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Structural and FTIR Analysis of Sm^{3+} ions doped Ytterbium Zinc Lithium Lead Sodalime Cadmium Phosphate Glasses

S.L. Meena

Ceramic Laboratory, Department of Physics, Jai Narain Vyas University, Jodhpur(Raj.) India

E-mail address:shankardiya7@rediffmail.com

ABSTRACT: Ytterbium zinc lithium lead sodalime cadmium phosphate glasses containing Sm^{3+} in $(30-x)\text{P}_2\text{O}_5:10\text{ZnO}:10\text{Li}_2\text{O}:10\text{PbO}:10\text{CaO}:10\text{Na}_2\text{O}:10\text{Yb}_2\text{O}_3:10\text{CdO}:x\text{Sm}_2\text{O}_3$ (where $x=1,1.5$ and 2 mol %) have been prepared by melt-quenching method. The amorphous nature of the prepared glass samples was confirmed by X-ray diffraction. The structural change in glass system doped with different concentrations of Sm^{3+} ions were investigated by FTIR spectroscopy. The Optical absorption, Excitation and Fluorescence spectra were recorded at room temperature for all glass samples. Judd-Ofelt intensity parameters were calculated and they followed $\Omega_2 > \Omega_4 > \Omega_6$ trends. These parameters helped to calculate radiative transition probability (A_{rad}), branching ratio (β), stimulated emission cross-section (σ_p) and radiative life time (τ_R). The emission spectra of Sm^{3+} have been recorded under the excitation wavelength 602nm and the emission peaks at 562 nm, 602nm, 645 nm and 705 nm for all glass samples. The value of stimulated emission cross-section (σ_p) is found to be maximum for the transition (${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$), Suggestion that transition is better compared to the other transitions.

KEYWORDS: Optical properties; X-ray diffraction; FTIR Spectra, Stimulated emission cross section

I. INTRODUCTION

Rare-earth doped glasses have promising properties for various applications such as laser technology, computer technology, amorphous thin film technology, optoelectronic devices, optical fibers and optical amplifiers [1-5]. Among different glasses, Phosphate glasses have excellent properties such as low phonon energy, thermal stability, high transparency, chemical durability, high refractive index, high density and optical stability [6-9]. Glasses containing phosphate oxide glasses have wide range application in the field of glass ceramics, layers for optical and electronic device, thermal and mechanical sensors, reflecting windows [10-12]. Due to their excellent thermal, physical and mechanical properties, they are used in fiber lasers, thermal imaging, spectral conversion, photo-voltaic solar cells, temperature sensors and optical coherence tomography [13-15]. Phosphate glasses containing Sm^{3+} ions good for candidates because of their strong absorption and emission in the UV and visible spectral regions [16-20]. Recently Sm^{3+} ions doped glasses found important in the area of wave guide laser, laser action and Telecommunications optical fibers [21-23].

I have studied on the Optical absorption, Excitation, Fluorescence and FTIR spectra of Sm^{3+} doped Ytterbium zinc lithium lead sodalime cadmium phosphate glasses. Using Judd-Ofelt intensity parameters the laser parameters such as branching ratio (β), radiative probability (A), radiative life time (τ_R) and stimulated emission cross section (σ_p) are calculated.

II. EXPERIMENTAL TECHNIQUES

Preparation of glasses

The following Sm^{3+} doped phosphate glass samples $(30-x)\text{P}_2\text{O}_5:10\text{ZnO}:10\text{Li}_2\text{O}:10\text{PbO}:10\text{CaO}:10\text{Na}_2\text{O}:10\text{Yb}_2\text{O}_3:10\text{CdO}:x\text{Sm}_2\text{O}_3$, (where $x=1,1.5$ and 2 mol%) have been prepared by melt-quenching method. The chemical used consist of $\text{P}_2\text{O}_5, \text{ZnO}, \text{Li}_2\text{O}, \text{PbO}, \text{CaO}, \text{Na}_2\text{O}, \text{CdO}, \text{Yb}_2\text{O}_3$ and Sm_2O_3 . All chemicals were powdered by agate pestle mortar. For preparation of Phosphate glasses Silicon Carbide Muffle furnace was heated to working temperature of 1072°C . For quenching the melts were quickly poured in to



moulds. After cooling the glass specimens were removed from moulds. The glass specimens were annealed at 350°C for two hours to remove stresses and give thermal stability to the glass samples. Finally polished using fine powders of cerium oxide so prepared glass specimens were transparent.

