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Spectral and Transmittance properties of Ho^{3+} ions doped Zinc LithiumLead Calcium Borophosphate Glasses

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Abstract
Glass of the system: (40-x)P_2O_5:10ZnO:10Li_2O:10PbO:10CaO:20B_2O_3:xHo_2O_3. (where x=1, 1.5,2 mol %) have been prepared by melt-quenching method.The amorphous nature of the prepared glasssamples was confirmed by X-ray diffraction. Optical absorption,Excitation, fluorescence and Transmittance spectra were recorded at roomtemperature for all glass samples.Judd-Ofeltintensity parameters Ω_λ (λ=2, 4 and6) are evaluated from theintensities of various absorption bands of opticalabsorption spectra. Using these intensity parameters variousradiative properties like spontaneous emission probability (A), branching ratio (β), radiative life time(τ_R) and stimulatedemission cross–section(σ_p)of various emission lines have been evaluated.

Keywords: *ZLLCBP Glasses,Optical Properties, Judd-Ofelt Theory, Transmittance Properties.*

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I. INTRODUCTION

Glass materials doped with rare earth ions are widely used mainly for near-infrared solid-state lasers,sensors, infrared detectors, up-conversion lasers[1-5].Some of the most popular applications of luminescence materials are solid light lasers, white light-emitting diodes (WLEDs) and plasma display panels (PDP)[6-9].Phosphate glasses are known as attractive host materials for rare earth ions with application in photonic, visible and infrared solid state lasers, wave guides and display devices.Phosphate Glasses are both scientifically and technologically important materials becausethey generally offer some unique physical properties better than other Glasses [10-17].Borophosphate glass systems exhibit high refractive indices, high gain density, high solubility and non-linear optical susceptibilities. They present superior properties that include high transparency, low melting point, high thermal stability and high solubility for rare-earth ions and low dispersion [18-21]. Among different rare-earth ions, the Ho^{3+}ions have been identified as the most efficient ion for obtaining the lasing action,frequency up-conversion and optical amplification [22-24].

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,excitation and emission properties of Ho^{3+}doped zinc lithium lead calcium 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 (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).

II. EXPERIMENTAL TECHNIQUES

Preparation of glasses

The following Ho^{3+}doped borophosphateglass samples (40-x)P_2O_5:10ZnO:10Li_2O:10PbO:10CaO:20B_2O_3:xHo_2O_3. (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_2O_5,ZnO, Li_2O, PbO,CaO,$B_2O_3$and Ho_2O_3. They were thoroughly mixed by using an agate pestle mortar. then melted at 1045^0C 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 250^0C 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 %)
ZLLCBP(UD)	$40P_2O_5$:10ZnO:$10Li_2O$:10PbO:10CaO:$20B_2O_3$
ZLLCBP (HO1)	$39P_2O_5$:10ZnO:$10Li_2O$:10PbO:10CaO:$20B_2O_3$:1 Ho_2O_3
ZLLCBP(HO1.5)	$38.5P_2O_5$:10ZnO:$10Li_2O$:10PbO:10CaO:$20B_2O_3$:$1.5Ho_2O_3$
ZLLCBP (HO2)	$38P_2O_5$:10ZnO:$10Li_2O$:10PbO:10CaO:$20B_2O_3$:$2Ho_2O_3$

ZLLCBP (UD) -Represents undopedZinc Lithium Lead Calcium Borophosphate glass specimen.
ZLLCBP(HO)-Represents Ho^{3+}dopedZinc Lithium Lead Calcium Borophosphateglass specimens.

III. THEORY

3.1Oscillator Strength

The intensity of spectral lines are expressed in terms of oscillator strengths using the relation [25].

$$f_{expt.} = 4.318 \times 10^{-9} \int \varepsilon (\nu)\, d\nu \qquad (1)$$

where, ε (*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 [26], using the modified relation:

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

where c is the molar concentration of the absorbing ion per unit volume, I is the optical path length, $\log I_0/I$ is optical density and $\Delta\upsilon_{1/2}$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> level and the terminal J' manifold $|4f^N$ (S',L') J'> is given by:

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

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

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

λ =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 (λ=2,4and 6) are known as Judd-Ofelt intensity.

