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# Spectral and Transmittance Properties of Sm<sup>3+</sup> Doped in Zinc Lithium Alumino Boromolybdate Glasses

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**ABSTRACT:** Glass of the system: (45-x)MoO<sub>3</sub>:10ZnO:10Li<sub>2</sub>O:10Al<sub>2</sub>O<sub>3</sub>:25B<sub>2</sub>O<sub>3</sub>: xSm<sub>2</sub>O<sub>3</sub> (where x=1,1.5,2 mol%) have been prepared by melt-quenching technique. (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 Optical absorption, Excitation and fluorescence spectra of three Sm<sup>3+</sup> doped zinc lithium aluminoboromolybdate glasses have been recorded at room temperature. The various interaction parameters like Slater-Condon parameters F<sub>k</sub> (k=2,4,6), Lande parameter (ξ<sub>4f</sub>), nephelauxetic ratio (β'), bonding parameters (b<sup>1/2</sup>) and Racah parameters E<sup>k</sup> (k=1,2,3) have been computed. Judd-Ofelt intensity parameters and laser parameters have also been calculated.

**KEYWORDS:** ZLABM Glasses, Transmission Properties, Optical properties, Judd-Ofelt analysis.

## I. INTRODUCTION

Rare-earth ion doped glasses have attracted much attention as potential materials for various optical devices such as lasers, up-converters, optical fibers, mechanical, optical and magnetic properties [1-8]. Among different glass hosts, molybdate glasses have unique properties. They have high thermal stability, high transparency, a low melting point, a high gain density, low dispersion rates, chemical properties and excellent optical properties [9-11]. Molybdate glass exhibit very important physical properties such as low melting temperature, low glass transition temperature and low softening temperature. Due to their good chemical durability, samarium-doped ceramic glasses are attractive materials for the fabrication of low-cost integrated optical amplifiers by using the ion-exchange technique [12-17]. The addition of network modifier (NWF) Li<sub>2</sub>O is to improve both electrical and mechanical properties of such glasses. Zinc oxide is added in the glass matrix to increase glass forming ability and to ensure low rates of crystallization in the glass system [18-20].

The aim of the present study is to prepare the Sm<sup>3+</sup> doped zinc lithium aluminoboromolybdate glass with different Sm<sub>2</sub>O<sub>3</sub> concentrations. The absorption spectra, Excitation, fluorescence spectra of Sm<sup>3+</sup> of the glasses were investigated. The Judd-Ofelt theory has been applied to compute the intensity parameters Ω<sub>λ</sub> (λ=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<sup>3+</sup> doped zinc lithium aluminoboromolybdate glass samples (45-x)MoO<sub>3</sub>:10ZnO:10Li<sub>2</sub>O:10Al<sub>2</sub>O<sub>3</sub>:25B<sub>2</sub>O<sub>3</sub>:xSm<sub>2</sub>O<sub>3</sub>. (where x = 1, 1.5, 2) have been prepared by melt-quenching method. Analytical reagent grade chemical used in the present study consist of MoO<sub>3</sub>, ZnO, Li<sub>2</sub>O, Al<sub>2</sub>O<sub>3</sub>, B<sub>2</sub>O<sub>3</sub> and Sm<sub>2</sub>O<sub>3</sub>. They were thoroughly mixed by using an agate pestle mortar, then melted at 1070°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 %)
ZLABM(UD)	45MoO <sub>3</sub> :10ZnO:10Li <sub>2</sub> O:10Al <sub>2</sub> O <sub>3</sub> :25B <sub>2</sub> O <sub>3</sub>
ZLABM(SM1)	44MoO <sub>3</sub> :10ZnO:10Li <sub>2</sub> O:10Al <sub>2</sub> O <sub>3</sub> :25B <sub>2</sub> O <sub>3</sub> :1Sm <sub>2</sub> O <sub>3</sub>
ZLABM (SM 1.5)	43.5MoO <sub>3</sub> :10ZnO:10Li <sub>2</sub> O:10Al <sub>2</sub> O <sub>3</sub> :25B <sub>2</sub> O <sub>3</sub> :1.5Sm <sub>2</sub> O <sub>3</sub>
ZLABM(SM 2)	43MoO <sub>3</sub> :10ZnO:10Li <sub>2</sub> O:10Al <sub>2</sub> O <sub>3</sub> :25B <sub>2</sub> O <sub>3</sub> :2Sm <sub>2</sub> O <sub>3</sub>

ZLABM(UD) -Represents undoped Zinc Lithium AluminoBoromolybdate Glass specimens.

