OPTICAL ABSORPTION AND JUDD-OFELT OF Pr^{3+} DOPED ANTIMONY BASED GLASSES

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ABSTRACT

Glass with the composition $79.75Sb_2O_3$-$10Li_2O$-$10WO_3$-$0.25PrF_3$ (mol%) were synthesized employing melt quenching technique. Visible and near infrared absorption spectra were measured the Judd–Ofelt (J-O) parameters Ω_2, Ω_4 and Ω_6, were determined for Pr^{3+} ions using the absorption bands and calculate the oscillator strengths, these parameters are used to calculated various radiative properties of different Pr^{3+} transitions.

KEYWORDS: Sb_2O_3-Li_2O-WO_3 glass, spectroscopic properties Pr^{3+}, Judd-Ofelt analysis.

1 INTRODUCTION

Glasses doped with rare earth (RE) ions draw much attention for the past few decades due to their potential applications in the field of science and technology such as optical fibers, detectors, sensors, solar concentrators, display monitors, lasing devices [1], solar cells, sensors and light emitting diodes (LEDs) [2]. Antimony based glasses are potentially important host materials for developing RE doped optical devices, is said to be one of the best glass formers, owing to its superior characteristic features such a the phonon energy close to 600 cm^{-1} is much lower compared to glasses containing lighter elements such as silicon, boron or phosphorus [3] and large optical non-linearity that is correlated to high refractive index, good mechanical properties and better chemical durability than that of fluoride or tellurite glasses [4]. Presence of lithium oxide (Li_2O) in a glass can enhance the chemical stability and modifies physical as well as chemical properties [5]. Addition of WO_3, as a network modifier or an intermediate oxide, to tellurite glasses provides several advantageous properties, such as doping with rare earth elements in a wide range, modifying the composition by a third, fourth, and even fifth component controlling the optical properties, enhancing the chemical stability and devitrification resistance [6,7]. It is also that recognized tungsten (W) ions are capable to influence the optical properties of rare earth ions in glasses, for the main reason that these ions can exist in various valence states such as W^{6+}, W^{5+} and W^{4+}, respectively.

Among all the rare earth ions, Pr^{3+} activated amorphous materials have attracted many researchers because of their potential applications in the field of ultraviolet laser, scintillator and lamp phosphor [8]. Furthermore, energy levels of Pr^{3+} ions demonstrate several meta-stable states and many of the researchers focus on the $^3P_0$-$^3H_4$ (blue) and $^1D_2$-$^3H_4$ (orange) laser transitions which offer emission in the visible region [9]. There are many studies of optical properties of Pr^{3+} ions in various environments we choice some of them in the following literature [10-13].

Since the publication by (J-O) theory [14,15], describes the origin of electric-dipole intensity for the nominally parity-forbidden f-f transitions between SLJ-multiplets ($^{2S+1}L_J$); it provides means of expressing the dipole-strength in terms of three material-dependent parameters (Ω_2, Ω_4, and Ω_6) [16]. However it is known that the application of (J-O) theory to Pr^{3+} ion doped in some host glasses leads to negative values of the phenomenological Ω_2 intensity parameter due to the small energy difference between the ground state configuration $4f^2$ and the first excited state configuration of $4f^15d^1$ [17]. To overcome this problem, the hypersensitive $^3H_4\rightarrow{}^3P_2$ transition has not been taken in account for calculating (J-O) parameters or modifications of the conventional (J-O) theory have been used [17].

In this paper, we have studied of $79.75Sb_2O_3$-$10Li_2O$-$10WO_3$-$0.25PrF_3$ glass and the present study includes optical absorption. We reported the spectroscopic and laser properties such as the phenomenological Judd-Ofelt (J-O) intensity parameters Ω_λ (λ=2, 4 and 6) used to calculate the various radiative properties. The results are then analyzed and compared to other findings.