The chemical composition of the glass samples are given in Table 1.

Table 1.

Chemical composition of the glasses

Sample	Glass composition (mol %)
YZLLSLCP (UD)	30P ₂ O ₅ :10ZnO:10Li ₂ O:10PbO:10CaO:10Na ₂ O:10Yb ₂ O ₃ :10CdO
YZLLSLCP (SM1)	29P ₂ O ₅ :10ZnO:10Li ₂ O:10PbO:10CaO:10Na ₂ O:10Yb ₂ O ₃ :10CdO:1 Sm ₂ O ₃
YZLLSLCP (SM 1.5)	28.5P ₂ O ₅ :10ZnO:10Li ₂ O:10PbO:10CaO:10Na ₂ O:10Yb ₂ O ₃ :10CdO:1.5 Sm ₂ O ₃
YZLLSLCP (SM 2)	28P ₂ O ₅ :10ZnO:10Li ₂ O:10PbO:10CaO:10Na ₂ O:10Yb ₂ O ₃ :10CdO:2 Sm ₂ O ₃

YZLLSLCP (UD) -Represents undoped Ytterbium Zinc Lithium Lead Sodalime Cadmium Phosphate glass specimen.

YZLLSLCP (SM)-Represents Sm³⁺ doped Ytterbium Zinc Lithium Lead Sodalime Cadmium Phosphate glass specimen.

III. THEORY

3.1 Oscillator Strength

The intensity of spectral lines are expressed in terms of oscillator strengths using the relation [24].

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

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 [25], using the modified relation:

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

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 [26] and Ofelt [27] 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:

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

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

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

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

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:

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

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

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

where, the sum is over all terminal manifolds.

The radiative life time is given by

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

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

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

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 [28, 29]. The Nephelauxetic Ratio is given by

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

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

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

IV. RESULT AND DISCUSSION

4.1 XRD Measurement

The glass samples of Sm^{3+} doped YZLLSLCP glass specimens have been presented in Figure 1. The absence of peak in general indicates almost amorphous nature of specimens.

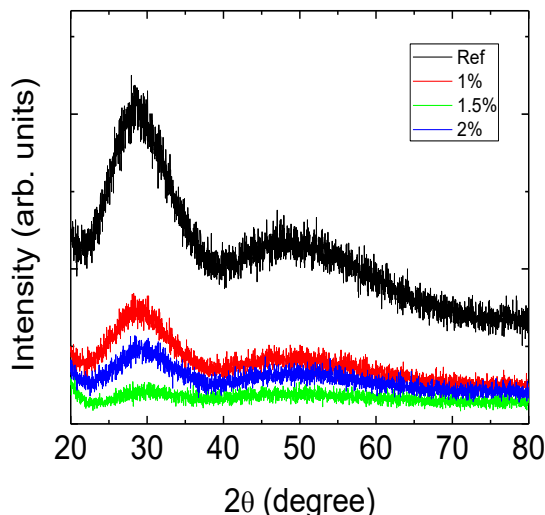


Fig. 1. X-ray diffraction pattern of YZLLSLCP SM glass samples.

4.2 FTIR Transmission spectra

The FTIR spectrum of YZLLSLCP glass samples in the wave number range 500-4000 cm^{-1} is presented in Fig.2 and the possible mechanism bands are tabulated in Table 2

