

Spectral, FTIR, Thermal and Interaction parameters of Ho^{3+} doped Bismuth Borate Glasses with Large Balaji-Shankar Relation

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Abstract

Zinc lithium niobium barium bismuth borate glasses containing Ho^{3+} in $(45-x) \text{Bi}_2\text{O}_3: 10\text{ZnO}:10\text{Li}_2\text{O}:10\text{Nb}_2\text{O}_5: 10\text{BaO}:15\text{B}_2\text{O}_3: x\text{Ho}_2\text{O}_3$. X-ray diffraction results show the glassy nature of the glass samples. The FTIR spectra confirmed that BO_3 and BO_4 are structural units. DTA curve was analysed to evaluate the glass transition temperature, crystallization and melting temperature. The absorption spectra of prepared glasses were recorded in the wavelength range 450-980 nm. The absorption spectra have been used to determine the Energy band gap, Urbach energy and Steepness parameter. The optical band gap decrease from 2.23 to 2.19 eV (direct band gap) and 2.13 to 2.06 eV (indirect band gap). Urbach energy increase from 0.4809 to 0.4857 eV with increasing Ho_2O_3 content. The estimated values of the refractive index by Balaji Formula. The resulted indicated that prepared glass system is suitable for laser and optical devices applications.

Keywords: ZLNBBB glasses, Steepness parameter, Refractive index, Thermal parameters.

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I. Introduction

Rare earth doped ceramic glasses have drawn much attention due to their potential applications in optical sensors, luminescence, optical amplifier, waveguide laser, frequency up conversion materials, solid state lasers and electronic devices [1-5]. Among all glasses, borate glasses have received more interest because their high transparency, good mechanical and thermal stabilities, low melting temperature, high refractive index, high gain density and low phonon energy [6-12]. Borate glasses are known for having good thermal and solubility, high chemical durability and high dielectric constant, exceptional linear-nonlinear optical properties, high physical and chemical stability [13-16]. Borate glasses containing ZnO, BaO oxides possess high transparency, lower the melting temperature and low phonon energy besides the Li_2O network modifier present in the glass system show low melting point and good solubility of rare earth ions. The network intermediate (Nb_2O_5) can be added to enhance the mechanical, chemical, electrical and thermal stability of borate glasses. Borate glasses have shown important applications in the area of optoelectronic devices, reflecting windows and ceramic glasses [17-20].

Ceramic glasses based on heavy metal oxides have received increased attention due to their important applications such as optoelectronic devices, reflecting window and development of photonic devices. The batch material should have low melting temperature, good thermal and chemical stability. Ho^{3+} ion doped glasses have wide ranging applications in laser, optical data storage, good thermo-mechanical properties and luminescent devices [21-24].

The present work reports on the preparation and characterization of bismuth borate glasses for lasing materials. I have studied on the absorption, structural and thermal properties of Ho^{3+} doped bismuth borate glasses. The FTIR spectra confirmed BO_3 and BO_4 are structural units present in batch material. Optical energy band gap (direct and indirect), Urbach energy and steepness parameters have also been calculated. The prepared glass samples exhibit high transparency, low interaction parameter, high thermal stability.

II. Theoretical Background

Energy band gap-based refractive index(n_{Eg})

The refractive index on the basis of energy band gap can be calculated using Balaji formula

$$n_{\text{B}} = 8 \left(\frac{E_{\text{g}}}{50} \right)^{\frac{1}{2}} \quad (1)$$

where E_{g} is indirect energy band gap.

III. Experimental Techniques

Glass preparation

The bismuth borate glasses with holmium doped $(45-x)\text{Bi}_2\text{O}_3: 10\text{ZnO}: 10\text{Li}_2\text{O}: 10\text{Nb}_2\text{O}_5: 10\text{BaO}: 15\text{B}_2\text{O}_3: x\text{Ho}_2\text{O}_3$ (where $x=1, 1.5$ and 2%) have been prepared by melt quenching method. The glass composition bismuth oxide (Bi_2O_3), zinc oxide (ZnO), lithium oxide (Li_2O), niobium pentoxide (Nb_2O_5), barium oxide (BaO), boron trioxide (B_2O_3), holmium oxide (Ho_2O_3) with high purity (CDH, 99.99%). All chemical oxides were mixed in an agate pestle mortar. The glass compositions (10g) were melted at 1060°C in electric furnace. For quenching the melt batch materials were quickly poured in to rectangular moulds. After cooling the prepared glass specimens were transferred in furnace at 355°C for 2 hour. Finally cerium oxide (CeO_2) using for fine polished so the prepared bismuth borate glasses were transparent.

