



# Luminescence features of Dy<sup>3+</sup>-doped B<sub>2</sub>O<sub>3</sub> and B<sub>2</sub>O<sub>3</sub>-P<sub>2</sub>O<sub>5</sub> hosts for optical devices: the linear and non-linear optical studies

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## Abstract

The glass with various concentrations of trivalent rare earth Dy<sup>3+</sup> in borate and boro-phosphate host structures, along with alkali and transition metals, was synthesized by the melt-quenching technique. An FT-IR and FT-Raman spectrum reveal the structural features by tracing out the various molecular vibrations of borate and phosphate units. UV-Vis-NIR spectral analysis was used to investigate the effect of RE concentration on linear ( $E_g$ ,  $\Delta E$ ,  $n_1$ , and  $\chi^{(1)}$ ) and non-linear ( $\chi^{(3)}$ ,  $n_2$ ) optical characteristics. The metal–oxygen bond character is explored in relation to the impact of alkali cations on the  $\chi^{(3)}$  of alkali borate and boro-phosphate host glasses. Photoluminescence emission spectra obtained in the optical zone show two intense band emissions traced at 482 and 574 nm, which correspond to  $^4F_{9/2} \rightarrow ^6H_{15/2}$  (electric-dipole transition) and  $^4F_{9/2} \rightarrow ^6H_{13/2}$  (magnetic-dipole transition), respectively, and one weak emission at 674 nm, which corresponds to  $^4F_{9/2} \rightarrow ^6H_{11/2}$ . The fluorescence decay time was measured and fitted with the characteristics of a non-exponential decay profile. The estimated color coordinates are in the white region of the CIE 1931 chromaticity curve and are quite close to the conventional white illuminant (0.330, 0.330). In addition, the computed yellow-to-blue (Y/B) ratio and correlated color temperature of the prepared photonic glasses could allow tweaking white light emission via varying concentrations and host environments, which might be advantageous for various optical-based device fabrications.

**Keywords** Urbach energy · Non-linear optics · Luminescence decay · I-H fit curve

## 1 Introduction

To develop laser science and technology, non-linear optical (NLO) active compounds can advantageously widen the few coherent spectrum sources currently available to far larger spectral regions. Despite its exceptional expandability, there is a strong need to investigate innovative NLO compounds with superior efficiency and performance for prospective photonic applications. The phosphate glasses were primarily studied as hosts for laser and optical amplifiers owing to their high gain properties in comparison to other glass

families [1]. The structure of borate glasses is especially fascinating due to the boron anomaly, and as a result, the impact of the network modifier and generated non-bridging oxygens on the linear and non-linear optical properties may be segregated. Because of their considerable effect on luminous efficiency, B<sub>2</sub>O<sub>3</sub>-based glasses are widely recognized for their photo-generated second-order NLO effects.

Borate glasses can generate superstructures of B<sub>2</sub>O<sub>3</sub> due to the triangular planer BO<sub>3</sub>, and tetrahedral BO<sub>4</sub> may interact to form diborate (B<sub>2</sub>O<sub>5</sub><sup>4-</sup>), tetraborate/pyroborate (B<sub>4</sub>O<sub>7</sub><sup>2-</sup>), pentaborate (B<sub>5</sub>O<sub>8</sub>), or boroxol ring, depending on the quantity of non-bridging oxygens (NBOs) or bridging oxygens (BOs). Alkali metals (Li, Na, and K) modify borate glass, and the emergence of different structural units causes a notable alteration in its physical characteristics [2]. Specifically, lithium borate glasses, which are primarily known as solid electrolytes, are the most adaptable glass systems. One can investigate and synthesize novel materials for optical purposes by adding a specified amount of rare earth oxide to these glasses for specific applications. Host materials with low maximal phonon energies are preferred

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to synthesize rare earth ions-doped glasses since rare earth ions consume less excitation energy during non-radiative transitions, which leads to relatively high luminous quantum efficiencies. Typically, the maximal phonon energies of the hosts with oxides, such as boron oxides ( $B_2O_3$ ) and phosphorous pentoxide ( $P_2O_5$ ), are higher than those of the hosts with halide and sulfur. However, the study found that the inclusion of alkali and alkaline metal ions in the oxide glasses substantially reduces phonon energy through energy transfer interactions.

Heavy metal oxide-based RE-infused glass networks have large lasing abilities due to their lower field strength and higher mass and polarizability [3]. The doping concentration of  $Dy^{3+}$  should be considered because it affects both the optical transition properties of the material and the concentration quenching of fluorescence intensity caused by the cross-relaxation between  $Dy^{3+}$  existents. Several researchers reported that concentration quenching occurs at high doping concentrations ( $\geq 1.0$  mol%) of  $Dy_2O_3$  in glasses, so many have studied the glass systems with only 1.0 mol% of  $Dy_2O_3$  [4–6]. In their ground state, lanthanide ions ( $Dy^{3+}$ ) have an electronic structure of  $[Xe] 4f^{10} 5s^2 5p^6$ , where the f-electrons are typically non-bonding since they are protected by the  $5s^2 5p^6$  orbitals. The optical characteristics are known to be primarily influenced by the configuration and number of 4f electrons. The NLO properties for lanthanide glasses are predicted to be improved because of the optical resonance phenomenon, which is followed by f–f electron transitions. According to reports, the inclusion of  $TiO_2$  in glass compositions improves the NLO factors of the glasses due to their empty d-shells and lone pair electrons [7]. The addition of  $Al_2O_3$  in a sample improves the optical sensitivity by changing the host structure and generating asymmetric sites that result in a 30% increase in the PL intensity when compared to non- $Al_2O_3$  samples [8].

The current paper deals with a study of the  $Dy^{3+}$  ion in borate and boro-phosphate host matrixes, together with heavy metal ions. The effects of the  $Dy^{3+}$  doping concentration on the transition, quantum effectiveness, quenching state, and luminescence features were studied. Finally, the values of the CIE chromaticity coordinates are assessed to confirm the w-LED application.

## 2 Experimental technique

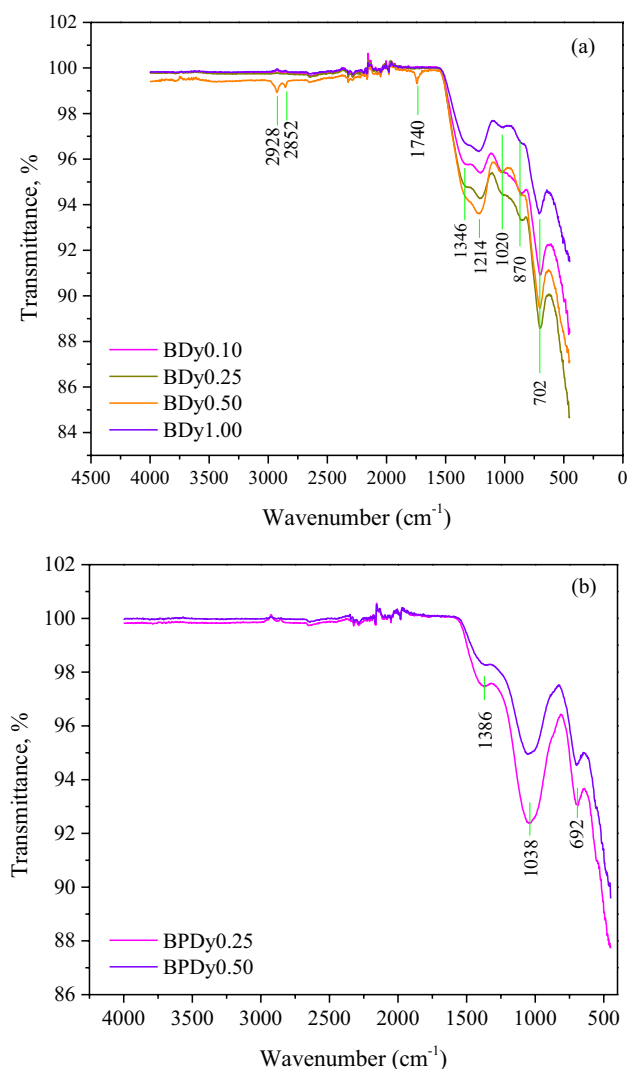
Glasses with the chemical compositions of  $(60 - x) B_2O_3 + 25SrCO_3 + 7TiO_2 + 5LiCO_3 + 3Al_2O_3 + xDy_2O_3$  ( $x = 0.10, 0.25, 0.50,$  and  $1.00$  mol%) and  $40B_2O_3 + 20SrCO_3 + 20H_6NO_4P + 7ZnO + 5LiCO_3 + (5 - x) P_2O_5 + 3Al_2O_3 + xDy_2O_3$  ( $x = 0.25$  and  $0.50$  mol%), respectively, were synthesized by melt quenching technique. The boron oxide ( $B_2O_3$ ) and phosphorous pentoxide ( $P_2O_5$ ) act

as a network former in the respective glasses;  $SrCO_3$  acts as a network modifier; aluminum oxide ( $Al_2O_3$ ) will increase network connectivity;  $ZnO$  will increase the mechanical stability of the glass. The measured quantity of AR-grade chemicals was taken and mixed thoroughly to attain homogeneity in an agate mortar. The blended chemicals were placed in an alumina crucible and heated in an electric furnace for 7 h at  $1125$  °C. After quickly pouring the melt onto a preheated brass metal plate, the samples were annealed at  $400$  °C to reduce internal stress and achieve structural stability. For convenience, the borate host glasses are designated as BDy0.10, BDy0.25, BDy0.50, and BDy1.0, and the boro-phosphate host glasses are designated as BPDy0.25- and BPDy0.50-based on the  $Dy^{3+}$  concentration.

