# Absorption Studies of Tripositive Neodymium Doped Lithium Bismuth Borate Glasses

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*Glasses having composition (60-x) $B_2O_3$-10$Bi_2O_3$-30$Li_2O$-x$Nd_2O_3$ with x =1, 1.5 and 2 were prepared by melt quenching technique. The amorphous nature of the prepared glass samples was confirmed by X-ray diffraction. The absorption spectra of three $Nd^{3+}$ lithium bismuth borate (LBB) glasses have been recorded at room temperature. From these spectral data various energy interaction parameters like Slater-Condon parameters $F_k$ (k=2, 4 and 6), Lande' parameters $\xi_{4f}$ and Racah parameters $E^k$ (k=2, 4 and 6) have been computed. Nephelauxetic ratio (β') and bonding parameters ($b^{1/2}$) have also been computed from these parameters to study the nature of bonding in doped glasses. The intensities of the f-f transitions in the absorption spectra have been analyzed by the application of the Judd-Ofelt theory.*

**Keywords**: $Nd^{3+}$: LBB Glasses, Energy Interaction Parameters, Absorption.

## 1. INTRODUCTION

Glass matrices exhibit several advantageous properties to recognize them as more useful host materials in the development of rare earth lasing systems due to their unique properties like preparation, fabrication and less optical homogeneity [1]. In recent years, much attention has been focused on the search for new rare earth doped materials to be used as hosts for optical devices. Rare earth doped heavy metal oxide (HMO) glasses such as $Bi_2O_3$, PbO, $GeO_2$ and $GaO_2$ [2,3,4,5] are transparent from ultraviolet to the infrared region. It is a known fact that $Bi_2O_3$ could be found as a network modifier and it is not considered as the glass network former (NWF) because of the small field strength of $Bi^{3+}$ ions, however, in combination with $B_2O_3$ in a relatively large composition range, there is a possibility for the glass formation [6]. $B_2O_3$ is familiar as network former (NWF) and is present in almost all important commercial glasses. Thus, $Bi_2O_3$ and $Li_2O$ are considered as network modifier when those are added to the $B_2O_3$ content. The advantages such as good solubility for rare earth ions, good mechanical and thermal stability, and the low cost of the glasses make them as the potential host materials for rare earth ions.

In the present work, we have studied on the absorption spectra of tripositive neodymium ions doped LBB glasses have been investigated. From these spectral data various energy interaction parameters like Slater-Condon parameters $F_k$ (k=2, 4 and 6), Lande' parameters $\xi_{4f}$ and Racah parameters $E^k$ (k=2, 4 and 6) have been computed. Nephelauxetic Ratio (β') and Bonding Parameters ($b^{1/2}$) have also been computed from these parameters to study the nature of bonding in doped glasses. The intensity of the

transition for the $RE^{3+}$ -ions has been calculated in term of Judd-Ofelt theory [7,8]. This theory known a set of three intensity parameter $\Omega_\lambda$ ($\lambda$ = 2, 4 and 6) which are useful to the environment of trivalent rare earth ions. To understand the laser efficiency of these materials, the value of spectroscopy quality factor ($\Omega_4/\ \Omega_6$) has been evaluated.

## 2. EXPERIMENTAL

$Nd^{3+}$: LBB glasses were prepared by melt quenching technique [9]. The starting chemicals used were reagent grade of $H_3BO_3$, $Bi_2O_3$, $Li_2CO_3$ and $Nd_2O_3$. The chemical compositions of the prepared glasses are as follows: (60-x) $B_2O_3$-10$Bi_2O_3$-30$Li_2O$-x$Nd_2O_3$ where x =1, 1.5 and 2 mol%. All weighed chemicals were powdered by using an Agate pestle mortar and mixed thoroughly before each batch (15g) was melted in alumina crucibles in silicon carbide based an electrical furnace for 2h at $1050^0$ C. The molten glass was then poured in a muffle furnace on to a steel mould and annealed at temperature of $300^0$ C for 60 minutes 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. 1, 1.5 and 2 mol% of $Nd^{3+}$: LBB glasses are labeled as LBB (1), LBB (1.5) and LBB (2) respectively.

X-ray diffractogram of glass sample was studied with PANalytical X′pert Pro MPD diffractometer of $CuK_\alpha$ radiation (1.5406 ) system. The absorption spectra of these glasses were recorded between wavelength ranges 400-900 nm with a Perkin-Elmer Lambda 750 UV/VIS/NIR Spectrophotometer at room temperature.