2 EXPERIMENTAL PROCEDURE

2.1 GLASS SYNTHESIS

The glass composition $79.75Sb_2O_3$-$10Li_2O$-$10WO_3$-$0.25PrF_3$ is chosen for the present study. The prepared glass along with symbol are as follows:

P25: $79.75Sb_2O_3$-$10Li_2O$-$10WO_3$-$0.25PrF_3$

Glass sample with are prepared by next technique: the batch of 5 g weight prepared by the proportional amounts of grade material powders with purity viz., Sb_2O_3 (99+%), ACROS ORGANICS, WO_3 (99.995%), Li_2O (99% Alfa Aesar), and PrF_3 (99.9% Sigma Aldrich) are homogenized and they were melted in silica crucible under heat flame in at temperature 850°C for 5-10 min in air. The resultant melt was swirled to ensure the homogeneity while a release of CO_2 was observed, due to the decomposition of lithium carbonate and then poured on a preheated brass plate and subsequently annealed in electrical furnace at 290°C for three hours to eliminate internal mechanical stress and polishing was implemented after annealing.

Densities, ρ, of the glasses were determined at room temperature by the Archimedes method and a digital balance of sensitivity 10-4 g. Density values obtained by five repeated measurements showed an error of 0.1%.The absorption spectra were measured onto polished samples by a Agilent Cary 5000 UV-Vis-NIR spectrophotometer operating between 175 and 3300 nm, with around 2 nm resolution.

3 RESULTS AND DISCUSSION

3.1 ABSORPTION SPECTRUM

Fig.1 shown the optical absorption of Pr^{3+} doped in $79.5Sb_2O_3$-$10Li_2O$-$10WO_3$, glass. Pr^{3+} ions reveal themselves in the absorption spectra through different absorption peaks [18], consists of four absorption peaks at 449, 473, 486 and 595 nm corresponding from the ground state 3H4 to the excited states $^3P_2$, $^3P_1$, $^3P_0$ and $^1D_2$ in the visible region and five absorption peaks at 1014, 1430, 1538, 1944 and 2263 nm corresponding from the ground state $^3H_4$ to the excited state $^1G_4$, $^3F_4$, $^3F_3$, $^3F_2$, and $^3H_6$ in the infrared region, respectively.

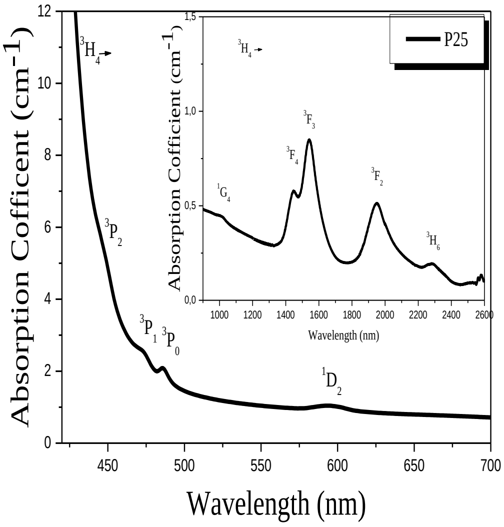

Figure 01: Absorption spectrum of P25 glass

3.2 JUDD-OFELT CALCULATIONS

The Judd–Ofelt (J-O) theory [14,15] is widely used to calculate 4f transition intensities of rare-earth ions in various hosts. Its application requires the computation of three parameters Ω_2, Ω_4 and Ω_6 by a fitting procedure of experimental data usually obtained from ground state absorption [19]. The experimental oscillator strength (f_{exp}) for the different bands was obtained from the equation following form:

$$f_{exp} = \frac{mc^2}{e^2 N \pi \lambda^2} \int \alpha(\lambda) d\lambda \quad (1)$$

where α(λ) is absorption coefficient, m is the mass of an electron, e is the charge of an electron, N is concentration of Pr^{3+} (N_{Pr}=0.29292×1020 ions/cm^3), n is the refractive index (n=2.0491), c is the velocity of light, λ is the mean wavelength of the absorption band.

The experimental oscillator strength is then used to obtain the (J-O) parameters Ω_2, Ω_4 and Ω_6 by solving the set of in the equations following form [20]:

$$f_{calc} = \frac{8\pi^2 mc\left(n^2+2\right)^2}{27nh(2J+1)\lambda} \sum_{\lambda=2,4,6} \Omega_\lambda \left| \left\langle (S,L)J \left\| U^\lambda \right\| (S',L')J' \right\rangle \right|^2 \quad (2)$$

where Ω_2, Ω_4, and Ω_6 are the Judd–Ofelt intensity parameters, and $|\langle(S,L)J\|U(\lambda)\|(S',L')J'\rangle|^2$ are the doubly reduced matrix elements. These elements are assumed to be independent of the host glass and here we use the elements determined by Weber for Pr^{3+} in LaF_3

[21].