## 3 Characterization techniques

### 3.1 FT-IR spectral analysis

Thin pellets of approximately 3 mm in thickness were made by mixing KBr, and powdered glass samples were subjected to FT-IR measurements. The spectra were taken at room temperature with a JASCO FTIR 6100 spectrometer with a resolution of  $0.5$   $cm^{-1}$  in the  $400$ – $4000$   $cm^{-1}$  wavenumber range. The FTIR spectra of borate glass systems are depicted in Fig. 1a. The peak at  $702$   $cm^{-1}$  confirmed the bending vibration of the B–O–B linkage of the penta-borate unit and the vibration of linking oxygen atoms ( $BO_2^-$ ). The presence of B–O bond stretching vibrations of tetrahedral  $BO_4$  units is responsible for the small band at  $870$   $cm^{-1}$  [9]. The peak at  $1020$   $cm^{-1}$  is caused by pentaborate as well as  $BO_4$  tetrahedral vibrations [10]. The peaks at  $1214$   $cm^{-1}$  and  $1346$   $cm^{-1}$  correspond to B–O asymmetric stretching vibrations in ortho-borate groups and B–O stretching vibrations of  $BO_3$  trigonal units in ortho-, pyro-, and meta-borate units, respectively. A change in the oxygen-to-boron ratio is enhanced by the substitution of  $B_2O_3$  with a significant concentration of metal oxides such as  $TiO_3$  and  $Al_2O_3$ , along with an increase in  $Dy_2O_3$  concentrations, resulting in stable  $BO_4$  units. The creation of  $BO_4$  units results in tightly packed glass structures, and it decreases the absorption intensity [11]. It was determined that there were no –OH molecules present in the BDy0.10, BDy0.25, and BDy1.0 glasses because this IR band of these materials did not explore the peak associated with the OH vibration position. In BDy0.5 glass, there are two distinct small-intense peaks at  $2852$  and  $2928$   $cm^{-1}$  showing the presence of a smaller quantity of OH molecules. The weakened OH content indicates improved quantum efficiency and lower non-radiative losses for the RE-doped luminous materials [12]. The presence of water



**Fig. 1** FT-IR spectral analysis of (a)  $\text{BDy}_x$  ( $x=0.10, 0.25, 0.50$ , and  $1.00$  mol%) and (b)  $\text{BPDy}_x$  ( $x=0.25$  and  $0.50$  mol%)

molecules or hydroxyl-related bands is further validated by the significant peak that occurred at  $1740\text{ cm}^{-1}$  in  $\text{BDy}0.5$  glass.

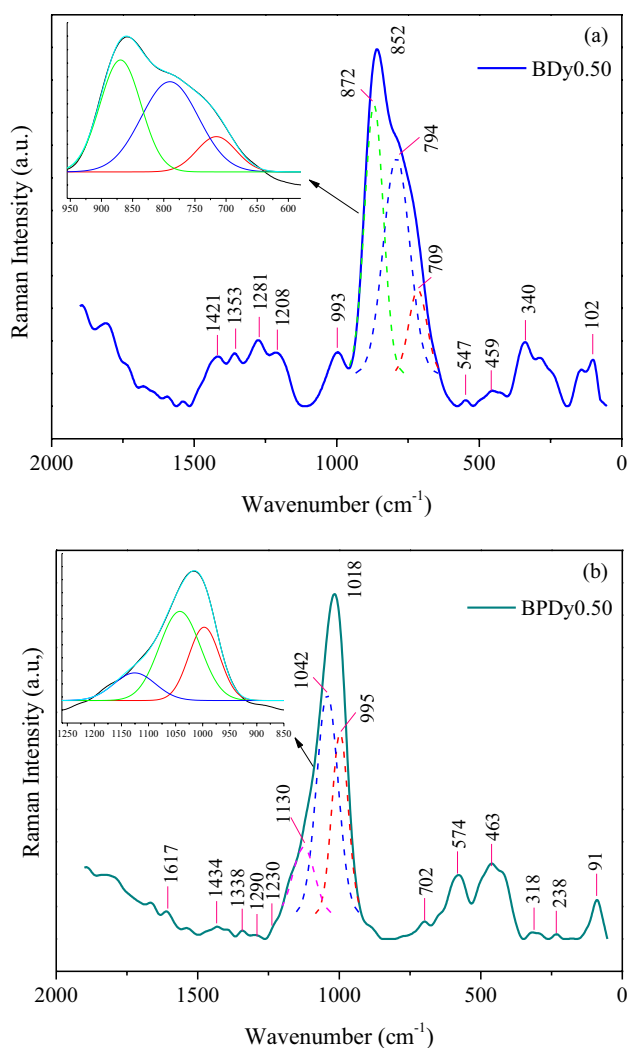
Figure 1b demonstrates the vibrational spectrum of  $\text{BPDy}0.25$  and  $\text{BPDy}0.50$  glass systems. The peak at  $692\text{ cm}^{-1}$  is instigated by P–O–P bending vibrations, and the band at  $1038\text{ cm}^{-1}$  is formed by the symmetric stretching vibrations of the B–O–P bridge between borate ( $\text{BO}_4$ ) and phosphate ( $\text{PO}_4$ ) units, as well as the B–O stretching in  $\text{BO}_4$  tetrahedra [13]. The well-defined small band at  $1386\text{ cm}^{-1}$  dominates the infrared spectrum of boro-phosphate glass matrix in the  $800$  to  $1600\text{ cm}^{-1}$  spectral range as the predominant contribution of  $Q^2$  ( $[\text{P}(\text{OP})_2(\text{O}^-)_2]$ ) and  $\text{BO}_3$  units. The minor peaks traced between  $2000$  and  $2500\text{ cm}^{-1}$  confirm the presence of a hydroxyl group.

### 3.2 Raman spectral analysis

The FT-Raman spectra were taken with a Bruker RFS 27 Multi-RAM spectrometer with a resolution of  $2\text{ cm}^{-1}$ . Because of their intriguing properties, such as the boron anomalies,  $\text{B}_2\text{O}_3$ -based glasses have received a lot of attention over the years. The Raman spectrum of  $\text{BDy}0.5$  glass reveals a significant number of bands caused by the vibration of  $\text{BO}_4$  and  $\text{BO}_3$  groups, as well as complexes including  $[\text{BO}_3]^{3-}$  and  $[\text{BO}_4]^{5-}$  groups. The spectrum, as shown in Fig. 2a, reveals a significant intensity band at  $102\text{ cm}^{-1}$ , which is associated with the symmetric stretching vibration of heavy metal ions, such as Sr–O and Ti–O bonds. The small-broaden peak at  $459\text{ cm}^{-1}$  is ascribed to B–O–B ring angle bending, which is seen at  $470\text{ cm}^{-1}$  for pure  $\text{B}_2\text{O}_3$ . The asymmetric structure of a highly intense peak centered at  $\sim 852\text{ cm}^{-1}$  is caused by B–O–B symmetric stretching in pyro-borate units [14]. Three peaks were detected through band deconvolution at  $709$ ,  $794$ , and  $870\text{ cm}^{-1}$ , respectively. The modest intensity band at  $709\text{ cm}^{-1}$  is accredited to the  $\gamma$ -vibration of  $[\text{BO}_3]^{3-}$ /symmetric breathing vibration of boroxol rings with two  $\text{BO}_4$  groups. The presence of a six-membered boron (boroxol) ring [15] in the prepared glass is indicated by the Raman band nearly observed at  $800\text{ cm}^{-1}$  (in our case  $794\text{ cm}^{-1}$ ). The  $794$  and  $870\text{ cm}^{-1}$  peaks were due to symmetric breathing vibrations of the boroxol ring containing one  $\text{BO}_x$  unit (penta-, tetra-, or tri borate), and the symmetric stretching of  $[\text{BO}_3]^{3-}$  units, respectively [16, 17]. Peaks at  $993$  and  $1281\text{ cm}^{-1}$  indicate that the borate host glass contains B–O $^-$  symmetric stretching vibrations in penta-borate ( $\text{B}_5\text{O}_8^-$ ) and pyro-borate ( $\text{B}_3\text{O}_5^{4-}$ ) anionic groups, respectively [14, 18].

The first band edge in the region of  $1200$ – $1450\text{ cm}^{-1}$ , specifically at  $1208\text{ cm}^{-1}$ , is accountable for the asymmetric stretching vibration of  $[\text{BO}_3]^{3-}$  units. The band shoulder at  $1353\text{ cm}^{-1}$  is associated with the  $\text{B}\emptyset_2\text{O}^-$  triangles linked to  $\text{B}\emptyset_4^-$  units ( $\text{O}^-$ : non-bridging oxygen atom, and  $\emptyset$ : bridging oxygen atom between the two boron atoms) [19]. The peak at  $1421\text{ cm}^{-1}$  is due to the B–O stretching vibrations of the meta-borate ( $\text{BO}_2^-$ ) chains and rings. The bands in the Raman spectra of  $\text{BDy}0.5$  glass may have widened due to the anharmonicity of the interaction between the  $\text{BO}_4$  groups in the borate rings and the Li-ions. A strong network structure is created because the emergence of  $\text{BO}_4$  units in the case of lithium ions is greatly influenced by the mass of the alkali ion [20]. The emergence of a peak after  $1900\text{ cm}^{-1}$  may be due to the presence of hydroxyl group vibration in the glass system.

The Raman spectrum of  $\text{BPDy}0.5$  is depicted in Fig. 2b, and when  $\text{P}_2\text{O}_5$  is replaced with  $\text{B}_2\text{O}_3$ , the structural network of this glass exhibits consequently significant modifications. A significant intense peak detected in the lower wavenumber region at  $\sim 91\text{ cm}^{-1}$  reveals the vibration of heavy metal ions



**Fig. 2** FT-Raman spectral analysis of (a) BDy0.50 and (b) BPDy0.50

with the binding oxygen, and it was observed nearly identical location in BDy0.5 glass. The weak vibrations brought on by  $\text{PO}_2$  bending and bending motions in O–P–O linkages are discernible at 238 and 318  $\text{cm}^{-1}$ , respectively [21]. The broad band with a sharp edge at 463  $\text{cm}^{-1}$  and the consecutive band at 574  $\text{cm}^{-1}$  were attributed to the independent di-borate group vibrations, and O–P–O bending vibrations of  $\text{PO}_4^{3-}$  tetrahedral units [22, 23]. The band at 713  $\text{cm}^{-1}$  originates from the P–O–P symmetric stretching vibrations, which are produced by the binding oxygen atoms connecting the neighboring  $\text{PO}_4$  tetrahedral units. The strong and intense peak at 1018  $\text{cm}^{-1}$  is responsible for the P–O symmetric stretching vibration of the  $\text{Q}^1$  terminal group. The observed band with a center at 1018  $\text{cm}^{-1}$  is asymmetric, and the bulge in the higher wavenumber zone is caused by the overlap of the  $\sim 1130 \text{ cm}^{-1}$  and  $\sim 1230 \text{ cm}^{-1}$  bands, which are the locations of the symmetric and asymmetric stretching of non-binding ( $\text{PO}_2$ ) of  $\text{Q}^2$  units, respectively [24].

The deconvoluted peak at 995  $\text{cm}^{-1}$  within a large band is designated to the orthophosphate ( $\text{PO}_4^{3-}$ ) group symmetric stretching vibration of the  $\text{Q}^0$  units [23]. The detectable band at 1290  $\text{cm}^{-1}$  may be associated with the combined vibration of boro-phosphate groups [24]. The moderate peak identified at 1338  $\text{cm}^{-1}$  is caused by the P=O stretching vibration of terminal oxygen. The tiny-band edge detected at 1434  $\text{cm}^{-1}$  is revealed for the stretching vibration of molecular oxygen bands and B–O in non-bridging oxygen of  $[\text{BO}_3]^{3-}$  triangles. The peak emergence in the higher region around 1600  $\text{cm}^{-1}$  is ascribed to the OH-bending vibration. Tables 1 and 2 show a description of the vibrational bands found in the FT-IR and FT-Raman spectra of the prepared glasses.

