

Photoluminescence and Energy Transfer Process in $\text{Bi}^{3+}/\text{Sm}^{3+}$ Co-Doped Phosphate Zinc Lithium Glasses

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Received 5 July 2014; revised 22 August 2014; accepted 7 September 2014

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Abstract

Present paper reports on luminescence characteristics of individually doped Bi^{3+} : PZL, Sm^{3+} : PZL and co-doped ($\text{Bi}^{3+}/\text{Sm}^{3+}$): PZL ($50\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiF}$) glasses prepared by a melt quenching method. The results revealed that Bi^{3+} : PZL glass exhibited a broad emission peak at 440 nm ($^3\text{P}_1 \rightarrow ^1\text{S}_0$) under excitation wavelength 300 nm ($^1\text{S}_0 \rightarrow ^3\text{P}_1$). Sm^{3+} : PZL doped glass has shown a prominent orange emission at 601 nm ($^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$) with an excitation wavelength 403 nm ($^6\text{H}_{5/2} \rightarrow ^4\text{F}_{7/2}$). Later on Bi^{3+} is added to Sm^{3+} : PZL glass by increasing its concentrations from 0.1 - 1.5 mol%. By co-doping Bi^{3+} to Sm^{3+} : PZL glass, Sm^{3+} emission intensity has been considerably enhanced till 1.0 mol% due to energy transfer from Bi^{3+} to Sm^{3+} and when its concentration exceeds this critical value (1.0 mol%) there has been a drastic decrease in Sm^{3+} emission which is explained accordingly from photoluminescence spectra, energy level diagram and lifetime measurements.

Keywords

Melt Quenching, Optical Glasses, Rare Earth Ions, Photoluminescence, Luminescence Quenching and Energy Transfer

1. Introduction

From the optical spectroscopy point of view, a remarkable progress has been observed in the development of rare earth ions as luminescent centres due to their narrow emission bands (f-f interactions) and high internal quantum efficiencies with suitable promising application in the field of photonics as solid state lasers and opto-

How to cite this paper: Reddy, C.P., Naresh, V., Babu, B.C. and Buddhudu, S. (2014) Photoluminescence and Energy Transfer Process in $\text{Bi}^{3+}/\text{Sm}^{3+}$ Co-Doped Phosphate Zinc Lithium Glasses. *Advances in Materials Physics and Chemistry*, 4, 165-171. <http://dx.doi.org/10.4236/ampc.2014.49019>

electronic devices [1]–[5]. In the present study, lithium zinc phosphate glass has been taken as a host because of its combined advantages of fluoride (low phonon energy) and oxide glasses (high mechanical and thermal stability and chemical durability) [6]. Such a stable base glass $50\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiF}$ (PZL) is initially incorporated with 0.5 mol% Sm^{3+} and 0.5 mol% Bi^{3+} separately and their luminescence properties were analyzed. Later on Bi^{3+} ion is co-doped to 1.0 mol% Sm^{3+} to study the enhancement in the luminescence through energy transfer from Bi^{3+} to Sm^{3+} ion. Here, we choose Sm^{3+} because it has efficient luminescence (strong orange emission) in the visible region and has wide range of applications such as hole burning, high-density optical storage, colour displays and alongside samarium doped glasses can also be used as a cladding for Nd-glass laser rods [7]. Bi^{3+} co-doped rare earth ions are considered as prospective materials for scintillators. Bi^{3+} is chosen because of its closed-shell 6S^2 configuration which influences the luminescence and it is considered as suitable material both as an activator and sensitizer for scintillators due to an intense and fast Bi^{3+} -related luminescence [8]. The energy transfer occurs between a sensitizer and an activator, if the energy difference between the excited state of sensitizer (S) and ground state activator (A) is equal. The sensitizer transfers all its energy non-radiatively to activator by quenching its luminescence and enhancing emission of activator. This resonance condition is called resonance transfer of energy. This resonance condition can be tested by the spectral overlap of emission band of sensitizer (S) and absorption band of activator (A) [9] [10]. Energy transfer between doped luminescent ions in the optical materials enhances the emission due to acceptor (Sm^{3+}) and quenches donor emission by transferring its excitation energy. So in the present work we have undertaken to co-dope Bi^{3+} & Sm^{3+} ions into the PZL glass matrix to investigate the possibility of energy transfer between these luminescent ions.