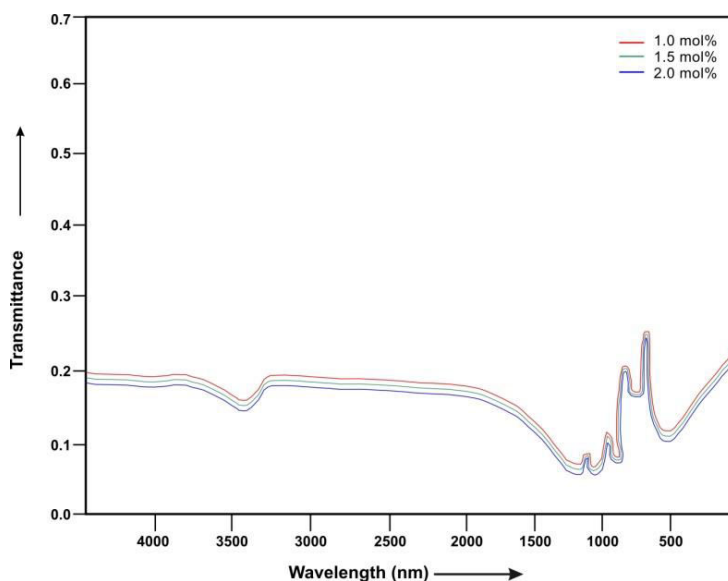


Fig.(2): FTIR spectrum of YZLLSLCP SM glasses.

The band at 526 cm^{-1} is assigned to the P-O-P bending vibrations [30].The Symmetric stretching vibration of P-O-P bonds is observed around 752 cm^{-1} [31].The observed band around 925 cm^{-1} is due to the Asymmetric stretching vibration of P-O-P bonds while the occurrence of band around 1112 cm^{-1} is assigned to the Asymmetric stretching mode of $(\text{PO}_3)^{2-}$ groups[32]. The observed band at 1236 cm^{-1} is attributed to Asymmetric stretching vibration of P=O and O-P-O bands [33]. The Symmetric stretching of the O-H bonds is observed around 3426 cm^{-1} [34].

Table2. Assignment of infrared transmission bands of YZLLSLCP SM glasses.

Peak position(cm ⁻¹)	Band Assignment
~526	Bending vibration of P-O-P bands
~752	Symmetric stretching vibration of P-O-P bonds
~925	Asymmetric stretching vibration of P-O-P bonds
~1112	Asymmetric stretching modes of (PO ₃) ²⁻ groups
~1236	Asymmetric stretching vibration of P=O and O-P-O bonds
~3426	Symmetric stretching of the O-H bonds

4.3 Absorption Spectrum

The absorption spectra of YZLLSLCP SM glass specimens have been shown in Figure 3. There are 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 YZLLSLCP SM glass specimens.

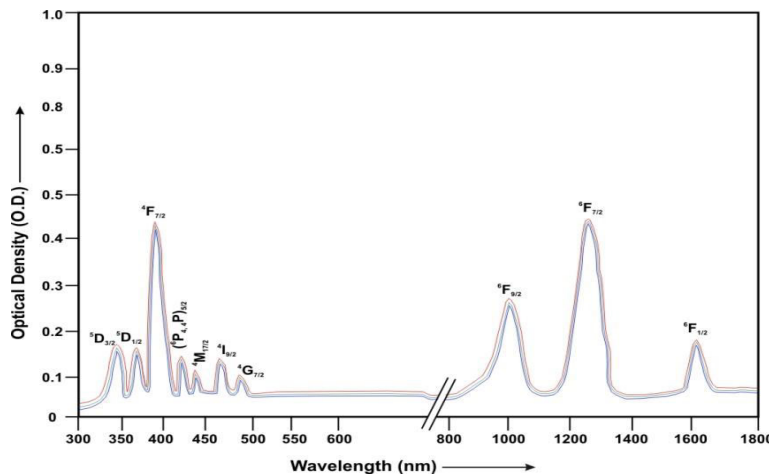


Fig. (3) Absorption spectrum of YZLLSLCP SM glass specimens.

The experimental and calculated oscillator strength for Sm³⁺ ions in YZLLSLCP glasses are given in Table 3.

Table 3: Measured and calculated oscillator strength (P_m×10⁺⁶) of Sm³⁺ ions in YZLLSLCP glasses.