### 3.3 UV–Vis spectral analysis: optical characterization

Optical absorption spectra of the  $\text{BDy}_x$  ( $x=0.10, 0.25, 0.50$ , and  $1.00 \text{ mol\%}$ ) and  $\text{BPDy}_x$  ( $x=0.25$  and  $0.50 \text{ mol\%}$ ) glasses were performed in the JASCO V-670 spectrometer at a wavelength between 190 and 1400 nm at room temperature. Figure 3a and b depicts the collected spectra of the  $\text{Dy}^{3+}$ -doped borate and boro-phosphate host glasses, respectively. The optical absorption spectra of  $\text{Dy}^{3+}$ -doped glasses are divided into two sets of manifolds in the UV–Vis (350–550 nm) and near-infrared (715–2000 nm) ranges [25]. The comparatively strong spin-allowed transitions that dominate the near-infrared region of the spectrum are its defining feature. Four absorption bands are originating from the ground state ( ${}^6\text{H}_{11/2}$ ) of the  $\text{Dy}^{3+}$  traced out in the intermediate spectral region of both the glass matrixes. The absence of bands in a lower wavelength region (UV–Vis) is caused by the strong absorption of the hosts via the closed packing of heavy metal ions. In addition, it was observed that the absorption peak intensity increased and resolved their positions while increasing the  $\text{Dy}^{3+}$  ion concentration in the glass matrices. The four observed dominant absorption band locations and their excited states are as follows: 801 nm ( ${}^6\text{F}_{5/2}$ ), 900 nm ( ${}^6\text{F}_{7/2}$ ), 1086 nm ( ${}^6\text{F}_{9/2}$ ), and 1262 nm ( ${}^6\text{F}_{11/2}$ ). The positions of the absorption bands in the two host glasses are nearly identical. The ligand environment has an impact on the location and intensities of several transitions of  $\text{Dy}^{3+}$  ions. The transition  ${}^6\text{H}_{11/2} \rightarrow {}^6\text{F}_{11/2}$  has a relatively high intensity compared to the others, and it is termed to be a hypersensitive transition that obeys the selection rules  $\Delta L \leq 2$ ,  $\Delta S = 0$ , and  $\Delta J \leq 2$ . The visible region's bands are spin-prohibited and have a weak cross-section, at least up to 400 nm, and thus pose a serious obstacle to the emergence of effective visible laser media.

The optical band gap energies are calculated using the diffused reflectance data of  $\text{BDy}_x$  and  $\text{BPDy}_x$  glasses in the Kubelka–Munk (K-M) theory [26]. The K-M function  $F(R)$  in this case is related to the absorption coefficient ( $\alpha$ ),

**Table 1** FT-IR and FT-Raman spectroscopic vibrational assignments for the BDy0.5 glass (borate host)

Wavenumber (cm <sup>-1</sup> )		Vibrational assignments
FT-IR	FT-Raman	
–	102	Symmetric stretching vibration of heavy metal ions including Sr–O and Ti–O bonds
702	–	Bending vibration of the B–O–B
–	709	$\gamma$ -vibration of [BO <sub>3</sub> ] <sup>3-</sup> /symmetric breathing vibration of boroxol rings with two BO <sub>4</sub> groups
–	794	Symmetric breathing vibrations of boroxol ring containing one BO <sub>4</sub> units (penta-, tetra-, or triborate) and the symmetric stretching of [BO <sub>3</sub> ] <sup>3-</sup> units
–	852	B–O–B symmetric stretching in pyroborate units
870	870	B–O bond stretching vibrations of tetrahedral BO <sub>4</sub> units and symmetric stretching of [BO <sub>3</sub> ] <sup>3-</sup> units
–	993	B–O <sup>-</sup> symmetric stretching vibrations in pentaborate (B <sub>5</sub> O <sub>8</sub> <sup>-</sup> ) and pyro-borate (B <sub>3</sub> O <sub>5</sub> <sup>4-</sup> ) anionic groups
1020	–	Pentaborate along with the BO <sub>4</sub> tetrahedral stretching vibrations
–	1208	Asymmetric stretching vibration of [BO <sub>3</sub> ] <sup>3-</sup> units
1214	–	B–O asymmetric stretching vibration in orthoborate group
–	1281	B–O <sup>-</sup> symmetric stretching vibrations in pyroborate (B <sub>3</sub> O <sub>5</sub> <sup>4-</sup> ) anionic groups
1346	–	B–O stretching vibrations of BO <sub>3</sub> trigonal units in ortho-, pyro-, and meta-borate units
–	1353	Stretching vibrations of BØ <sub>2</sub> O <sup>-</sup> triangles linked to BØ <sub>4</sub> <sup>-</sup> units
–	1421	B–O stretching vibrations of the meta-borate (BO <sub>2</sub> <sup>-</sup> ) chains and rings
1740	–	Bending vibrations of the hydroxyl group
2852, 2928	–	Stretching vibration of –OH molecules

**Table 2** FT-IR and FT-Raman spectroscopic vibrational assignments for the BPDy0.5 glass (boro-phosphate host)

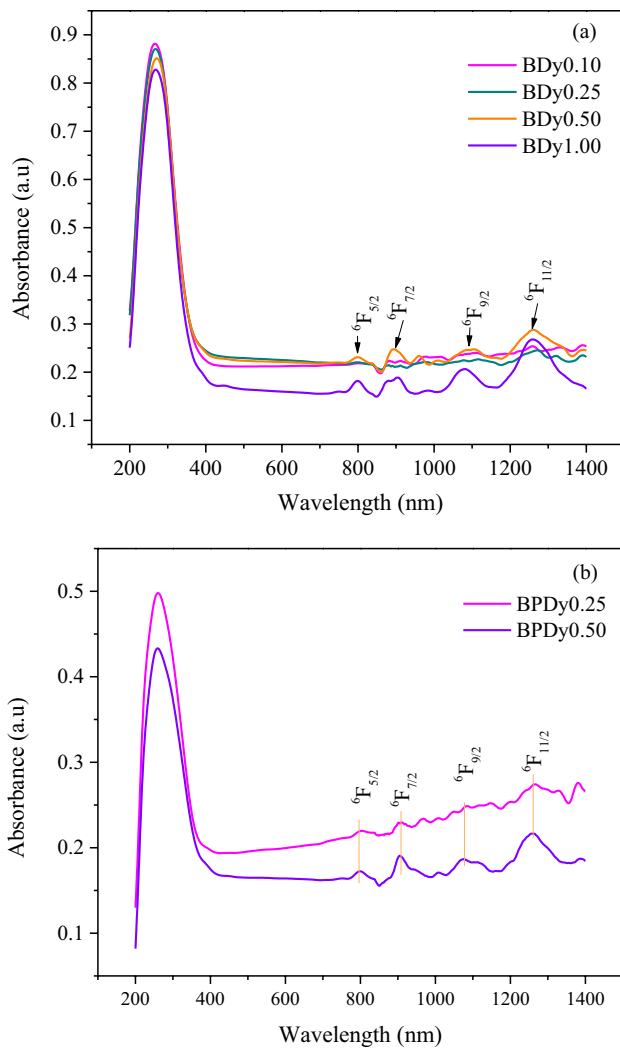
Wavenumber (cm <sup>-1</sup> )		Vibrational assignments
FT-IR	FT-Raman	
–	91	Vibration of heavy metal ions including Sr–O and Ti–O bonds
–	238	PO <sub>2</sub> bending vibration
–	318	Bending motions in O–P–O linkages
–	463	Independent di-borate groups vibrations,
–	574	O–P–O bending vibrations of PO <sub>4</sub> <sup>3-</sup> tetrahedral units
692	–	P–O–P bending vibrations
–	713	P–O–P symmetric stretching vibrations
–	995	Orthophosphate (PO <sub>4</sub> <sup>3-</sup> ) group symmetric stretching vibration of the Q <sup>0</sup> units
–	1018	P–O symmetric stretching vibration of Q <sup>1</sup> terminal group
1038	–	Combined vibrations about the symmetric stretching of B–O–P bridge between BO <sub>4</sub> and PO <sub>4</sub> units and B–O stretching vibrations in BO <sub>4</sub> tetrahedra
–	1130, 1230	Symmetric and asymmetric stretching of non-binding (PO <sub>2</sub> ) of Q <sup>2</sup> units
–	1290	Combined vibration of borate and phosphate groups
–	1338	P=O stretching vibration of terminal oxygen
1386	–	Stretching vibrations of Q <sup>2</sup> ([P(OP) <sub>2</sub> (O <sup>-</sup> ) <sub>2</sub> ] and BO <sub>3</sub> units
–	1434	Stretching vibration of molecular oxygen bands and B–O in non-binding oxygen of [BO <sub>3</sub> ] <sup>3-</sup> triangles
–	1617	OH <sup>-</sup> bending vibration
2000–2500	–	hydrogen bonding in B–OH and P–OH units caused by the presence of hydroxyl group

diffusion coefficient ( $S$ ), and reflectance ( $R$ ) of an infinitely thick layer, as given below:

$$F(R) = \frac{\alpha}{S} = \frac{(1-R)^2}{2R} \quad (1)$$

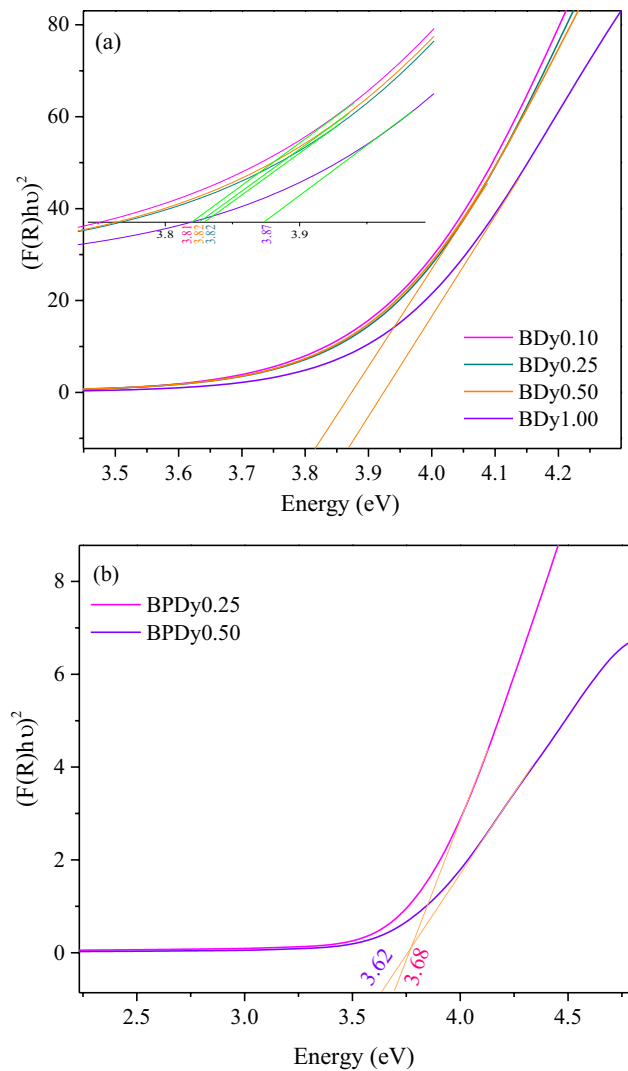
$$F(R) hv = B(hv - E_g)^\gamma \quad (2)$$

