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Judd-Ofelt analysis of Gd^{3+} Doped in Lead Lithium Antimony Alumino Sodalime Silicate Glasses

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Abstract

Glass sample of Lead Lithium Antimony Alumino Sodalime Silicate (35-x) SiO_2:10PbO:10Li_2O:10Sb_2O_3:10Al_2O_3:10CaO:15Na_2O:xGd_2O_3. (where x=1,1.5 and 2 mol%) have been prepared by melt-quenching technique. The amorphous nature of the prepared glass samples was confirmed by X-ray diffraction. The physical parameters like density, dielectric constant and electrical susceptibility have been evaluated. Dielectric constant, refractive index, Optical basicity varies with increasing mole% of Gd_2O_3 respectively. Optical absorption was recorded at room temperature for all glass samples. Judd-Ofelt intensity parameters Ω_λ (λ=2, 4 and 6) are evaluated from the intensities of various absorption bands of optical absorption spectra.

Keywords: LLAASLS Glasses, Physical Properties, Judd-Ofelt Theory, Rare earth ions.

Introduction:

Silicate glasses are very important because of their technological and scientific application [1-3]. Rare earth doped transparent silicate glasses and glass ceramics are of increasing interests in various optical applications, because of their superior optical, physical, liner and electrical properties [4,5]. Among oxide glasses, silicate glasses are the most popular glass hosts for making optical fiber lasers and amplifiers [6-9].

Glasses based on high content of silica possess very good thermal, mechanical and chemical stability [10,11].Therefore they are more compatible with the fabrication process in the development of optical devices .Silica glasses possess low refractive index and low dispersion and so they are good candidates to minimize self-focusing effects of laser radiation [12-15].

In the present work, the absorption spectra of Gd^{3+} doped lead lithium antimony alumino sodalime silicate glass with different Gd_2O_3 concentrations have been investigated and the effect of Gd_2O_3content on the various physical parameters such as density, refractive index and molar refractivity. In addition the optical basicity and Electrical susceptibility were theoretically determined.

Experimental:

Preparation of glasses:

The following Gd^{3+} doped silicate glass samples (35-x) SiO_2:10PbO:10Li_2O: 10Sb_2O_3: 10Al_2O_3:10CaO:15Na_2O:xGd_2O_3. (where x=1,1.5 and 2 mol%) have been prepared by melt-quenching method. Analytical reagent grade chemical used in the present study consist of SiO_2, PbO, Li_2O, Sb_2O_3, Al_2O_3, CaO, Na_2O, and Gd_2O_3. They were thoroughly mixed by using an agate pestle mortar. then melted

at 850^{0}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^{0}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 %)
LLAASLS (UD)	$35SiO_2$:10PbO:$10Li_2O$:$10Sb_2O_3$:$10Al_2O_3$:10CaO:$15Na_2O$
LLAASLS (GD1)	$34SiO_2$:10PbO:$10Li_2O$:$10Sb_2O_3$:$10Al_2O_3$:10CaO:$15Na_2O$:$1Gd_2O_3$.
LLAASLS (GD 1.5)	$33.5SiO_2$:10PbO:$10Li_2O$:$10Sb_2O_3$:$10Al_2O_3$:10CaO:$15Na_2O$:1.5 Gd_2O_3.
LLAASLS (GD 2)	$33SiO_2$:10PbO:$10Li_2O$:$10Sb_2O_3$:$10Al_2O_3$:10CaO:$15Na_2O$:2 Gd_2O_3.

LLAASLS (UD) -Represents undoped Lead Lithium Antimony Alumino Sodalime Silicate glass specimens.
LLAASLS (GD) -Represents Gd^{3+} doped Lead Lithium Antimony Alumino Sodalime Silicate glass specimens.

Physical Properties:

Density measurement:

The density of all glasses was measured by using Archimedes principle with xylene as immersing liquid. The relation used is

$$\rho(gm/cm^3) = \frac{W_a}{W_a - W_b} \times \rho_b \qquad (1)$$

Where W_a is the weight of glass sample in air, W_b is the weight of glass sample when immersed in xylene and ρ_b is the density of xylene (0.86gm/cm^3).

The molar volume of the glass samples can be calculated from following expression:

$$V_m = \frac{M_T}{\rho} \qquad (2)$$

Where ρ is the density of the sample and M_T is the total molecular weight of the multi-component glass system given by

$$M_T = X_{SiO2} Z_{SiO2} + X_{PbO} Z_{PbO} + X_{Li2O} Z_{Li2O} + X_{Sb2O3} Z_{Sb2O3} + X_{Al2O3} Z_{Al2O3} + X_{CaO} Z_{CaO} + X_{Na2O} Z_{Na2O} + X_{Gd2O3} Z_{Gd2O3} \qquad (3)$$

Where X_{SiO2}, X_{PbO}, X_{Li2O}, X_{Sb2O3}, X_{Al2O3}, X_{CaO}, X_{Na2O}, X_{Gd2O3}, are the molar fraction of the constituent oxides and Z_{SiO2}, Z_{PbO}, Z_{Li2O}, Z_{Sb2O3}, Z_{Al2O3}, Z_{CaO}, Z_{Na2O}, X_{Gd2O3}, are the molar weights of the constituent oxides.

