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► **To cite this version:**

M Reddi Babu, N Madhusudhana Rao, A Mohan Babu. Spectral Analysis of Nd 3+ Doped Lead Borosilicate Glasses for Efficient Broadband Laser Amplification . Mechanics, Materials Science & Engineering MMSE Journal. Open Access, 2017, 9, <10.2412/mmse.59.84.971provided>. <hal-01502478>

**HAL Id: hal-01502478**

**<https://hal.science/hal-01502478v1>**

Submitted on 5 Apr 2017

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# Spectral Analysis of Nd<sup>3+</sup> Doped Lead Borosilicate Glasses for Efficient Broadband Laser Amplification

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DOI 10.2412/mmse.59.84.971 provided by [Seo4U.link](#)

**Keywords:** lead borosilicate glasses, JO analysis, intensity parameters, emission analysis, concentration quenching.

**ABSTRACT.** In this investigation, Nd<sup>3+</sup> doped lead borosilicate glasses (LBS) were prepared with chemical composition of (30-x) PbO – 40 H<sub>3</sub>BO<sub>3</sub> – 10 SiO<sub>2</sub> – 10 Al<sub>2</sub>O<sub>3</sub> – 10 LiF – x Nd<sub>2</sub>O<sub>3</sub> (where x varies from 0.0, 0.1, 0.5, 1.0 and 2.0 mol%) by conventional melt-quenching method. The spectroscopic analysis can be done using absorption, emission and decay measurements. The oscillator strengths ( $f_{exp}$  and  $f_{cal}$ ) and the evaluated Judd-Ofelt (JO) intensity parameters ( $\Omega_\lambda$ ,  $\lambda = 2, 4$  and  $6$ ) determined from the absorption spectrum. From the emission spectra, three NIR bands observed at 903, 1060 and 1334 nm corresponding to the  $^4F_{3/2} \rightarrow ^4I_{9/2}$ ,  $^4F_{3/2} \rightarrow ^4I_{11/2}$  and  $^4F_{3/2} \rightarrow ^4I_{13/2}$  transitions, respectively for which the effective bandwidths ( $\Delta\lambda_p$ ), radiative transition probabilities ( $A_R$ ) branching ratios ( $\beta_R$ ) and stimulated emission cross-sections ( $\sigma_{sc}$ ) are also evaluated. The intensities of emission bands increased with the increase of Nd<sup>3+</sup> ions concentration upto 1.0 mol% and then decreased at higher concentrations due to the concentration quenching. From the analysis of emission properties, it is concluded that the Nd<sup>3+</sup> doped LBS glasses could be useful for various photonic applications in different fields.

**Introduction.** In the present scenario, the multicomponent oxide glasses have been attracted the several researchers and technologists due to their unique properties such as trouble-free casting, good transparency, solubility of rare earth ions and long term stability [1]. However, due to the increasing demand of the rare earth doped multi-component oxide laser glasses in various applications such as higher order harmonic generation [2], time-resolved laser spectroscopy [3], plasma generation [4] and in many areas, a good host is highly essential to improve the quantum efficiency to meet the aforesaid applications. In the process of searching various oxide glasses, the host glasses with heavy metallic components such as lead oxide, aluminum oxide and silicon dioxide are found to have the ultrafine transparency, high thermal stability, low melting point, infrared transparency, corrosion resistance and also good solubility of rare earth ions [5]. In these multiconstituents, the role of lead in the glass network is to reduce the phonon energy of borate ( $\sim 1400\text{cm}^{-1}$ ) and also to increase the mechanical stability by lowering the melting temperature. Hence, in the present study lead borosilicate glasses (LBS) are chosen as a host matrix to meet the specified applications. Among the available rare earth ions, the neodymium (Nd<sup>3+</sup>) ion is identified as one of the most efficient ions for solid state lasers with the emission wavelength at 1060 nm as well as the possibility for lasing action at other wavelengths such as 1800, 1350 and 880 nm which are be useful for broadband laser amplifiers and other photonic applications.

**Experimental.** High purity reagents of PbO, H<sub>3</sub>BO<sub>3</sub>, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, LiF and Nd<sub>2</sub>O<sub>3</sub> (99.99) were selected as raw materials for the preparation of Nd<sup>3+</sup> doped lead borosilicate (LBS) glasses by the melt-quench procedure.

