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Optical and Judd Ofelt analysis of Europium ion induced SZB glasses for Red Emission

S. Hima Bindu, Ch. Prameela, Ch. Rani, M. Hemalatha, Ch. Linga Raju

[1]*Department of Basic Science and Humanities, Chalapathi Institute of Technology, A.R.Nagar, Mothadaka, Guntur, Andhra Pradesh, 522016, India*
[2]*Department of Basic Science and Humanities, Kallam Haranadha Reddy Institute of Technology, Chowdavaram, Guntur, Andhra Pradesh, 522019, India.*
[3]*Department of Physics, Kakaraparthi Bhavanarayana College, Kothapeta, Vijayawada, Andhra Pradesh, 520001, India.*

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ABSTRACT

Eu^{3+} doped SZB glass system has been prepared by melt quenching technique and characterized. From absorption spectra the bonding parameter (δ=+0.35) have been calculated which specifies the covalent nature of Eu-O bonds in the present glass system, it also confirmed from the Ω_2 intensity parameter. The J-O parameters and the asymmetry ratio derived from the f-f transitions of the Eu^{3+} ion gives information on the local structure around the impurity ions. The predicted values of radiative lifetime (τ_R) and luminescence intensity branching ratio (β_R) are compared with the measured values for $^5D_0$ level. The potentiality of present Eu doped SZB glasses to produce red luminescence at 612 nm corresponding to $^5D_0 \rightarrow {}^7F_2$ emission level in laser host materials is confirmed by its spectroscopic properties and also specifies the significance importance in the development of emission rich optical systems.

I. INTRODUCTION

In the present century photonics plays an important role in telecommunications. The unique applications of photonics lead to the developments in the fields of quantum optics, opto mechanics, electro optics, optoelectronics and quantum electronics [1].The trivalent europium ion is the best choice for use in photonic applications. To know the effect of chemical environment on the optical properties of rare earth ions, glasses are best as hosts. Also for efficient luminescence of rare earth ions, oxide hosts are suitable [2]. A host of borate rich glasses containing alkaline earth oxides along with ZnO, SrO, and BaO acts as glass modifiers are optimistic materials for their probable applications in the

fields of optical communications (optical fibers), laser hosts, optical filters, X- and γ-ray absorbers, photonic devices, and so forth [3]. Zinc oxide with excellent electro-optic properties is good for opto electronic materials [4].

The present work reports the structural and optical properties of Eu $^{3+}$ doped SZB glasses by measuring XRD, optical absorption, and luminescence spectra measurements. When the divalent cation zinc is incorporated in the glass network it could be found that it becomes network modifier and the oxygen becomes non-bridging.

2.Experimental:

The trivalent Eu^{3+} doped SZB glasses were prepared by melt quenching technique using high purity analytical grade chemicals (99.99%) H_3BO_3, ZnO, SrO, Eu_2O_3 with the following molar composition.

20 SrO – 40 ZnO – 40 B_2O_3 - SZB
20 SrO - (39) ZnO - 40 B_2O_3 – (1) Eu_2O_3 - SZB Eu_1
20 SrO - (38) ZnO - 40 B_2O_3 – (2) Eu_2O_3 - SZB Eu_2
20 SrO - (37) ZnO - 40 B_2O_3 – (3) Eu_2O_3 - SZB Eu_3
20 SrO - (36) ZnO - 40 B_2O_3 – (4) Eu_2O_3 - SZB Eu_4
20 SrO - (35) ZnO - 40 B_2O_3 – (5) Eu_2O_3 - SZB Eu_5

About 10 grams batches of the above chemical composition were thoroughly mixed in an agate mortar to attain homogeneity and the mixture is taken into a silica crucible, melted in an electric furnace at the temperature of 1200^{0}C for about 30 min. The melts were air quenched by pouring them on preheated brass plate.

3.Characterization:

The amorphous nature of the prepared glass is confirmed through X-ray diffraction technique at room temperature. The optical absorption spectra were recorded using Hitachi U- 3400 UV-VIS spectrophotometer in the wavelength region 350-2400 nm.

4.Results and discussion:

4.1. XRD analysis: The broad diffuse scattering at lower angles is the characteristic of long range structural disorder which confirms the amorphous nature of the glass [2].