2. Experimental Studies

Lithium zinc phosphate glasses with base composition $50\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiF}$ containing singly doped Bi^{3+} , Sm^{3+} ions and together doped ($\text{Sm}^{3+}/\text{Bi}^{3+}$) ions in different sets were prepared by a melt quenching method. The chemical compositions are listed below:

- 1) $50\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiF}$ (PZL) (host glass).
- 2) $(50 - x)\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiF-xBi}_2\text{O}_3$ ($x = 0.5$ mol%).
- 3) $(50 - y)\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiF-ySm}_2\text{O}_3$ (where $y = 0.5$ mol%).
- 4) $(50 - x)\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiF-xBi}_2\text{O}_3\text{-ySm}_2\text{O}_3$ (where $x = 0.1, 0.5, 1.0, 1.5$ mol% and $y = 0.5$ mol%).

Reagent grade chemicals $\text{NH}_4\text{H}_2\text{PO}_4$, ZnCO_3 , LiF , Sm_2O_3 , and Bi_2O_3 were used for the preparation of glasses. All those chemicals were weighed separately in a 10 g batch, thoroughly mixed and finely powdered using an agate mortar and pestle. Each batch of chemical mix was transferred into porcelain crucibles and each of those was sintered in an electric furnace for an hour at 950°C separately. These melts were quenched in between two smooth surfaced brass plates to obtain circular glass discs of 2 - 3 cm in diameter with 0.3 cm in thickness. The reference PZL glass was transparent and colourless. Sm^{3+} : PZL glass did exhibit an *orange* emission, $\text{Sm}^{3+}/\text{Bi}^{3+}$: PZL co-doped glasses have displayed an enhanced bright *reddish-orange* emission under an UV source.

3. Measurements

The optical absorption spectra of PZL glasses doped with Sm^{3+} and Bi^{3+} were recorded at room temperature in the spectral range of 250 nm - 2500 nm on a *Varian-Cary-Win* Spectrometer (*JASCO V-570*). The excitation and emission spectra of singly doped Sm^{3+} , Bi^{3+} and co-doped ($\text{Sm}^{3+}/\text{Bi}^{3+}$) glasses were recorded at room temperature on a *SPEX Fluorolog-3 (Model-II)* spectrophotometer, attached with an Xe-arc lamp (450 W) as the excitation source. This system is employed with a Datamax software package for acquiring the spectral data and decay-curve (lifetime measurement) data using a phosphorimeter and a Xe-flash lamp.

4. Results and Discussion

4.1. Photoluminescence Spectrum of Bi^{3+} : PZL Glass

Figure 1(a) and **Figure 1(b)** represent excitation and emission spectra of 0.5 mol% Bi^{3+} : PZL glass. The luminescence properties of Bi^{3+} (6S^2) were attributed to radiative decay of triplet relaxed state of Bi^{3+} centres, $^3\text{P}_1 \rightarrow ^1\text{S}_0$ and $^3\text{P}_0 \rightarrow ^1\text{S}_0$ originating from the S-P inter-configurational transition [11] [12]. Usually the excitation occurs from the ground state $^1\text{S}_0$ to the excited state $^3\text{P}_0$, $^3\text{P}_1$, $^3\text{P}_2$ and $^1\text{P}_1$ in the sequence of increasing energy. The transitions from $^1\text{S}_0 \rightarrow ^3\text{P}_J$ where $J = 0$ and 1 are spin forbidden. However, the $^1\text{S}_0 \rightarrow ^3\text{P}_1$ is partially allowed by mixing with the singlet and triplet states of Bi^{3+} ions. In the present glass matrix $^1\text{S}_0 \rightarrow ^3\text{P}_1$ transition is

