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Spectroscopic and Photoluminescence Properties of Ho^{3+}doped Borate Glasses with NIR Light Emission Applications

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Abstract
Glasses samples containing Ho^{3+} in Zinc Lithium Alumino Cadmium Sodalime Bismuth Borate Glasses (25-x)Bi_2O_3: 10ZnO: 10Li_2O: 10Al_2O_3: 10CdO: 10CaO: 10Na_2O :15B_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 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 (λ=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: *ZLACSLBB Glasses, Optical Properties, Judd-Ofelt Theory, Radiative Properties.*

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I. Introduction

Rare earth glasses have attracted much attention, because they have potential applications in many fields, such as electro-luminescent devices, memory devices optical fibers, sensors, glass lasers, optical fiber amplifiers, up-conversion lasers, waveguide laser and optical fiber amplifiers [1–5].Among different glasses borate glasses have unique properties. They have high transparency, high refractive index and low dispersion rates with good chemical and thermal stability [6-8].The up-conversion of borate glasses is also compressed because of their relatively large phonon energy. Borate glasses can be a good matrix for fiber lasers because they exhibit good mechanical property [9-12]. Borate glasses are both scientifically and technologically important materials because they generally offer some unique physical, optical and spectral properties better than other glasses [13-15]. The addition of network modifier (NWF) Li_2O 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 [16-18]. Among active rare-earth ions Ho^{3+} exhibits high solubility in ceramic glasses, which also possess excellent Thermal, physical and optical properties [19, 20].

Recently borate based glasses have a wide range of potential applications in optical data transmission, laser technologies, detection, sensing and photonic applications [21-23].

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, fluorescence and FTIR spectra of Ho^{3+} doped zinc lithium alumino cadmium sodalime bismuth borate 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, λ=2,4 and 6).

II. Experimental Techniques

Preparation of glasses

The following Ho^{3+}doped borate glass samples (25-x)Bi_2O_3: 10ZnO: 10Li_2O: 10Al_2O_3: 10CdO: 10CaO: 10Na_2O :15B_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 Bi_2O_3, ZnO, Li_2O, Al_2O_3, CdO, CaO,Na_2O, B_2O_3 and Ho_2O_3. They were thoroughly mixed by using an agate pestle mortar. then melted at 960^0C 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 %)
ZLACSLBB (UD)	$25Bi_2O_3$: 10ZnO: $10Li_2O$: $10Al_2O_3$: 10CdO: 10CaO: $10Na_2O$:$15B_2O_3$
ZLACSLBB (HO1)	$24Bi_2O_3$: 10ZnO: $10Li_2O$: $10Al_2O_3$: 10CdO: 10CaO: $10Na_2O$:$15B_2O_3$:$1Ho_2O_3$
ZLACSLBB (HO1.5)	$23.5Bi_2O_3$: 10ZnO: $10Li_2O$: $10Al_2O_3$: 10CdO: 10CaO: $10Na_2O$:$15B_2O_3$:1.5 Ho_2O_3
ZLACSLBB (HO2)	$23Bi_2O_3$: 10ZnO: $10Li_2O$: $10Al_2O_3$: 10CdO: 10CaO: $10Na_2O$:$15B_2O_3$:2 Ho_2O_3

ZLACSLBB (UD) -Represents undoped Zinc Lithium Alumino Cadmium Sodalime Bismuth Borate glass specimen.
ZLACSLBB (HO)-Represents Ho^{3+} doped Zinc Lithium Alumino Cadmium Sodalime Bismuth Borate glass specimens.

III. Theory

3.1 Oscillator Strength

The intensity of spectral lines are expressed in terms of oscillator strengths using the relation [24].

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

where, ε (*ν)* 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 [25], using the modified relation:

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

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 [26] and Ofelt [27] 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:

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

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

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

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 (λ=2,4and 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:

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

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

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

where, the sum is over all terminal manifolds.

The radiative life time is given by

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

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

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

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 [28, 29]. The Nephelauxetic Ratio is given by

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

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

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

IV. Result and Discussion

4.1 XRD Measurement

Figure 1 presents the XRD pattern of the sample contain – B_2O_2 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.