Table 1. Chemical Composition ratio for glass specimens.

Glass Samples	Bi_2O_3	ZnO	Li_2O	Nb_2O_5	BaO	B_2O_3	Ho_2O_3	
ZLNBBB HO (1.0)	44	10		10		10	15	1.0
ZLNBBB HO (1.5)	43.5		10	10		10	15	1.5
ZLNBBB HO (2.0)	43	10	10	10	10	15	2.0	

IV. Result and Discussion

4.1 X-ray Diffraction

X-ray diffractometer ($\text{CuK}\alpha$, $1.54\text{\AA}$) were used to record XRD pattern of prepared glass samples. The X-ray diffraction pattern for the glass samples are shown in Fig.1. XRD pattern do not show any sharp peaks, which confirm the amorphous nature of glass specimens.

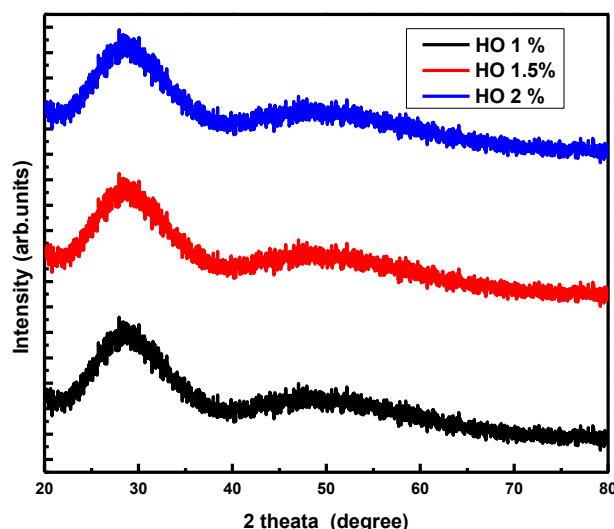


Fig.1: X-ray diffraction pattern of ZLNBBB HO glass samples.

4.2 FTIR Transmission Spectra

The FTIR spectrum in the wave length $4015\text{--}575\text{ cm}^{-1}$ for glass specimens is shown in Fig.2. The possible vibrational bands are depicted in Table 2. The bands nearly 690 cm^{-1} is due to the vibration mode of B-O-B linkage bending in glass network. Vibrations nearly 880 and 1352 cm^{-1} are due to stretching vibrations of the B-O bond of tetrahedral and trigonal units respectively.

Table 2. FTIR bands of ZLNBBB HO glasses.

FTIR bands (cm^{-1})	Assignment
~ 690	Vibrations of B-O-B linkage [25]
~ 880	Stretching vibrations of the B-O bond of tetrahedral units [26]
~ 1352	Stretching vibrations of the B-O bond of trigonal units [27]

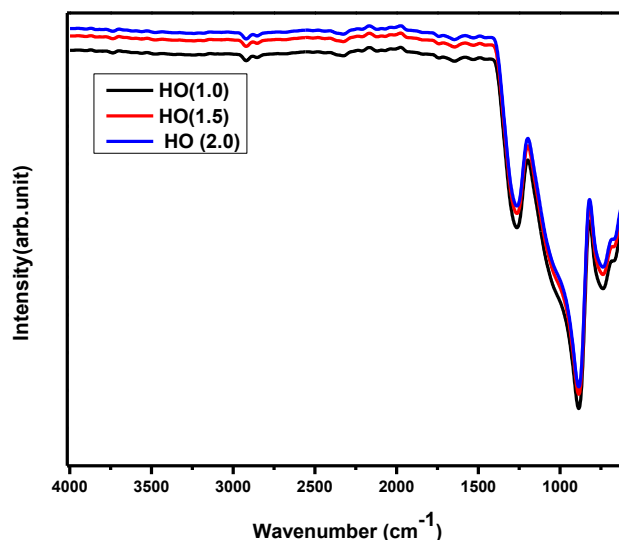


Fig.2: FTIR Spectrum of ZLNBBB HO glasses.