## 3. RESULT AND DISCUSSION

The X-ray diffractogram shown in Figure 1 showed no prominent peaks indicating the absence of a crystalline phase.

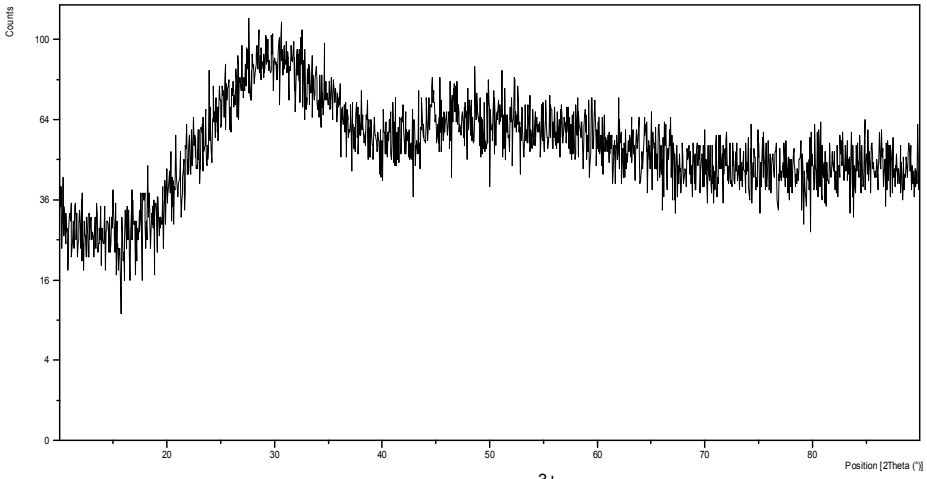


**Fig. 1:** XRD Pattern of (1 mol%) $Nd^{3+}$ ions doped glass.

The absorption spectra of $Nd^{3+}$ doped LBB glass specimens have been presented in Figure 2 in terms of relative absorption ($I_0/I$) versus wavelength (nm), where I and $I_0$ are intensities of the radiation transmitted through doped specimens and undoped

specimens of equal thickness. Eleven absorption bands have been observed from the ground state $^4I_{9/2}$ to excited states $^4F_{3/2}$, $^4F_{5/2}$, $^4F_{7/2}$, $^4F_{9/2}$, $^2H_{11/2}$, $^4G_{5/2}$, $^4G_{7/2}$, $^4G_{9/2}$, $^2G_{9/2}$, $^4G_{11/2}$, and $^2D_{5/2}$ for $Nd^{3+}$ doped LBB glasses.

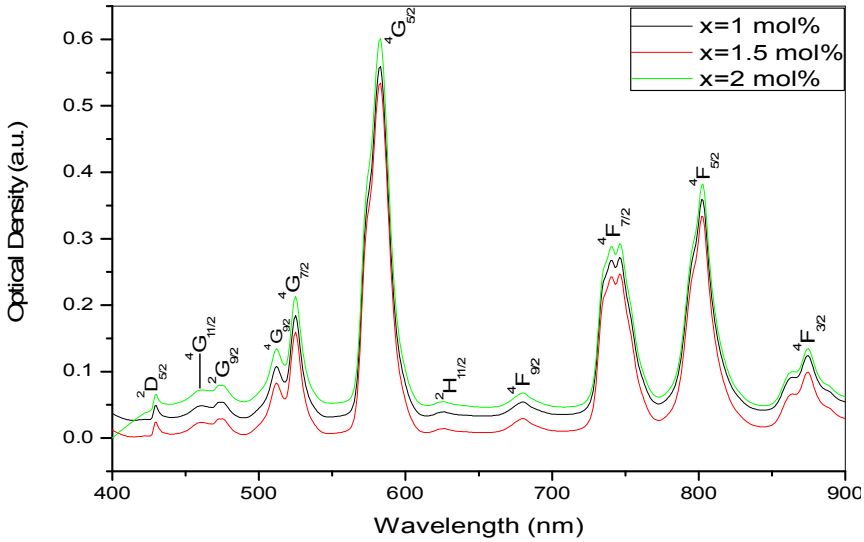


**Fig. 2:** Vis-NIR absorption spectra of $Nd^{3+}$:LBB glasses.

**Table 1:** Experimental energy ($E_{exp}$) and calculated energy ($E_{cal}$) with their different ($\Delta E$) for various absorption bands of $Nd^{3+}$ doped LBB glass specimens of different doping concentrations