Refractive index measurement:

The refractive index were measured by using an Abbe refractometer with sodium vapor lamp as the light source emitting the light at a wavelength λ of 589.3nm and having mono-bromonaphthalene as the contact layer between the sample and prism of the refractometer.

Reflection loss:

The reflection loss from the glass surface was computed from the refractive index using Fresnel's formula [16]

$$R_L = \left[\frac{(n-1)}{(n+1)}\right]^2 \quad (4)$$

Where n is the refractive index.

Molar refraction:

The molar refractivity of the glass samples were calculated using the formula which is well known as Volf and Lorentz-Lorentz formula [17]

$$R_m = \left[\frac{(n^2-1)}{(n^2+2)}\right] \times V_m \quad (5)$$

Where n is the refractive index of the glass sample, V_m is the molar volume.

Energy gap:

According to Duffy the energy gap is given by [18]

$$E_g = 20\left(1 - \frac{R_m}{V_m}\right)^2 \quad (6)$$

Molar electronic polarizability:

The molar electronic polarizability of the material can be calculated from following expression [19]

$$\alpha_m = \frac{R_m}{2.52} \quad (7)$$

Dielectric constant:

The dielectric constant was calculated using refractive index of the glass [20]

$$\varepsilon = n^2 \quad (8)$$

Where n is the refractive index.

Optical dielectric constant:

The optical Dielectric Constant refractive index of the glass [21]

$$p\frac{dt}{dp} = (\varepsilon - 1) = n^2 - 1 \quad (9)$$

Where ϵ is the dielectric constant.

Electronic polarizability:

The electronic polarizability was cal calculated using the formula [22]

$$\alpha_e = \frac{3(n^2-1)}{4\Pi A_V(n^2+2)} \quad (10)$$

Where A_V is the Avogadro number.

Ionic concentrations:

The ionic concentrations of the glass samples are determined using the following relation [23]

$$N\ (\text{ions}/\text{cm}^3) = \frac{(Avogadro's\ number)(glass\ density)}{(Average\ molecular\ weight)} \times (mol\%\ of\ rare\ earth) \quad (11)$$

Polaron radius:

The polaron radius was calculated using the formula [24]

$$R_p = \frac{1}{2} \times \left(\frac{\Pi}{6N} \right)^{\frac{1}{3}} \quad (12)$$

Where N is the ionic concentrations.

Inter-ionic distance:

Inter-ionic distance of the glass samples is given as [24]

$$R_i = \left(\frac{1}{N} \right)^{\frac{1}{3}} \quad (13)$$

Where R_i is the ionic concentrations.

Field strength:

The field strength was calculated using the formula [25]

$$F\ (cm^3) = \left(\frac{Z}{R_p^{\ 2}} \right) \quad (14)$$

Where Z is the thickness of the samples.

Oxygen packing density:

The oxygen packing density of the glass samples were calculated using the following relation [26]

$$O.P.D. = n\left(\frac{\rho}{M} \right) \times 1000 \quad (15)$$

Where ρ the density of desired glass samples, M is the molecular weight of the sample and n is the number of oxygen atoms in the composition.

Insulating nature:

According to the Herzfeld theory of metallization, If $R_m/V_m > 1$ and $R_m/V_m < 1$ predicting metallic or insulating [27]. Subtracting by 1 gives the metallization (M)

$$M = \left(1 - \frac{R_m}{V_m} \right) \quad (16)$$

Optical basicity:

The optical basicity addresses the ability of oxide glass in contributing the negative charges in the glass matrix. In other words it defines the electron donating power of the oxygen in the oxides glass. The theoretical optical basicity can be calculated by the equation proposed by Duffy and Ingram [28]

$$\Lambda_{th} = X_1 \Lambda_1 + X_2 \Lambda_2 + X_3 \Lambda_3 + X_4 \Lambda_4 + X_5 \Lambda_5 + X_6 \Lambda_6 + X_7 \Lambda_7 + X_8 \Lambda_8 \quad (17)$$

Where X_1, X_2, X_3 ,X_4, X_5, X_6, X_7, X_8 are equivalent fraction based on the amount of oxygen each oxide contributes to the overall glass stoichometry and Λ_1, Λ_2 , Λ_3,Λ_4, Λ_5, Λ_6, Λ_7, Λ_8 are basicities assigned to the individual oxides. The values of optical basicity of each oxide are: Λ (SiO_2) =0.48, Λ (PbO) =1.18, Λ (Li_2O) =1.00, Λ (Sb_2O_3) =1.18, Λ (Al_2O_3) =0.40, Λ (CaO) =1.00, Λ (Na_2O) =1.11, Λ (Gd_2O_3)=0.955.