Here, the values of  $\gamma=2$  and  $1/2$  for indirect and direct transitions are measured by extrapolating the linear sections of the curve to intersect the  $h\nu$  axis at  $(F(R)h\nu)^{1/2}=0$  and  $(F(R)h\nu)^2=0$ , respectively, as shown in Figs. 4 and



**Fig. 3** UV–Vis spectral analysis: optical absorption of (a)  $\text{BDy}_x$  ( $x=0.10, 0.25, 0.50,$  and  $1.00$  mol%) and (b)  $\text{BPDy}_x$  ( $x=0.25$  and  $0.50$  mol%)

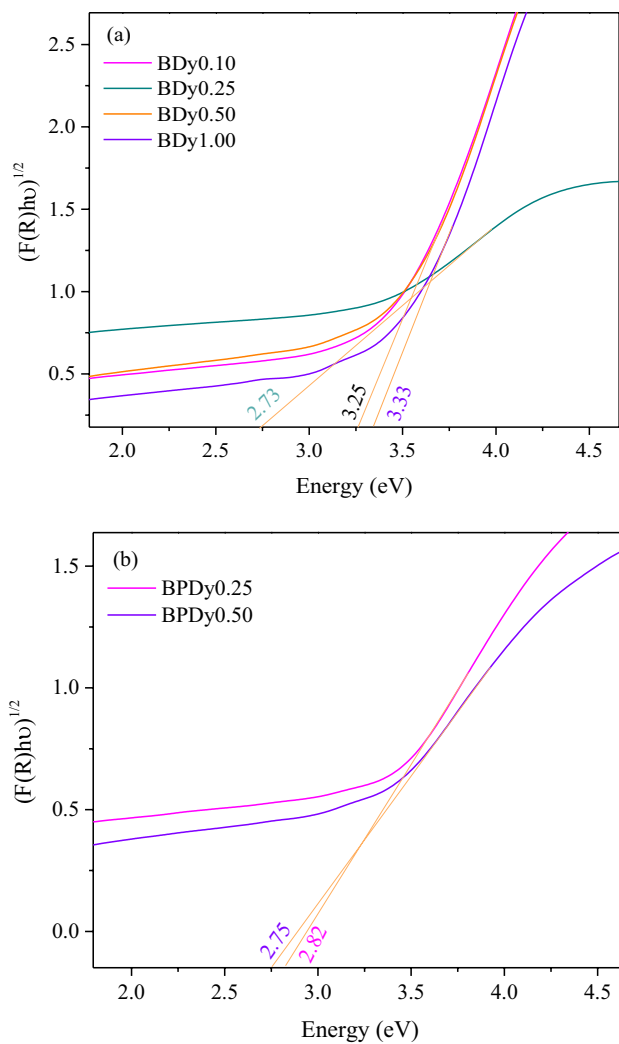
5. The calculated values of direct and indirect band gap energy of the glasses vary from 3.81 to 3.87 eV and 2.73 to 3.33 eV, respectively, with an increase in  $\text{Dy}^{3+}$  concentration in borate host matrixes ( $\text{BDy}_x$ ), as shown in Figs. 4a and 5a. At initial, the band gap was low as  $\text{Dy}_2\text{O}_3$  generated non-bridging oxygens in the borate host matrix; the optical band gap was increased with the further addition of  $\text{Dy}_2\text{O}_3$ , which was formed by the substitution of boron ions (0.85 Å) with greater atomic radii (0.91 Å)  $\text{Dy}^{3+}$  ions. The optical characteristics of the glasses changed when  $\text{Dy}^{3+}$  ions were added to the lattice owing to more energy levels being generated within the band gap. In the borate glass matrix, the band gap energy increases with the  $\text{Dy}^{3+}$  ion concentration (Table 3) by contributing additional conduction electrons, which then occupy the conduction band at energies greater than the conduction band minimum. As a result, the larger



**Fig. 4** Kubelka–Munk plot: direct band-gap energy of (a)  $\text{BDy}_x$  ( $x=0.10, 0.25, 0.50,$  and  $1.00$  mol%) and (b)  $\text{BPDy}_x$  ( $x=0.25$  and  $0.50$  mol%)

energy differences between conduction and valence bands also influenced the absorption and emission. The energy difference between these bands in an indirect band gap material also includes the change of electron momentum, which is associated with the presence of phonon.

This behavior change could be associated with structural changes in the matrix that modify binding strength. However, as the concentrations of  $\text{Dy}^{3+}$  in the boro-phosphate host matrixes increase, the band gap energies decrease from 3.68 to 3.62 eV (direct band gap) and 2.82 to 2.75 eV (indirect band gap), as shown in Figs. 4b and 5b. According to Safeya et al., the rate of conversion of  $[\text{BO}_3] \rightarrow [\text{BO}_4]$  increases the inclusion of modifier oxides into the borate host structure [27]. Whereas in the phosphate host, the inclusion of modifier oxides results in the high formation of



**Fig. 5** Kubelka–Munk plot: indirect band-gap energy of (a)  $\text{BDy}_x$  ( $x=0.10, 0.25, 0.50$ , and  $1.00$  mol%) and (b)  $\text{BPDy}_x$  ( $x=0.25$  and  $0.50$  mol%)

negative non-bridging oxygen, which is balanced with the positive cations.

In an amorphous structure, the absorption edge indicates the presence of inter-band transition tails, and Urbach energy ( $\Delta E$ ) is a crucial factor that determines the level of disordering in the networks; it was calculated using the relation:

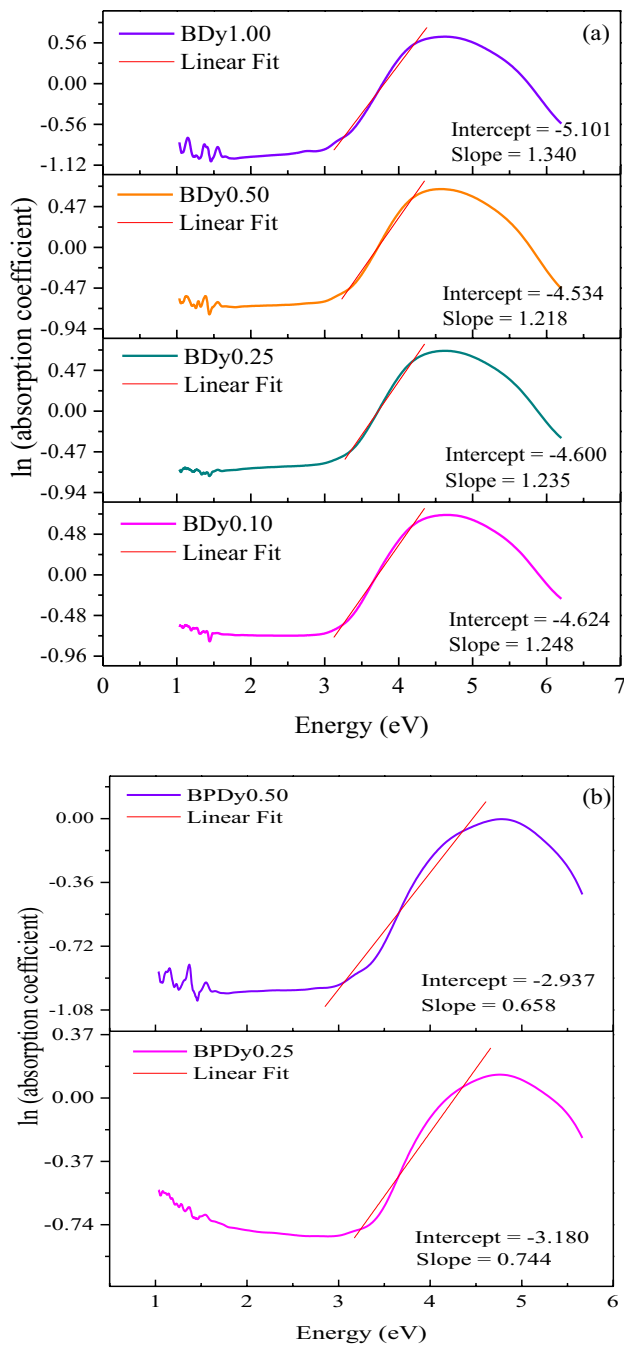
$$\alpha(\nu) = \alpha_0 \exp(h\nu / \Delta E) \quad (3)$$

The quantity of  $\Delta E$  signifies the width of localized states, including vacancies, defects due to concentrations, and dangling bonds. As a result, determining the Urbach energy and characterizing the degree of material imperfection are made possible by analyzing the optical absorption spectrum region corresponding to the density of state tails. The Urbach energy measures the steepness of the absorption beginning close to the band edge and, in consequence, the width of the density states. The Urbach energy ( $\Delta E$ ) values ( $\sim 0.80$  eV) were measured from the slope of the linear section of both the glass matrixes and are displayed in Fig. 6a and b. Table 3 displays the measured Urbach's energy values of both  $\text{BDy}_x$  and  $\text{BPDy}_x$  glass matrixes. The measured  $\Delta E$  value decreases in borate host glasses, whereas its value increases in boro-phosphate glasses with an increase in  $\text{Dy}^{3+}$  ion concentrations. The low value of  $\Delta E$  values suggested that the prepared borate host structures have a relatively high organized structure, and it is indicated by a stronger absorption onset. Since  $h$  energy and the band gap energy  $E_g$  have an inverse relationship, a sample with a larger band gap is anticipated to have a smaller band tail. Furthermore, the lower Urbach energy of the borate-host glasses implies a reduced disorder of phonon states, hence enhancing the emission features including its purity. The increase in  $\Delta E$  values in  $\text{BPDy}_x$  glasses is due to the increased quantity of non-bridging oxygens and is ascribed to the short-range order of their structure.