The root-mean-square deviation r.m.s between the experimental and the calculated oscillator strengths can be calculated based on the following relationship [22]:

$$r.m.s = \left[\frac{\sum \left(f_{\exp} - f_{calc} \right)^2}{(q-p)} \right]^{\frac{1}{2}} \quad (3)$$

Here, q is the number of analyzed spectral bands, and p is the number of the parameters sought, which in this case are three.

Table 01: Absorption band assignments (from the ground state, $^3H_4$), wavelengths (nm), energy (cm^{-1}), experimental (f_{exp}) and calculated (f_{cal}) oscillator strengths ($\times10^{-6}$), r.m.s deviations, spectroscopic quality factor (χ), $\Sigma\Omega_t$ and J-O parameters obtained by including and excluding the $^3H_4 \rightarrow {}^3P_2$ hypersensitive transition in P25 glass

Levels	Wavelength (nm)	Energy (cm^{-1})	Line strengths and Oscillator strengths			
			With (Including) $^3P_2$ level		Without (Excluding) $^3P_2$ level	
			f_{exp} ($\times10^{-6}$)	f_{cal} ($\times10^{-6}$)	f_{exp} ($\times10^{-6}$)	f_{cal} ($\times10^{-6}$)
$^3P_2$	449	22271.71	1.08	5.35	-	-
$^3P_1$	474	21097.05	1.60	2.20	1.60	1.99
$^3P_0$	486	20576.13	2.24	1.99	2.24	1.97
$^1D_2$	595	16806.72	3.21	1.63	3.21	1.65
$^1G_4$	1015	9852.22	1.55	0.48	1.55	0.49
$^3F_4+^3F$	1542	6485.08	14.79	14.54	14.79	14.68
$^3F_2$	1950	5128.20	5.25	5.27	5.25	5.25
$^3H_6$	2293	4361.09	0.37	1.07	0.37	1.08
r.m.s. deviation ($\times10^{-6}$ cm^2)			2.1280		1.0408	
J-O parameters ($\times10^{-20}$ cm^2)						
Ω_2			4.788		4.762	
Ω_4			2.234		2.207	
Ω_6			6.464		6.549	
Spectroscopic quality factor (χ)				0.34	0.33	
$\Sigma\Omega_\lambda$				13.48	13.51	

The nature of Pr^{3+} ligand bond in both P25 can be understood by evaluating the bonding parameter (δ) [23], are obtained via:

$$\delta = \left[\frac{1-\beta'}{\beta'} \right] \times 100 \quad (4)$$

where β' represents the average of the nephelauxetic ratios for all the noted transitions in the absorption spectrum. relied on the sign of the bonding parameter, the Pr^{3+} ligand bond can be covalent or ionic in nature. For the present glass the bonding parameter is positive value shows covalent nature of Pr^{3+} ligand bond in P25=0.9061.

The calculated J-O intensity parameters, r.m.s, χ and $\Sigma\Omega_\lambda$ of P25 glass are presented in Table.1. When all the f_{exp} values of transitions are included in the fitting procedure, vast r.m.s will be observed between experimental and calculated oscillator strengths, which is mainly caused by the inclusion of hypersensitive transition ($^3H_4 \rightarrow {}^3P_2$) only [24]. Such kind of transitions are termed as hypersensitive transition which obey the transition rules, $|\Delta S| = 0$, $|\Delta L| \leq 2$ and $|\Delta J| \leq 2$ in the case of Pr^{3+} ion [25]. Therefore, in the present work the Judd–Ofelt parameters have been recalculated by excluding $^3H_4 \rightarrow {}^3P_2$ transition, because to reduce the r.m.s. For the prepared glass, the J-O intensity parameters follows the trend as $\Omega_6 > \Omega_2 > \Omega_4$ similar results were reported in literature [25-28]. The value of Ω_2 is related with the asymmetry of the glass, in the P25 glass system, the value Ω_2 is $4.762\times10^{-20}cm^2$ showing a strong asymmetrical and covalent environment around Pr^{3+} ions. The value of Ω_6 is inversely proportional to the covalence of Pr–F bond. Moreover, the values of Ω_4 and Ω_6 are related to the viscosity and rigidity of host materials in which the ions are situated [27]. The sum of the three J-O intensity parameters ($\Sigma\Omega_\lambda$) can also provide information about the covalence of the RE-F(or O) bond. Comparison of J-O intensity parameters (Ω_2, Ω_4, Ω_6), r.m.s and spectroscopic quality factor of Pr^{3+} doped in various host matrices are given in Table.2.