Electronic polarizability of oxide ions polarizability:

The Electronic polarizability of oxide ions has been calculated using the equation proposed by Dimitrov and Sakka [29]

$$\alpha_O^{2-(n)} = \left[\frac{R_m}{2.52} - \sum \alpha_i\right]\left(N_o^{2-}\right)^{-1} \quad (18)$$

Where $\sum \alpha_i$ In the above equation is molar cation polarizability and N_O^{2-} is the number of oxide ions in the chemical formula. The molar cation Polarizability (α) values are α_{Si}=0.033Å^3, α_{Pb}=3.623Å^3, α_{Li} =0.024 Å^3 ,α_{Sb}=1.111 Å^3, α_{Al} =0.052 Å^3 , α_{Ca}= 0.47 Å^3, α_{Na} =0.179 Å^3 and α_{Gd} =1.08 Å^3

Electrical susceptibility (χ):

The Electrical susceptibility was calculated using the formula [30]

$$\chi = \left(\frac{n^2-1}{4\pi}\right) \quad (19)$$

Table 2: The physical and optical properties of SiO_2:PbO:Li_2O:Sb_2O_3:Al_2O_3:10CaO:Na_2O:Gd_2O_3 glasses

Physical properties	**LLAASLS (UD)**	**LLAASLS (GD01)**	**LLAASLS (GDU1.5)**	**LLAASLS (GDU02)**
Refractive Index (n)	1.760	1.762	1.763	1.764
Density (ρ) (gm/cm^3)	3.162	3.242	3.365	3.454
Thickness(Z)	0.245	0.245	0.245	0.245
AveragemolecularweightM(gm)	100.58	103.61	105.12	106.64
Rare earth ions concentratio(N)	--	1.885	1.720	2.355
Dielectric Constant (ϵ)	3.098	3.105	3.108	3.112
Optical Dielectric Constant (pdt/dp)	2.098	2.105	2.108	2.112
MolarVolume V_m (gm/cm^3)	31.809	31.960	31.240	30.874
Reflection losses(R_L)	7.582	7.611	7.626	7.640
Molar refractivity (R_m)	13.089	13.177	12.893	12.754
Polaron radius R_p(A^0)	--	3.263	2.829	2.560
Interionic distance R_i (A^0)	--	0.8096	0.7019	0.6352
Electronic polarizability (α_e)	0.1632	0.1635	0.1637	0.1638
Field strength(F)	--	0.2302	0.3062	0.3739
Molarpolarizability(α_m)	5.194	5.229	5.116	5.061
Oxygen packing density(OPD)	55.016	55.070	56.497	57.331
Metallization criterion (M)	0.5885	0.5877	0.5873	0.5869
Energy gap(E_g)	6.927	6.908	6.898	6.889
Optical basicity	0.7306	0.7372	0.7406	0.7438
Oxideion polarizability	2.555	2.554	2.477	2.433
Electrical susceptibility (χ)	0.1670	0.1675	0.1678	0.1680

Theory:

Oscillator Strength:

The intensity of spectral lines are expressed in terms of oscillator strengths using the relation [31].

$$f_{expt.} = 4.318 \times 10^{-9} \int \varepsilon(\nu)\, d\nu \qquad (20)$$

where, ε (*ν)* 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 [32], using the modified relation:

$$P_m = 4.6 \times 10^{-9} \times \frac{1}{cl} \log \frac{I_0}{I} \times \Delta\upsilon_{1/2} \qquad (21)$$

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.

Judd-Ofelt Intensity Parameters:

According to Judd [33] and Ofelt [34] 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:

$$\frac{8\Pi^2 mc\bar{\upsilon}}{3h(2J+1)} \frac{1}{n}\left[\frac{(n^2+2)^2}{9}\right] \times S(J, J') \qquad (22)$$

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

$$S(J, J') = e^2 \sum \Omega_\lambda \langle 4f^N (S, L)\, J \| U^{(\lambda)} \| 4f^N (S', L')\, J' \rangle^2 \qquad (23)$$

$$\lambda = 2, 4, 6$$

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 (λ=2,4and 6) are known as Judd-Ofelt intensity parameters.

Results and Discussion:

XRD Measurement:

Figure 1 presents the XRD pattern of the sample contain - SiO_2 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

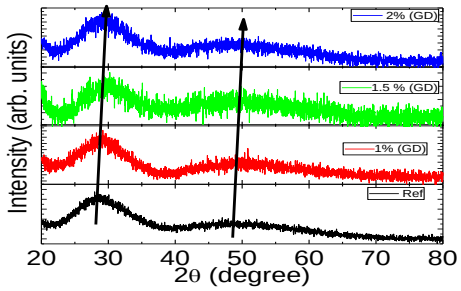

Fig.1: X-ray diffraction pattern of SiO_2: PbO: Li_2O: Sb_2O_3: Al_2O_3: CaO: Na_2O: Gd_2O_3.