3.3Radiative 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'> to a final manifold $|4f^N$ (S,L) J >| is given by:

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

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'> to a final many fold $|4f^N$ (S,L)J> is given by

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

S L J
where, the sum is over all terminal manifolds.

The radiative life time is given by

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

S L J

where, the sum is over all possible terminal manifolds.The stimulated emission cross -section for a transition from an initial manifold $|4f^N (S', L') J'\rangle$ 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 [29, 30]. 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

4.1XRD 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.

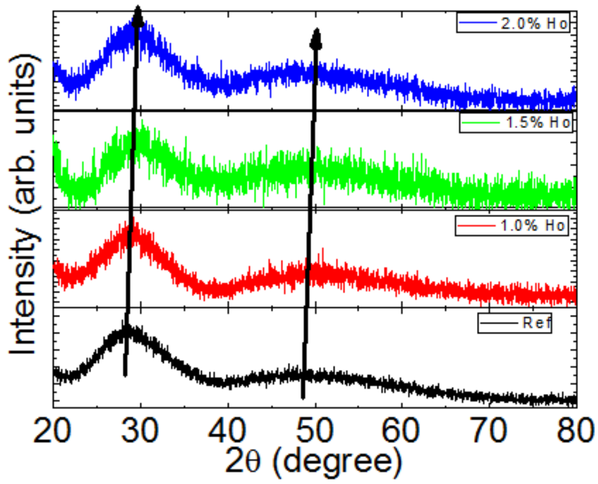

Fig. 1 X-ray diffraction pattern of P_2O_5:ZnO:Li_2O:PbO:CaO:B_2O_3:Ho_2O_3

4.2Transmittance Spectrum

The Transmittance spectrum of Ho^{3+}doped in zinc lithium lead calcium borophosphateglass is shown in Figure 2.

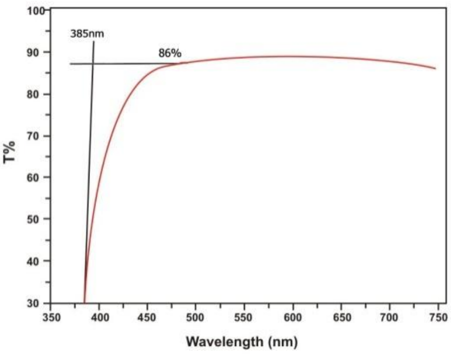

Fig. (2) Transmittance spectrum of Ho^{3+}doped ZLLCBPglasses.

4.3 Absorption Spectrum

The absorption spectra of Ho^{3+}doped ZLLCBP glass specimens have been presented in Figure 3 in terms of optical density versus wavelength. Twelve absorption bands have been observed from the ground state $^5I_8$to excited states $^5I_5$, $^5I_4$, $^5F_5$, $^5F_4$, $^5F_3$, $^3K_8$, $^5G_6$, $(^5G,^3G)_5$, $^5G_4$, $^5G_2$, $^5G_3$, and$^3F_4$for Ho^{3+} doped ZLLCBP glasses.

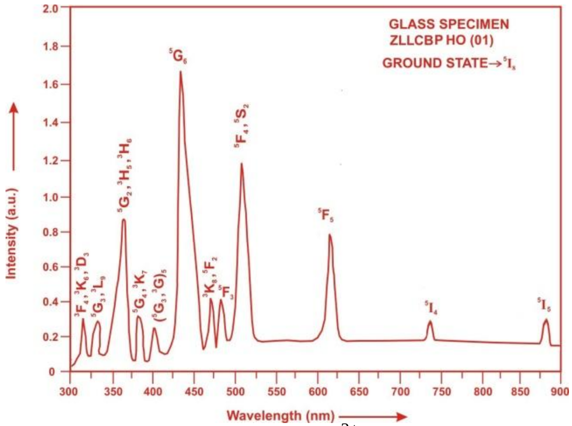

Fig. (3) Absorption spectrum of Ho^{3+}doped ZLLCBPglasses.