ZLABM(SM) -Represents Sm<sup>3+</sup>doped Zinc Lithium AluminoBoromolybdate Glass specimens.

### III. THEORY

#### 3.1 Oscillator Strength

The intensity of spectral lines are expressed in terms of oscillator strengths using the relation [21].

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

where,  $\varepsilon(\nu)$  is molar absorption coefficient at a given energy  $\nu$  (cm<sup>-1</sup>), 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[22], 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[23] and Ofelt[24] 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:

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

Where, the line strength  $S(S', L')$  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,  $\nu$  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,  $\Omega_\lambda$  ( $\lambda = 2, 4, 6$ ) are known as Judd-Ofelt intensity parameters .

#### 3.3 Radiative Properties

The  $\Omega_\lambda$  parameters obtained using the absorption spectral results have been used to predict radiative properties such as spontaneous emission probability ( $A$ ) and radiative life time ( $\tau_R$ ), and laser parameters like fluorescence branching ratio( $\beta_R$ ) and stimulated emission cross section ( $\sigma_p$ ).

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:

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



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

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

where, the sum is over all terminal manifolds.

The radiative life time is given by

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

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

### 3.4 Nephelauxetic Ratio ( $\beta$ ) and Bonding Parameter ( $b^{1/2}$ )

The nature of the R-O bond is known by the Nephelauxetic Ratio ( $\beta$ ) and Bonding Parameters ( $b^{1/2}$ ), which are computed by using following formulae [25, 26]. The Nephelauxetic Ratio is given by

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

where,  $\nu_a$  and  $\nu_g$  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  $-MoO_3$  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.

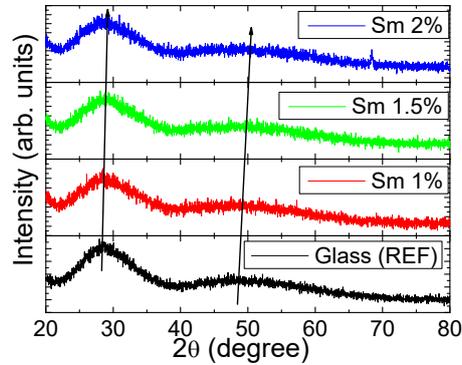


Fig. 1: X-ray diffraction pattern of MoO<sub>3</sub>:ZnO:Li<sub>2</sub>O:Al<sub>2</sub>O<sub>3</sub>:B<sub>2</sub>O<sub>3</sub>

**4.2 Transmittance Spectrum**

The Transmittance spectrum of Sm<sup>3+</sup> doped in zinc lithium aluminoboromolybdate glass is shown in Figure 2.

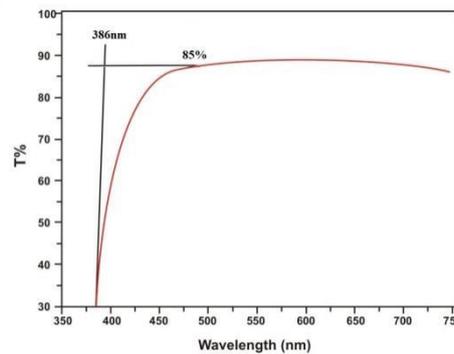


Fig. (2) Transmittance spectrum of Sm<sup>3+</sup> doped ZLABM (01) glasses.