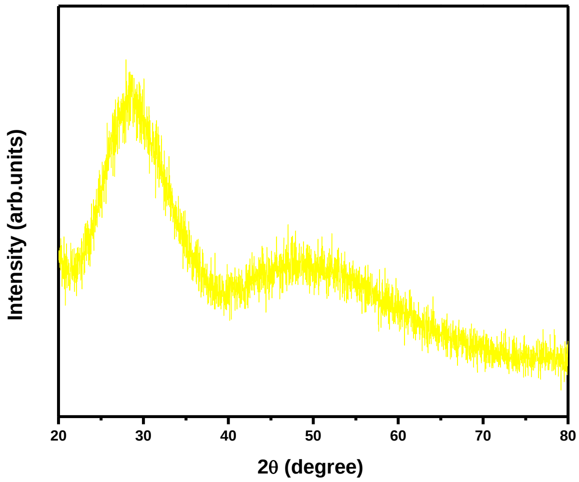

Fig. 1: X-ray diffraction pattern of ZLACSLBB HO (01) glass.

4.2 Absorption Spectrum

The absorption spectra of Ho^{3+}doped ZLACSLBB glass specimens have been presented in Figure 2 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 ZLACSLBB glasses.

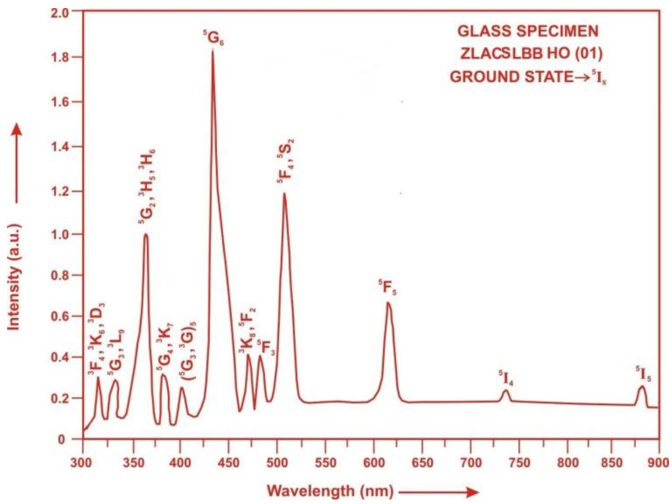

Fig. 2: Absorption spectrum of ZLACSLBB HO (01) glass.

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

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

Energy level from $^5I_8$	**Glass ZLACSLBB (HO01)**		**Glass ZLACSLBB (HO1.5)**		**Glass ZLACSLBB (HO02)**	
	$P_{exp.}$	$P_{cal.}$	$P_{exp.}$	$P_{cal.}$	$P_{exp.}$	$P_{cal.}$
$^5I_5$	0.46	0.24	0.43	0.25	0.40	0.24
$^5I_4$	0.06	0.02	0.05	0.02	0.04	0.02
$^5F_5$	3.66	2.81	3.62	2.77	3.58	2.75
$^5F_4$	4.7	4.36	4.65	4.30	4.61	4.27
$^5F_3$	1.55	2.42	1.51	2.39	1.47	2.37
$^3K_8$	1.45	1.99	1.41	1.95	1.35	1.92
$^5G_6$	25.75	25.73	24.35	24.35	23.42	23.44
$(^5G,^3G)_5$	3.88	1.72	3.84	1.69	3.79	1.67
$^5G_4$	0.08	0.62	0.07	0.60	0.05	0.59
$^5G_2$	5.72	5.47	5.67	5.21	5.62	5.04
$^5G_3$	1.48	1.40	1.45	1.37	1.40	1.35
$^3F_4$	1.36	4.22	1.32	4.14	1.28	4.10
r.m.s. deviation	±1.1221		±1.1193		±1.1196	