Table 02: Judd-Ofelt intensity parameters ($\Omega_\lambda \times 10^{-20}$ cm^2), r.m.s ($\times 10^{-6}$) and spectroscopic quality factor (χ) of Pr^{3+} -doped in different host matrices

Host	Ω_2	Ω_4	Ω_6	$\chi(\Omega_4/\Omega_6)$	r.m.s	Reference
Pr25	4.762	2.207	6.549	0.33	1.040	Present work
$Sr_{0.5}Ca_{0.5}TiO_3$ phosphor	0.02479	0.00103	0.0325	0.031	0.032	[25]
SLBiB	18.31	22.32	32.72	0.69	1.23	[26]
LiBaNaPb	5.11	4.87	21.72	0.22		[27]
BPbZnLi	8.06	6.13	9.79	0.63		[28]

In addition, spectroscopic quality factor ($\chi = \Omega_4 / \Omega_6$) are greatly useful for active laser medium [29]. From table.2, it is observed the values of χ for Pr^{3+} doped studied glass is 0.33, which is like than that of χ (=0.3) of the Nd^{3+}: YAG standard laser crystal grown by Czochralski method [30].

For Pr^{3+}-doped glass, using J-O parameters (Ω_λ) that are evaluated without $^3P_2$ level, the radiative properties such as predicted radiative transition probabilities (A_{rad}) can be calculated as follow:

$$A_{rad}(J \rightarrow J') = A_{ed} + A_{md} = \frac{64\pi^4 e^2}{3h(2J+1)\lambda^3}\left[\left[\frac{n(n^2+2)^2}{9}\right]S_{ed} + n^3 S_{md}\right] \quad (5)$$

Where S_{ed} and S_{md} are the electric-dipole line strength and the magnetic-dipole line strength, respectively, can be calculated with the expression:

$$S_{ed} = \Omega_2 U^2 + \Omega_4 U^4 + \Omega_6 U^6 \quad (6)$$

$$S_{md} = \left[\frac{h^2}{16\pi^2 m^2 c^2}\right]\left|\langle (S,L)J \| L+2S \| (S',L')J' \rangle\right|^2 \quad (7)$$

where $|\langle (S,L)J \| L+2S \| (S',L')J' \rangle|^2$ are the reduced matrix elements of the operator L+2S. The radiative lifetime is expressed as:

$$\tau_{rad} = \frac{1}{\sum A_{rad}(J \rightarrow J')} \quad (8)$$

The branching ratio β is given by:

$$\beta(J \rightarrow J') = \frac{A_{rad}(J \rightarrow J')}{\sum A_{rad}(J \rightarrow J')} = A_{rad}(J \rightarrow J')\tau_{rad} \quad (9)$$

The values of integrated (Σ) is related to the radiative transition probability (A_{rad}) and can expressing from them as:

$$\Sigma = \left[\frac{\lambda_p^2}{8\pi c n^2}\right] A_{rad} \quad (10)$$

where λ_p is the transition peak wavelength.