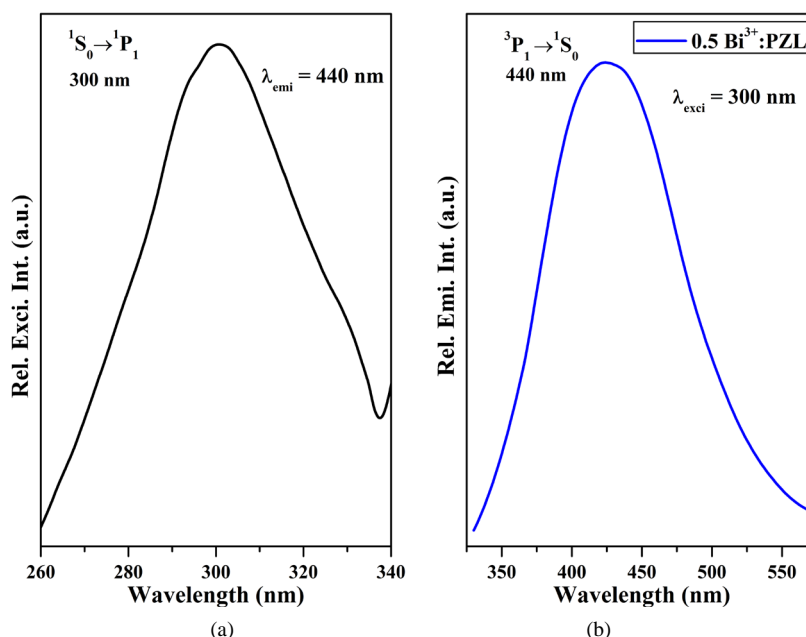


Figure 1. (a) Excitation and (b) emission spectra of Bi^{3+} : PZL glass.

noticed. In the excitation spectrum a broad band in the range of 280 nm to 330 nm centred at 300 nm which corresponds to $^1\text{S}_0 \rightarrow ^1\text{P}_1$ electronic transition of Bi^{3+} , which is partially allowed due to mixing of triplet $^3\text{P}_1$ state with singlet $^1\text{P}_1$ state by the spin-orbit interaction. The emission spectrum is measured by monitoring at 300 nm as excitation wavelength, a broad band with a maxima centred at 440 nm assigned to $^3\text{P}_1 \rightarrow ^1\text{S}_0$ is obtained [13]. The emission of Bi^{3+} ion is assigned to its electro-covalence, which arose from the S-P interactions [12] [14].

4.2. Emission Analysis of Sm^{3+} : PZL Glass

In **Figure 2(a)** and **Figure 2(b)** the excitation and emission spectra of Sm^{3+} : PZL glasses for 0.5 mol% Sm^{3+} are shown. The spectra is measured in the range of 325 nm to 575 nm exhibiting bands at $^6\text{H}_{5/2} \rightarrow ^4\text{H}_{9/2}$ (345 nm), $^4\text{D}_{5/2}, ^6\text{P}_{5/2}$ (363 nm), $^4\text{D}_{1/2}$ (376 nm), $^4\text{F}_{7/2}$ (403 nm), $^4\text{M}_{19/2}$ (418 nm), $^4\text{G}_{9/2}$ (439 nm), $^4\text{I}_{13/2}$ (463 nm), $^4\text{I}_{11/2}$ (471 nm), $^4\text{G}_{7/2}$ (501 nm), $^4\text{F}_{3/2}$ (528 nm), $^4\text{G}_{5/2}$ (563 nm) attributed to 4f-4f transition of Sm^{3+} [15]. Among all the transitions, the prominent excitation transition $^6\text{H}_{5/2} \rightarrow ^4\text{F}_{7/2}$ at 403 nm has been chosen for the recording emission spectra of Sm^{3+} doped glasses. Upon exciting, Sm^{3+} ions are pumped to upper energy level $^4\text{H}_{9/2}$ and from where they relax non-radiatively to $^4\text{G}_{5/2}$ metastable state through $^4\text{F}_{7/2}$, $^4\text{G}_{7/2}$, and $^4\text{F}_{3/2}$ levels. As the energy levels $^4\text{F}_{7/2}$ and $^4\text{G}_{5/2}$ are very close fast non-radiative relaxations takes place. The photoluminescence spectra consists of four emission bands assigned to their electronic transitions $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{(2j+1)/2}$ where $j = 2, 3, 4$, and 5 at (565 nm: yellow), (602 nm: orange), (647 nm: orange reddish), and (709 nm: red). Of all these transitions, $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$ (602 nm) is the most dominant transition with intense orange emission which is partially MD- and partially ED-allowed with selection rule $\Delta J = \pm 1$, therefore it can be considered to be suitable for laser emission. $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$ (565 nm) transition is a forbidden magnetic dipole transition ($\Delta J = 0$ i.e., $J \neq 0 \leftrightarrow 0$ values), $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{9/2}$ (647 nm) is purely electric dipole transition with $\Delta J = \pm 2$ having moderate intensity and $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{11/2}$ (709 nm) is forbidden transition with $\Delta J = \pm 3$ having feeble intensity [16]-[18]. The intensity ratio (R) between ED and MD transitions elucidates the asymmetry nature Sm^{3+} ion in the glass matrix. Higher is the intensity of the ED transition greater is the asymmetric nature. In the present work, $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{9/2}$ (ED) transition is less intense compared to $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$ (MD) transition suggesting symmetric nature of Sm^{3+} in the host glass. The assignment of luminescent bands are made on the basis of energy level diagram given by Dieke [19] and from Carnall and his co-workers [15].

