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Spectral, Transmittance and luminescencent Properties ofNd^{3+} Doped Borosilicate Glasses for Laser applications

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Abstract: Glass of the system :(25-x)SiO_2: 10ZnO: 10Li_2O: 10Na_2O: 10CaO: 10MgO: 10WO_3: 15B_2O_3:xNd_2O_3. (where x=1, 1.5,2 mol %) have been prepared by melt-quenching method. The amorphous nature of the glasses was confirmed by X-ray diffraction studies. Optical absorption, Excitation, fluorescence and transmittance spectra were recorded at room temperature for all glass samples. Slater-Condon parameters F_k (k=2, 4, 6), Lande parameter ξ_{4f} and Racah parameters E^k (k=1, 2, 3) have been computed. Using these parameters energies and intensities of these bands has been calculated. Judd-Ofelt intensity parameters Ω_λ (λ=2, 4, 6) are evaluated from the intensities of various absorption bands of optical absorption spectra. Using these intensity parameters various radiative properties like spontaneous emission probability (A), branching ratio (β_R), radiative life time (τ_R) and stimulated emission cross–section (σ_p) of various emission lines have been evaluated.
Keywords: ZLSLMTBS Glasses, Optical Properties, Judd-Ofelt Theory, luminescencent Properties.

I. INTRODUCTION

Rare- earth ion doped glasses have attracted much attention because they have various important properties such as high transparency, high thermal stability, a low melting point, low dispersion rates, a high gain density, excellent physical, optical, chemical, mechanical and mechanical properties [1-5].Among different ceramic glasses silicate glasses is attributed to their favorable properties such as, high density, high refractive index, large third order nonlinear optical susceptibility, good transmittance and luminescent properties [5-8].Silicate glass is an extremely promising material for reflecting windows, laser, mechanical sensors and nonlinear applications in optics. They have high thermal stability, low phonon energy, high transparency, high solubility for rare-earth ions, high chemical and thermal stability [9-12].Thus, it can be widely used in visible and infrared laser, fiber amplifier and optical data storage devices. The host matrix compose of ZnO, a glass modifier/glass farmer a heavy metal oxide along with Na_2O, CaO,,MgO,$WO_3$and B_2O_3.The addition of Li_2O to the glass mixture improves the rare earth ion solubility leading to the possibility of using even higher concentrations of ions [15-18].

The present work reports on the absorption and emission properties of Nd^{3+} doped zinc lithium sodalime magnesium tungsten borosilicate glass. The intensities of the transitions for the rare earth ions have been estimated successfully using the Judd-Ofelt theory, The laser parameters such as radiative probabilities (A), branching ratio (β_R), radiative life time (τ_R) and stimulated emission cross section(σ_p) are evaluated using J.O. intensity parameters (Ω_λ, λ=2,4 and 6).

II. EXPERIMENTAL TECHNIQUES

A. Preparation of Glasses

The following Nd^{3+} doped borosilicate glass samples :(25-x)SiO_2: 10ZnO: 10Li_2O: 10Na_2O: 10CaO: 10MgO: 10WO_3: 15B_2O_3:xNd_2O_3. (where *x* =1, 1.5, 2) have been prepared by melt-quenching method. Analytical reagent grade chemical used in the present study consist of SiO_2, ZnO, Li_2O, Na_2O, CaO, MgO, WO_3, B_2O_3 and Nd_2O_3. They were thoroughly mixed by using an agate pestle mortar. Then melted at 1025˚C by an electrical muffle furnace for 2 hours. After complete melting, the melts were quickly poured in to a preheated stainless steel mould and annealed at temperature of 250˚C for 2 h to remove thermal strains and stresses. Every time fine powder of cerium oxide was used for polishing the samples. The glass samples so prepared were of good optical quality and were transparent. The chemical compositions of the glasses with the name of samples are summarized in **Table 1.**

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Table 1.