Energy level from ⁶ H _{5/2}	Glass YZLLSLCP (SM01)		Glass YZLLSLCP (SM1.5)		Glass YZLLSLCP (SM02)	
	P _{exp.}	P _{cal.}	P _{exp.}	P _{cal.}	P _{exp.}	P _{cal.}
⁶ F _{1/2}	1.72	1.75	1.68	1.71	1.65	1.69
⁶ F _{7/2}	5.52	5.55	5.48	5.53	5.44	5.50
⁶ F _{9/2}	3.88	3.88	3.84	3.85	3.81	3.83
⁴ G _{7/2}	0.22	0.12	0.20	0.12	0.17	0.12
⁴ I _{9/2} , ⁴ M _{15/2} , ⁴ I _{11/2}	1.24	1.91	1.21	1.89	1.18	1.88
⁴ M _{17/2} , ⁴ G _{9/2} , ⁴ I _{15/2}	0.32	0.25	0.29	0.25	0.27	0.24
(⁶ P, ⁴ P) _{5/2} , ⁴ L _{13/2}	1.38	1.31	1.35	1.31	1.31	1.30
⁴ F _{7/2} , ⁶ P _{3/2} , ⁴ K _{11/2}	5.67	5.64	5.62	5.62	5.58	5.60
⁴ D _{1/2} , ⁶ P _{7/2} , ⁴ L _{17/2}	2.55	2.46	2.52	2.44	2.47	2.43
⁴ D _{3/2} , (⁴ D, ⁶ P) _{5/2}	2.68	3.49	2.64	3.47	2.60	3.46
r.m.s. deviation	0.3367		0.3435		0.3518	

The experimental and calculated oscillator strength for Sm³⁺ ions in YZLLSLCP glasses are given in Table 4.



Table 4: Measured and calculated absorption line strength and Goodness of fit for Sm³⁺ doped YZLLSLCP glasses are given in Table 4.

Energy level from ⁶ H _{5/2}	Glass		Glass		Glass	
	YZLLSLCP (SM01)		YZLLSLCP (SM1.5)		YZLLSLCP (SM02)	
	S _m (x10 ⁻²⁰)	S _m (x10 ⁻²⁰)	S _m (x10 ⁻²⁰)	S _m (x10 ⁻²⁰)	S _m (x10 ⁻²⁰)	S _m (x10 ⁻²⁰)
⁶ F _{1/2}	1.02	1.04	1.00	1.02	0.98	1.01
⁶ F _{7/2}	2.48	2.50	2.46	2.48	2.44	2.47
⁶ F _{9/2}	1.55	1.55	1.53	1.54	1.52	1.53
⁴ G _{7/2}	0.04	0.02	0.04	0.02	0.03	0.02
⁴ I _{9/2} , ⁴ M _{15/2} , ⁴ I _{11/2}	0.21	0.33	0.21	0.33	0.20	0.33
⁴ M _{17/2} , ⁴ G _{9/2} , ⁴ I _{15/2}	0.05	0.04	0.05	0.04	0.04	0.04
(⁶ P, ⁴ P) _{5/2} , ⁴ L _{13/2}	0.21	0.20	0.21	0.20	0.20	0.20
⁴ F _{7/2} , ⁶ P _{3/2} , ⁴ K _{11/2}	0.83	0.83	0.83	0.83	0.82	0.82
⁴ D _{1/2} , ⁶ P _{7/2} , ⁴ L _{17/2}	0.35	0.34	0.35	0.33	0.34	0.33
⁴ D _{3/2} , (⁴ D, ⁶ P) _{5/2}	0.35	0.46	0.35	0.47	0.34	0.46
Goodness of fit	0.03743		0.03914		0.04142	

In the Ytterbium Zinc Lithium Lead Sodalime Cadmium Phosphate glasses (YZLLSLCP) Ω₂, Ω₄ and Ω₆ parameters decrease with the increase of x from 1 to 2 mol%.

The values of Judd-Ofelt intensity parameters are given in Table 5.

Table 5: Judd-Ofelt intensity parameters for Sm³⁺ doped YZLLSLCP glass specimens.

Glass Specimen	Ω ₂ (pm ²)	Ω ₄ (pm ²)	Ω ₆ (pm ²)	Ω ₄ /Ω ₆	Ref.
YZLLSLCP (SM01)	5.382	4.620	4.256	1.086	[P.W.]
YZLLSLCP (SM 1.5)	5.273	4.601	4.222	1.090	[P.W.]
YZLLSLCP (SM 02)	5.195	4.582	4.193	1.093	[P.W.]
LBLB(SM)	6.81	4.43	2.58	1.717	[35]
PZABP(EU)	6.52	2.29	0.98	2.337	[36]

4.4 Excitation Spectrum

Fig.4 shows the excitation spectra of YZLLSLCP SM glass specimens at room temperature under emission wavelength at 602 nm. There are seven excitation band at 337,360,375,401,411,435and477 nm corresponding to the transitions ⁴H_{9/2}, ⁴D_{3/2}, ⁶P_{7/2}, ⁴F_{7/2}, ⁴M_{19/2}, ⁴G_{9/2} and ⁴I_{11/2}+⁴I_{13/2} respectively. The prominent transition is ⁴F_{7/2} corresponding to the excitation band centered at 401nm.