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

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

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

Glass Specimen	**Ω_2(pm^2)**	**Ω_4(pm^2)**	**Ω_6(pm^2)**	**Ω_4 /Ω_6**	**Ref.**
ZLACSLBB (HO 01)	6.290	1.368	2.213	0.618	P.W.
ZLACSLBB (HO1.5)	5.893	1.342	2.184	0.614	P.W.
ZLACSLBB (HO 02)	5.633	1.326	2.165	0.612	P.W.
ZPCPNP (HO)	5.998	1.329	2.201	0.604	[30]
BAP (HO)	5.77	0.32	0.58	0.552	[31]
LCASO (HO)	5.606	1.175	1.052	1.117	[32]

4.3 Excitation Spectrum

The Excitation spectrum of ZLACSLBB (HO 01) 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, 473and 486 nm are 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 452nm.So this is to be chosen for excitation wavelength.

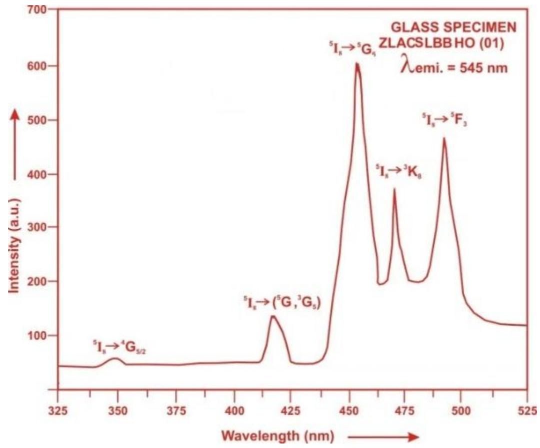

Fig. 3: Excitation spectrum of ZLACSLBB HO (01) glass.

4.4 Fluorescence Spectrum

The fluorescence spectrum of Ho^{3+}doped in zinc lithium alumino cadmium sodalime bismuth borate glass is shown in Figure 4. There are eleven broad bands observed in the Fluorescence spectrum of Ho^{3+}doped zinc lithium alumino cadmium sodalime bismuth borate 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 ($^5I_7 \rightarrow {}^5I_8$) transition.