Sample	Glass composition (mol %)
ZLSLMTBS (UD)	$25SiO_2$: 10ZnO: $10Li_2O$: $10Na_2O$: 10CaO: 10MgO: $10WO_3$: $15B_2O_3$
ZLSLMTBS ND(1.0)	$24SiO_2$: 10ZnO: $10Li_2O$: $10Na_2O$: 10CaO: 10MgO: $10WO_3$: $15B_2O_3$:$1Nd_2O_3$
ZLSLMTBS ND(1.5)	$23.5SiO_2$: 10ZnO: $10Li_2O$: $10Na_2O$: 10CaO: 10MgO: $10WO_3$: $15B_2O_3$:$1.5Nd_2O_3$
ZLSLMTBS ND(2.0)	$23SiO_2$: 10ZnO: $10Li_2O$: $10Na_2O$: 10CaO: 10MgO: $10WO_3$: $15B_2O_3$: $2Nd_2O_3$

ZLSLMTBS (UD) - Represents undoped Zinc Lithium Sodalime Magnesium Tungsten Borosilicate glass specimen.
ZLSLMTBS (ND) **-** Represents Nd^{3+} doped Zinc Lithium Sodalime Magnesium Tungsten Borosilicate glass specimens.

III. THEORY

A. Oscillator Strength

The intensity of spectral lines is expressed in terms of oscillator strengths using the relation [19].

$$f_{expt.} = 4.318 \times 10^{-9} \int \varepsilon(\nu)\, d\nu \qquad (1)$$

where, ε (*v)* is molar absorption coefficient at a given energy *v* (cm^{-1}), to be evaluated from Beer–Lambert law.
Under Gaussian Approximation, using Beer–Lambert law, the observed oscillator strengths of the absorption bands have been experimentally calculated [20], using the modified relation:

$$P_m = 4.6 \times 10^{-9} \times \frac{1}{cl} \log \frac{I_0}{I} \times \Delta\upsilon_{1/2} \qquad (2)$$

Where c is the molar concentration of the absorbing ion per unit volume, I is the optical path length, $\log I_0/I$ is optical density and $\Delta\upsilon_{1/2}$ is half band width.

B. Judd-Ofelt Intensity Parameters

According to Judd [21] and Ofelt [22] theory, independently derived expression for the oscillator strength of the induced forced electric dipole transitions between an initial J manifold $|4f^N (S, L) J>$ level and the terminal J' manifold $|4f^N (S', L') J'>$ is given by:

$$\frac{8\Pi^2 mc\bar{\upsilon}}{3h(2J+1)}\frac{1}{n}\left[\frac{(n^2+2)^2}{9}\right] \times S(J,J') \qquad (3)$$

Where, the line strength S (J, J') is given by the equation

$$S(J, J') = e^2 \sum_{\lambda=2,4,6} \Omega_\lambda <4f^N(S, L) J \| U^{(\lambda)} \| 4f^N (S', L') J'>2 \qquad (4)$$

In the above equation m is the mass of an electron, c is the velocity of light, *v* is the wave number of the transition, h is Planck's constant, n is the refractive index, J and J' are the total angular momentum of the initial and final level respectively, Ω_λ (λ=2,4and 6) are known as Judd-Ofelt intensity parameters.

C. Radiative Properties

The Ω_λ parameters obtained using the absorption spectral results have been used to predict radiative properties such as spontaneous emission probability (A) and radiative life time (τ_R), and laser parameters like fluorescence branching ratio (β_R) and stimulated emission cross section (σ_p).
The spontaneous emission probability from initial manifold $|4f^N (S', L') J'>$ to a final manifold $|4f^N (S, L) J>|$ is given by:

$$A[(S', L') J'; (S, L) J] = \frac{64\pi^2\nu^3}{3h(2J'+1)}\left[\frac{n(n^2+2)^2}{9}\right] \times S(J', \bar{J}) \qquad (5)$$

Where, $S(J', J) = e^2 [\Omega_2 \| U^{(2)} \|^2 + \Omega_4 \| U^{(4)} \|^2 + \Omega_6 \| U^{(6)} \|^2]$