4.3. Energy Transfer between Luminescent Bi^{3+} and Sm^{3+} Ions

Energy transfer is very important mechanism in the study of luminescent properties in co-activated glassy sys-

tems. According to Dexter's theory the probability of energy transfer is proportional to spectral overlap of sensitizer emission and activator absorption/excitation [9] [10]. The spectral overlap of Bi^{3+} (sensitizer) emission and Sm^{3+} (acceptor) excitation is shown in the Figure 3. The primary condition for energy transfer is fulfilled from the spectral overlap of emission band of Bi^{3+} (440 nm; $^3\text{P}_1 \rightarrow ^1\text{S}_0$) appreciably with the excitation bands of Sm^{3+} ($^6\text{H}_{5/2} \rightarrow ^4\text{F}_{7/2}$ (403 nm); $^4\text{G}_{9/2}$ (439 nm); $^4\text{I}_{13/2}$ (463 nm), $^4\text{I}_{11/2}$ (471 nm). Spectral overlap is not only the main criteria for energy transfer, even if the relative fluorescence levels are matched or if sensitizer excitation state is above the emission level of the activator ion energy transfer occurs [20].

In Figure 4(a), the emission spectra for co-doped ($\text{Bi}^{3+}/\text{Sm}^{3+}$): PZL glasses are shown. The glass samples under study are prepared by taking Bi^{3+} at various concentrations (0.1 - 1.5 mol%) and Sm^{3+} at a definite concentration of 0.5 mol%. The effect of co-doping Bi^{3+} ion on photoluminescence emission of Sm^{3+} is studied at excitation wavelength 403 nm. The emission spectra displayed four peaks ($^4\text{G}_{5/2} \rightarrow ^6\text{H}_J$ where $J = 5/2, 7/2, 9/2, 11/2$) attributed to Sm^{3+} with an additional band centred at (440 nm; $^3\text{P}_1 \rightarrow ^1\text{S}_0$) which arises from Bi^{3+} . It is also

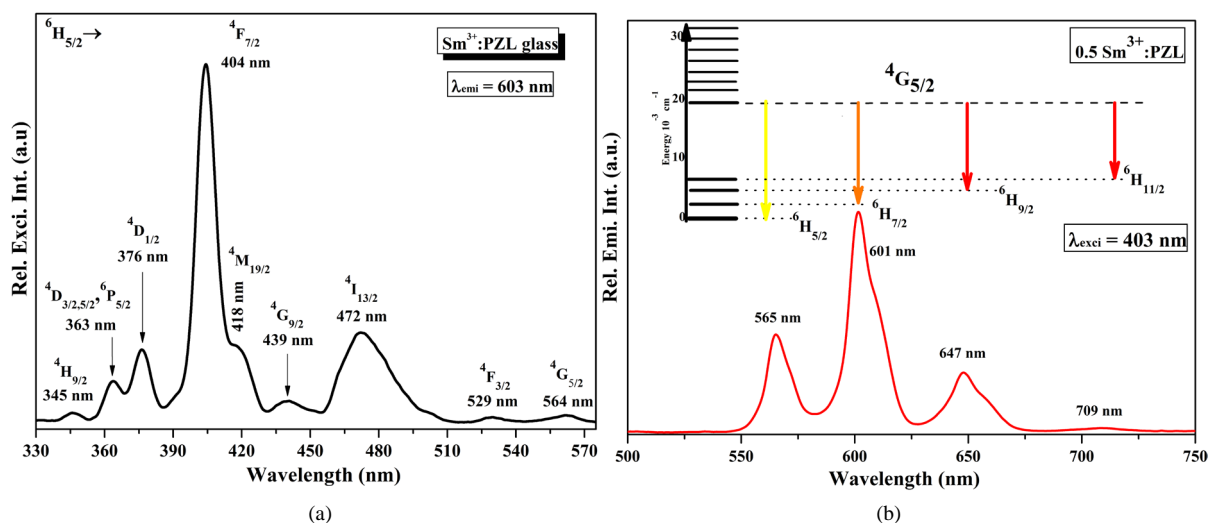


Figure 2. (a) Excitation spectrum of Sm^{3+} : PZL glass; (b) Emission spectra and energy level diagram of (0.5 mol%) Sm^{3+} : PZL glass.