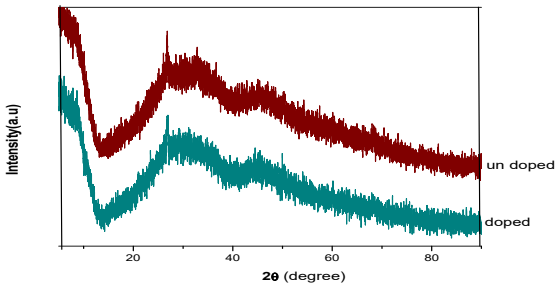
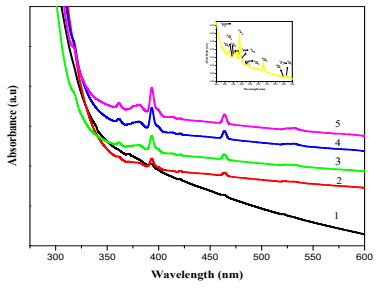


Fig 1: XRD pattern of and pure, Europium doped SZB glass system.

4.2. Optical Absorption spectra analysis:

The optical absorption bands in the spectra are due to in homogeneously broadened 4f-4f optical excitations from the ground to the excited states of Eu^{3+} ions [5]. From the spectra we observe 12 peaks. Out of which, 10 are in UV-VIS region and 2 in NIR region. All these bands along with their band assignments are represented in the inset figure of Fig.2(a) and Fig. 2(b). The $F_J \rightarrow D_J$ absorption and emission transitions are spin forbidden and hence they are very weak **[1]**. From figure 1, we observe the hypersensitive electronic transition at $^7F_0 \rightarrow {}^5D_2$ and is more intense than $^7F_0 \rightarrow {}^5D_1$ magnetic dipole transition. Also the transition $^7F_0 \rightarrow {}^5L_6$ is found to be more intense than other bands and although it is forbidden by ΔS and ΔL rules, but allowed by ΔJ selection rule. The transition $^7F_0 \rightarrow {}^5D_3$ is not observed as it is a spin forbidden **[2].** The band position of $^7F_0 \rightarrow {}^7F_6$ and $^7F_1 \rightarrow {}^7F_6$ transitions reveals the small energy gap which is comparable to other Eu^{+3}doped glasses [6]. This small gap results the population of $^7F_1$ and $^7F_0$ states. [5].



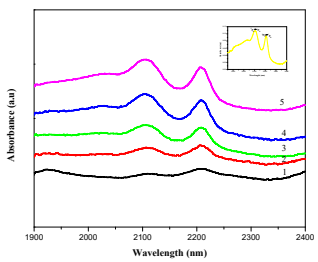


Fig. 2: Optical absorption spectra Europium doped SZB glasses in (a) (UV-VIS) and (b) NIR regions.

4.3. Optical band gap and Urbach's energy:

The optical band gap has been computed for understanding optically induced transitions and also determines the electrical conductivity of a solid material. [5]. The optical band gap values for Eu^{+3} doped glasses in table 3 are found to be nearly 3.36 and 3.35eV for direct and indirect transitions respectively. The increase of band gaps with the

increase of concentration of dopant ion results the decrease of formation of non bridging oxygen, leads to structural changes in the host glass. So the oxide ion polarizability is the cause of variations in the band gap [7]. Further it increases the donor centers there by increases the degree of localization of electrons within the host matrix. The smaller values of Urbach's energy indicate the presence of minimum number of defects in the prepared glass [8]. The optical basicity of an oxide glass will reflect the ability of the glass to donate negative charge to the probe ion. It can be predicted from the composition of the glass and the basicity moderating parameters of the various cations present [8, 9]. It increases with the increase of Eu^{3+} content. This could be related to the formation of chemical bonds with increased covalence in the glass structure [11].

Table 2: Optical energy band gap values of Eu^{3+} doped glass system.

Glass system	Direct band gap (eV)	Indirect band gap (eV)	Urbach energy	Optical basicity
Eu1	3.53	3.49	0.42	0.64
Eu2	3.56	3.54	0.49	0.63
Eu3	3.89	3.83	0.52	0.63
Eu4	4.26	4.35	0.58	0.64
Eu5	4.58	4.55	0.63	0.65

4.4. Oscillator strengths:

The oscillator strengths are used to know the intensities of an absorption bands. The results in table 4 clearly show the dependence of oscillatory strength values of different bands on the intensity of the spin forbidden transition $^7F_1 \rightarrow {}^5D_1$, whose intensity is higher than the magnetic dipole transition and also on glass composition [1]. It is found to be good agreement of experimental and calculated values.

4.5. Judd-Ofelt analysis and radiative properties:

The J-O parameters provide information about the bonding nature of dopant ion with its surrounding ligands and the local structure around the RE ion site. The calculated J-O values of the prepared glass are tabulated. The larger Ω_2 values specify the existence of strong covalence between the Eu^{+3} ion and their surrounding ligands and lower symmetry which is confirmed through the bonding parameter **[11].** The higher asymmetry is further confirmed through the presence of $^5D_0 \rightarrow {}^7F_0$ non-degenerate transition in the emission spectra.

Table 3: Experimental and Calculated values of Eu^{3+} doped glass system.