4.3 Thermal Property

Differential thermal analysis checks the heat absorbed by glass samples during heating or cooling. Fig. 2 depicts the DTA thermogram of powdered ZLNBBB HO (1.0) sample. The glass transition temperature (T_g), onset crystallization temperature (T_c), crystallization temperature (T_p), melting temperature (T_m), thermal stability (T_s), Balaji Parameter (B_P), Hurbe's criterion (H_R), Shankar parameter (K_S), reduced glass transition temperature (T_{rg}) and Balaji-Shankar relation were calculated. All the determined thermal parameters are given in table 3.

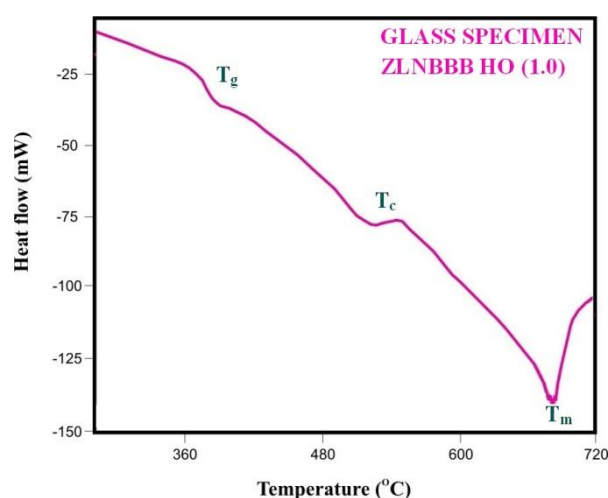


Fig.3: DTA curve of ZLNBBB HO (1.0) glass.

Table 3. Thermal parameters determined from the DTA traces of ZLNBBB HO glasses.

Sample Name	$T_g(^{\circ}\text{C})$	$T_c(^{\circ}\text{C})$	$T_p(^{\circ}\text{C})$	$T_m(^{\circ}\text{C})$	$T_s(^{\circ}\text{C})$	$B_P(^{\circ}\text{C})$	$H_R(^{\circ}\text{C})$	$K_S(^{\circ}\text{C})$	$T_{rg}(^{\circ}\text{C})$	$B_P \times K_S$
ZLNBBB HO (1.0)	375	512	550	686	137	3.605	0.218	34.749	0.547	125.270
ZLNBBB HO (1.5)	380	513	552	688	133	3.410	0.223	33.830	0.552	115.360
ZLNBBB HO (2.0)	385	516	555	691	130	3.250	0.222	33.111	0.557	107.611

The thermal stability of the glass samples can be calculated by difference between onset crystallization temperature and transition temperature [28].

$$\text{Thermal Stability } (T_s) = T_c - T_g \quad (2)$$

Balaji Parameter can be calculated using [28].

$$\text{Balaji Parameter (B}_p\text{)} = [(T_c - T_g)/(T_p - T_c)] \quad (3)$$

Hruby's criterion is calculated using the Hurby's relation [28].

$$\text{Hruby's criterion (H}_R\text{)} = [(T_p - T_c)/(T_m - T_c)] \quad (4)$$

Reduced glass transition temperature is given as [28].

$$\text{Reduced glass transition temperature (T}_{rg}\text{)} = T_g/T_m \quad (5)$$

Shankar Parameter is given as [28].

$$K_s = [(T_m - T_c) (T_c - T_g)/T_m] \quad (6)$$

Balaji-Shankar relation is given by

$$(B_p \times K_s) = [(T_c - T_g)^2 (T_m - T_c) / T_m (T_p - T_c)] \quad (7)$$

4.4 Energy band gap-based refractive index (n_{eg})

The refractive index of prepared glass specimens on the basis of energy band gap are presented in Table 4.

Table 4. The refractive index of prepared glass specimens on the basis of energy band gap	
Glass samples	Refractive index
ZLNBBB HO (1.0)	1.651
ZLNBBB HO (1.5)	1.647
ZLNBBB HO (2.0)	1.624

4.5 Optical properties

4.5.1 Absorption and Transmittance spectra

The absorption bands are centred at 489, 542 and 651, corresponding to the transitions from ground state $^4I_{11/2}$ to excited states 5F_3 , 5F_4 and 5F_5 respectively (Fig.4).

