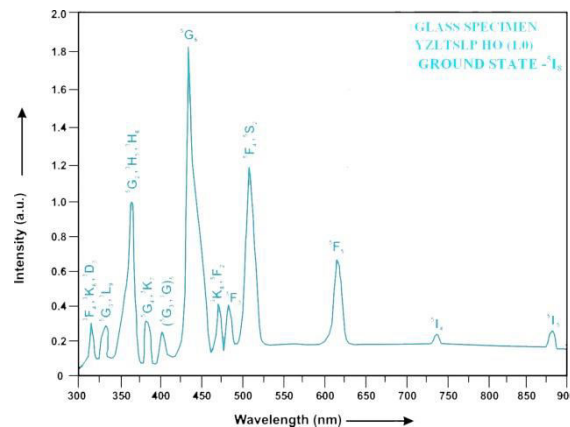


Fig. 2: Absorption spectrum of YZLTSLP HO (1.0) glass.

The experimental and calculated oscillator strength for Ho³⁺ ions in YZLTSLP glasses are given in Table 2.

Table 2: Measured and calculated oscillator strength ($P_m \times 10^{+6}$) of Ho^{3+} ions in YZLTSLP glasses.

Energy level from $^5\text{I}_8$	Glass YZLTSLP HO(1.0)		Glass YZLTSLP HO(1.5)		Glass YZLTSLP HO(2.0)	
	$P_{\text{exp.}}$	$P_{\text{cal.}}$	$P_{\text{exp.}}$	$P_{\text{cal.}}$	$P_{\text{exp.}}$	$P_{\text{cal.}}$
$^5\text{I}_5$	0.46	0.24	0.43	0.25	0.39	0.24
$^5\text{I}_4$	0.05	0.02	0.04	0.02	0.03	0.02
$^5\text{F}_5$	3.68	2.81	3.65	2.79	3.61	2.76
$^5\text{F}_4$	4.72	4.36	4.69	4.33	4.65	4.29
$^5\text{F}_3$	1.55	2.43	1.52	2.41	1.48	2.39
$^3\text{K}_8$	1.48	1.98	1.45	1.96	1.41	1.93
$^5\text{G}_6$	24.86	24.84	23.72	23.72	22.65	22.67
$(^5\text{G}_3, ^3\text{G})_5$	3.82	1.70	3.78	1.68	3.75	1.66
$^5\text{G}_4$	0.07	0.61	0.06	0.60	0.05	0.59
$^5\text{G}_2$	5.86	5.31	5.83	5.11	5.79	4.91
$^5\text{G}_3$	1.53	1.39	1.50	1.37	1.46	1.34
$^3\text{F}_4$	1.44	4.18	1.41	4.13	1.38	4.09
r.m.s. deviation	± 1.1018		± 1.1030		± 1.1077	

Computed values of F_2 , Lande' parameter (ξ_{4f}), Nephelauxetic ratio (β') and bonding parameter ($b^{1/2}$) for Ho^{3+} ions in YZLTSLP glass specimen are given in Table 3.

Table 3: F_2 , ξ_{4f} , β' and $b^{1/2}$ parameters for Holmium doped glass specimen.

Glass Specimen	F_2	ξ_{4f}	β'	$b^{1/2}$
Ho^{3+}	358.82	1258.16	0.9337	0.1821

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

Table 4: Judd-Ofelt intensity parameters for Ho^{3+} doped YZLTSLP glass specimens.

Glass Specimen	$\Omega_2(\text{pm}^2)$	$\Omega_4(\text{pm}^2)$	$\Omega_6(\text{pm}^2)$	Ω_4 / Ω_6	Ref.
YZLTSLP HO (1.0)	5.817	1.302	2.140	0.6084	P.W.
YZLTSLP HO (1.5)	5.506	1.282	2.125	0.6033	P.W.
YZLTSLP HO (2.0)	5.212	1.269	2.104	0.6031	P.W.
LBTAf (TB)	13.230	1.230	2.960	0.4155	[24].
ZLLCBP (HO)	5.690	1.342	2.195	0.6114	[25].