**Results and Discussion.** Due to the absorption of energy by the ground state ( $^4I_{9/2}$ ) Nd<sup>3+</sup> ions, they get excited to the various excited levels. The absorption spectrum of Nd<sub>1.0</sub>:LBS glass recorded in the

wavelength region of 400 - 950 nm is shown in the Fig. 1. It shows eleven absorption bands at 430, 460, 477, 513, 526, 583, 627, 682, 747, 804 and 875 nm are identified corresponding to the transitions  ${}^4I_{9/2} \rightarrow {}^2P_{1/2}$ ,  ${}^4G_{11/2}$ ,  ${}^2G_{9/2}$ ,  ${}^4G_{9/2}$ ,  ${}^4G_{7/2}$ ,  ${}^4G_{5/2}$ ,  ${}^2H_{11/2}$ ,  ${}^4F_{9/2}$ ,  ${}^4F_{7/2}$ ,  ${}^4F_{5/2}$  and  ${}^4F_{3/2}$ , respectively. The assignments of the transitions have been done based on the reports by the Carnal et.al [6]. Among these energy levels, the  ${}^2P_{1/2}$ ,  ${}^4G_{11/2}$ ,  ${}^2G_{9/2}$ , and  ${}^2H_{11/2}$  levels are not well resolved due to the strong absorption of the host composition. The observed absorption bands in the present work are similar to those of other  $Nd^{3+}$  doped glasses, which indicate homogeneous incorporation of the rare earth ions in the glass network. It is also noticed that the bands at 526, 583, 747 and 804 nm are more intense than other bands. In the case of rare earths ions certain transitions are sensitive to ligand environment of the host and such transitions are called as hypersensitive transitions, obeying the selection rules  $\Delta J \leq 2$ ,  $\Delta L \geq 2$  and  $\Delta S = 0$ . These transitions possess higher reduced matrix elements ( $\|U^2\|$ ) and also higher oscillator strengths with respect to other transitions. In the case of  $Nd^{3+}$  ion, the  ${}^4I_{9/2} \rightarrow {}^4G_{5/2}$  is the hypersensitive transition and possesses higher oscillator strength.

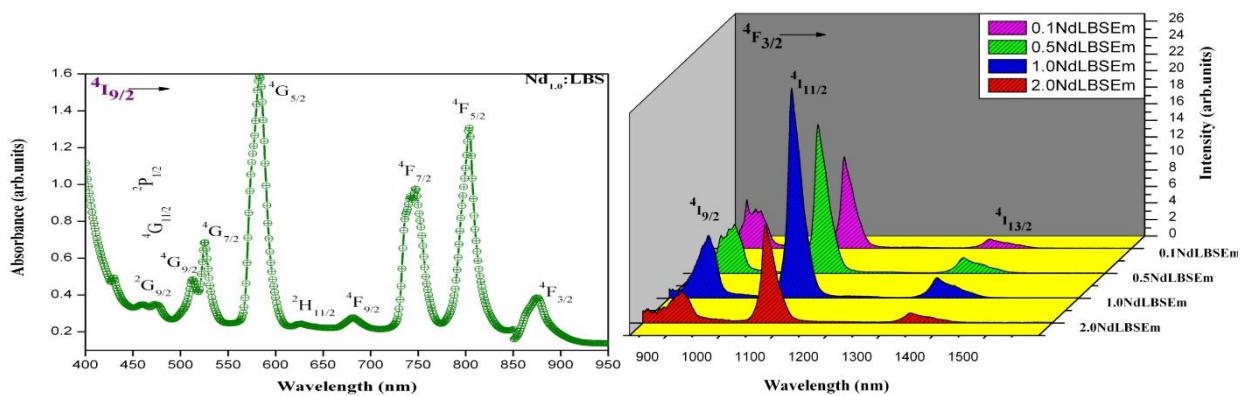


Fig. 1.(a) Absorption and (b) Emission spectra of  $Nd_x$ :LBS glasses

**Judd-Ofelt analysis.** To investigate the nature of bonding exists between the rare earth ions and the surrounding ligands and as well as the symmetry around the rare earth ions, the JO analysis has been adopted. The JO theory enables the evaluation of intensity parameters  $\Omega_\lambda$  ( $\lambda = 2, 4$  and  $6$ ), which are useful to know the nature of the bonding, asymmetry and also in the prediction of certain radiative parameters. To evaluate the JO intensity parameters, the experimental ( $f_{exp}$ ) and calculated ( $f_{cal}$ ) oscillator strengths are determined using the expressions given in earlier reports by Reddi Babu et al. [1] and are reported in Table 1. The evaluated intensity parameters  $\Omega_\lambda$  ( $\lambda = 2, 4$  and  $6$ ) of the  $Nd_{1.0}$ :LBS glass are compared with those of other  $Nd^{3+}$  doped glasses as presented in the Table 2 [7-10].