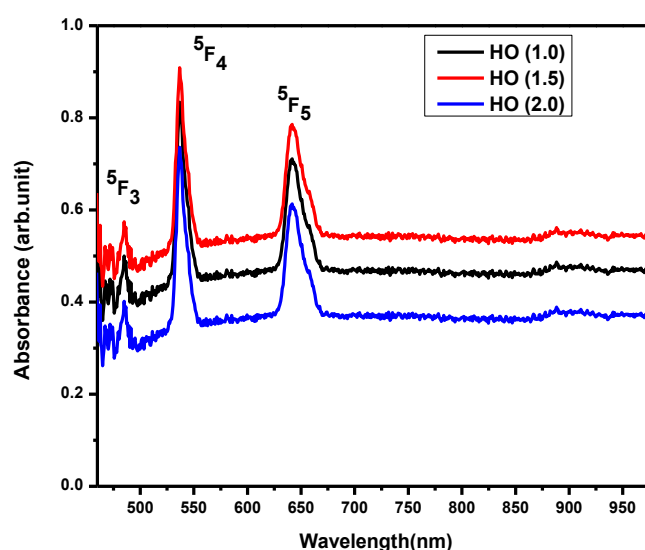


Fig.4: Absorption spectrum of ZLNBBB HO glasses.

4.5.2 Optical energy band gap (direct and indirect), Urbach energy and Steepness parameter

The energy difference between the conduction and valance band is called energy band gap. It can be explain the characteristics and structure properties of prepared glass samples. It can be determined using Davis and Mott equation [29].

$$\alpha = \frac{B(h\nu - E_{opt})^p}{h\nu} \quad (8)$$

where α is the optical absorption coefficient which is depend on the absorbance and thickness. The band tailing parameter is represent by constant B, $h\nu$ represent the photon energy and p is the index number. For direct and indirect transitions the values of p are 1/2 and 2 respectively. The direct and indirect band gap were determined using Tauc's plots,

The Urbach energy (ΔE) can be calculated using the following relation [30].

$$\alpha = \beta \exp\left(\frac{h\nu}{\Delta E}\right) \quad (9)$$

where β is a constant. The values of ΔE can be determined by the reciprocal slope of the Urbach plot, Fig. (5-9). The steepness parameter (S) can be determined using the formula [31].

$$S = \frac{kT}{\Delta E} \quad (10)$$

where k is Boltzmann constant, T is the room temperature and ΔE is the Urbach energy.

The results for direct and indirect optical band gap, Urbach energy and steepness parameter of prepared glass specimens are presented in Table 5.

Table 5. The direct and indirect band gap, Urbach energy and steepness parameter of the prepared glass samples

Glass samples parameter(s)	Direct band gap (eV) , Indirect band gap (eV)		Urbach energy (e V)	Steepness
ZLNBBB HO (01)	2.23	2.13	0.4809	0.096
ZLNBBB HO (1.5)	2.20	2.12	0.4837	0.095
ZLNBBB HO (02)	2.19	2.06	0.4857	0.094

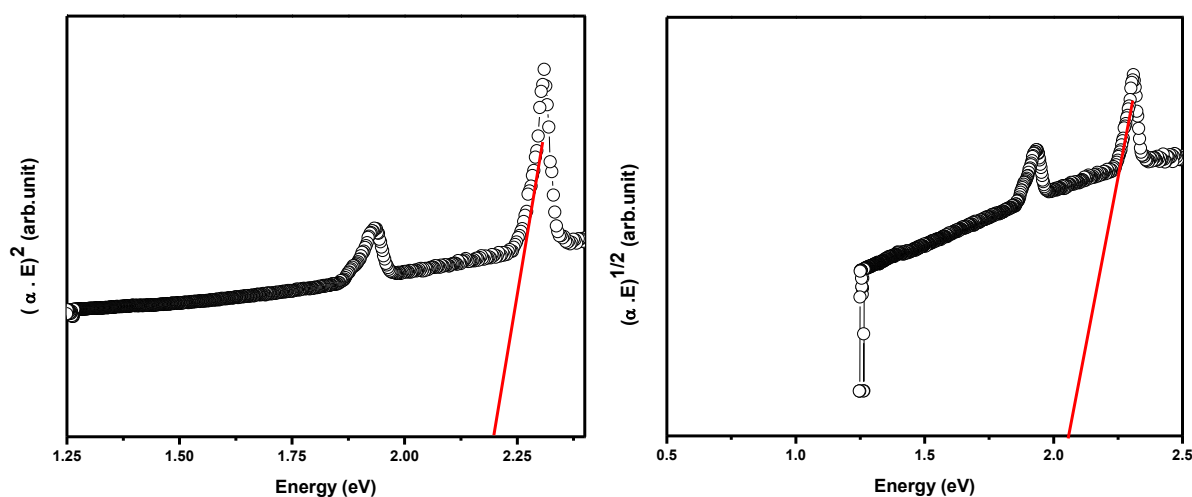


Fig.5: Tauc's plot for ZLNBBB HO (01) (a) Direct band gap (b)Indirect band gap.