**4.3 Absorption Spectrum**

The absorption spectra of Sm<sup>3+</sup> doped ZLABM(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 <sup>6</sup>H<sub>5/2</sub> to excited states <sup>6</sup>F<sub>1/2</sub>, <sup>6</sup>F<sub>7/2</sub>, <sup>6</sup>F<sub>9/2</sub>, <sup>4</sup>G<sub>7/2</sub>, <sup>4</sup>I<sub>9/2</sub>, <sup>4</sup>M<sub>7/2</sub>, (<sup>6</sup>P, <sup>4</sup>P)<sub>5/2</sub>, <sup>4</sup>F<sub>7/2</sub>, <sup>4</sup>D<sub>1/2</sub>, and (<sup>4</sup>D, <sup>6</sup>P)<sub>5/2</sub> for Sm<sup>3+</sup> doped ZLABM glasses.

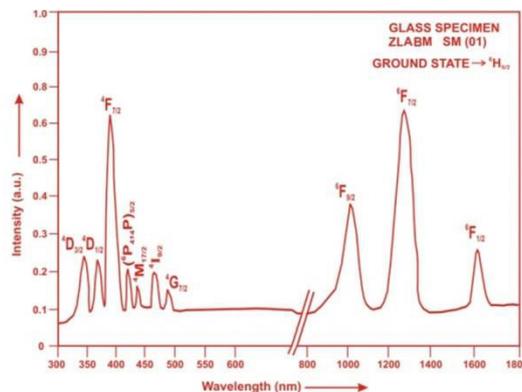


Fig.3: Absorption spectrum of Sm<sup>3+</sup> doped ZLABM(01)glass.



The experimental and calculated oscillator strengths for Sm<sup>3+</sup> ions in zinc lithium aluminoboromolybdate glasses are given in Table 2.

**Table 2:** Measured and calculated oscillator strength ( $P_m \times 10^{+6}$ ) of Sm<sup>3+</sup> ions in ZLABM glasses.

Energy level from <sup>6</sup> H <sub>5/2</sub>	Glass ZLABM (SM01)		Glass ZLABM (SM1.5)		Glass ZLABM (SM02)	
	P <sub>exp.</sub>	P <sub>cal.</sub>	P <sub>exp.</sub>	P <sub>cal.</sub>	P <sub>exp.</sub>	P <sub>cal.</sub>
<sup>6</sup> F <sub>1/2</sub>	1.70	1.73	1.68	1.72	1.64	1.69
<sup>6</sup> F <sub>7/2</sub>	5.54	5.58	5.51	5.45	5.48	5.53
<sup>6</sup> F <sub>9/2</sub>	3.88	3.90	3.84	3.88	3.81	3.86
<sup>4</sup> G <sub>7/2</sub>	0.22	0.12	0.20	0.12	0.17	0.12
<sup>4</sup> I <sub>9/2</sub> , <sup>4</sup> M <sub>15/2</sub> , <sup>4</sup> I <sub>11/2</sub>	1.25	1.91	1.21	1.90	1.18	1.89
<sup>4</sup> M <sub>17/2</sub> , <sup>4</sup> G <sub>9/2</sub> , <sup>4</sup> I <sub>15/2</sub>	0.35	0.25	0.32	0.24	0.28	0.25
( <sup>6</sup> P, <sup>4</sup> P) <sub>5/2</sub> , <sup>4</sup> L <sub>13/2</sub>	1.40	1.31	1.39	1.30	1.36	1.30
<sup>4</sup> F <sub>7/2</sub> , <sup>6</sup> P <sub>3/2</sub> , <sup>4</sup> K <sub>11/2</sub>	5.60	5.61	5.54	5.59	5.51	5.60
<sup>4</sup> D <sub>1/2</sub> , <sup>6</sup> P <sub>7/2</sub> , <sup>4</sup> L <sub>17/2</sub>	2.52	2.47	2.48	2.46	2.42	2.45
<sup>4</sup> D <sub>3/2</sub> , ( <sup>4</sup> D, <sup>6</sup> P) <sub>5/2</sub>	2.67	3.48	2.64	3.47	2.61	3.46
r.m.s. deviation	0.3364		0.3453		0.3549	

Computed values of F<sub>2</sub>, Lande' parameter ( $\xi_{4f}$ ), Nephelauxetic ratio ( $\beta'$ ) and bonding parameter ( $b^{1/2}$ ) for Sm<sup>3+</sup> doped ZLABM glass specimen are given in Table 3.

