

Spectral and Raman Analysis of Sm^{3+} Doped in Zinc Lithium Soda lime Alumino Silicate Glasses

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Abstract

Glass of the system: $(40-x)\text{SiO}_2:10\text{ZnO}:10\text{Li}_2\text{O}:10\text{CaO}:10\text{Na}_2\text{O}:20\text{Al}_2\text{O}_3:x\text{Sm}_2\text{O}_3$ (where $x=1, 1.5, 2$ mol %) have been prepared by melt-quenching method (where $x=1, 1.5, 2$ mol%) have been prepared by melt-quenching technique. The amorphous nature of the prepared glass samples was confirmed by X-ray diffraction. The absorption spectra of three Sm^{3+} doped zinc lithium soda lime alumino silicate glasses have been recorded at room temperature. The various interaction parameters like Slater-Condon parameters F_k ($k=2, 4, 6$), Lande parameter (ζ_{4f}), nephelauxetic ratio (β'), bonding parameters ($b^{1/2}$) and Racah parameters E^k ($k=1, 2, 3$) have been computed. Judd-Ofelt intensity parameters and laser parameters have also been calculated.

Keywords: ZLSLAS Glasses, Optical properties, Judd-Ofelt analysis, Raman Analysis.

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I. INTRODUCTION

Rare earth doped silicate glasses are being extensively studied due to their technological importance and their applications in solid state lasers, optical fibers and optical amplifiers [1-5]. Among rare earth ion doped glasses, borosilicate glasses are popular host materials due to their good transparency and easy to draw into fibers for different laser applications [6-8]. Silicate glasses possess excellent optical properties, chemical durability and better mechanical properties. Silicate glasses possess unique and interesting optical, thermal, and electrical properties which make them candidates for a wide range of applications [9-12]. Due to their good chemical durability, samarium-doped soda-lime silicate glasses are attractive materials for the fabrication of low-cost integrated optical amplifiers by using the ion-exchange technique [13-15]. Silicate Glasses are both scientifically and technologically important materials because they generally offer some unique physical properties better than other Glasses [16-18]. Among active rare-earth ions Sm^{3+} exhibits high solubility in ceramic glasses, which also possess excellent optical and physical properties [19-22].

The aim of the present study is to prepare the Sm^{3+} doped zinc lithium soda lime alumino silicate glass with different Sm_2O_3 concentrations. The absorption spectra, fluorescence spectra of Sm^{3+} of the glasses were investigated. The Judd-Ofelt theory has been applied to compute the intensity parameters Ω_λ ($\lambda=2, 4, 6$). These intensity parameter have been used to evaluate optical properties such as spontaneous emission probability, branching ratio, radiative life time and stimulated emission cross section. Large stimulated emission cross section is one of the most important parameters required for the design of high peak power solid state lasers.

II. EXPERIMENTAL TECHNIQUES

Preparation of glasses

The following Sm^{3+} doped zinc lithium soda lime alumino silicate glass samples $(40-x)\text{SiO}_2:10\text{ZnO}:10\text{Li}_2\text{O}:10\text{CaO}:10\text{Na}_2\text{O}:20\text{Al}_2\text{O}_3:x\text{Sm}_2\text{O}_3$ (where $x=1, 1.5, 2$) have been prepared by melt-quenching method. Analytical reagent grade chemical used in the present study consist of $\text{SiO}_2, \text{ZnO}, \text{Li}_2\text{O}, \text{CaO}, \text{Na}_2\text{O}, \text{Al}_2\text{O}_3$ and Sm_2O_3 . They were thoroughly mixed by using an agate pestle mortar. then melted at 1055°C by an electrical muffle furnace for 2h., After complete melting, the melts were quickly poured in to a preheated stainless steel mould and annealed at temperature of 250°C for 2h 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.

Table 1

Chemical composition of the glasses

Sample	Glass composition (mol %)
ZLSLAS (UD)	40SiO ₂ :10ZnO:10Li ₂ O:10CaO:10Na ₂ O:20Al ₂ O ₃
ZLSLAS M(SM1)	39SiO ₂ :10ZnO:10Li ₂ O:10CaO:10Na ₂ O:20Al ₂ O ₃ :1 Sm ₂ O ₃
ZLSLAS(SM 1.5)	38.5SiO ₂ :10ZnO:10Li ₂ O:10CaO:10Na ₂ O:20Al ₂ O ₃ :1.5 Sm ₂ O ₃
ZLSLAS(SM 2)	38 SiO ₂ :10ZnO:10Li ₂ O:10CaO:10Na ₂ O:20Al ₂ O ₃ : 2 Sm ₂ O ₃

ZLSLAS(UD) -Represents undopedZinc Lithium Soda Lime AluminoSilicate glass specimens.