The experimental and calculated oscillator strength for Ho^{3+}ions in ZLLCBP glasses are given in **Table 2.**

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

Energy level from $^5I_8$	**Glass ZLLCBP(HO01)**		**Glass ZLLCBP(HO1.5)**		**Glass ZLLCBP(HO02)**	
	P_{exp}.	P_{cal}.	P_{exp}.	P_{cal}.	P_{exp}.	P_{cal}.
$^5I_5$	0.43	0.24	0.38	0.24	0.32	0.24
$^5I_4$	0.05	0.02	0.04	0.02	0.02	0.02
$^5F_5$	3.68	2.81	3.62	2.80	3.58	2.77
$^5F_4$	4.69	4.36	4.64	4.34	4.57	4.30
$^5F_3$	1.56	2.42	1.52	2.41	1.48	2.39
$^3K_8$	1.42	1.97	1.38	1.96	1.32	1.93
$^5G_6$	24.25	24.25	23.98	24.01	22.86	22.94
$(^5G,^3G)_5$	3.85	1.71	3.79	1.71	3.68	1.67
$^5G_4$	0.07	0.61	0.05	0.60	0.04	0.59
$^5G_2$	5.68	5.21	5.61	5.16	5.57	4.96
$^5G_3$	1.48	1.38	1.42	1.37	1.38	1.35
$^3F_4$	1.36	4.19	1.34	4.18	1.29	4.11
r.m.s. deviation	±1.1204		±1.1115		±1.1027	

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

Table 3: F_2,$\xi_{4f,}$ 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

In theZinc Lithium Lead Calcium Borophosphateglasses (ZLLCBP)$\Omega_{2,}$ Ω_4 and $\Omega_6$parameters decrease with the increase of x from 1 to 2 mol%. The order ofmagnitude of Judd-Ofelt intensity parameters is $\Omega_2>\Omega_6>\Omega_4$for all the glass specimens. The high valuesobtained for $\Omega_2$in all glasses indicate that the Ho^{3+} ion is subjected to higher covalency with low symmetry. The spectroscopic quality factor (Ω_4/Ω_6) related with the rigidity of the glass system has been found to lie between 0.6072 and 0.6152 in the present glasses.
The values of Judd-Ofelt intensity parameters are given in **Table 4.**

Table4:Judd-Ofelt intensity parameters for Ho^{3+} doped ZLLCBP **glass specimens.**

Glass Specimen	**$\Omega_2(pm^2)$**	**$\Omega_4(pm^2)$**	**$\Omega_6(pm^2)$**	**Ω_4/Ω_6**	**Ref.**
ZLLCBP (HO01)	5.840	1.353	2.212	0.6117	P.W.
ZLLCBP (HO1.5)	5.768	1.351	2.196	0.6152	P.W.
ZLLCBP (HO02)	5.462	1.322	2.177	0.6072	P.W.
PHOSPHATE	6.28	1.03	1.39	0.7410	[31]
LBTAF	13.23	1.23	2.96	0.4155	[32]

4.4Excitation Spectrum

The Excitation spectra of Ho^{3+}doped ZLLCBP glass specimens have been presented in Figure 4 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 at349,419, 452, 473and 486 nmare attributed to the $^5G_3$,(^5G, $^3G)_5$,$^5G_6$,$^3K_8$and$^5F_3$ transitions, respectively. The highest absorption level is $^5G_6$ and is at 450nm.So this is to be chosen for excitation wavelength.

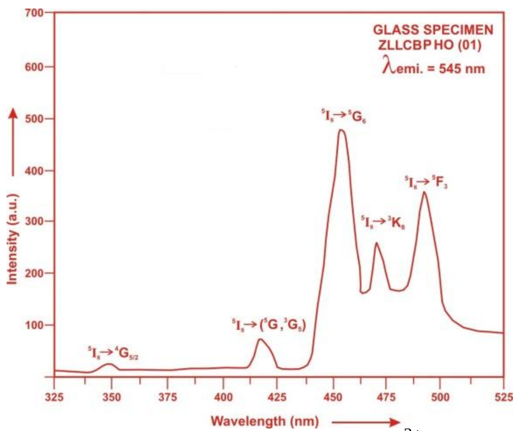

Fig. (4) Excitation spectrum of doped with Ho^{3+}ZLLCBP glasses.