**Table 3.** F<sub>2</sub>,  $\xi_{4f}$ ,  $\beta'$  and  $b^{1/2}$  parameters for Samarium doped glass specimen.

Glass Specimen	F <sub>2</sub>	$\xi_{4f}$	$\beta'$	$b^{1/2}$
Sm <sup>3+</sup>	358.82	1258.16	0.9337	0.1821

Judd-Ofelt intensity parameters  $\Omega_\lambda$  ( $\lambda=2,4,6$ ) 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  $\Omega_\lambda$  parameters follow the trend  $\Omega_2 > \Omega_4 > \Omega_6$ . The spectroscopic quality factor ( $\Omega_4/\Omega_6$ ) related with the rigidity of the glass system has been found to lie between 1.073 and 1.083 in the present glasses.

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

**Table 4:** Judd-Ofelt intensity parameters for Sm<sup>3+</sup> doped ZLABM glass specimens.

Glass Specimen	$\Omega_2(\text{pm}^2)$	$\Omega_4(\text{pm}^2)$	$\Omega_6(\text{pm}^2)$	$\Omega_4/\Omega_6$	Ref.
ZLABM (SM01)	5.797	4.998	4.659	1.073	P.W.
ZLABM (SM1.5)	5.748	4.983	4.628	1.077	P.W.
ZLABM (SM02)	5.638	4.986	4.603	1.083	P.W.

#### 4.4 Excitation Spectrum

Excitation spectra of ZLABMSM (01) glass recorded at the emission wavelength 602 nm is depicted as figure 4. The excitation spectra consists of seven peaks corresponding to the transitions from the ground state <sup>6</sup>H<sub>5/2</sub> to the various excited states <sup>4</sup>H<sub>9/2</sub>, <sup>4</sup>D<sub>3/2</sub>, <sup>6</sup>P<sub>7/2</sub>, <sup>4</sup>F<sub>7/2</sub>, <sup>4</sup>M<sub>19/2</sub>, <sup>4</sup>G<sub>9/2</sub> and <sup>4</sup>I<sub>11/2</sub>+<sup>4</sup>I<sub>13/2</sub> at the wavelengths of 335, 361, 375, 401, 412, 435 and 478 nm respectively. Among these, a prominent excitation band at 401 nm has been selected for the measurement of emission spectrum of Sm<sup>3+</sup> glass.

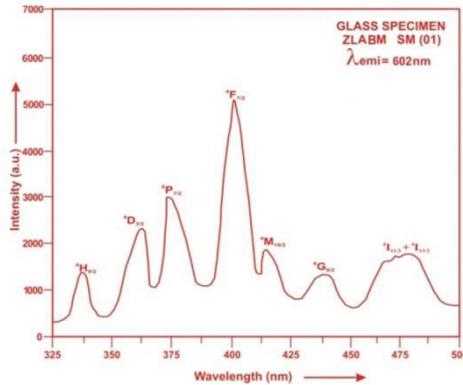


Fig.4: Excitation spectrum of Sm<sup>3+</sup>doped ZLABM (01) glass

**4.4. Fluorescence Spectrum**

The fluorescence spectrum of Sm<sup>3+</sup>doped in zinc lithium aluminoboromolybdateglass is shown in Figure 5. There are nine broad bands observed in the Fluorescence spectrum of Sm<sup>3+</sup>dopedzinc lithium aluminoboromolybdateglass. The wavelengths of these bands along with their assignments are given in Table 5. Fig. (5).Shows the fluorescence spectrum with nine peaks (<sup>4</sup>G<sub>5/2</sub>→<sup>6</sup>H<sub>5/2</sub>), (<sup>4</sup>G<sub>5/2</sub>→<sup>6</sup>H<sub>7/2</sub>), (<sup>4</sup>G<sub>5/2</sub>→<sup>6</sup>H<sub>9/2</sub>),(<sup>4</sup>G<sub>5/2</sub>→<sup>6</sup>H<sub>11/2</sub>),(<sup>4</sup>G<sub>5/2</sub>→<sup>6</sup>H<sub>13/2</sub>), (<sup>4</sup>G<sub>5/2</sub>→<sup>6</sup>F<sub>3/2</sub>),(<sup>4</sup>G<sub>5/2</sub>→<sup>6</sup>F<sub>5/2</sub>),(<sup>4</sup>G<sub>5/2</sub>→<sup>6</sup>F<sub>7/2</sub>) and (<sup>4</sup>G<sub>5/2</sub>→<sup>6</sup>F<sub>9/2</sub>), respectively for glass specimens.