4.3 Excitation Spectrum

The Excitation spectrum of YZLTSLP HO(1.0) glass has been presented in Figure 3 in terms of Excitation Intensity versus wavelength. The excitation spectrum was recorded in the spectral region 325–525 nm fluorescence at 545nm having different excitation band centered at 349,419, 452, 473 and 486 nm are attributed to the $^5\text{G}_3$, $(^5\text{G}, ^3\text{G})_5$, $^5\text{G}_6$, $^3\text{K}_8$ and $^5\text{F}_3$ transitions, respectively. The highest absorption level is $^5\text{G}_6$ and is at 452nm. So this is to be chosen for excitation wavelength.

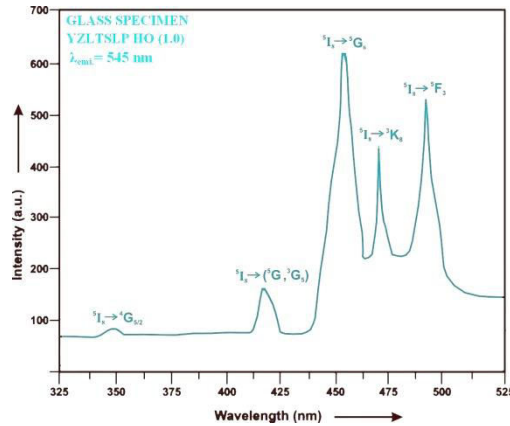


Fig. 3: Excitation spectrum of YZLTSLP HO (1.0) glass.

4.4 Fluorescence Spectrum

The fluorescence spectrum of Ho³⁺ doped in ytterbium zinc lithium tungsten sodalime phosphate glass is shown in Figure 4. There are eleven broad bands observed in the Fluorescence spectrum of Ho³⁺ doped ytterbium zinc lithium tungsten sodalime phosphate glass. The wavelengths of these bands along with their assignments are given in Table 5. The peak with maximum emission intensity appears at 2035 nm and corresponds to the (⁵I₇→⁵I₈) transition.