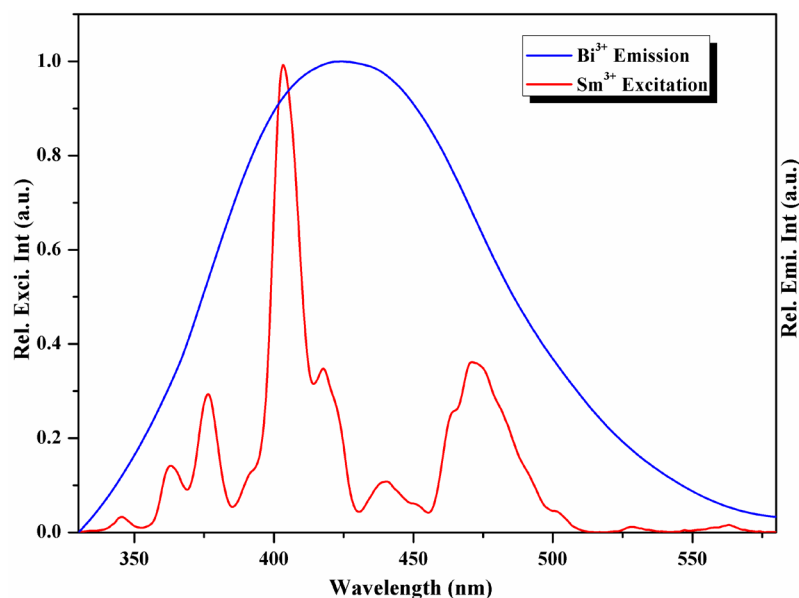


Figure 3. Spectral overlap of Bi^{3+} emission spectrum and Sm^{3+} excitation spectrum.

noticed from the spectra, with increasing the concentration of Bi^{3+} ion in the co-doped glasses, the emission intensity of Sm^{3+} has been enhanced greatly suggesting the energy transfer due to sensitization effect of Bi^{3+} . In addition to this broad emission band of Bi^{3+} peaking at 440 nm has also been increased [21]–[23]. Beyond 1.0 mol% of Bi^{3+} (*i.e.* for 1.5 mol%) there is quenching in the emission intensity of $\text{Bi}^{3+}+\text{Sm}^{3+}$ co-doped glass system. At lower concentrations of Bi_2O_3 , the average distance between the $\text{Bi}^{3+}-\text{Bi}^{3+}$ ions is high so the interactions between them is negligible as a result more amount of energy would be migrated from Bi^{3+} to Sm^{3+} . Whereas at higher concentration of Bi_2O_3 , the distance between $\text{Bi}^{3+}-\text{Bi}^{3+}$ ions is low resulting in high interactions between the ions. Due to these high interactions, lower amount of energy would be transferred from Bi^{3+} to Sm^{3+} . Therefore at higher concentrations of Bi^{3+} , luminescence quenching is observed in case of 1.5 $\text{Bi}^{3+} + 0.5 \text{Sm}^{3+}$ co-doped glass.

Figure 4(b) shows the excitation spectrum of the co-doped glass which exhibits the same trend as shown by the emission spectrum. The mechanism for energy transfer from Bi^{3+} to Sm^{3+} is explained from energy level diagram for the ($\text{Bi}^{3+}/\text{Sm}^{3+}$) co-doped glass is shown in **Figure 5**. Under near UV irradiation Bi^{3+} ions are pumped to $^1\text{P}_1$ level from ground state and these ions cascade rapidly to $^3\text{P}_2$, $^3\text{P}_1$ levels and finally reach to ground state with emission of radiations. This emitted radiation energy of Bi^{3+} is partially reabsorbed by the Sm^{3+} ions in the ground state and jumps to the higher energy levels and there by relaxes non-radiatively to the ground state with enhanced red-orange emission. The energy transfer mechanism is further understood from excitation spectra and lifetime measurements for the co-doped PZL glass matrices.