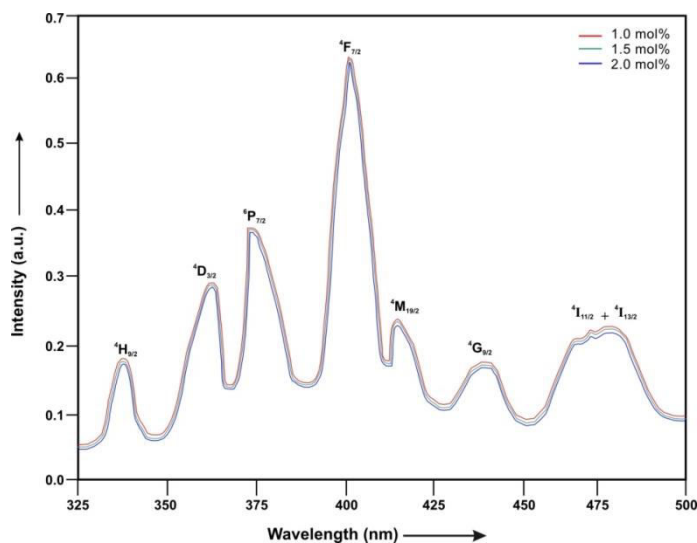


Fig. (4) Excitation spectrum of YZLLSLCP SM specimens glass.

4.5 Fluorescence Spectrum

The fluorescence spectrum of YZLLSLCP SM glass specimens at room temperature have been shown in Figure 5. There are four transitions (${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{5/2}$), (${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$), (${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{9/2}$) and (${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{11/2}$) corresponding to the emission intensity 562,602,645 and 705 nm respectively. The maximum emission intensity at 602 nm corresponding the transition (${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$). The wavelengths of bands along with their transitions are given in Table 7.

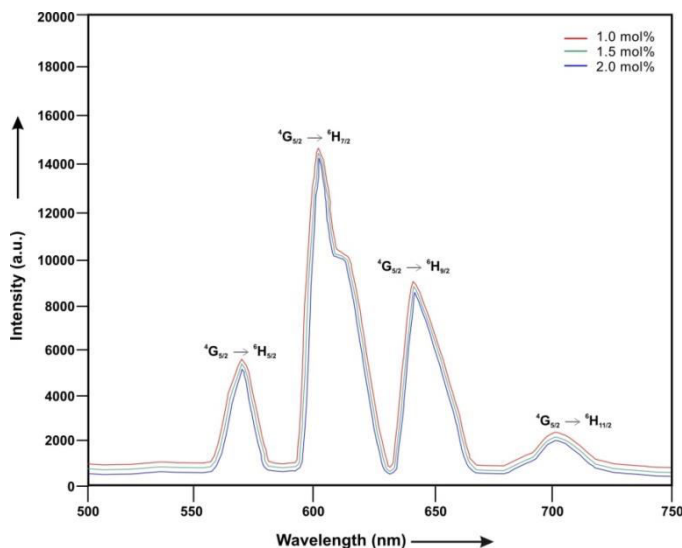


Fig. (5). Fluorescence spectrum of YZLLSLCP SM glass specimens.

4.6 Energy level diagram and Up Conversion Mechanism

The energy level diagram of glass specimens has been shown in fig.6(a). This figure represents the absorption transition from the lower energy level ${}^6\text{H}_{5/2}$ to the upper energy levels. Figure 6(b) illustrates possible up conversion mechanism under 488 nm excitation.