The fluorescence branching ratio for the transitions originating from a specific initial manifold $|4f^N (S', L') J'>$ to a final many fold $|4f^N (S, L) J>$ is given by

$$\beta [(S', L') J'; (S, L) J] = \frac{A[(S' L)]}{A[(S' L') J'(\bar{S} L)]} \qquad (6)$$

S L J

The radiative life time is given by

$_{rad}$ $A[(S', L') J'; (S,L)] = A_{Total}^{-1}$
(7)

S L J

Where, the sum is over all possible terminal manifolds. The stimulated emission cross -section for a transition from an initial manifold $|4f^N (S', L') J'>$ to a final manifold
$|4f^N (S, L) J>|$ is expressed as

$$\sigma_p(\lambda_p) = \left[\frac{\lambda_p^4}{8\pi c n^2 \Delta\lambda_{eff}}\right] \times A[(S', L') J'; (\bar{S}, \bar{L})\bar{J}] \qquad (8)$$

Where, λ_p the peak fluorescence wavelength of the emission band and $\Delta\lambda_{eff}$ is the effective fluorescence line width.

D. Nephelauxetic Ratio (β) and Bonding Parameter ($b^{1/2}$)

The nature of the R-O bond is known by the Nephelauxetic Ratio (β') and Bonding Parameters ($b^{1/2}$), which are computed by using following formulae [23]. The Nephelauxetic Ratio is given by

$$\beta' = \frac{\nu_g}{\nu_a} \qquad (9)$$

where, ν_a and ν_g refer to the energies of the corresponding transition in the glass and free ion, respectively. The values of bonding parameter $b^{1/2}$ are given by

$$b^{1/2} = \left[\frac{1-\beta'}{2}\right]^{1/2} \qquad (10)$$

IV. RESULT AND DISCUSSION

A. XRD Measurement

Figure 1 presents the XRD pattern of the samples shows no sharp Bragg's peak, but only a broad diffuse hump around low angle region. This is the clear indication of amorphous nature within the resolution limit of XRD instrument.

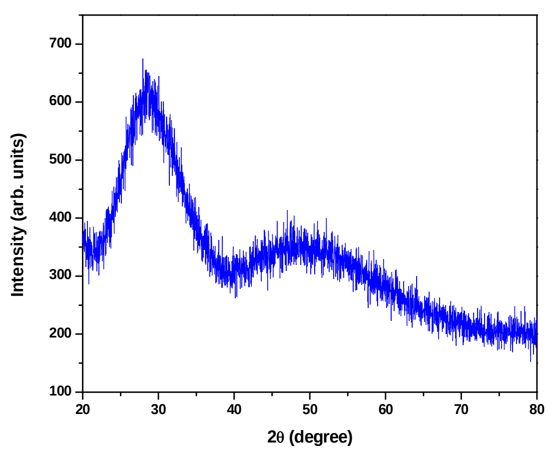

Fig.1: X-ray diffraction pattern of ZLSLMTBS ND (1.0) glass.

B. Transmittance spectra

The Transmittance spectrum of ZLSLMTBS ND (1.0) glass is shown in Figure 2.

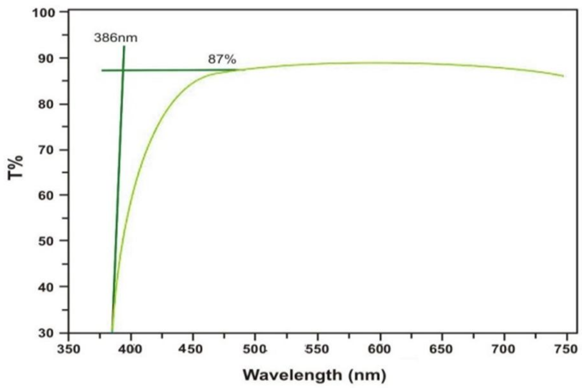

Fig. 2: Transmittance spectrum of ZLSLMTBS ND(1.0) glass.