**Emission analysis.** The emission spectra of  $Nd_x$ :LBS ( $x=0.1, 0.5, 1.0$  and  $2.0$  mol %) glasses recorded in the region 800-1500 nm with an excitation wavelength of 808 nm are shown in the Fig 1(b). It is also observed that the emission intensities increases with the increase of  $Nd^{3+}$  ions concentration upto 1.0 mol% and then decreases at higher concentrations. It may be due to concentration quenching as well as the energy transfer between the  $Nd^{3+}$  ions. Among the emission transitions, the  ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$  transition at 1060 nm is found be more intense than the other transitions. In order to predict the lasing efficiency of  $Nd^{3+}$  doped LBS glasses, the evaluated Judd-Ofelt intensity parameters  $\Omega_\lambda$  ( $\lambda = 2, 4$  and  $6$ ) are used to calculate the radiative properties such as spontaneous emission probabilities ( $A_R$ ), total radiative transition probability ( $A_T$ ), radiative lifetimes ( $\tau_R$ ) and branching ratios ( $\beta_R$ ) for the  ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ ,  ${}^4I_{11/2}$  and  ${}^4I_{13/2}$  emission transitions using the expressions available in previous reports [1] and are shown in Table 2. From the data, the higher experimental branching ratios are observed for the  ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$  transition at 1060 nm for all the  $Nd^{3+}$  doped LBS glasses which is useful for high gain broadband amplification applications. Also, certain laser characteristic parameters such as stimulated emission cross-sections ( $\sigma_e$ ), optical gain bandwidths ( $\sigma_e \times \Delta_{eff}$ ) and optical gain

parameters ( $\sigma_e \times \tau_R$ ) are calculated using the expressions [1] and are presented in Table 3. From the tabulated data, the Nd<sub>0.5</sub>:LBS glass possess higher values of  $\sigma_e$ , ( $\sigma_e \times \Delta_{eff}$ ) and ( $\sigma_e \times \tau_R$ ) for the  $^4F_{3/2} \rightarrow ^4I_{11/2}$  transition observed at 1060 nm.

Table 1. Comparison of Judd-Ofelt intensity parameters (in  $10^{-20} \text{ cm}^2$ ) of the Nd<sub>1.0</sub>:LBS glass with other Nd<sup>3+</sup> doped host glasses.

Glass matrix	$\Omega_2$	$\Omega_4$	$\Omega_6$	Trend
Nd <sub>1.0</sub> :LBS [Present work]	10.96	1.62	19.87	$\Omega_6 > \Omega_2 > \Omega_4$
30B <sub>2</sub> O <sub>3</sub> -70PbO[7]	3.52	2.98	5.48	$\Omega_6 > \Omega_2 > \Omega_4$
30PbO-70B <sub>2</sub> O <sub>3</sub> [7]	3.96	3.77	4.68	$\Omega_6 > \Omega_2 > \Omega_4$
ZBLAN [8]	5.09	3.12	7.16	$\Omega_6 > \Omega_2 > \Omega_4$
Silicate[9]	4.71	4.54	5.05	$\Omega_6 > \Omega_2 > \Omega_4$
Vitreous Borate[10]	4.3	3.6	4.7	$\Omega_6 > \Omega_2 > \Omega_4$

Table 2. Different radiative parameters of the Nd<sub>x</sub>:LBS (x= 0.1, 0.5, 1.0 and 2.0mol%) glasses.

Transition	Property	Nd <sub>0.1</sub> :LBS	Nd <sub>0.5</sub> :LBS LBS	Nd <sub>1.0</sub> :LBS LBS	Nd <sub>2.0</sub> :LBS
$^4F_{3/2} \rightarrow ^4I_{9/2}$	A <sub>R</sub>	1368	5117	4356.1	5942
	A <sub>T</sub>	7221	13054	11013	13083
	$\beta_m$	0.35	0.26	0.25	0.45
	$\beta_R$	0.19	0.39	0.4	0.21
	$\tau_R(\mu\text{s})$	138	95	82	76
$^4F_{3/2} \rightarrow ^4I_{11/2}$	A <sub>R</sub>	4623	6542	5492	5999
	A <sub>T</sub>	7221	13054	11013	13083
	$\beta_m$	0.55	0.65	0.66	0.46
	$\beta_R$	0.64	0.50	0.50	0.69
	$\tau_R(\mu\text{s})$	138	95	82	76
$^4F_{3/2} \rightarrow ^4I_{13/2}$	A <sub>R</sub>	1172	1328	1109	1087
	A <sub>T</sub>	7221	13054	11013	13083
	$\beta_m$	0.1	0.09	0.09	0.1
	$\beta_R$	0.16	0.10	0.10	0.10
	$\tau_R(\mu\text{s})$	138	95	82	76

Table 3. Laser characteristic parameters of the Nd<sub>x</sub>:LBS (x= 0.1, 0.5, 1.0 and 2.0 mol%) glasses.