| Absorption levels | LBB (1) ($cm^{-1}$) | | | LBB (1.5) ($cm^{-1}$) | | | LBB (2) ($cm^{-1}$) | | |
|---|---|---|---|---|---|---|---|---|---|
| | $E_m$ | $E_c$ | $\Delta E$ | $E_m$ | $E_c$ | $\Delta E$ | $E_m$ | $E_c$ | $\Delta E$ |
| $^4F_{3/2}$ | 11435 | 11327 | +108 | 11436 | 11329 | 107 | 11434 | 11326 | 108 |
| $^4F_{5/2}$ | 12461 | 12463 | -2 | 12463 | 12464 | -1 | 12460 | 12461 | -1 |
| $^4F_{7/2}$ | 13399 | 13353 | +46 | 13399 | 13355 | 44 | 13398 | 13352 | 46 |
| $^4F_{9/2}$ | 14704 | 14886 | -182 | 14703 | 14886 | -183 | 14702 | 14884 | -182 |
| $^2H_{11/2}$ | 15967 | 15950 | +17 | 15967 | 15950 | 17 | 15967 | 15950 | 17 |
| $^4G_{5/2}$ | 17162 | 16991 | 171 | 17161 | 16994 | 167 | 17162 | 16990 | 172 |
| $^4G_{7/2}$ | 19048 | 18990 | 58 | 19051 | 18993 | 58 | 19048 | 18990 | 58 |
| $^4G_{9/2}$ | 19527 | 19484 | 43 | 19531 | 19487 | 44 | 19527 | 19483 | 44 |
| $^2G_{9/2}$ | 21160 | 21100 | 60 | 21160 | 21101 | 59 | 21160 | 21099 | 61 |
| $^4G_{11/2}$ | 21706 | 21694 | 12 | 21716 | 21696 | 20 | 21701 | 21692 | 9 |
| $^2D_{5/2}$ | 23278 | 23549 | -271 | 23283 | 23552 | -269 | 23278 | 23550 | -272 |
| rms deviation ($\sigma$) | ±120.21 | | | ±119.33 | | | ±120.54 | | |

The absorption spectra of lanthanide ions lie in the visible and near infrared regions and correspond to transitions from the ground state to various excited states of $4f^N$ configuration. The relevant theoretical background along with the interpretation of the observed absorption spectra of $Nd^{3+}$ doped LBB glasses in terms of the energy interaction parameters and Judd-Ofelt intensity parameters are given in Table 1.

The experimental and calculated energy band positions are given in Table 1 for $Nd^{3+}$ doped LBB glass specimens. The value of $F_k$, $E^k$ and $\xi_{4f}$ parameters have been computed by using the observed energies of the bands, $E_{0j}$ and partial derivatives with the help of partial regression method [10] and given in Table 2.

**Table 2:** Calculated values of Slater-Condon, Lande', Racah, Nephelauxetic ratio and bonding parameters of $Nd^{3+}$ doped LBB glass specimens of different doping concentrations.

| Parameters | Free ion | LBB (1) | LBB (1.5) | LBB (2) |
|---|---|---|---|---|
| $F_2$ ($cm^{-1}$) | 331.16 | 329.83 | 329.72 | 329.66 |
| $F_4$ ($cm^{-1}$) | 50.71 | 49.40 | 49.45 | 49.46 |
| $F_6$ ($cm^{-1}$) | 5.154 | 5.260 | 5.254 | 5.253 |
| $\xi_{4f}$ ($cm^{-1}$) | 884.0 | 891.39 | 890.92 | 890.91 |
| $E^1$ ($cm^{-1}$) | 5024.0 | 5003.435 | 5002.397 | 5002.139 |
| $E^2$ ($cm^{-1}$) | 23.90 | 24.271 | 24.237 | 24.230 |
| $E^3$ ($cm^{-1}$) | 497.0 | 488.97 | 489.07 | 488.99 |
| $F_4/F_2$ | 0.1531 | 0.1497 | 0.1499 | 0.1500 |
| $F_6/F_2$ | 0.0155 | 0.015947 | 0.0159340 | 0.0159369 |
| $E^1/E^3$ | 10.1086 | 10.2325 | 10.2284 | 10.2295 |
| $E^2/E^3$ | 0.0481 | 0.049638 | 0.049558 | 0.049552 |
| β' | - | 0.9959 | 0.9956 | 0.9954 |
| $b^{1/2}$ | - | 0.0447 | 0.0466 | 0.0474 |