ZLSLAS(SM) -Represents Sm³⁺ dopedZinc Lithium Soda Lime Alumino Silicateglass specimens.

III. THEORY

3.1 Oscillator Strength

The intensity of spectral lines are expressed in terms of oscillator strengths using the relation [23].

$$f_{\text{expt.}} = 4.318 \times 10^{-9} \int \epsilon(\nu) d\nu \quad (1)$$

where, $\epsilon(\nu)$ is molar absorption coefficient at a given energy ν (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 [24], using the modified relation:

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

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

3.2. Judd-Ofelt Intensity Parameters

According to Judd [25] and Ofelt [26] theory, independently derived expression for the oscillator strength of the induced forced electric dipole transitions between an initial J manifold $|4f^N(S, L) J\rangle$ level and the terminal J' manifold $|4f^N(S', L') J'\rangle$ is given by:

$$\frac{8\pi^2 mc \bar{\nu}}{3h(2J+1)n} \left[\frac{(n^2+2)^2}{9} \right] \times S(J, J') \quad \text{Where, the line strength } S(S', L') \quad (3)$$

is given by the equation

$$S(J, J') = e^2 \sum \Omega_\lambda \langle 4f^N(S, L) J \| U^{(\lambda)} \| 4f^N(S', L') J' \rangle^2$$

$\lambda = 2, 4, 6$

In the above equation m is the mass of an electron, c is the velocity of light, ν 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 ($\lambda = 2, 4, 6$) are known as Judd-Ofelt intensity parameters .

3.3 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 $|4f^N(S', L') J'\rangle$ to a final manifold $|4f^N(S, L) J\rangle$ is given by:

$$A [(S', L') J'; (S, L) J] = \frac{64 \pi^2 \nu^3}{3h(2J'+1)} \left[\frac{n(n^2+2)^2}{9} \right] \times S(J', J) \quad (4)$$

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

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

$$\beta[(S', L') J'; (S, L) J] = \sum \frac{A[(S' L)]}{A[(S' L') J' (S L)]} \quad (5)$$

S L J

where, the sum is over all terminal manifolds.

The radiative life time is given by

$$\tau_{rad} = \sum A[(S', L') J'; (S, L) J] = A_{Total}^{-1} \quad (6)$$

S L J

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

$$\sigma_p(\lambda_p) = \left[\frac{\lambda_p^4}{8\pi c n^2 \Delta\lambda_{eff}} \right] \times A[(S', L') J'; (\bar{S}, \bar{L}) \bar{J}] \quad (7)$$

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

3.4 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 [27, 28]. The Nephelauxetic Ratio is given by

$$\beta' = \frac{\nu_g}{\nu_a} \quad (8)$$

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

$$b^{1/2} = \left[\frac{1-\beta'}{2} \right]^{1/2} \quad (9)$$

IV. RESULT AND DISCUSSION

4.1 XRD Measurement

Figure 1 presents the XRD pattern of the sample contain - SiO₂ which is show 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.

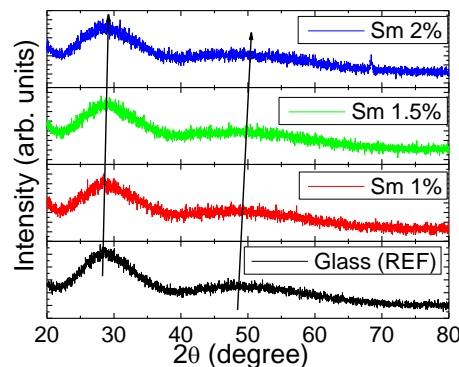


Fig. 1: X-ray diffraction pattern of SiO₂:ZnO:Li₂O:CaO:Na₂O:Al₂O₃:Sm₂O₃

4.2 Raman spectra

The Raman spectrum of Zinc Lithium Soda Lime Alumino Silicate (ZLSLAS) glass specimens is recorded and is shown in Fig. 2. The spectrum peaks located at 600, 795 and 1205 cm⁻¹. The band at 600 and 795 cm⁻¹ assigned to Si–O–Si symmetric stretching and bending vibration, respectively. The band at 1205 cm⁻¹ assigned to Si–O–Si asymmetric stretching.