Transition	Lasing parameter	Nd <sub>0.1</sub> :LBS	Nd <sub>0.5</sub> :LBS	Nd <sub>1.0</sub> :LBS	Nd <sub>2.0</sub> :LBS
<sup>4</sup> F <sub>3/2</sub> → <sup>4</sup> I <sub>9/2</sub>	λ <sub>p</sub>	903	902	904	902
	Δλ <sub>eff</sub>	58.55	53.15	52.26	45.87
	σ <sub>e</sub>	0.89	3.57	3.08	4.62
	σ <sub>e</sub> × Δλ <sub>eff</sub>	5.21	18.98	16.09	21.20
	σ <sub>e</sub> × τ <sub>R</sub>	1.23	3.50	2.53	3.51
<sup>4</sup> F <sub>3/2</sub> → <sup>4</sup> I <sub>11/2</sub>	λ <sub>p</sub>	1059	1059	1059	1060
	Δλ <sub>p</sub>	35.81	39.54	39.90	39.91
	σ <sub>e</sub>	9.32	11.64	9.56	10.22
	σ <sub>e</sub> × Δλ <sub>eff</sub>	33.38	46.02	38.15	40.79
	σ <sub>e</sub> × τ <sub>R</sub>	12.86	11.05	7.83	7.76
<sup>4</sup> F <sub>3/2</sub> → <sup>4</sup> I <sub>13/2</sub>	λ <sub>p</sub>	1334	1334	1337	1334
	Δλ <sub>p</sub>	67.53	56.36	53.44	63.57
	σ <sub>e</sub>	3.16	4.18	3.67	2.92
	σ <sub>e</sub> × Δλ <sub>eff</sub>	21.35	23.56	19.62	18.57
	σ <sub>e</sub> × τ <sub>R</sub>	4.36	3.97	3.00	2.21

**Summary.** In the present work, different concentrations of the Nd<sup>3+</sup> doped LBS glasses were prepared by the melt-quench method and are characterized by using the absorption and emission measurements. Using the absorption studies, the JO intensity parameters (Ω<sub>λ</sub>) were determined from the absorption spectrum of Nd<sub>1.0</sub>: LBS glass and their trend has been observed as Ω<sub>6</sub> > Ω<sub>2</sub> > Ω<sub>4</sub>. The higher Ω<sub>6</sub> parameter of the LBS glass indicates more rigidness of the glass and also strong electron-phonon coupling strengths between the Nd<sup>3+</sup> ions and anion ligands. The radiative properties such as transition probabilities (A<sub>R</sub>), total transition probabilities (A<sub>T</sub>), branching ratios (β<sub>R</sub>), radiative lifetimes (τ<sub>R</sub>) and peak stimulated emission cross sections (σ<sub>e</sub>), optical gain bandwidths (σ<sub>e</sub> × Δλ<sub>eff</sub>) and optical gains (σ<sub>e</sub> × τ<sub>exp</sub>) were calculated for all the emission transitions of the Nd<sup>3+</sup> doped LBS glasses. The higher values of stimulated emission cross sections, optical gain bandwidth and optical gain observed, for the <sup>4</sup>F<sub>3/2</sub> → <sup>4</sup>I<sub>11/2</sub> transition at 1060 nm suggests that, the prepared glasses are highly useful for efficient broadband laser amplification applications. From the decay profiles of the <sup>4</sup>F<sub>3/2</sub> → <sup>4</sup>I<sub>11/2</sub> transition of Nd<sup>3+</sup> ion, it is concluded that, lifetimes (τ<sub>exp</sub>) of the <sup>4</sup>F<sub>3/2</sub> level decreased with the increase of Nd<sup>3+</sup> ion concentrations.

**Acknowledgements.** One of the authors Dr. A. Mohan Babu would like to thank DAE – BRNS, Mumbai, (Letter No.2012/34/72/BRNS) and DST-SERB, New Delhi, (Letter. No. SR/FTP/PS-109/2012) for the sanction of research projects. Also, Mr. M. Reddi Babu is thankful to DAE – BRNS, Mumbai for the financial support in the form of SRF in the above project. The authors also grateful to Prof. C. K. Jayasankar, Department of Physics, Sri Venkateswara University, Tirupati for providing lab facilities to carry our the research work.

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Cite the paper

M. Reddi Babu, N. Madhusudhana Rao, A. Mohan Babu (2017). [Spectral Analysis of Nd<sup>3+</sup> Doped Lead Borosilicate Glasses for Efficient Broadband Laser Amplification](#). *Mechanics, Materials Science & Engineering*, Vol 9. Doi [10.2412/mmse.59.84.971](#)