Table.3 presents, the radiative lifetimes of excited states, the integrated emission cross-sections and branching ratios of all transitions for P25 glass. Generally, the higher values of Arad indicate the stronger luminescence intensity of the transitions [31]. The values of the integrated emission cross-sections obtained are than 43.31×10^{-18} cm of P25 glass at $^1D_2 \rightarrow ^3F_2$ level, which indicates a possibility of lasing action. In the present investigation the radiative lifetime (τ_{rad}) of trend $^3P_1$, $^3P_0$,$^1D_2$ and $^3F_3$ states are found to be 14.41, 14.03 and 112.08 μs for P25 respectively. Comparison of radiative lifetime of $^3P_1$, $^3P_0$ and $^1D_2$ states of Pr^{3+} doped different host matrices are collected in Table.4.

Table 03: Radiative transition probabilities (A_{rad}) (s^{-1}), total radiative transition probabilities (ΣA_{rad}) (s^{-1}), branching ratios (β_{cal}), radiative lifetimes (τ_{rad}) (μs) and integrated emission cross-section (Σ) ($\times 10^{-18}$ cm) of certain excited states of Pr^{3+} doped studied glass

Transition	P25				
	A_{rad} (s^{-1})	ΣA_{rad} (s^{-1})	β	τ_{rad} (μs)	Σ (cm)
$^3P_1 \rightarrow {}^3P_0$	0		0		0
$\rightarrow {}^1D_2$	69		0.099		1.10
$\rightarrow {}^1G_4$	591		0.85		1.44
$\rightarrow {}^3F_4$	3241		4.66		5.31
$\rightarrow {}^3F_3$	21341	69400	30.75	14.41	30.98
$\rightarrow {}^3F_2$	11318		16.30		13.74
$\rightarrow {}^3H_6$	8171		11.77		9.13
$\rightarrow {}^3H_5$	17211		24.79		15.27
$\rightarrow {}^3H_4$	7457		10.74		5.29
$^3P_0 \rightarrow {}^1D_2$	23		0.032		0.51
$\rightarrow {}^1G_4$	943		1.32		2.63
$\rightarrow {}^3F_4$	3761		5.28		6.41
$\rightarrow {}^3F_3$	0		0		0
$\rightarrow {}^3F_2$	32553	71226	45.70	14.03	43.31
$\rightarrow {}^3H_6$	12940		18.16		15.45
$\rightarrow {}^3H_5$	0		0		0
$\rightarrow {}^3H_4$	21006		29.49		15.67
$^1D_2 \rightarrow {}^1G_4$	1138.8		12.76		7.11
$\rightarrow {}^3F_4$	3885.4		43.55		12.15
$\rightarrow {}^3F_3$	300.5		3.36		0.86
$\rightarrow {}^3H_6$	488.3	8921.6	5.47	112.08	1.002
$\rightarrow {}^3H_5$	30.1		0.33		0.044
$\rightarrow {}^3H_4$	2514.4		28.18		2.74

Table 04: Comparison of radiative lifetime (τ_{rad} in μs) of excited states of Pr^{3+} doped in different host matrices.

Host	$^3P_1$	$^3P_0$	$^1D_2$	Ref.
P25	14.41	14.03	112.08	Present work
$Li_2B_4O_7$-BaF_2-NaF- MgO	12	13	122	[27]
TeO_2-WO_3-ZnO-TiO_2-Na_2O	23	21	143	[31]
TeO_2-WO_3-PbO-Lu_2O_3	-	8.08	70.1	[32]
ZnBPr	12.79	45.59	191	[33]
P_2O_5-CaO-BaO-SrO	26	25	287	[34]

4 CONCLUSION

An overview, optical properties of the Pr^{3+} doped 79.75Sb_2O_3-10Li_2O-10WO_3 (mol%) glass. The absorption spectrum was analyzed using the (J–O) theory with the transition $^3H_4 \rightarrow {}^3P_2$ excluding results in the least deviation. (J-O) intensity parameters displayed $\Omega_6 > \Omega_2 > \Omega_4$ trend for the prepared glass. The J–O parameters ($\Omega_2 > \Omega_4$) for P25 confermed the higher covalent character of Pr^{3+} bond and lower symmetry at the Pr^{3+} site. Utilizing these J-O parameters we have evaluated different radiative properties such as predicted radiative transition probabilities (A_{rad}), the branching ratios (β_{cal}), and the radiative lifetime (τ_{rad}) integrated emission cross-section (Σ) of Pr^{3+} -doped studied glass.

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