The linear refractive index ( $n$ ) was calculated from the indirect optical band gap energy ( $E_g$ ) of the glasses using:

**Table 3** Linear ( $E_g$ ,  $n_1$ ) and non-linear optical ( $\chi^{(1)}$ ,  $\chi^{(3)}$ , and  $n_2$ ) characteristics of  $\text{BDy}_x$  ( $x=0.10, 0.25, 0.50$ , and  $1.00$ ) and  $\text{BPDy}_x$  ( $x=0.25$  and  $0.50$ )

Glass	$E_{g\text{-dir}}$ (eV)	$E_{g\text{-indir}}$ (eV)	$\Delta E$ (eV)	$n_1$	TPA (cm/GW)	$\chi^{(1)}$ esu	$\chi^{(3)} \times 10^{-12}$ esu	$n_2 \times 10^{-11}$
Dy doped in borate host								
BDy0.10	3.81	3.25	0.801	2.332	10.435	0.3533	2.6509	4.2821
BDy0.25	3.82	2.73	0.809	2.473	14.645	0.4073	4.6786	7.1286
BDy0.50	3.82	3.25	0.821	2.332	10.435	0.3533	2.6509	4.2821
BDy1.00	3.87	3.33	0.746	2.365	9.787	0.3657	3.0406	3.4047
Dy doped in boro-phosphate host								
BPDy0.25	3.68	2.82	1.344	2.447	13.918	0.3971	4.2279	6.5103
BPDy0.50	3.62	2.75	1.519	2.467	14.485	0.4049	4.5711	6.9818
DFBP [53]	2.88	2.69	0.418	2.48				



**Fig. 6** Urbach energy of (a)  $\text{BDy}_x$  ( $x=0.10, 0.25, 0.50,$  and  $1.00$  mol%) and (b)  $\text{BPDy}_x$  ( $x=0.25$  and  $0.50$  mol%)

$$\frac{n^2 - 1}{n^2 + 2} = 1 - \sqrt{\frac{E_g}{20}} \quad (4)$$

The refractive index dispersion of  $\text{BDy}_x$  and  $\text{BPDy}_x$  glasses is shown in Table 3. It is noticeable that increasing the concentration of  $\text{Dy}^{3+}$  ions increased the refractive index. According to Wemple, materials with a covalent

nature display high microscopic oscillator strength due to the substantial band-broadening of the orbitals of molecules, which results in a high refractive index [28]. Therefore,  $\text{BDy0.25}$  and  $\text{BPDy0.50}$  in borate and borophosphate host matrixes have a high refractive index compared to other prepared glasses in their hosts. Fanderlik [29] reported that the refractive index of the glass is directly related to the glass density, and the glass containing  $\text{Al}_2\text{O}_3$  has a comparatively large ion size as well as acting as a glass modifier and not a glass former in the studied glasses. The presence of  $\text{Al}^{3+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Ti}^{2+}$ , and  $\text{Sr}^{3+}$  ions is responsible for the greater levels of density and electronic polarizability shown in these glass systems. For the boro-phosphate ( $\text{B}_2\text{O}_3\text{-P}_2\text{O}_5$ ) host glasses with free  $-\text{OH}$  groups, a higher index of refraction and more non-bridging oxygen generation lead to a denser structure. Many authors discovered that the system containing  $\text{Li}_2\text{CO}_3$  exhibits the highest degree of refraction. Because the refractive index is affected not only by polarizability but also by the molar volume  $M/\rho$ , and as a result, a dense structure is predicted as the molar volume decreases [30, 31]. The high refractive index of transition metal-containing glasses including  $\text{Sr}_2\text{O}_3$  and  $\text{Ti}_2\text{O}$  may be due to these ions' ability to reduce the average oxygen coordination number. This causes the structure to become more compact, which is then reflected in the rise in refractive index.

The two-photon absorption (TPA) coefficient is an important metric that may be determined by employing indirect optical band gap energy values, and it was used to evaluate the optical switching performance in waveguides [32].

$$\text{TPA} = 36.76 - 8.1E_g \text{ (cm/GW)} \quad (5)$$

TPA coefficient of composition  $\text{BDy1.0}$  glass is low (9.787 cm/GW), implying their application in waveguides, and measured TPA values (presented in Table 3) demonstrate that TPA may be modified by influencing the optical band gap [33].

### 3.4 Non-linear optical characterization

Non-linear optical features are caused by non-linear electric polarization, which can be described as follows:

$$P = \chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \dots \quad (6)$$

where  $P$  represents the electric polarization, and  $\chi$  represents optical susceptibility. In a glass structure with inversion symmetry,  $\chi^{(1)}$  is theoretically zero. As a result, the non-linear optical response in glass is dominated by  $\chi^{(3)}$ . The addition of RE ions to the glass matrix affords them a non-linear optical characteristic. The following equation relates first-order linear optical susceptibility  $\chi^{(1)}$  and third-order



non-linear optical susceptibilities  $\chi^{(3)}$  and linear refractive indices ( $n_1$ ) and non-linear refractive indices ( $n_2$ ) [34]:

$$\chi^{(1)} = \frac{n_1^2 - 1}{4\pi} \quad (7)$$

$$\chi^{(3)} \cong \text{Re } \chi^{(3)} = [\chi^{(1)}]^4 \times 1.7 \times 10^{-10} \text{ esu} \quad (8)$$

$$n_2 = \frac{12\pi \text{Re } \chi^{(3)}}{n_1} \quad (9)$$

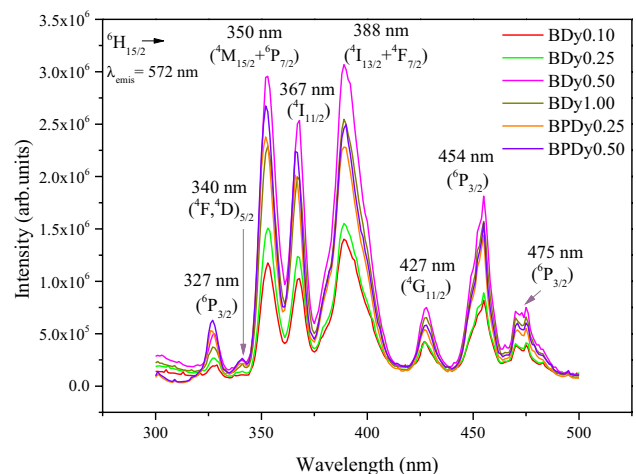
Figure 5b depicts the variation of  $\chi^{(3)}$  and  $n_2$  with the various concentrations of  $\text{Dy}^{3+}$  ions in the BDyx glasses. Miller's rule states that the greater the refractive index, the greater the value of  $\chi^{(3)}$  [35]. As a result, the high  $\chi^{(3)}$  in BDy0.25 from the borate host and BPDy0.50 from the boro-phosphate is caused by the covalent nature of the metal–oxygen bond. The non-linear behavioral patterns of the curves suggested that the concentration of  $\text{Dy}^{3+}$  influenced the non-linear optical characteristics of prepared glass systems. Alkali cations can influence the total  $\chi^{(3)}$  value in alkali borate and boro-phosphate glasses because  $\chi^{(3)}$  for pure  $\text{B}_2\text{O}_3$  glass is quite small ( $= 2.88 \times 10^{-14}$  esu) [36], as shown in Table 3. Adir et al. expected that the glasses containing ions with high polarizability would exhibit a substantial  $\chi^{(3)}$  value. The fact that the value of  $\chi^{(3)}$  steadily rises as  $E_g$  falls suggests that  $E_g$  is the key factor of the  $\chi^{(3)}$  value of all the glasses in the present work. Due to the reduced band gap, there can be an impact caused by optical resonance enhancement. Dysprosium-doped boro-phosphate glasses exhibit higher optical non-linearity than borate glasses due to one-photon absorption [37]. The combined contribution of the  $\text{Dy}^{3+}$  ion and heavy-metal oxide ions ( $\text{Ti}^{2+}$  and  $\text{Sr}^{3+}$ ) with high polarizability could further increase  $\chi^{(3)}$ .

The two types of resonance effects that rise in  $\chi^{(3)}$  value are the pre-resonance effect and the rigorous-resonance effect. The rigorous resonance effect termed as  $\chi^{(3)}$  values is enhanced dramatically due to the large transition moment to the first excited state. According to Nasu et al. [38], the field strength of rare-earth cations has a significant impact on the non-linearity. The large optical non-linearity is caused by the small field strength ions' inability to bind valence electrons as tightly. The optical band gap value for these glasses is substantially higher than the energy of a photon in the third harmonic beam ( $E = 1.96$  eV). As a result, the pre-resonance effect can be the primary factor in increasing the  $\chi^{(3)}$  in both the host glasses. Table 3 illustrates the linear and non-linear optical parameters of BDy<sub>x</sub> ( $x = 0.10, 0.25, 0.50,$  and  $1.00$ ) and BPDy<sub>x</sub> ( $x = 0.25$  and  $0.50$ ) glasses. The calculated non-linear refractive index ( $n_2$ ) value of BDy<sub>0.25</sub>, BPDy<sub>0.25</sub> and BPDy<sub>0.50</sub> glasses were found to be higher than

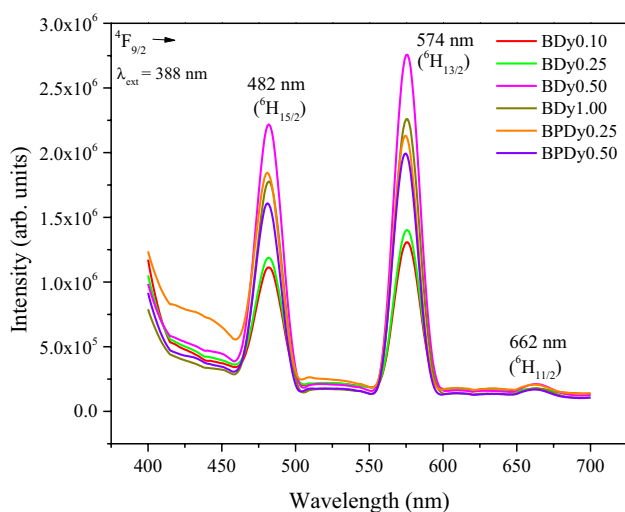
the other prepared glasses which suggests that these glasses can be utilized in light-regulated optical devices.