C. *Absorption spectra*

The absorption spectra of ZLSLMTBS ND(1.0) glass, consists of absorption bands corresponding to the absorptions from the ground state $^4I_{9/2}$ of Nd^{3+} ions. Nine absorption bands have been observed from the ground state $^4I_{9/2}$ to excited states $^4F_{3/2}$, $^4F_{5/2}$, $^4F_{7/2}$, $^4F_{9/2}$, $^2H_{11/2}$, $^4G_{5/2}$, $^4G_{7/2}$, $^4G_{9/2}$, and $^2G_{9/2}$ for Nd^{3+} doped ZLSLMTBS ND(1.0) glass.

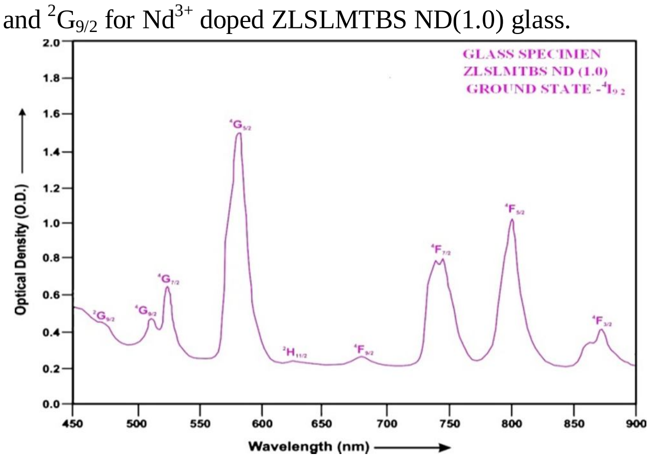

Fig.3: Absorption spectra of ZLSLMTBS ND(1.0) glass.

The experimental and calculated oscillator strength for Nd^{3+} ions in ZLSLMTBS glasses are given in **Table 2.**

Table 2. Measured and calculated oscillator strength ($P^m \times 10^{+6}$) of Nd^{3+} ions in ZLSLMTBS glasses.

Energy level from $^4I_{9/2}$	Glass ZLSLMTBS ND(1.0)		Glass ZLSLMTBS ND(1.5)		Glass ZLSLMTBS ND(2.0)	
	$P_{exp.}$	$P_{cal.}$	$P_{exp.}$	$P_{cal.}$	$P_{exp.}$	$P_{cal.}$
$^4F_{3/2}$	3.48	3.43	3.42	3.41	3.38	3.39
$^4F_{5/2}$	8.35	8.37	8.30	8.32	8.25	8.29
$^4F_{7/2}$	9.56	9.85	9.51	9.82	9.47	9.80
$^4F_{9/2}$	0.64	0.52	0.62	0.52	0.60	0.52
$^2H_{11/2}$	0.25	0.15	0.22	0.15	0.19	0.15
$^4G_{5/2}$	25.26	25.51	24.35	24.61	23.42	23.71
$^4G_{7/2}$	4.22	5.03	4.17	4.95	4.12	4.88
$^4G_{9/2}$	2.28	2.17	2.25	2.16	2.21	2.14
$^2G_{9/2}$	0.96	2.75	0.94	2.73	0.91	2.72
r.m.s.deviation	0.6703		0.6670		0.6705	

Table3. Computed values of Slater-Condon, Lande, Racah, nephelauexetic ratio and bonding parameter for Nd^{3+} doped ZLSLMTBS glass specimens.

Parameter	Free ion	ZLSLMTBS ND(1.0)	ZLSLMTBS ND(1.5)	ZLSLMTBS ND(2.0)
$F_2(cm^{-1})$	331.16	324.67	324.60	324.61
$F_4(cm^{-1})$	50.71	50.71	50.71	50.71
$F_6(cm^{-1})$	5.154	5.043	5.039	5.040
$\xi_{4f}(cm^{-1})$	884.0	882.74	882.88	882.80
$E^1(cm^{-1})$	5024.0	4948.45	4947.19	494.74
$E^2(cm^{-1})$	23.90	23.09	23.08	23.08
$E^3(cm^{-1})$	497.0	489.58	489.57	489.58
F_4/F_2	0.1531	0.1562	0.1562	0.1562
F_6/F_2	0.0155	0.0155	0.0155	0.0155
E^1/E^3	10.1086	10.1076	10.1051	10.1054
E^2/E^3	0.0481	0.0472	0.0472	0.0471
β'		0.9961	0.9958	0.9959
$b^{1/2}$		0.04425	0.05798	0.05549

The values of Judd-Ofelt intensity parameters are given in Table 4.