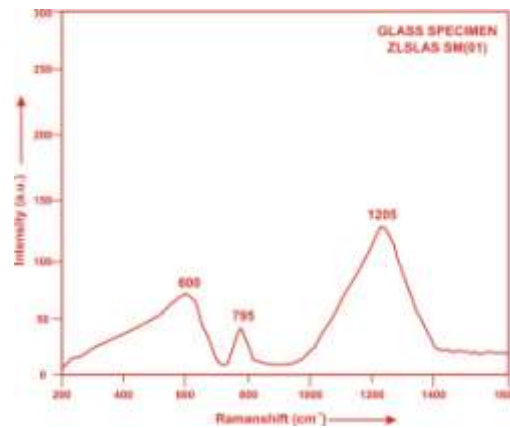


Fig.2 Raman spectrum of ZLSLAS SM (01) glass.

4.3 Absorption Spectrum

The absorption spectra of Sm^{3+} doped ZLSLAS(SM 01) glass specimen has been presented in Figure 3 in terms of optical density versus wavelength (nm). Ten absorption bands have been observed from the ground state $^6H_{5/2}$ to excited states $^6F_{1/2}$, $^6F_{7/2}$, $^6F_{9/2}$, $^4G_{7/2}$, $^4I_{9/2}$, $^4M_{7/2}$, $(^6P, ^4P)_{5/2}$, $^4F_{7/2}$, $^4D_{1/2}$, and $(^4D, ^6P)_{5/2}$ for Sm^{3+} doped ZLSLAS glasses

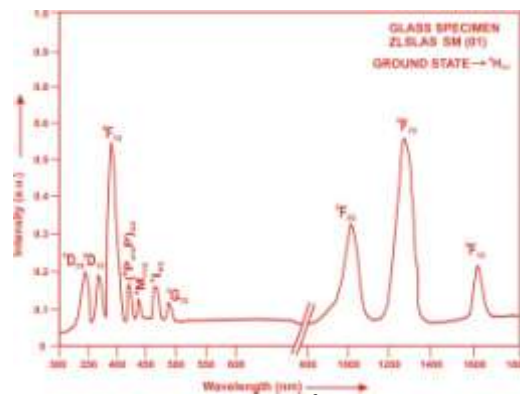


Fig.3: Absorption spectrum of Sm^{3+} doped ZLSLAS(01) glass

The experimental and calculated oscillator strengths for Sm^{3+} ions in zinc lithium soda lime alumino silicate glasses are given in Table 2

Table2: Measured and calculated oscillator strength ($P_m \times 10^6$) of Sm^{3+} ions in ZLSLAS glasses.

Energy level from $^6H_{5/2}$	Glass ZLSLAS (SM01)		Glass ZLSLAS (SM1.5)		Glass ZLSLAS (SM02)	
	P_{exp}	P_{cal}	P_{exp}	P_{cal}	P_{exp}	P_{cal}
$^6F_{1/2}$	1.58	1.59	1.54	1.56	1.50	1.53
$^6F_{7/2}$	5.46	5.47	5.41	5.43	5.37	5.40
$^6F_{9/2}$	3.80	3.82	3.75	3.78	3.71	3.76
$^4G_{7/2}$	0.18	0.12	0.16	0.12	0.14	0.12
$^4I_{9/2}, ^4M_{15/2}, ^4I_{11/2}$	1.16	1.87	1.11	1.85	1.07	1.84
$^4M_{17/2}, ^4G_{9/2}, ^4I_{15/2}$	0.30	0.24	0.27	0.24	0.24	0.24
$(^6P, ^4P)_{5/2}, ^4L_{13/2}$	1.33	1.29	1.29	1.29	1.24	1.28
$^4F_{7/2}, ^6P_{3/2}, ^4K_{11/2}$	5.65	5.54	5.62	5.54	5.57	5.52
$^4D_{1/2}, ^6P_{7/2}, ^4L_{17/2}$	2.44	1.88	2.41	1.86	2.37	1.85
$^4D_{3/2}, (^4D, ^6P)_{5/2}$	2.68	3.43	2.63	3.41	2.59	3.40
r.m.s. deviation	0.3736		0.3838		0.3901	

Computed values of F_2 , Lande' parameter (ξ_{4f}), Nephelauxetic ratio (β') and bonding parameter ($b^{1/2}$) for Sm^{3+} doped ZLSLAS glass specimen are given in Table 3.