### 3.5 Photoluminescence

The excitation spectra of all the samples were monitored for the emission wavelength of 574 nm. The spectra reveal three predominant peaks at 350, 367, and 388 nm and other minor peaks at 327, 340, 427, 454, and 475 nm, which correspond to transitions of  $\text{Dy}^{3+}$  ions from the  ${}^6\text{H}_{15/2}$  state to the  ${}^4\text{M}_{15/2} + {}^6\text{P}_{7/2}$ ,  ${}^4\text{I}_{11/2}$ ,  ${}^4\text{I}_{13/2} + {}^4\text{F}_{7/2}$ ,  ${}^6\text{P}_{3/2}$ ,  $({}^4\text{F}, {}^4\text{D})_{5/2}$ ,  ${}^4\text{G}_{11/2}$ ,  ${}^4\text{I}_{15/2}$ , and  ${}^6\text{F}_{9/2}$  states, respectively, as shown in Fig. 7. The emission spectra were recorded in the 400–700 nm range for the excitation wavelength of 388 nm and are shown in Fig. 8. There are a large number of f-block energy states in  $\text{Dy}^{3+}$  with  ${}^4\text{F}_9$  meta-stable configuration. The luminescent spectrum reveals two strong emission bands at 482 nm due to magnetic-dipole (MD) transition  ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{15/2}$  (blue), and 574 nm corresponds to the electric-dipole (ED) transition  ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{13/2}$  (yellow) strongly influenced by the surrounding environment. The emission at 662 nm is attributed to  ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{11/2}$  (red) transitions and is found to be relatively weak. It was discovered that the emission intensity of the ED transition is greater than the MD transition for the present glasses, and it follows the selection rules  $\Delta L = \pm 2$  and  $\Delta j = \pm 2$ . However, in comparison to the other two identified emission bands, the emission band in the 662 nm has a low intensity. Among those observed, the  ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{13/2}$  transition is hypersensitive, and the ligand field surrounding the  $\text{Dy}^{3+}$  ion site has a significant impact on its strength. Typically, the host has a minute influence on the intensity of the hypersensitive transition, or it changes only slightly from host to host, whereas the intensity of the magnetic-dipole



**Fig. 7** Photo-excitation spectra of BDy<sub>x</sub> ( $x = 0.10, 0.25, 0.50,$  and  $1.00$  mol%) and BPDy<sub>x</sub> ( $x = 0.25$  and  $0.50$  mol%)



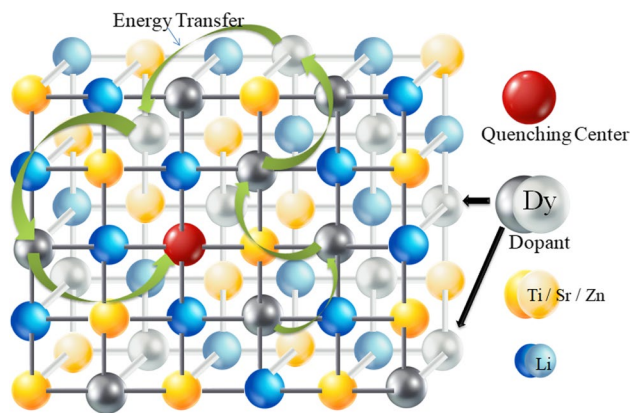
**Fig. 8** Photoluminescence spectra of  $\text{BDy}_x$  ( $x=0.10, 0.25, 0.50$ , and  $1.00$  mol%) and  $\text{BPDy}_x$  ( $x=0.25$  and  $0.50$  mol%)

transition varies significantly from host to host [39]. Additionally, the emission of white light has a greater influence on the intensity variation between transitions. The color of the luminescence is nearly white, if the emission intensities of the two transitions ( ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{15/2}, {}^6\text{H}_{13/2}$ ) are identical, for instance, in the systems  $\text{Ba}_3\text{Bi}(\text{PO}_4)_3:\text{Dy}^{3+}$ ,  $\text{BaY}_2\text{ZnO}_5:\text{Dy}^{3+}$ , and  $(\text{Gd}0.98\text{Dy}0.02)(\text{BO}_2)_3$  [40, 41].

In borate glass system, the intensity of luminescence emission is directly related to their indirect band gap energy. The prepared glasses containing 0.5 and 1 mol% of  $\text{Dy}^{3+}$  ion concentration have a relatively large band gap. These samples absorb large quantities of high-energy photons, which cause the high-intensity emission. The wider energy gap below the  ${}^4\text{G}_{5/2}$  state of  $\text{Dy}^{3+}$  ion-doped glass substantially reduces the multi-phonon transition rate, even in glass with high-phonon energy. Moreover, the elevated energy states of  $\text{Dy}^{3+}$  ions impact three emission bands, leading to emission within the visible region.

From Fig. 8, it can be observed that the luminescence intensity rises as the  $\text{Dy}^{3+}$  ion concentration rises to 0.5 wt% and 0.25 wt% in borate and boro-phosphate host glasses, respectively. As the concentration of  $\text{Dy}^{3+}$  ions increases in the glass network, the inter-ionic distance decreases gradually, and the exchange interaction among the  $\text{Dy}^{3+}$  ions becomes robust. When the inter-ionic distance of  $\text{Dy}^{3+}$ - $\text{Dy}^{3+}$  ions is smaller than the critical distance, the tunneling effect is easily activated, leading to cascade energy transfer and luminescence quenching [42]. Figure 9 illustrates the schematic representation of the concentration quenching mechanism in  $\text{Dy}^{3+}$ -doped host glasses.

Z. Zhu et al. reported that a multi-polar interaction plays an essential role in the concentration quenching mechanism of  $\text{Dy}^{3+}$  [43]. According to Van Uitert's report, the



**Fig. 9** Schematic representation of concentration quenching mechanism of  $\text{Dy}^{3+}$ -doped glass matrix

relationship between activator ion concentration and their emission intensity can be represented as follows [44]:

$$I/x = k(1 + \beta(x)^{\theta/3})^{-1} \quad (10)$$

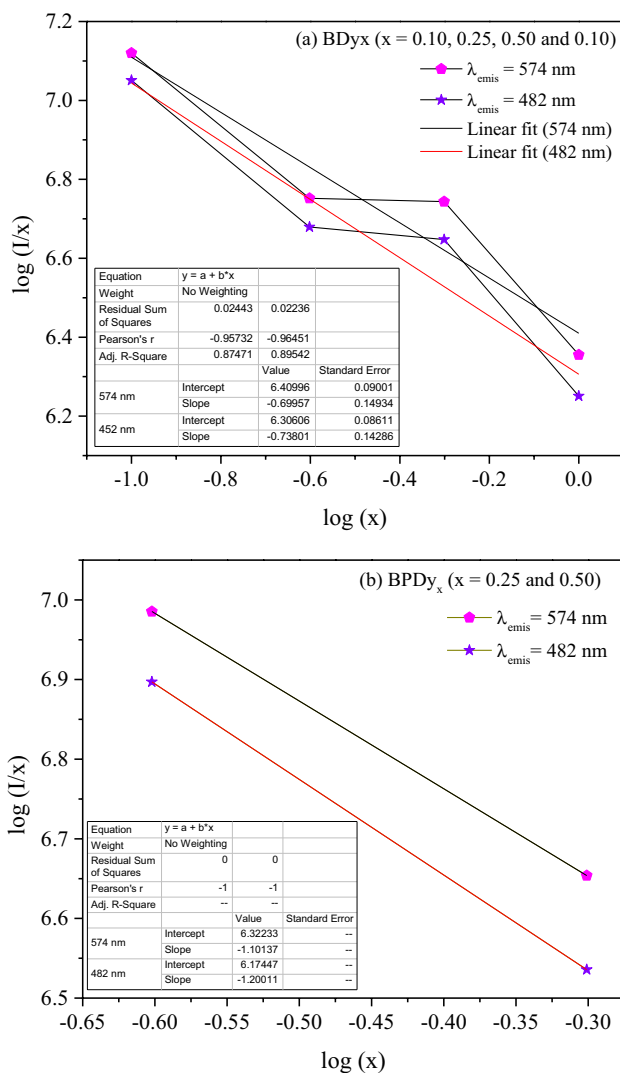
where  $x$  = concentration of activator ion;  $I$  = intensity of emission;  $k$  and  $\beta$  are constant for a given host network under the identical excitation condition; and  $\theta=6, 8$ , and  $10$  values correspond to the different multi-polar interactions. Considering that  $\beta(x)\theta/3 \gg 1$ , then Eq. (10) can be rewritten as:

$$\text{Log}(I/x) = K' - (\theta/3) \log(x) \text{ and } K' = \log k - \log \beta \quad (10)$$

The relation between  $\log(x)$  and  $\log(I/x)$  and the accompanying straight line has a slope that signifies  $-\theta/3$ , as shown in Fig. 10a and b for  $\text{BDy}_x$  and  $\text{BPDy}_x$  glass, respectively. Herein, the computed average value (both the emission) of  $\theta/3$  is 0.718, and  $\theta$  can be measured as 2.155 for  $\text{BDy}_x$  glass, whereas the value is 1.150, and  $\theta$  can be measured as 3451 for  $\text{BPDy}_x$  glass. The  $\theta$  value for both glass matrixes was found to be less than 6. The finding indicates that the dipole–dipole (D–D) interaction influences the concentration quenching mechanism of  $\text{Dy}^{3+}$  emission in borate and boro-phosphate hosts [43, 45]. Table 4 specifies the magnitude of  $-\theta/3$  for the individual emission bands of both host glasses.

### 3.6 Y/A ratio and decay profile

The dominance of yellow emission is the result of a hypersensitive transition, which specifies that the  $\text{Dy}^{3+}$  ions are positioned at low-symmetry sites without an inversion center. The Y/B value is proportional to the  $\text{Dy}^{3+}$  concentrations, and it is found to be 1.233–1.352 and 1.218–1.298 for the  $\text{BDy}_x$  ( $x=0.10, 0.25, 0.50$ , and  $1.00$ ) and  $\text{BPDy}_x$  ( $x=0.25$  and  $0.50$ ) glasses, respectively. According to Arunkumar et al. [46], a rise in the covalency between  $\text{Dy}^{3+}$ - $\text{O}^{2-}$  ions and the atoms surrounding them in the glasses matrix is



**Fig. 10** (a, b) Linear fitting of  $\log(x)$  vs.  $\log(I/x)$  for borate ( $\text{BDy}_x$ ,  $x = 0.10, 0.25, 0.50$ , and  $1.00$ ) and boro-phosphate ( $\text{BPDy}_x$ ,  $x = 0.25$  and  $0.50$ ) glasses

indicated by a ratio of Y/B increases. As a result, while the concentration of  $\text{Dy}^{3+}$  ions in the prepared glasses increases, it consequently increases the covalency of the Dy–O bond. The value of Y/B changes when  $\text{Dy}^{3+}$  is introduced in place of an element with a different valency ( $\text{Al}^{3+/2+}$ ,  $\text{Li}^{3+}$ ,  $\text{Sr}^{2+}$ ,  $\text{Ti}^{2+}$ , and  $\text{Zn}^{2+}$ ), and this change is proportional to variations in the concentration of  $\text{Dy}^{3+}$ . The local symmetry of  $\text{Dy}^{3+}$  will change with an increase in  $\text{Dy}^{3+}$  concentration because defects are produced in this circumstance. According to the gradual increase in Y/B ratios, the present glasses can produce blue or white light depending on the concentration of  $\text{Dy}^{3+}$  [47, 48].