The transfer of energy taking place from Bi^{3+} to Sm^{3+} is further explained from the lifetime measurements. Luminescence decay analysis is very useful for understanding the energy transfer mechanism and luminescence quenching. Generally, luminescent materials possess an exponential decay so that it is convenient to express the time constant as $1/e$ (time to decay 37% of the initial intensity) and the decay times are noticed in the range of milliseconds to several hundred nanoseconds. The emission decay lifetime for the co-doped ($\text{Bi}^{3+}/\text{Sm}^{3+}$) is shown in the **Figure 6**. The life time value of decay curves are calculated from the first order exponential decay method by using the equation: $I = I_0 \exp(-t/\tau)$ [6]. Measured emission decay lifetimes are found to be 1.23 ms, 1.27 ms, 1.30 ms, 1.32 ms, 1.37 ms. With increasing concentration of Bi^{3+} all the profiles of decay curves exhibited non-exponential nature due to energy migration depending on the types of interaction and on the average distance between the donor and acceptor ions. The reason for this non-exponential decay trend could be explained as follow: when the donor ion are excited in the presence of activator ions in a co-doped glass matrix, donors near to the acceptors decay first due to fast migration of energy so the decay is fast at initial stages whereas donors at far apart transfers their excitation energy for a long time to acceptors so the decay is slow and finally donors decay with their own lifetimes showing a non-exponential nature in the decay curve.

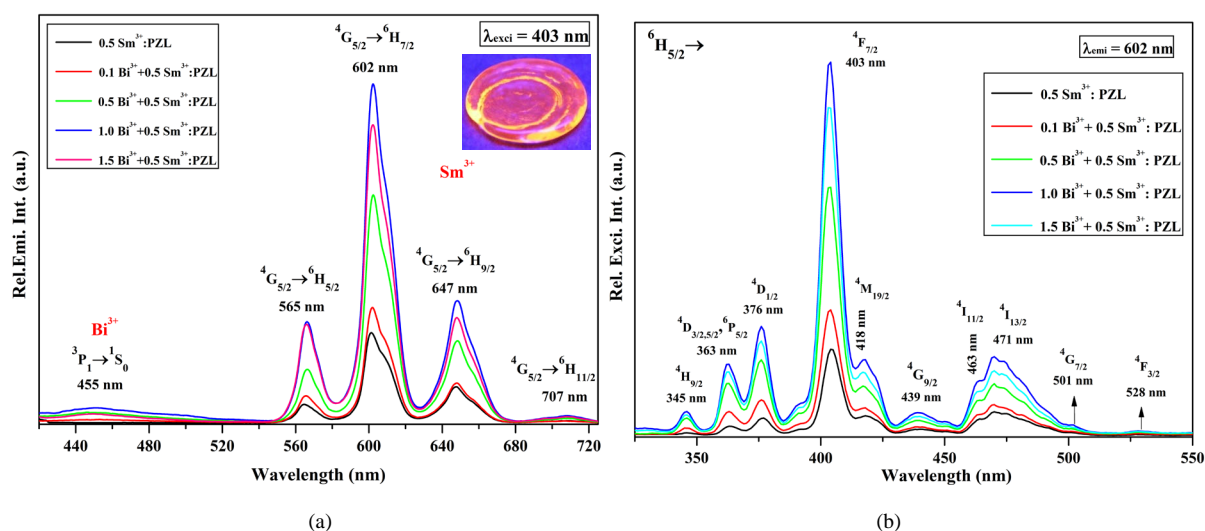


Figure 4. (a) Energy transfer based emission spectra of co-doped (0.5 mol%) $\text{Sm}^{3+} + (0.1 - 1.5 \text{ mol\%}) \text{Bi}^{3+}$: PZL glasses. (b) Excitation spectrum of co-doped (0.5 mol%) $\text{Sm}^{3+} + (0.1 - 1.5 \text{ mol\%}) \text{Bi}^{3+}$: PZL glasses.