Table 3. F_2, ζ_{4f}, β' and $b^{1/2}$ parameters for Samarium doped glass specimen.

Glass Specimen	F_2	ζ_{4f}	β'	$b^{1/2}$
Sm^{3+}	358.82	1258.16	0.9337	0.1821

Judd-Ofelt intensity parameters Ω_λ ($\lambda=2,4,6$) were calculated by using the fitting approximation of the experimental oscillator strengths to the calculated oscillator strengths with respect to their electric dipole contributions. In the present case the three Ω_λ parameters follow the trend $\Omega_2 > \Omega_4 > \Omega_6$. The spectroscopic quality factor (Ω_4/Ω_6) related with the rigidity of the glass system has been found to lie between 1.0825 and 1.0996 in the present glasses.

The value of Judd-Ofelt intensity parameters are given in **Table 4**

Table 4: Judd-Ofelt intensity parameters for Sm^{3+} doped ZLSLAS glass specimens

Glass Specimen	$\Omega_2(\text{pm}^2)$	$\Omega_4(\text{pm}^2)$	$\Omega_6(\text{pm}^2)$	Ω_4/Ω_6	Ref.
ZLSLAS (SM01)	4.845	4.488	4.146	1.0825	P.W.
ZLSLAS (SM1.5)	4.742	4.486	4.099	1.0944	P.W.
ZLSLAS (SM02)	4.641	4.473	4.068	1.0996	P.W.
TNS (SM)	1.584	0.544	0.394	1.3807	[29]
LBSG(SM)	10.225	7.385	7.443	0.9922	[30]

4.4. Fluorescence Spectrum

The fluorescence spectrum of Sm^{3+} doped in zinc lithium soda lime alumino silicate glass is shown in Figure 4. There are nine broad bands observed in the Fluorescence spectrum of Sm^{3+} doped zinc lithium soda lime alumino silicate glass. The wavelengths of these bands along with their assignments are given in Table 5. Fig. (4). Shows the fluorescence spectrum with nine peaks (${}^4G_{5/2} \rightarrow {}^6H_{5/2}$), (${}^4G_{5/2} \rightarrow {}^6H_{7/2}$), (${}^4G_{5/2} \rightarrow {}^6H_{9/2}$), (${}^4G_{5/2} \rightarrow {}^6H_{11/2}$), (${}^4G_{5/2} \rightarrow {}^6H_{13/2}$), (${}^4G_{5/2} \rightarrow {}^6F_{3/2}$), (${}^4G_{5/2} \rightarrow {}^6F_{5/2}$), (${}^4G_{5/2} \rightarrow {}^6F_{7/2}$) and (${}^4G_{5/2} \rightarrow {}^6F_{9/2}$), respectively for glass specimens.