Absorption Spectrum:

The absorption spectra of Gd^{3+}doped LLAASLS glass specimens have been presented in Figure 2 in terms of optical density versus wavelength. Nine absorption bands have been observed from the ground state $^8S_{7/2}$ to excited states $^6P_{7/2}$, $^6P_{5/2}$, $^6I_{7/2}$, $^6I_{9/2}$, $^6I_{11/2}$, $^6I_{13/2}$, $^6D_{9/2}$, $^6D_{7/2}$ and $^6D_{3/2}$ for Gd^{3+} doped LLAASLS glasses.

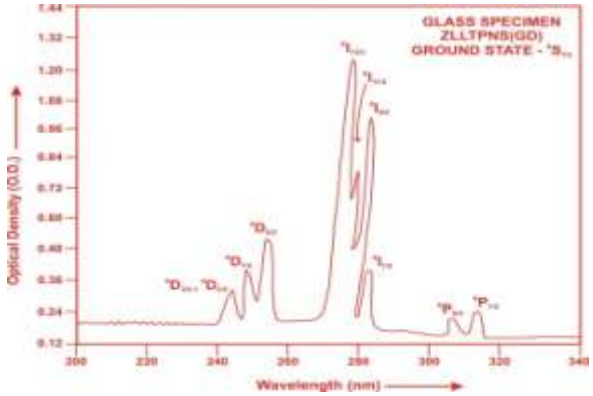

Fig. 2: Absorption spectrum of Gd^{3+}doped LLAASLS glasses.

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

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

Energy level from $^8S_{7/2}$	**Glass LLAASLS (GD01)**		**Glass LLAASLS (GD1.5)**		**Glass LLAASLS (GD02)**	
	$P_{exp.}$	$P_{cal.}$	$P_{exp.}$	$P_{cal.}$	$P_{exp.}$	$P_{cal.}$
$^6P_{7/2}$	0.565	0.514	0.545	0.509	0.522	0.478
$^6P_{5/2}$	0.245	0.209	0.225	0.208	0.200	0.195
$^6I_{7/2}$	3.25	3.09	2.64	3.032	1.76	2.95
$^6I_{9/2}$	25.75	24.23	24.95	23.78	23.75	23.16
$^6I_{11/2}$	12.48	13.57	11.85	13.32	10.95	12.97
$^6I_{13/2}$	38.75	39.34	37.88	38.60	36.78	37.59
$^6D_{9/2}$	5.75	5.75	4.75	5.03	3.88	4.54
$^6D_{7/2}$	3.15	2.86	2.76	2.84	1.85	2.67
$^6D_{3/2}$	1.76	2.16	1.45	2.15	1.22	2.01
r.m.s. deviation	0.6760		0.7275		0.9572	

*Low r.m.s.deviation values clearly indicate the accuracy of fitting.

The order of magnitude of Judd-Ofelt intensity parameters is $\Omega_6>\Omega_2>\Omega_4$ for all the glass specimens. The high values obtained for Ω_2 in all glasses indicate that the Gd^{3+} ion is subjected to higher covalency with low symmetry. The values of Judd-Ofelt intensity parameters are given in **Table 4.**

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

Glass Specimen	$\Omega_2(pm^2)$	$\Omega_4(pm^2)$	$\Omega_6(pm^2)$	Ω_4/Ω_6	Ref.
LLAASLS (GD01)	5.450	0.01831	7.162	0.00255	P.W.
LLAASLS (GD1.5)	5.398	0.01237	7.022	0.00176	P.W.
LLAASLS (GD02)	5.064	0.01004	6.834	0.00147	P.W.
ZBLAN (GD)	2.49	----	2.30	----	[35]
ZBLAK(GD)	4.66	----	1.88	----	[35]

Conclusion:

In the present study, the glass samples of composition: $(35\text{-}x)SiO_2$:10PbO:$10Li_2O$:$10Sb_2O_3$:$10Al_2O_3$:10CaO:$10Na_2O$:xGd_2O_3. (where x =1, 1.5and 2mol %) have been prepared by melt-quenching method. The density, refractive index and optical basicity increases with an increase in concentration of Gd_2O_3. The Judd–Ofelt intensity parameters determined from the room temperature absorption spectrum are: $\Omega_2=5.304(pm^2)$, $\Omega_4=0.01357(pm^2)$ and $\Omega_6=7.006(pm^2)$. The parameter values are comparable with those in other glasses, but larger than in LLAASLS glasses.

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