The PL decay curves of  $\text{BDy}_x$  ( $x = 0.25$  and  $0.50$ ) and  $\text{BPDy}_x$  ( $x = 0.25$  and  $0.50$ ) glasses are displayed in Fig. 11. For all the prepared glass networks, the decay rates of

emission exhibit a non-exponential configuration. The  $\text{Dy}^{3+}$  non-exponential decay curves were fitted to the subsequent tri-exponential function:

$$I(t) = A_1 \exp\left(-\frac{t}{\tau_1}\right) + A_2 \exp\left(-\frac{t}{\tau_2}\right) + A_3 \exp\left(-\frac{t}{\tau_3}\right) \quad (12)$$

where,  $\tau_1$ ,  $\tau_2$ , and  $\tau_3$  are the fluorescence lifetime components that undergo calculating the average lifespan, while  $A_1$ ,  $A_2$ , and  $A_3$  stand for the amplitudes of the corresponding decay components. The expression that follows can be used to determine the average lifespan for a non-exponential with three components [49].

$$\tau_{avg} = \frac{A_1 \tau_1^2 + A_2 \tau_2^2 + A_3 \tau_3^2}{A_1 \tau_1 + A_2 \tau_2 + A_3 \tau_3} \quad (13)$$

The  $\text{Dy}^{3+}$  ion concentration influences the  ${}^4\text{F}_{9/2}$  fluorescence lifetime. For  $\text{BDy}_x$  glasses, the mean lifespan values decreased from 1.490 to 1.190 ms with an increase in  $\text{Dy}^{3+}$  dopant concentration; however, the value of 1.082 ms remained the same for  $\text{BPDy}_x$  glasses. Effective lifetimes tend to be shortened as  $\text{Dy}^{3+}$  concentration rises; this is likely due to energy transfer within luminescence quenching centers, via the cross-relaxation (CR) process, and energy transfer from impurities [50]. In the present study, this behavior is explained by a substantial quenching as the dopant concentration increases and the distance between consecutive ions decreases. Therefore, there is a higher chance that the resonant interaction of energy within the  $\text{Dy}^{3+}$  ions will increase, leading to quicker fluorescence decay [51]. The non-radiative transmission of energy via cross-relaxation is schematically represented in Fig. 12. Three cross-relaxation channels could be discovered based on the energy resonance [2]. The excited  $\text{Dy}^{3+}$  ions relaxed from  ${}^4\text{F}_{9/2}$  level to  ${}^6\text{F}_{11/2}$  (CH-1),  ${}^6\text{F}_{5/2}$  (CH-2), or  ${}^6\text{F}_{3/2}$  (CH-3) level by transmitting its energy non-radiatively to another  $\text{Dy}^{3+}$  ions in  ${}^6\text{H}_{15/2}$  level (ground state) and the energy acquired  $\text{Dy}^{3+}$  ions excite to  ${}^6\text{F}_{3/2}$  (CH-1),  ${}^6\text{F}_{9/2}$  (CH-2), or  ${}^6\text{F}_{11/2}$  (CH-3) state. All the  $\text{Dy}^{3+}$  ions will relax to their ground state non-radiatively which results in the luminescence quenching.

The Inokuti-Hirayama model was used to evaluate the decay curve to explicate the energy migration between  $\text{Dy}^{3+}$ - $\text{Dy}^{3+}$  ions [26].

$$I(t) = I_0 \exp\left[-\frac{t}{\tau} - Q\left(\frac{t}{\tau}\right)^{3/5}\right] \quad (14)$$

$I_0$  is the initial emission intensity;  $\tau$  represents the intrinsic lifetime of donors, and  $Q$  is the energy transfer parameter, which is expressed as follows:

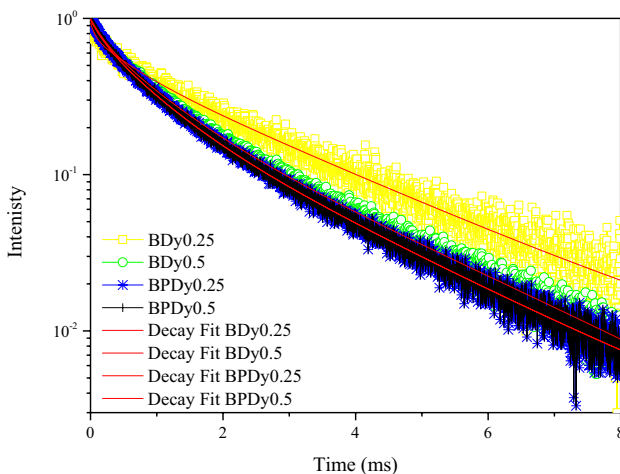
$$Q = \frac{4}{3} \pi \Gamma \left(1 - \frac{3}{5}\right) N_A R_c^3 \quad (15)$$

**Table 4** Van Uitert parameter ( $\theta$ ) and Y/B ratio of borate (BDy<sub>x</sub>,  $x=0.10, 0.25, 0.50,$  and  $1.00$ ) and boro-phosphate (BPDy<sub>x</sub>,  $x=0.25$  and  $0.50$ ) host network glasses

Glass	Dy <sup>3+</sup> (wt%)	$\lambda_{em}=482$ nm		$\lambda_{em}=574$ nm		Y/B ratio
		$-\theta/3$	$\theta$	$-\theta/3$	$\theta$	
Borate host						
BDy0.10	0.10	0.738	2.241	0.699	2.099	1.233
BDy0.25	0.25					1.262
BDy0.50	0.50					1.288
BDy1.00	1.00					1.352
Boro-phosphate host						
BPDy0.25	0.25	1.200	3.600	1.101	3.303	1.218
BPDy0.50	0.50					1.298
Dy0.1 [54]	0.10					1.66
Dy0.5 [54]	0.50					1.67
Dy1.0 [54]	1.00					1.70
DFBP [53]	1.00					1.447
BPAblLiDy 1.0 [55]	1.00					0.945

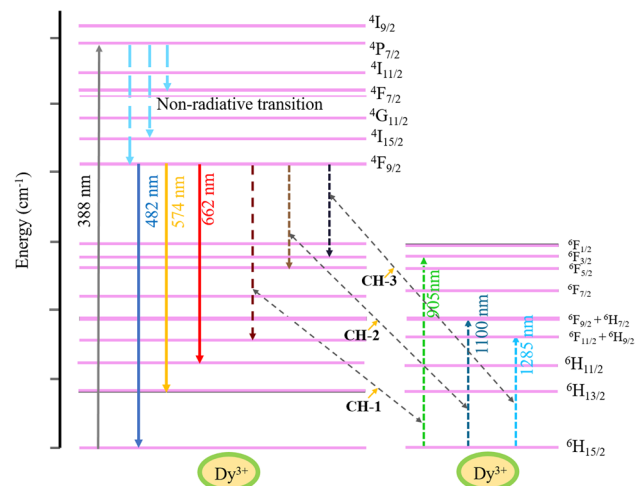
where  $\Gamma$  is Euler's gamma function, and  $S$  is the multipolar interaction parameter.  $N_A$  represents Dy<sup>3+</sup> concentration, and  $R_c$  denotes critical transfer distance. Figures 13 and 14 show the Inokuti-Hirayama fit for the decay profile of the BDy<sub>x</sub> and BPDy<sub>x</sub> ( $x=0.25$  and  $0.50$ ) glasses. The donor–acceptor ion interaction parameter ( $C_{DA}$ ) and the shortest distance between the randomly distributed RE ions can be determined from the following equations [52]:

$$C_{DA} = \frac{R_c^S}{\tau} \quad (16)$$

**Fig. 11** Fluorescence decay profile of BDy<sub>x</sub> and BPDy<sub>x</sub> glasses, where  $x=0.25$  and  $0.50$  mol%

$$D_{random} = 2 \left( \frac{3}{4\pi N_T} \right)^{1/3} \quad (17)$$

The cross-relaxation between donor and acceptor could be the predominant reason for non-mono exponential behavior in all the samples.  $D_{random} \gg C_{DA}$  suggests the cross-relaxation and energy transfer probably occur among the Dy<sup>3+</sup> clusters ions and not out of randomly distributed ions. Table 5 shows the lifetime value, energy transfer, and other related parameters derived from the PL Decay curve of BDy<sub>x</sub> and BPDy<sub>x</sub> ( $x=0.25$  and  $0.50$ ) glasses.

**Fig. 12** Energy level structure of Dy<sup>3+</sup> ion: cross-relaxation pathways through the energy resonance

### 3.7 CIE chromaticity coordinates

The color coordinates are one of the crucial components for assessing the effectiveness of the glasses' color rendering activity.

$$x = \frac{X}{X + Y + Z}, y = \frac{Y}{X + Y + Z} \text{ and } z = \frac{Z}{X + Y + Z} \quad (18)$$

where X, Y, and Z stand for the three tristimulus values and can be obtained by:

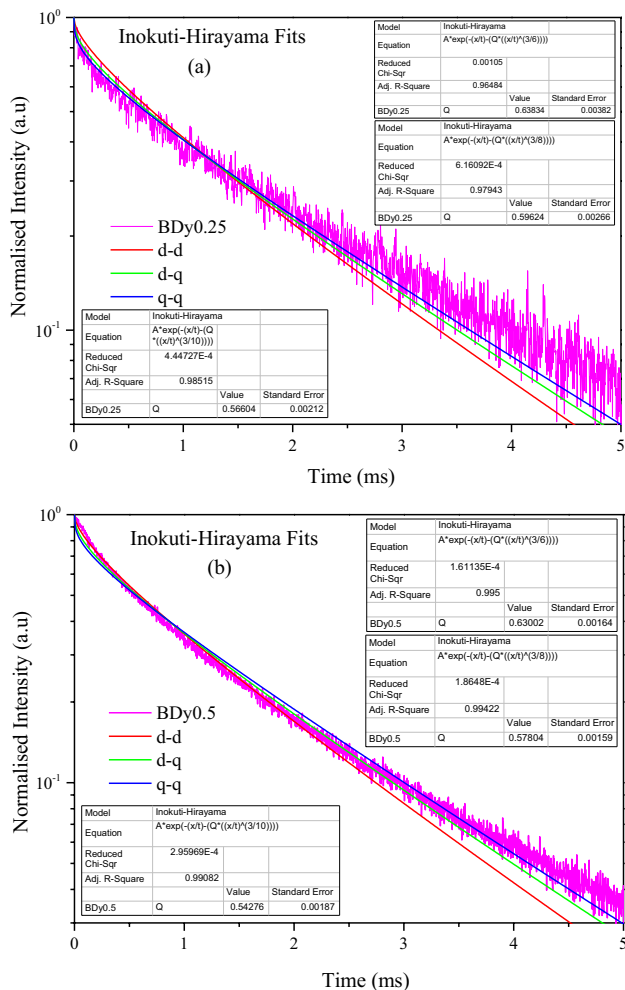
$$X = \int \bar{x}(\lambda)P(\lambda)d\lambda, Y = \int \bar{y}(\lambda)P(\lambda)d\lambda \text{ and } Z = \int \bar{z}(\lambda)P(\lambda)d\lambda \quad (19)$$

where  $\bar{x}(\lambda)$ ,  $\bar{y}(\lambda)$ , and  $\bar{z}(\lambda)$  are color-matching functions,  $\lambda$  is the wavelength of the analogous monochromatic light, and  $P(\lambda)$  is a spectral power distribution. Another significant