4.5 Fluorescence Spectrum

The fluorescence spectrum of Ho^{3+}doped in zinc lithium lead calcium borophosphateglass is shown in Figure 5. There are nine broad bands observed in the Fluorescence spectrum ofHo^{3+}dopedzinc lithium lead calcium borophosphateglass. The wavelengths of these bands along with their assignments are given in Table 5. The peak with maximum emission intensity appears at 555nm and corresponds to the ($^5F_4 \rightarrow {}^5I_8$) transition.

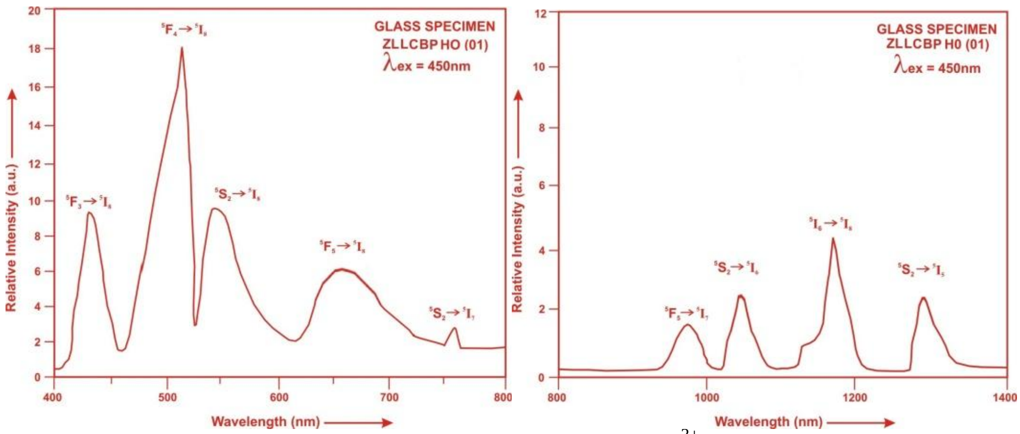

Fig. (5). Fluorescence spectrum of doped with Ho^{3+}ZLLCBPglasses

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 Ho^{3+} doped ZLLCBP glasses.

Transition		ZLLCBP (HO 01)				ZLLCBP (HO 1.5)				ZLLCBP (HO 02)			
	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)
$^5F_3 \rightarrow ^5I_8$	435	4197.35	0.2501	0.598		4175.28	0.2499	0.584		4158.50	0.2504	0.566	
$^5F_4 \rightarrow ^5I_8$	501	6667.63	0.3973	1.239		6638.44	0.3973	1.217		6595.58	0.3971	1.184	
$^5S_2 \rightarrow ^5I_8$	555	1752.29	0.1044	0.433		1744.02	0.1044	0.426		1735.81	0.1045	0.416	
$^5F_5 \rightarrow ^5I_8$	652	1897.58	0.1131	0.745		1891.48	0.1132	0.732		1875.78	0.1129	0.717	
$^5S_2 \rightarrow ^5I_7$	761	1330.06	0.0792	1.137	5958.01	1323.06	0.0792	1.123	5985.33	1316.84	0.0793	1.104	6020.69
$^5F_5 \rightarrow ^5I_7$	995	440.55	0.0262	1.205		438.91	0.0263	1.185		433.58	0.0261	1.155	
$^5I_6 \rightarrow ^5I_8$	1032	203.89	0.0121	0.705		203.06	0.0122	0.692		201.86	0.0122	0.677	
$^5S_2 \rightarrow ^5I_5$	1195	232.65	0.0139	1.222		231.44	0.0139	1.201		229.94	0.0138	1.172	
$^5S_2 \rightarrow ^5I_6$	1310	62.14	0.0037	0.631		61.83	0.0037	0.618		61.50	0.0037	0.600	

V. CONCLUSION

In the present study, the glass samples of composition (40-x)P_2O_5:10ZnO:10Li_2O:10PbO:10CaO:20B_2O_3:xHo_2O_3. (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 ($^5F_4 \rightarrow ^5I_8$) for glass ZLLCBP(HO 01), suggesting that glass ZLLCBP (HO 01) is better compared to the other two glass systems ZLLCBP (HO1.5) and ZLLCBP(HO02).On the basis of spectrophotometric, transmittance reaches about 86% for all phosphate glasses doped with Ho^{3+} ions.

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