The relation among different $F_k$ parameters is found to be $F_2>F_4>F_6$ in $Nd^{3+}$ doped LBB glass specimens. The value of $F_2$ ~ (329.6-329.8 $cm^{-1}$) is nearly same as ~ 328.62 $cm^{-1}$ and 330.8 $cm^{-1}$ for aluminoboro phosphate and fluoro glasses [11,12]. The decrease in its value from aquo ion is 0.41% for $Nd^{3+}$ doped LBB glass specimens. $F_4/F_2$ ~ (0.150-0.149) and $F_6/F_2$ ~ (0.016-0.015) are nearly same as reported in other glasses [13].

The values of $E^1$, $E^2$ and $E^3$ parameters have been given in Table 2. The ratio of $E^1/E^3$ ~ (10.22-10.23) and $E^2/E^3$ ~ (0.0496-0.0495) are found to remain almost const over the entire range of $Nd^{3+}$ doping concentrations and are in good agreement with the corresponding hydrogenic ratios. Bonding in the glasses can be inferred from the nephelauxetic Ratio (β') defined [14] by the relation,

$$\beta' = F_2 \text{ (glass)}/F_2 \text{ (free ion)}$$

where $F_2$ (glass) is the Slater-Condon energy interaction parameters and $F_2$ (free ion) is the same parameter for free ion. If β' is less than one it indicates covalent bonding, while its value greater than one indicates ionic bonding. Henrie and Choppin [15] have defined another bonding parameter ($b^{1/2}$) in terms of β', given by $b^{1/2} = [(1- \beta')/2]^{1/2}$ .

A real value of $b^{1/2}$ indicates covalent bonding. This parameter is very useful for comparative study of bonding between the central R.E. ion and the surrounding glass matrix. The values of nephelauxetic ratio (β') and bonding parameters ($b^{1/2}$) for $Nd^{3+}$ ion in LBB glasses are ~ 0.99 and ~ (0.047-0.044) respectively as shown in Table 2.

Small values of r.m.s. deviation 'σ' between experimental energy ($E_{exp}$) and calculated energy ($E_{cal}$) of absorption levels in neodymium ion doped LBB glass specimens justify the suitability of the use of Taylor series expansion method [16,17]. The intensities of the observed bands have been measured in terms of line strength, $S_m$, calculated from the observed oscillator strength of the absorption bands. The calculated line strengths ($S_c$) have been compared with the experimentally measured line strengths and collected in Table 3. The success of Judd-Ofelt theory has been shown by low value of goodness of fit between the measured ($S_m$) and calculated ($S_c$) line strengths.

**Table 3:** Measured ($S_m$) and calculated line strength ($S_c$) with their differences ($\Delta S'$) for various absorption bands of $Nd^{3+}$ doped LBB glass specimens of different doping concentrations.

| **Absorption Levels** | **LBB (1)** ($10^{-20}$) $cm^2$ | | | **LBB (1.5)** ($10^{-20}$) $cm^2$ | | | **LBB (1)** ($10^{-20}$) $cm^2$ | | |
|---|---|---|---|---|---|---|---|---|---|
| | $S_m$ | $S_c$ | $\Delta S$ | $S_m$ | $S_c$ | $\Delta S$ | $S_m$ | $S_c$ | $\Delta S$ |
| $^4F_{3/2}$ | 1.093 | 0.976 | +0.117 | 0.911 | 0.719 | +0.192 | 0.855 | 0.668 | +0.187 |
| $^4F_{5/2}$ | 3.082 | 3.151 | -0.069 | 2.097 | 2.197 | -0.100 | 1.719 | 1.802 | -0.083 |
| $^4F_{7/2}$ | 3.298 | 3.136 | +0.162 | 2.299 | 2.137 | +0.162 | 1.817 | 1.651 | +0.166 |
| $^4F_{9/2}$ | 0.195 | 0.223 | -0.028 | 0.126 | 0.153 | -0.027 | 0.124 | 0.121 | +0.003 |
| $^2H_{11/2}$ | 0.049 | 0.056 | -0.007 | 0.040 | 0.039 | +0.001 | 0.022 | 0.031 | -0.009 |
| $^4G_{5/2}$ | 7.441 | 7.387 | +0.054 | 5.168 | 5.114 | +0.054 | 4.247 | 4.181 | +0.066 |
| $^4G_{7/2}$ | 1.245 | 1.032 | +0.213 | 0.768 | 0.736 | +0.032 | 0.679 | 0.642 | +0.037 |
| $^4G_{9/2}$ | 0.392 | 0.482 | -0.090 | 0.430 | 0.343 | +0.087 | 0.479 | 0.296 | +0.183 |
| $^2G_{9/2}$ | 0.293 | 0.176 | +0.117 | 0.196 | 0.127 | +0.069 | 0.157 | 0.115 | +0.042 |
| $^4G_{11/2}$ | 0.303 | 0.169 | +0.134 | 0.165 | 0.125 | +0.040 | 0.181 | 0.118 | +0.063 |
| $^2D_{5/2}$ | 0.071 | 0.010 | +0.061 | 0.068 | 0.007 | +0.061 | 0.055 | 0.005 | +0.050 |
| Goodness of fit | 0.01718 | | | 0.01194 | | | 0.01461 | | |