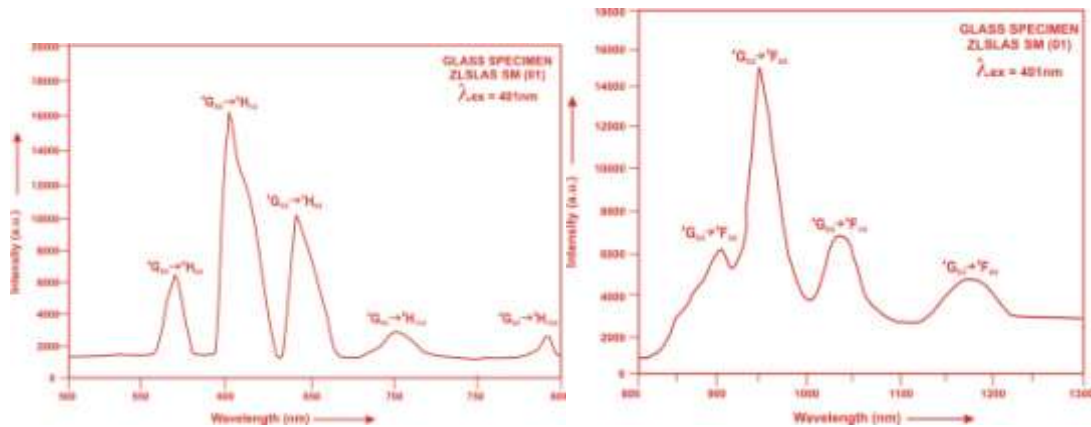


Fig.4: fluorescence spectrum of Sm^{3+} doped ZLSLAS(01) glass.

Table 5. Emission peak wave lengths (λ_p), radiative transition probability (A_{rad}), branching ratio (β), stimulated emission cross-section (σ_p) and radiative life time (τ_R) for various transitions in Sm^{3+} doped ZLSLAS glasses.

Transition	ZLSLAS SM 01					ZLSLAS SM 1.5				ZLSLAS SM 02			
	λ_{em} (nm)	$A_{rad}(s^{-1})$	β	σ_p (10^{-20}cm^2)	$\tau_R(\mu s)$	$A_{rad}(s^{-1})$	β	σ (10^{-20}cm^2)	$\tau_R(\mu s)$	$A_{rad}(s^{-1})$	β	σ_p (10^{-20}cm^2)	$\tau_R(\mu s)$
${}^4G_{5/2} \rightarrow {}^6H_{5/2}$	562	10.56	0.0367	0.00398	3479.71	10.58	0.0368	0.00434	3473.02	10.45	0.0369	0.00476	3535.17
${}^4G_{5/2} \rightarrow {}^6H_{7/2}$	602	112.02	0.3898	0.0448		112.19	0.3896	0.0486		111.12	0.3928	0.0514	
${}^4G_{5/2} \rightarrow {}^6H_{9/2}$	645	106.41	0.3703	0.0423		106.68	0.3705	0.0448		103.95	0.3675	0.0456	
${}^4G_{5/2} \rightarrow {}^6H_{11/2}$	705	27.45	0.0955	0.0133		27.49	0.0955	0.0139		27.32	0.0966	0.0145	
${}^4G_{5/2} \rightarrow {}^6H_{13/2}$	786	2.62	0.0091	0.00174		2.63	0.0091	0.00181		2.58	0.0091	0.00186	
${}^4G_{5/2} \rightarrow {}^6F_{3/2}$	915	4.37	0.0152	0.00692	4.38	0.0152	0.0072	4.20	0.0149	0.00728			
${}^4G_{5/2} \rightarrow {}^6F_{5/2}$	955	19.70	0.0686	0.0306	19.74	0.0686	0.0318	19.06	0.0674	0.0320			
${}^4G_{5/2} \rightarrow {}^6F_{7/2}$	1036	2.47	0.0086	0.00448	2.47	0.0086	0.00465	2.47	0.0087	0.0048			
${}^4G_{5/2} \rightarrow {}^6F_{9/2}$	1180	1.77	0.0062	0.00459	1.78	0.0062	0.00473	1.71	0.0061	0.00468			

V. CONCLUSION

In the present study, the glass samples of composition $(40-x)\text{SiO}_2:10\text{ZnO}:10\text{Li}_2\text{O}:10\text{CaO}:10\text{Na}_2\text{O}:20\text{Al}_2\text{O}_3:x\text{Sm}_2\text{O}_3$. (where $x=1, 1.5, 2\text{mol } \%$) have been prepared by melt-quenching method. The Judd-Ofelt theory has been applied to calculate the oscillator strength and intensity parameters Ω_λ ($\lambda=2, 4, 6$). The radiative transition probability, branching ratio are highest for (${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$) transition and hence it is useful for laser action. The stimulated emission cross section (σ_p) has highest value for the transition (${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$) in all the glass specimens doped with Sm^{3+} ion. This shows that (${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$) transition is most probable transition. The value of stimulated emission cross-section (σ_p) is found to be maximum for the transition (${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$) for glass ZLSLAS (SM 02), suggesting that glass ZLSLAS (SM 02) is better compared to the other two glass systems ZLSLAS (SM 01) and ZLSLAS (SM 1.5).

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