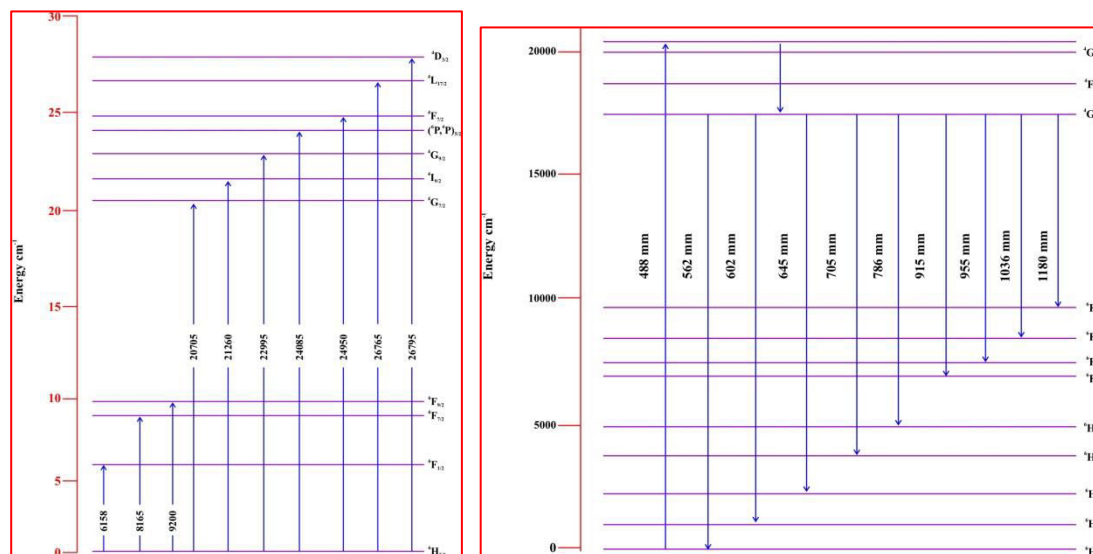


Fig. 6(a) Energy level diagram of doped YZLLSLCP (SM) glasses; (b) Up-conversion mechanism of doped YZLLSLCP (SM) glasses.

Table6: 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 YZLLSLCP glasses.

Transition	YZLLSLCP (SM 01)					YZLLSLCP (SM 1.5)					YZLLSLCP (SM 02)			
	λ_{max} (nm)	$A_{rad}(s^{-1})$	β	$\sigma_p (10^{-20} cm^2)$	$\tau_R(\mu s)$	$A_{rad}(s^{-1})$	β	$\sigma_p (10^{-20} cm^2)$	$\tau_R (\mu s)$	$A_{rad}(s^{-1})$	β	$\sigma_p (10^{-20} cm^2)$	$\tau_R (10^{-20} cm^2)$	
$^4G_{5/2} \rightarrow ^6H_5$	562	10.60	0.041	0.0042		10.52	0.0444	0.0044		10.47	0.041	0.0050		
$^4G_{5/2} \rightarrow ^6H_7$	602	109.97	0.428	0.0461		109.51	0.4995	0.0489		109.12	0.430	0.0539		
$^4G_{5/2} \rightarrow ^6H_9$	645	109.47	0.426	0.0460	3891.3	108.08	0.4774	0.0483	3921.8	107.08	0.422	0.0504	3945.34	
$^4G_{5/2} \rightarrow ^6H_1$	705	26.95	0.105	0.0133	0	26.87	0.1413	0.0136	3	26.78	0.105	0.0147		
													7	

V. CONCLUSION

In the present study, the glass samples of composition $(30-x)P_2O_5:10ZnO:10Li_2O:10PbO:10CaO:10Na_2O:10Yb_2O_3:10CdO: x Sm_2O_3$ (where $x = 1, 1.5$ and 2 mol %) have been prepared by melt-quenching method. The success of Judd-Ofelt theory has been shown by low value of goodness of fit between the measured (S_m) and calculated (S_c) line strengths. The value of stimulated emission cross-section (σ_p) is found to be maximum for the transition ($4G_{5/2} \rightarrow 6H_7/2$), Suggesting that transition is better compared to the transitions. The large stimulated emission cross section in phosphate glasses suggests the possibility of utilizing these systems as laser materials. The FTIR of glasses provide information about bonds present in glass samples such as P-O-P, O-P-O, (PO₃)₂- and O-H.

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