The computed values of $\Omega_\lambda$ ($\lambda$ = 2, 4, and 6) parameters are very important since they are used in the calculation of laser parameters. In the case of $Nd^{3+}$ doped LBB glass specimen eleven bands in the visible and NIR regions have been used in the computation of $\Omega_\lambda$ parameters. For $Nd^{3+}$ doped LBB glasses, $\Omega_\lambda$ values vary as $\Omega_2>\Omega_6>\Omega_4$. The $\Omega_2$ parameters is affected by the covalency, the $\Omega_6$ parameters is related to the rigidity of the glass hosts, and $\Omega_4$ parameters is determined by $\Omega_2$ and $\Omega_6$ parameters [18,19]. In $Nd^{3+}$ doped glasses the contribution to $\Omega_2$ parameters are mainly due to the hypersensitive transition $^4I_{9/2}\rightarrow{}^4G_{5/2}$. The bonding environment surrounding the rare earth ion has been discussed on the basis of the value of $\Omega_\lambda$ parameters. The Judd-Ofelt intensity parameters $\Omega_2$, $\Omega_4$ and $\Omega_6$ found by least square fit method have been represented in Table 4.

**Table 4:** Judd-Ofelt intensity parameters, $\Omega_\lambda$ for $Nd^{3+}$ doped LBB glass specimens of different doping concentration and compare with other similar glasses.

| Glass Specimen | $\Omega_2(pm^2)$ | $\Omega_4(pm^2)$ | $\Omega_6(pm^2)$ | $\Omega_4/\Omega_6$ | Ref. |
|---|---|---|---|---|---|
| LBB (1) | 5.158 | 3.170 | 4.531 | 0.699 | p |
| LBB (1.5) | 3.454 | 2.399 | 3.071 | 0.781 | P |
| LBB (2) | 2.602 | 2.354 | 2.339 | 1.006 | p |
| CdBiB | 4.893 | 2.976 | 5.789 | 0.514 | 22 |
| Phosphate | 3.6 | 5.0 | 5.5 | 0.909 | 23 |

Weber and Jacob [20] have reported that the ratio $\Omega_4/\Omega_6$ known as the spectroscopic quality factor characterizes the glass concerned. The values of $\Omega_4/\Omega_6$ for glasses studied are given in Table 4. $Nd^{3+}$ doped LBB glasses are having larger values ($\Omega_4/\Omega_6$) compared to CdBiB [21] and phosphate [22].

## 4. CONCLUSION

It is summarized that good optical glasses with 1, 1.5 and 2 mol% of $Nd^{3+}$ doped LBB glasses were prepared. Any further increase in the concentration of $Nd^{3+}$ ion in the above glass matrix does not provide good transparency with these glasses. The calculated energies of the transitions using least square fit method are exactly coinciding with the experimentally obtained energies of the transitions, reflecting the good accuracy of the experimentation. The hypersensitive transition $^4I_{9/2}\rightarrow{}^4G_{5/2}$ exhibits high intensity as a function of Judd-Ofelt intensity ($\Omega_2$) parameter in the case of 1 mol% $Nd^{3+}$ glass system. Spectroscopic quality factor ($\Omega_4/\Omega_6$) characterizes the glass concerned. The values of $\Omega_4/\Omega_6$ having larger compared to other oxide glasses.

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