Table 4. Judd-Ofelt intensity parameters for Nd^{3+} doped ZLSLMTBS glass specimens.

Glass Specimen	$\Omega_2(pm^2)$	$\Omega_4(pm^2)$	$\Omega_6(pm^2)$	Ω_4/Ω_6	Ref.
ZLSLMTBS ND(1.0)	2.950	7.324	3.653	2.005	P.W.
ZLSLMTBS ND(1.5)	2.685	7.259	3.642	1.993	P.W.
ZLSLMTBS ND(2.0)	2.395	7.219	3.632	1.988	P.W.
LBZF (SM)	0.270	2.520	2.470	1.020	[24].
PKAMZ (SM)	0.410	2.990	2.670	1.120	[25].

D. Excitation Spectrum

The Excitation spectra of Nd^{3+}doped ZLSLMTBS ND (1.0) glass specimen has been presented in Figure 3 in terms of Excitation Intensity versus wavelength. The excitation spectrum was recorded in the spectral region 700–1000 nm fluorescence at 1065nm having different excitation band centred at 808 nm and 887 nm are attributed to the ($^4I_{9/2}\rightarrow{}^4F_{5/2}$) and ($^4I_{9/2}\rightarrow{}^4F_{3/2}$) transitions, respectively. The highest absorption level is ($^4I_{9/2}\rightarrow{}^4F_{5/2}$) and is at 808 nm. So this is to be chosen for excitation wavelength.

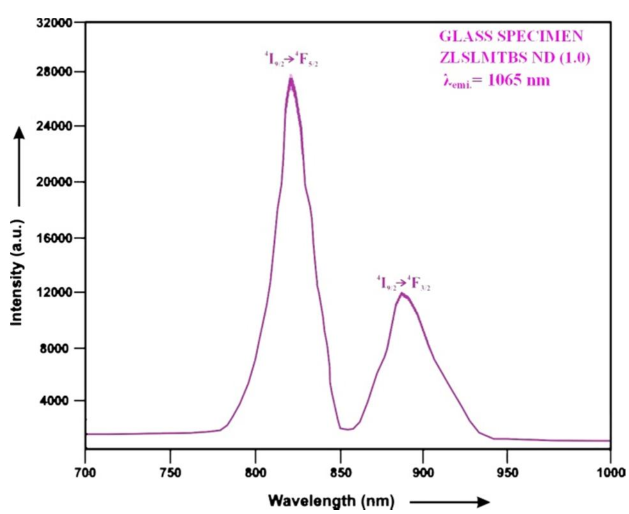

Fig.4: Excitation spectra of ZLSLMTBS ND(1.0) glass.

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E. Fluorescence Spectrum

The fluorescence spectrum of Nd^{3+}doped in zinc lithium sodalime magnesium tungsten borosilicate is shown in Figure 4. There are six broad bands ($^4G_{7/2}\rightarrow{}^4I_{9/2}$), ($^4G_{7/2}\rightarrow{}^4I_{11/2}$), ($^4F_{3/2}\rightarrow{}^4I_{9/2}$) ($^4F_{3/2}\rightarrow{}^4I_{11/2}$), ($^4F_{3/2}\rightarrow{}^4I_{13/2}$) and ($^4F_{3/2}\rightarrow{}^4I_{15/2}$) respectively for glass specimens.