Transitions	Energy (cm^{-1})	Eu_1		Eu_2	
		f_{exp}	f_{cal}	f_{exp}	f_{exp}
$^7F_0 \rightarrow {}^5D_4$.	27700	0.23	0.229	0.206	0.205
$^7F_0 \rightarrow {}^5G_4$.	26666	0.133	0.179	0.078	0.75
$^7F_0 \rightarrow {}^5G_2$.	26316	0.260	0.203	0.197	0.186
$^7F_0 \rightarrow {}^5L_6$.	25445	0.846	0.797	0.644	0.805
$^7F_1 \rightarrow {}^5L_6$.	25000	0.052	0.132	0.234	0.172
$^7F_1 \rightarrow {}^5D_3$.	24154	0.043	0.082	0.514	0.715
$^7F_1 \rightarrow {}^5D_2$.	21598	0.364	0.221	0.204	0.203
$^7F_0 \rightarrow {}^5D_1$.	19047	0.222	1.23	0.013	0
$^7F_1 \rightarrow {}^5D_1$.	18832	0.017	0.409	0.277	0.292
$^7F_0 \rightarrow {}^5F_6$.	47734543	3	0.54	0.59	1.68
$^7F_1 \rightarrow {}^5F_6$.		0	1.37	1.50	1.41

4.6.Photoluminescenceanalysis:

The photoluminescence characterization is useful to study the optical properties of the samples [12]. Fig.3, shows the excitation spectra of SZB glass system. The peaks are due to the intra 4f transitions of Eu^{3+} ions. The same is also observed for another ZnO phosphor containing Eu^{2+} and Eu^{+3} ions [13]. In excitation spectrum among 6 bands the prominent excitation band $F_0 \rightarrow L_6$ at 393nm has been chosen to measure the emission spectra of Eu^{3+} glass [5].

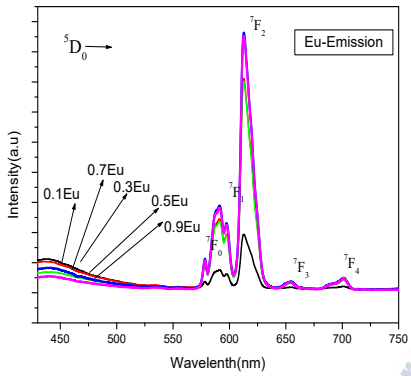


Fig 3: Excitation spectra of Eu^{3+} ion doped SZB glasses.

The spectra mainly specifying two transitions. The transition $^5D_0 \rightarrow ^7F_1$ belongs to magnetic dipole and it could be used for the estimation of transition probabilities of different excited levels and independent of the crystal field strength around Eu^{3+} ion [2]. The transition $^5D_0 \rightarrow ^7F_2$ is due to electric dipole, indicating that Eu^{3+} ions are not in the symmetry center of the host matrix. It is known that Eu^{3+} ions generally occupy the substitution sites of Zn^{+2} ions, permitting $^5D_0 \rightarrow ^7F_1$,$^5D_0 \rightarrow ^7F_2$ transitions, is due to having same total angular momentum. [4,3]. Among 5 observed emission bands, $^5D_0 \rightarrow ^7F_2$ transition has shown bright red emission and it is a hypersensitive transition following $\Delta J=2$ selection rule. The transition $^5D_0 \rightarrow ^7F_3$ has low intensity due to J-mixing effect between multiple's and electric dipole transition [5].

Fig.4, shows the emission spectrum of SZB glass system. In emission spectrum, we observe a broad band emission resulting from 5d-4f transition. The effective excitation in the blue region is very much useful for application in white light LEDs [14].

Transitions	Energy (cm^{-1})	Eu_3		Eu_4		Eu_5	
		f_{cal}	f_{cal}	f_{exp}	f_{cal}	f_{exp}	f_{cal}
$^7F_0 \rightarrow ^5D_4$.	27700	0.25	0.24	0.21	0.25	0.26	0.26
$^7F_0 \rightarrow ^5G_4$.	26666	2	7	6	5	6	5
$^7F_0 \rightarrow ^5G_2$.	26315	0.13	0.19	0.19	0.19	0.06	0.08
$^7F_0 \rightarrow ^5L_6$.	25445	8	0	1	5	2	1
$^7F_1 \rightarrow ^5L_6$.	25000	0.20	0.12	0.11	0.25	0.36	0.32
$^7F_1 \rightarrow ^5D_3$.	24154	1	6	2	9	0.94	0.87
$^7F_1 \rightarrow ^5D_2$.	21598	0.33	0.70	0.68	0.70	7	9
$^7F_0 \rightarrow ^5D_1$.	19047	4	5	1	1	0.11	0.15
$^7F_1 \rightarrow ^5D_1$.	18832	0.13	0.15	0.09	0.15	1	0
$^7F_0 \rightarrow ^5F_6$.	4773	0	3	4	2	0.06	0.11
$^7F_1 \rightarrow ^5F_6$.	4543	0.11	0.07	0.26	0.10	3	9
		3	2	9	2	0.35	0.35
		0.18	0.13	0.28	0.28	7	6
		8	8	4	3	0.01	0
		0.03	0	0.03	0	9	0.33
		5	0.23	8	0.26	0.47	6
		0.20	1	0.26	7	8	0.39
		3	1.31	9	0.40	0.39	1.34
		1.52	1.27	0.89	5	2	7
		1.49		3	0.16	1.41	
				0.56		2	
				1			