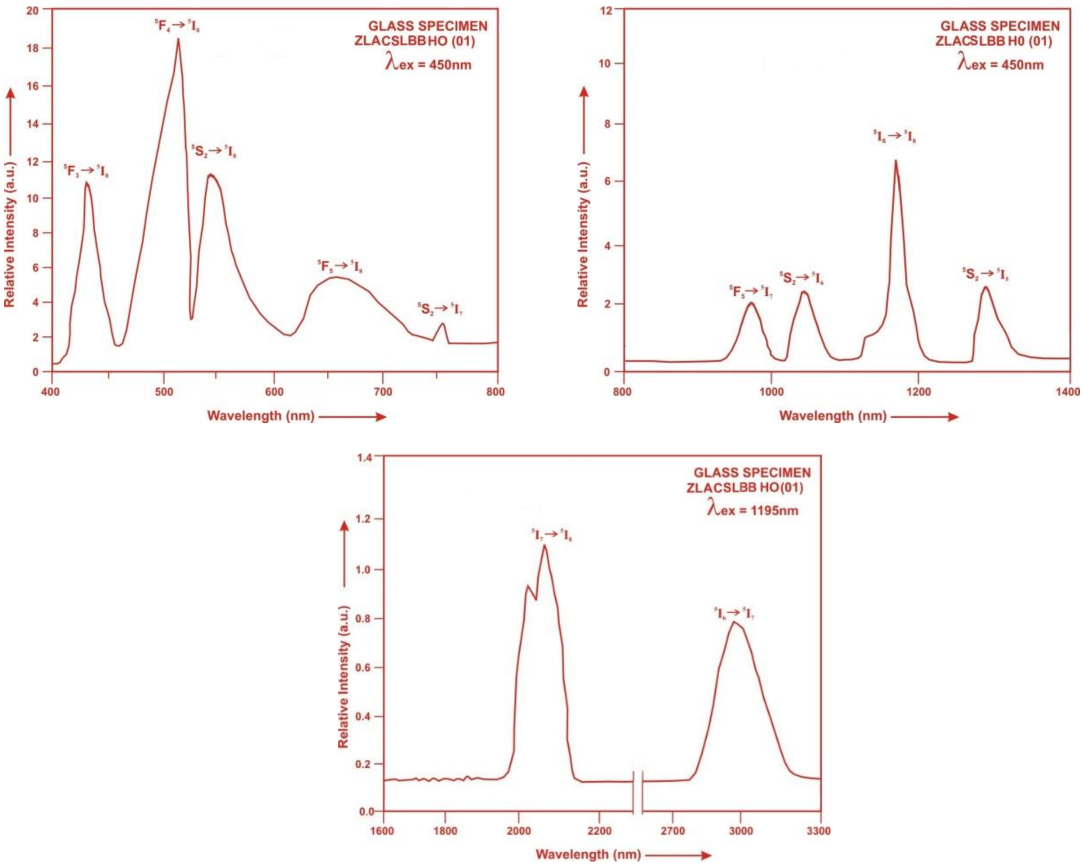

Fig. 4: Fluorescence spectrum of ZLACSLBB HO (01) glass.

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 Ho^{3+} doped ZLACSLBB glasses.

Transition		ZLACSLBB (HO 01)				ZLACSLBB (HO 1.5)				ZLACSLBB (HO 02)			
	λ_{max} (nm)	$A_{rad}(s^{-1})$	β	σ_p (10^{-20} cm^2)	$\tau_R(\mu s)$	$A_{rad}(s^{-1})$	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	4177.15	0.2481	0.612		4130.62	0.2483	0.589		4102.83	0.2484	0.570	
$^5F_4\rightarrow{}^5I_8$	501	6641.18	0.3945	1.238		6561.83	0.3945	1.207		6514.81	0.3945	1.176	
$^5S_2\rightarrow{}^5I_8$	555	1743.60	0.1036	0.442		1724.17	0.1037	0.430		1712.58	0.1037	0.421	
$^5F_5\rightarrow{}^5I_8$	652	1894.75	0.1126	0.746		1870.11	0.1124	0.725		1855.63	0.1124	0.709	
$^5S_2\rightarrow{}^5I_7$	761	1322.74	0.0786	1.129		1308.01	0.0786	1.097	6011.68	1299.21	0.0787	1.071	6054.93
$^5F_5\rightarrow{}^5I_7$	995	442.12	0.0263	1.228	5940.31	434.65	0.0261	1.185		430.12	0.0260	1.158	
$^5I_6\rightarrow{}^5I_8$	1032	203.10	0.0121	0.704		200.72	0.0121	0.686		199.31	0.0121	0.668	
$^5S_2\rightarrow{}^5I_5$	1195	232.01	0.0138	1.230		228.99	0.0138	1.193		227.17	0.0138	1.163	
$^5S_2\rightarrow{}^5I_6$	1310	61.82	0.0037	0.630		61.12	0.0037	0.613		60.70	0.0037	0.595	
$^5I_7\rightarrow{}^5I_8$	2035	92.17	0.0055	4.769		90.93	0.0055	4.610		90.17	0.0055	4.509	
$^5I_6\rightarrow{}^5I_7$	2925	23.51	0.0014	3.969		23.14	0.0014	3.846		22.91	0.0014	3.714	

V. Conclusion

In the present study, the glass samples of composition (25-x)Bi_2O_3: 10ZnO: 10Li_2O: 10Al_2O_3: 10CdO: 10CaO: 10Na_2O :15B_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 ($^5I_7\rightarrow{}^5I_8$) for glass ZLACSLBB (HO 01), suggesting that glass ZLACSLBB (HO 01) is better compared to the other two glass systems ZLACSLBB (HO1.5) and ZLACSLBB (HO 02). The large stimulated emission cross section in borate glasses suggests the possibility of utilizing these systems as laser materials. The results show that the Ho^{3+} doped bismuth borate glasses could be potential candidates for NIR light emission applications.

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