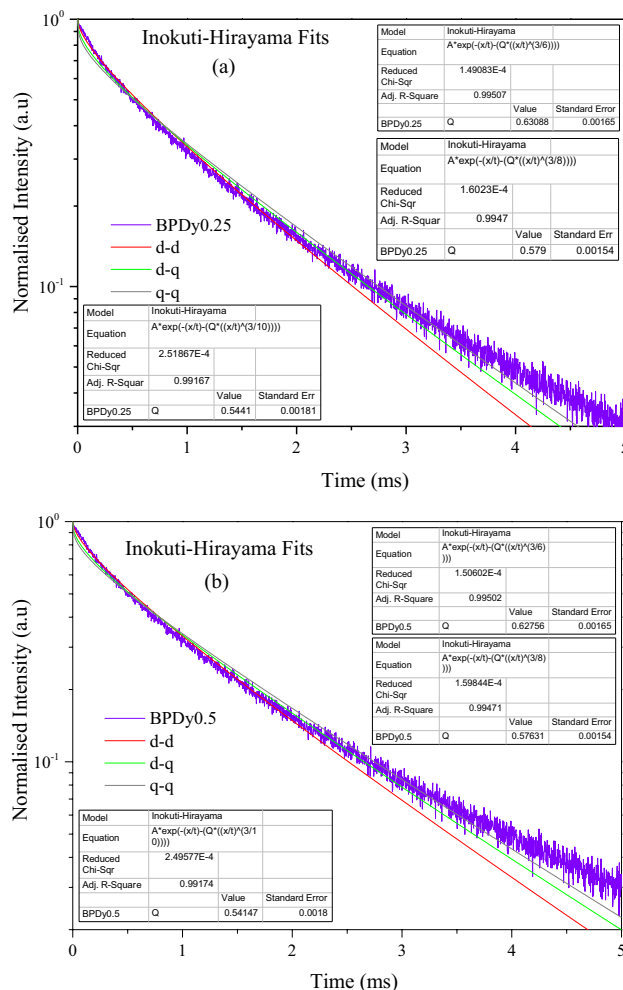
factor for better understanding sources of light for LED applications is color purity or color saturation.

$$P_c = \frac{\sqrt{((x_s - x_i)^2 + (y_s - y_i)^2)}}{\sqrt{((x_d - x_i)^2 + (y_d - y_i)^2)}} \times 100\% \quad (20)$$

where  $(x, y)$ ,  $(x_i, y_i)$ , and  $(x_d, y_d)$  represent the luminescent chromaticity coordinates, the standard white emission CIE coordinates (0.330, 0.330), and the dominant wavelength coordinates, respectively. The CIE chromaticity coordinates  $(x, y)$  of the present glasses with various concentrations of  $Dy^{3+}$  have been measured from the luminescence spectra and are discovered to (0.312, 0.314), (0.310, 0.313), (0.323, 0.335), and (0.328, 0.344) for  $BDy_x$  ( $x=0.10, 0.25, 0.50$ , and 1.00), whereas the values are (0.293, 0.293) for  $BPDy_{0.25}$  and (0.317, 0.332) for  $BPDy_{0.50}$  glass. The color purity ( $P_c$ ) of conventional white light is 0%; the  $P_c$  of the  $BDy_x$  glasses was found to vary between 9.5 and 2.2%, whereas  $P_c$  of



**Fig. 13** Inokuti-Hirayama fit of borate host: (a) BDy0.25 and (b) BDy0.50 glasses



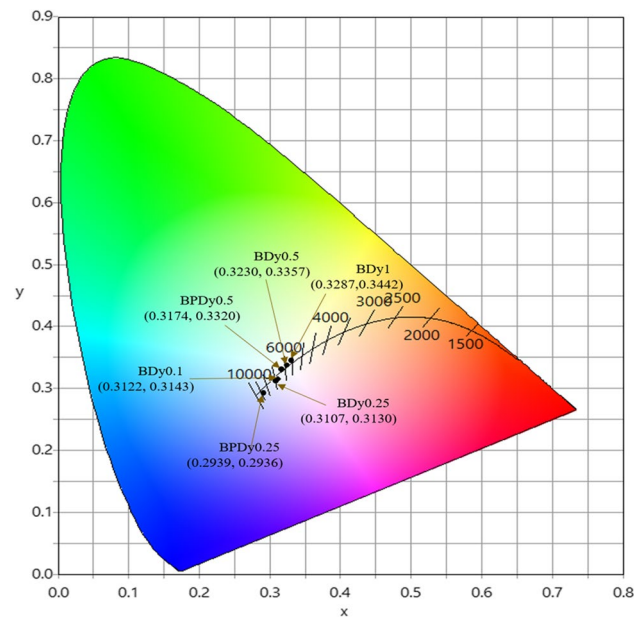
**Fig. 14** Inokuti-Hirayama fit of boro-phosphate host: (a) BPDy0.25 and (b) BPDy0.50 glasses

**Table 5** Experimental life time ( $T_{\text{exp}}$ ), concentration of the acceptor ( $N_A$ ), critical transfer distance ( $R_c$ ), donor–acceptor ions interaction parameter ( $C_{DA}$ ), and shortest distance between the randomly distributed RE ions ( $D_{\text{random}}$ ) value of BDy<sub>x</sub> and BPDy<sub>x</sub> ( $x=0.25$  and  $0.50$ ) glasses

Glass code	$T_{\text{exp}}$ (ms)	$N_A$ (ions/cm <sup>3</sup> )	$Q$	$R_c$ (Å)	$C_{DA}$ (cm <sup>6</sup> /s)	$D_{\text{random}}$ (Å)
BDy0.25	1.49	$1.112 \times 10^{20}$	0.638	9.181	$4.02 \times 10^{-40}$	25.8
BDy0.5	1.19	$2.206 \times 10^{20}$	0.63	7.276	$1.25 \times 10^{-40}$	20.5
BPDy0.25	1.08	$0.952 \times 10^{20}$	0.631	9.633	$7.40 \times 10^{-40}$	27.2
BPDy0.5	1.08	$1.887 \times 10^{20}$	0.627	7.669	$1.88 \times 10^{-40}$	21.6
Dy 0.1 [54]	0.673	-	-	-	-	-
Dy 0.5 [54]	0.521	-	-	-	-	-
Dy 1.0 [54]	0.4207	-	-	-	-	-

BPDy<sub>x</sub> glasses varied between 17.5 and 5.5%. Furthermore, the  $P_c$  value of the glasses was found to increase as Dy concentrations increased.

Figure 15 depicts the distribution of chromaticity coordinates in the CIE 1931 chromaticity diagram. The chromaticity coordinates move to the yellow-white zone when Dy<sup>3+</sup> concentration increases, indicating that warm white light is generated. Lower quantities of Dy<sup>3+</sup> in a glass demonstrated that emission occurred in the blue-white region, as validated by their predominated wavelengths ( $\lambda_d$ ) as well as the color purity ( $P_c$ ). Correlated color temperature (CCT) is an essential technique for illuminant critical features. The point of white light area achieved for BDy0.5 may be also made to correspond with reference white light by adjusting the excitation wavelengths and calcination temperature. The CCT of prepared BDy<sub>x</sub> and BPDy<sub>x</sub> glasses was computed using McCamy's relation from the chromaticity coordinates ( $x$  and  $y$ ):  $\text{CCT} = -437 n^3 + 3601 n^2 - 6861 n + 5514.31$ .



**Fig. 15** CIE 1931 chromaticity coordinates of borate (BDy<sub>x</sub>,  $x=0.10$ ,  $0.25$ ,  $0.50$ , and  $1.00$ ) and boro-phosphate (BPDy<sub>x</sub>,  $x=0.25$  and  $0.50$ ) glasses

Where  $n = (x - 0.3320)/(y - 0.1858)$ ;  $x$  and  $y$  are the CIE chromaticity coordinates. Natural white light can be classified into three broad zones based on CCT values: cool white ( $> 5000$  K), pure white ( $3700$ – $5000$  K), and warm white ( $< 3700$  K). A CCT value greater than  $5000$  K suggests cool white light from the glasses. Table 6 shows that the CCT values of borate host glasses range from  $5664$  to  $6769$  K, and those of boro-phosphate host glasses vary from  $6237$  to  $8432$  K. CCT values of more than  $5000$  K were found in Dy<sup>3+</sup>-doped borate and boro-phosphate glass networks; when compared with recent literature, the prepared glasses showed a cool white light emission.

## 4 Conclusions

Dy<sup>3+</sup> ions doped in two different hosts; borate (BDy<sub>x</sub>) and boro-phosphate (BPDy<sub>x</sub>) glasses were prepared by the melt-quenching technique. The vibrational analysis of glasses confirmed that an increase in the concentration of Dy<sup>3+</sup> ions increases the conversion of BO<sub>3</sub> units into BO<sub>4</sub> tetrahedral units, which leads to a decrease in the maximum phonon energy of the glass system and an increase in the intensity of luminescence. The BDy0.5 and BDy1.0 glasses exhibited a lower Urbach energy, indicating a

**Table 6** CIE chromaticity coordinates, color coordinates, CCT, and dominant wavelength of prepared BDy<sub>x</sub> ( $x=0.10$ ,  $0.25$ ,  $0.50$ , and  $1.00$ ) and BPDy<sub>x</sub> ( $x=0.25$  and  $0.50$ ) glasses

Glasses	$x$	$y$	CCT (K)	$\lambda_d$ (nm)	$P_c$ (%)
BDy0.10	0.3122	0.3143	6661	478.7	8.9
BDy0.25	0.3107	0.3130	6769	478.8	9.5
BDy0.50	0.323	0.3357	5942	494.7	3.3
BDy1.00	0.3287	0.3442	5664	526.7	2.2
BPDy0.25	0.2939	0.2936	8432	476.8	17.5
BPDy0.50	0.3174	0.332	6237	490.7	5.5
Dy 0.1 [54]	0.376	0.402	4247	-	-
Dy 0.5 [54]	0.374	0.397	4298	-	-
Dy 1.0 [54]	0.376	0.399	4407	-	-
DFBP [53]	0.383	0.419	4203	571.6	47.1

lesser phonon state disruption that leads to excellent emission characteristics. The resonance processes in the borate host matrix cause the BDy0.5 glass to react more prominently in non-linear optical characteristics. The PL spectra showed that a dipole–dipole interaction influences the concentration quenching over 0.5 wt% and 0.25 wt% of Dy<sup>3+</sup> dopant in a borate and boro-phosphate glass network, respectively. The Inokuti–Hirayama fit also confirms the dominance of dipole–dipole interaction energy transfer between acceptor–donor ions. With increasing Dy<sup>3+</sup> concentration, the bluish-white color of the BDy<sub>x</sub> and BPDy<sub>x</sub> glasses shifted to yellowish-white with extreme color purity. Among all the prepared glasses, BDy0.5 had a CCT value of 5942 K and color coordinates of (0.323, 0.335), indicating it is a promising cool-white light emitting glass for optical lighting applications.

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**Data availability** All data generated or analyzed during this study are included in this published article.

## Declarations

**Conflict of interest** The authors declare no competing interests.

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