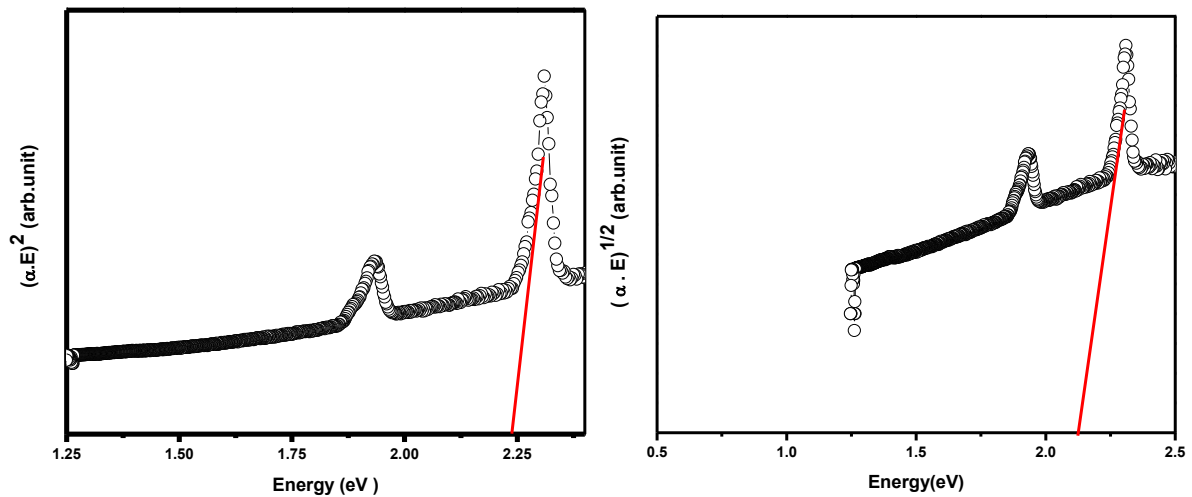


Fig.6: Tauc's plot for ZLNBBB HO (1.5) (a) Direct band gap (b) Indirect band gap.

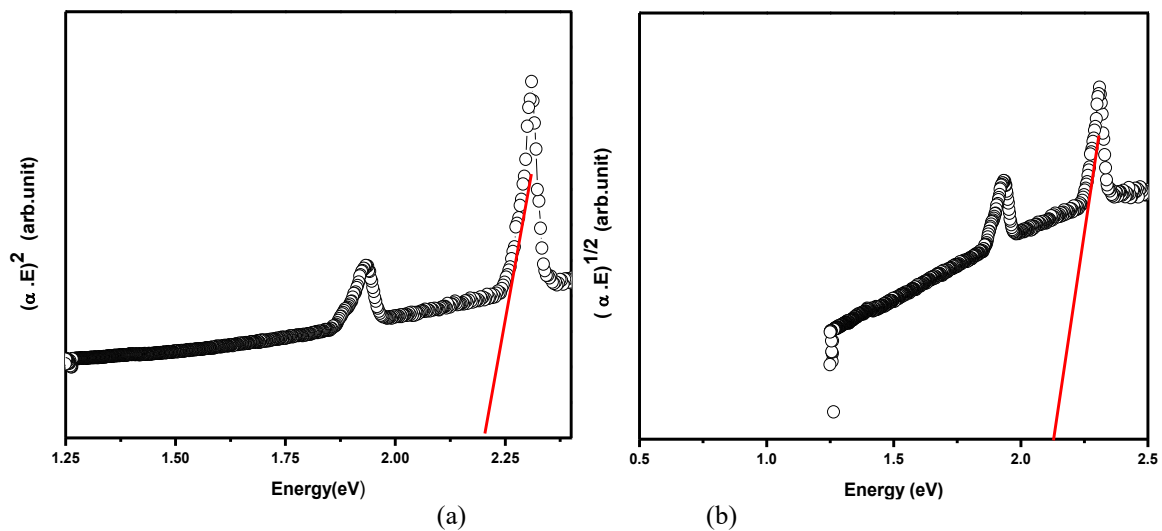


Fig.7: Tauc's plot for ZLNBBB HO (2.0) (a) Direct band gap (b) Indirect band gap.