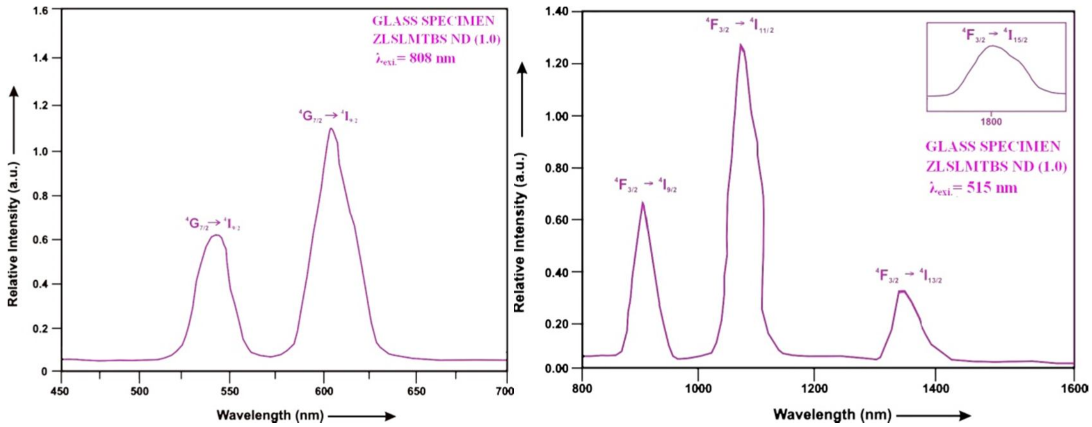

Fig.5: Fluorescence spectrum of ZLSLMTBS ND(1.0) glass.

The wavelengths of these bands along with their assignments are given in **Table 5**.

Table 5. Emission peak wave lengths (λ_p), radiative transition probability (A_{rad}), branching ratio (β_R), stimulated emission crosssection (σ_p), and radiative life time (τ_R) for various transitions in Nd^{3+} doped ZLSMTBS glasses.

Transition		ZLSLMTBS ND(1.0)				ZLSLMTBS ND(1.5)				ZLSLMTBS ND(2.0)			
	λ_{max} (nm)	$A_{rad}(s^{-1})$	β	σ_p (10^{-20} cm^2)	τ_R(μs)	$A_{rad}(s^{-1})$	β	σ_p (10^{-20} cm^2)	τ_R (μs)	$A_{rad}(s^{-1})$	β	σ_p (10^{-20} cm^2)	τ_R (10^{-20} cm^2)
$^4G_{7/2}\rightarrow{}^4I_{9/2}$	532	2934.88	0.4119	0.447	140.33	2884.47	0.4154	0.455	144.01	2838.32	0.4197	0.469	147.88
$^4G_{7/2}\rightarrow{}^4I_{11/2}$	595	2966.74	0.4163	1.146		2841.19	0.4092	1.131		2708.73	0.4006	1.165	
$^4F_{3/2}\rightarrow{}^4I_{9/2}$	905	713.91	0.1002	0.716		709.54	0.1022	0.737		707.26	0.1046	0.755	
$^4F_{3/2}\rightarrow{}^4I_{11/2}$	1075	434.75	0.0610	1.971		433.26	0.0624	2.135		432.44	0.0639	2.261	
$^4F_{3/2}\rightarrow{}^4I_{13/2}$	1320	73.85	0.0104	0.427		73.77	0.0106	0.441		73.72	0.0109	0.456	
$^4F_{3/2}\rightarrow{}^4I_{15/2}$	1800	1.76	0.0002	0.026		1.76	0.0003	0.026		1.76	0.0003	0.027	

V. CONCLUSION

In the present study, the glass samples of composition :(25-x)SiO_2: 10ZnO: 10Li_2O: 10Na_2O: 10CaO: 10MgO: 10WO_3: 15B_2O_3:xNd_2O_3. (where x =1, 1.5, 2 mol %) have been prepared by melt-quenching method. The stimulated emission cross section (σ_p) has highest value for the transition ($^4F_{3/2}\rightarrow{}^4I_{11/2}$) in all the glass specimens doped with Nd^{3+} ion. This shows that ($^4F_{3/2}\rightarrow{}^4I_{11/2}$) transition is most probable transition. The results show that the Nd^{3+} doped borosilicate glasses could be potential candidates for laser applications.

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