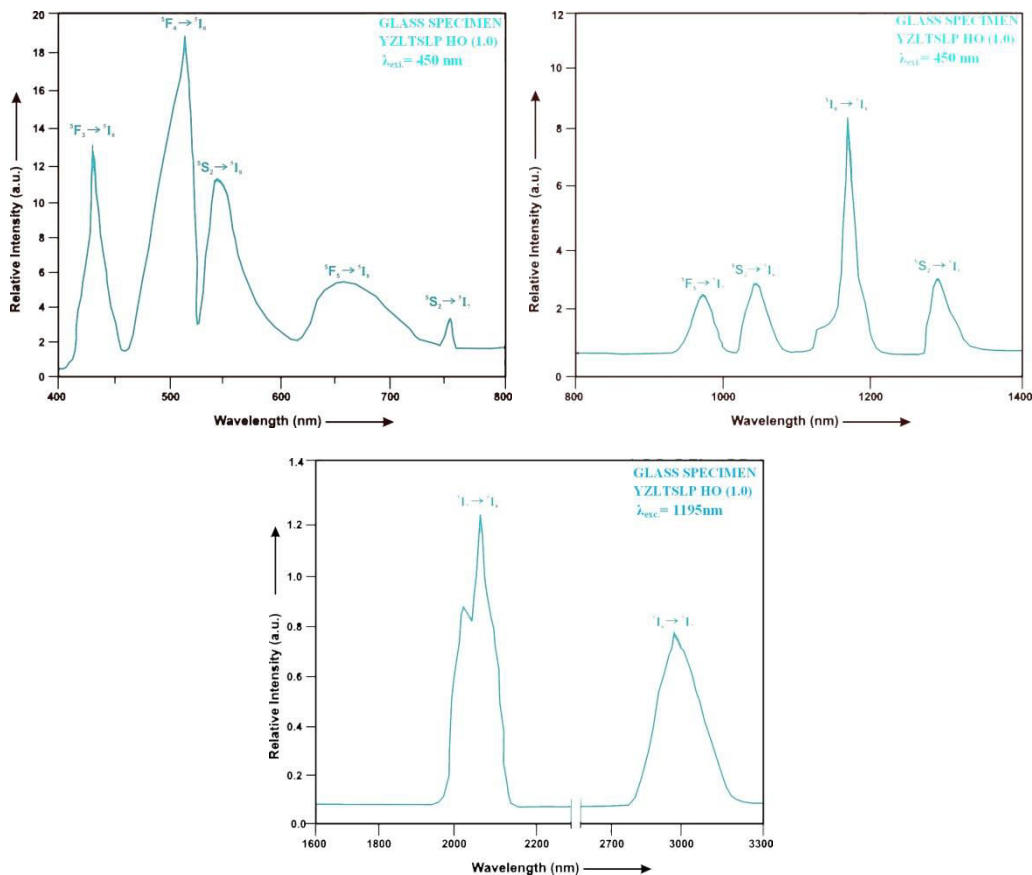


Fig. 4: Fluorescence spectrum of YZLTSLP HO (1.0) glass.

Table5: Emission peak wave lengths (λ_p),radiative transition probability (A_{rad}),branching ratio (β),stimulated emission cross-section(σ_p) and radiative life time(τ_R) for various transitions in Ho³⁺ doped YZLTSLP glasses.

Transition	YZLTSLP HO (1.0)					YZLTSLP HO (1.5)				YZLTSLP HO (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)
⁵ F ₃ → ⁵ I ₈	435	426	0.2	0.51	582 9.2 3	440	0.2	0.5	565 2.1 8	436	0.2	0.57	569 8.19
⁵ F ₄ → ⁵ I ₈	501	676	0.3	1.11		697	0.3	1.2		691	0.3	1.18	
⁵ S ₂ → ⁵ I ₈	555	177	0.1	0.40		183	0.1	0.4		182	0.1	0.41	
⁵ F ₅ → ⁵ I ₈	652	192	0.1	0.67		198	0.1	0.7		196	0.1	0.70	
⁵ S ₂ → ⁵ I ₇	761	134	0.0	1.00		139	0.0	1.1		138	0.0	1.07	
⁵ F ₅ → ⁵ I ₇	995	448.	0.0	1.07		459.	0.0	1.1		454	0.0	1.13	
⁵ I ₆ → ⁵ I ₈	1032	206.	0.0	0.62		213.	0.0	0.6		211	0.0	0.66	
⁵ S ₂ → ⁵ I ₅	1195	236.	0.0	1.10		243.	0.0	1.1		241	0.0	1.14	
⁵ S ₂ → ⁵ I ₆	1310	6.30	0.0	0.53		65.1	0.0	0.6		64.	0.0	0.59	
⁵ I ₇ → ⁵ I ₈	2035	9.39	0.0	4.13		96.6	0.0	4.5		95.	0.0	4.45	
⁵ I ₆ → ⁵ I ₇	2925	2.39	0.0	3.71	24.5	0.0	3.8	24.	0.0	3.72			

V. CONCLUSION

In the present study, the glass samples of composition (35-x)P₂O₅: 10ZnO: 10Li₂O: 10WO₃: 10Na₂O: 15CaO: 10Y₂O₃:xHo₂O₃. (where x =1, 1.5and 2mol %) have been prepared by melt-quenching method. The value of stimulated emission cross-section (σ_p) is found to be maximum for the transition (⁵I₇→⁵I₈) for glass YZLTSLP HO(1.5), suggesting that glass YZLTSLP HO (1.5) is better compared to the other two glass systems YZLTSLP HO(1.0) and YZLTSLP HO (2.0). The large stimulated emission cross section in borate glasses suggests the possibility of utilizing these systems as laser materials and photonic applications.

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