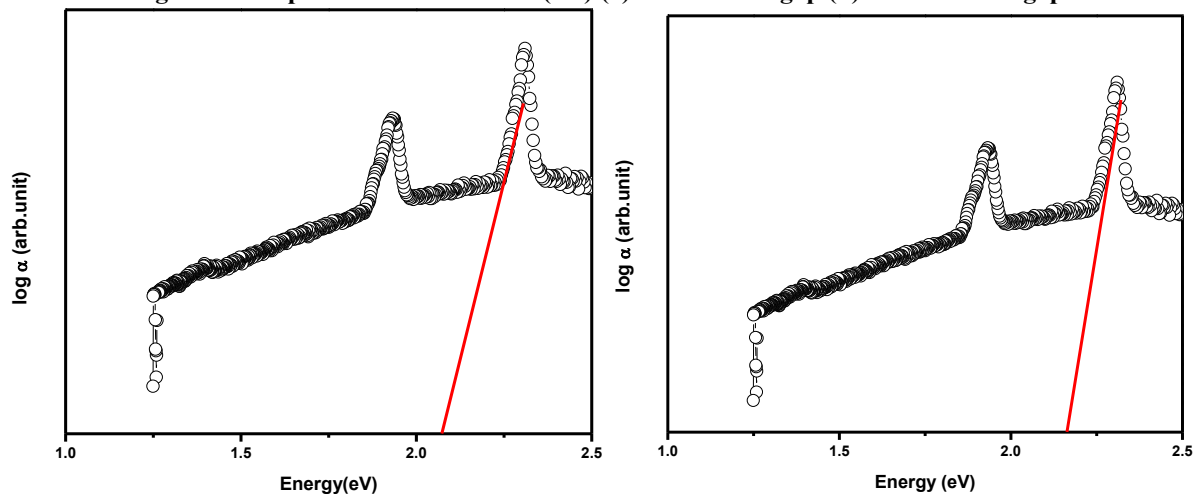


Fig.8: Variation of energy with $\log \alpha$ for prepared glasses (a) ZLNBBB HO (1.0) and (b) ZLNBBB HO (1.5).

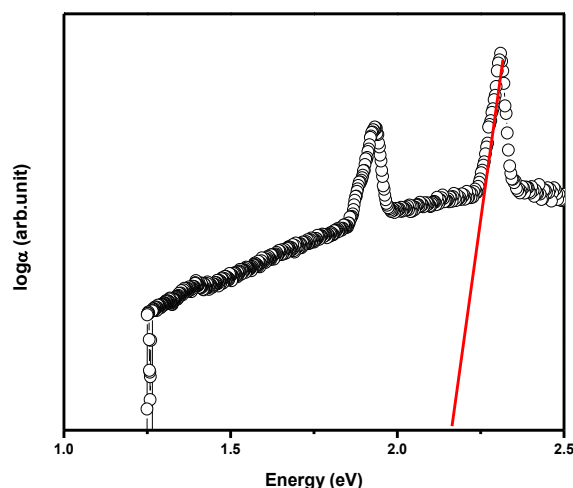


Fig.9: Variation of energy and $\log \alpha$ for glass sample ZLNBBB HO (2.0)

The direct energy band gap is found to be in the range of 2.19 to 2.23 eV, while indirect band gap lies between 2.06 to 2.13 eV. The Urbach's energy is in the range 0.4809 to 0.4857 eV. The direct and indirect energy band gap are found to be decrease with increasing Ho^{3+} ions concentration due to the shifting of absorption edge. Urbach energy increase with Ho_2O_3 content due to increase non-bridging oxygen. The values of steepness parameter lies between 0.94 and 0.96.