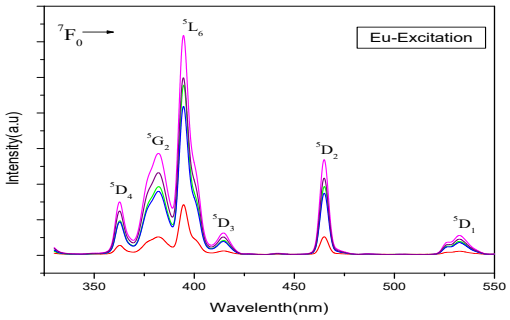


Fig .4: Emission spectra of Eu^{3+} ion doped SZB glasses.

Table 4: ω(2,4,6) values for Eu^{3+} doped glass system.

Glass System	Judd Ofelt parameters			Trend	Spectroscopic quality factor X= Ω_4 / Ω_6
	Ω_2	Ω_4	Ω_6		
	5.64	3.87	2.19	$\Omega_2 > \Omega_4 > \Omega_6$	1.76
Eu_1	7.31	4.2	2.35	$\Omega_2 > \Omega_4 > \Omega_6$	1.72
	8.71	4.27	2.50	$\Omega_2 > \Omega_4 > \Omega_6$	1.70
Eu_2	7.44	5.07	1.28	$\Omega_2 > \Omega_4 > \Omega_6$	3.96
	7.06	5.20	1.43	$\Omega_2 > \Omega_4 > \Omega_6$	3.63
Eu_3					
Eu_4					
Eu_5					

4.7. FTIR analysis:

The Infrared Spectra of present glasses contains several broad and moderate bandwidth peaks in which the broad peaks are as a result of highly degenerate vibration states, thermal boarding of the lattice dispersion and mechanical scattering of the powdered samples. Fig shows the IR spectra of all the samples. Since borate is a good glass former, the addition of alkaline earth and heavy metal ions results the formation of many borate groups. The FTIR spectra exhibited four broad absorption band regions, which are due to vibrations, stretching and bending of bridging oxygen atoms [10]. The band around 420-425 cm^{-1} was attributed to Zn-O stretching vibration of ZnO_4 units [11]. The band in the range 441-457 cm^{-1} is due to the stretching vibration of alkaline earth metal ions like Sr^{+3} [12]. The moderately broad band at around 689-702 cm^{-1} can be attributed to bending of B-O-B linkage between trigonal boran atoms [13]. The broad band around 1050 cm^{-1} is due to the vibrations of boran atoms attached to non bridging oxygen atoms in the form of BO_4 units. The absorption band at nearly 1250 cm^{-1} was assigned to B-O asymmetric stretching vibrations in BO_3 units from pyro to ortho borate groups [14]. The peak at around 415 cm^{-1} owing the manifestation of the Eu-O bonds [14]. There is a small change in the spectral pattern due to the change of alkaline elements and dopant concentration. These assignments are comparable to other borate glasses.

The positions of these bands are slightly shifted towards the lower wave number side with changing concentration of RE ion in the host structure. The shift may be attributed to (i) increase in bond length of B-O groups and the formation of BO_4 units [11]. (ii) change in the mass of network former (Zn

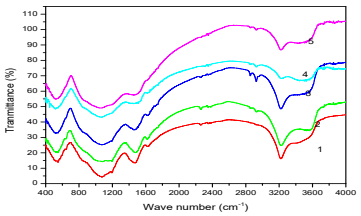


Fig 5: The FTIR spectra of Eu^{3+} doped SZP glasses.

Conclusions:

- No sharp Bragg's peaks confirm the amorphous nature as well as long range structural disorder of present glasses.
- The peaks observed in the UV-Visible spectra reveal the dependence dopant ion.
- The JO parameter Ω_2 specifies the hypersensitive behavior of $^5D_0 \rightarrow {}^7F_2$ transition which indicates that the Eu^{3+} ions are located within a highly polarizable chemical environment.

Conflict of interest statement

Authors declare that they do not have any conflict of interest.

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