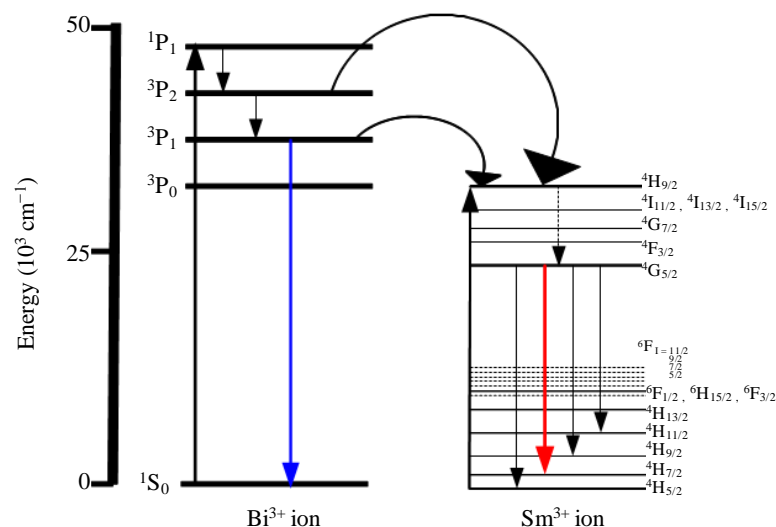


Figure 5. Energy level diagram for co-doped $\text{Sm}^{3+} + \text{Bi}^{3+}$ ions in PZL glass.

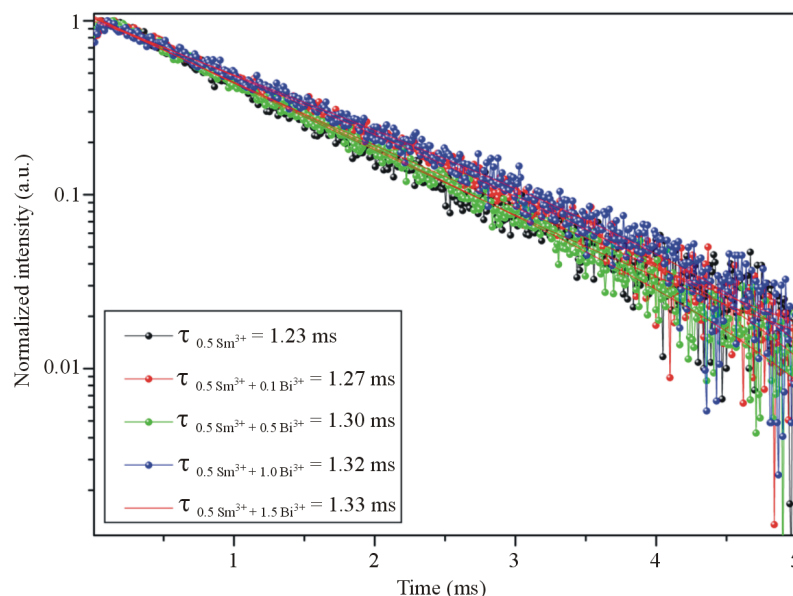


Figure 6. Emission lifetime decay curve of co-doped (0.5 mol%) Sm^{3+} + (0.1 - 1.5 mol%) Bi^{3+} : PZL glasses at 402 nm excitation.

5. Conclusion

In conclusion it is summarized that, stable and transparent glasses in the chemical composition 50P₂O₅-30ZnO-20LiF containing Bi³⁺, Sm³⁺ ions in single combination and (Bi³⁺/Sm³⁺) dual combinations are prepared separately by employing a melt quenching method. Bi³⁺ glass demonstrated a broad emission peak at 440 nm (³P₁→¹S₀) under 300 nm (¹S₀→³P₁) excitation while Sm³⁺ glass had shown an intense orange emission at 601 nm (⁴G_{5/2}→⁶H_{7/2}) at an excitation 403 nm (⁶H_{5/2}→⁴F_{7/2}). Energy transfer taking place from Bi³⁺ to Sm³⁺ has been realized from spectral overlap of Bi³⁺ emission spectrum and Sm³⁺ excitation spectrum. With the addition of Bi³⁺ to Sm³⁺: PZL glass, emission due to Sm³⁺ has been enhanced till 1.0 mol% of Bi³⁺ and beyond concentration quenching in luminescence is observed. The sensitization effect of Bi³⁺ has been explained in terms of emission spectrum of (Bi³⁺/Sm³⁺) co-doped glasses and also from energy level diagram and emission decay curves. Such glasses could be potentially useful as orange light emitting devices in the fields of photonics and optoelectronic devices.

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