4.5.3 Interaction parameter

The interaction parameter of the prepared glass samples is given by [32].

$$A = X_{\text{Bi}_2\text{O}_3} \frac{(3.921 - \alpha_{\text{O}^{2-}})}{(\alpha_{\text{Bi}^{3+}} + 3.921)(\alpha_{\text{Bi}^{3+}} + \alpha_{\text{O}^{2-}})} + X_{\text{ZnO}} \frac{(3.921 - \alpha_{\text{O}^{2-}})}{(\alpha_{\text{Zn}^{2+}} + 3.921)(\alpha_{\text{Zn}^{2+}} + \alpha_{\text{O}^{2-}})} + X_{\text{Li}_2\text{O}} \frac{(3.921 - \alpha_{\text{O}^{2-}})}{(\alpha_{\text{Li}^{2+}} + 3.921)(\alpha_{\text{Li}^{2+}} + \alpha_{\text{O}^{2-}})} + \\ X_{\text{Nb}_2\text{O}_5} \frac{(3.921 - \alpha_{\text{O}^{2-}})}{(\alpha_{\text{Nb}^{5+}} + 3.921)(\alpha_{\text{Nb}^{5+}} + \alpha_{\text{O}^{2-}})} + X_{\text{BaO}} \frac{(3.921 - \alpha_{\text{O}^{2-}})}{(\alpha_{\text{Ba}^{2+}} + 3.921)(\alpha_{\text{Ba}^{2+}} + \alpha_{\text{O}^{2-}})} + X_{\text{B}_2\text{O}_3} \frac{(3.921 - \alpha_{\text{O}^{2-}})}{(\alpha_{\text{B}^{3+}} + 3.921)(\alpha_{\text{B}^{3+}} + \alpha_{\text{O}^{2-}})} + \\ + X_{\text{Ho}_2\text{O}_3} \frac{(3.921 - \alpha_{\text{O}^{2-}})}{(\alpha_{\text{Ho}^{3+}} + 3.921)(\alpha_{\text{Ho}^{3+}} + \alpha_{\text{O}^{2-}})} \quad (11)$$

where 3.921 Å is Pauling value for the electronic polarizability of free oxide ion, $X_{\text{Bi}_2\text{O}_3}$, X_{ZnO} , $X_{\text{Li}_2\text{O}}$, $X_{\text{Nb}_2\text{O}_5}$, X_{BaO} , $X_{\text{B}_2\text{O}_3}$, $X_{\text{Ho}_2\text{O}_3}$ are equivalent fractions of Bi_2O_3 , ZnO , Li_2O , Nb_2O_5 , BaO , B_2O_3 , Ho_2O_3 respectively. Theoretical interaction parameter of the glass samples is calculated using the relation [33].

$$A_{\text{th}} = X_{\text{Bi}_2\text{O}_3} A_{\text{Bi}_2\text{O}_3} + X_{\text{ZnO}} A_{\text{ZnO}} + X_{\text{Li}_2\text{O}} A_{\text{Li}_2\text{O}} + X_{\text{Nb}_2\text{O}_5} A_{\text{Nb}_2\text{O}_5} + X_{\text{BaO}} A_{\text{BaO}} + X_{\text{B}_2\text{O}_3} A_{\text{B}_2\text{O}_3} + X_{\text{Ho}_2\text{O}_3} A_{\text{Ho}_2\text{O}_3} \quad (12)$$

where $A_{\text{Bi}_2\text{O}_3}$, A_{ZnO} , $A_{\text{Li}_2\text{O}}$, $A_{\text{Nb}_2\text{O}_5}$, A_{BaO} , $A_{\text{B}_2\text{O}_3}$, $A_{\text{Ho}_2\text{O}_3}$ are the interaction parameters corresponding oxides. Calculated values of A and A_{th} are tabulated in Table 6.

Table 6. The Interaction parameter of the prepared glass samples.

Glass samples	Interaction parameter (A)		Interaction parameter (A_{th})
ZLNBBB HO (1.0)	0.0659		0.0660
ZLNBBB HO (1.5)	0.0679		0.0680
ZLNBBB HO (2.0)	0.0698		0.0699

The interaction parameter is found to be increase with increasing Ho_2O_3 content. It is found that the theoretical interaction parameter and calculated interaction parameters both values are nearly equal.

V. Conclusions

The zinc lithium niobium barium bismuth borate glasses doped with Ho_2O_3 were investigated using melt quenching technique. Amorphous nature of the prepared glass specimens was confirmed by X-ray diffraction. The Fourier transform infrared spectra show the structural groups in glass samples such as BO_3 and BO_4 . The value of refractive index nearly 1.625 for glass ZLNBBB HO (2.0), suggesting that glass ZLNBBB HO (2.0) is better transmitted compared to other two glass systems ZLNBBB HO (1.0) and ZLNBBB HO (1.5).

The large values of Balaji–Shankar relation indicate that the prepared glass have good thermal stability. Hence the new glass specimens have potential applications for laser, optical, fiber fabrication and electronic devices.

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