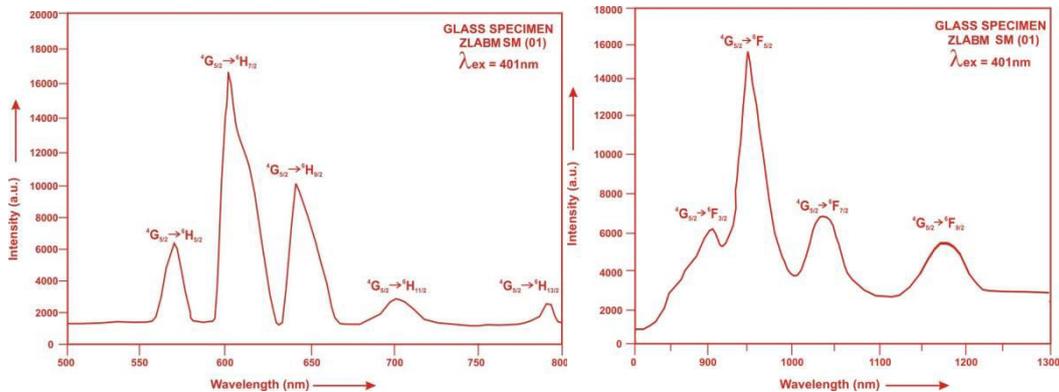


Fig.5: fluorescence spectrum of Sm<sup>3+</sup>doped ZLABM(01) glass



**Table 5.** Emission peak wave lengths ( $\lambda_p$ ), radiative transition probability ( $A_{rad}$ ), branching ratio ( $\beta$ ), stimulated emission cross-section ( $\sigma_p$ ) and radiative life time ( $\tau_R$ ) for various transitions in  $Sm^{3+}$  doped ZLABM glasses.

Transition	$\lambda_{max}$ (nm)	ZLABM (SM 01)				ZLABM (SM 1.5)				ZLABM (SM 02)			
		$A_{rad}(s^{-1})$	$\beta$	$\sigma_p(10^{-20} cm^2)$	$\tau_R(\mu s)$	$A_{rad}(s^{-1})$	$\beta$	$\sigma_p(10^{-20} cm^2)$	$\tau_R(\mu s)$	$A_{rad}(s^{-1})$	$\beta$	$\sigma_p(10^{-20} cm^2)$	$\tau_R(\mu s)$
$^4G_{5/2} \rightarrow ^6H_{5/2}$	562	9.35	0.0365	0.0039	3909.67	9.32	0.0366	0.0041	3923.90	9.30	0.0367	0.0045	3944.86
$^4G_{5/2} \rightarrow ^6H_{7/2}$	602	97.67	0.3819	0.0453		97.43	0.3823	0.0486		97.38	0.384	0.0534	
$^4G_{5/2} \rightarrow ^6H_{9/2}$	645	96.52	0.3774	0.0417		96.07	0.3770	0.0433		95.13	0.375	0.0448	
$^4G_{5/2} \rightarrow ^6H_{11/2}$	705	23.90	0.0934	0.0129		23.84	0.0936	0.0134		23.86	0.0941	0.0138	
$^4G_{5/2} \rightarrow ^6H_{13/2}$	786	2.29	0.0090	0.00172		2.29	0.0090	0.00176		2.28	0.0090	0.0018	
$^4G_{5/2} \rightarrow ^6F_{3/2}$	915	4.08	0.0159	0.00701		4.05	0.0159	0.00726		3.98	0.0157	0.0076	
$^4G_{5/2} \rightarrow ^6F_{5/2}$	955	18.19	0.0711	0.0313		18.09	0.0709	0.0320		17.82	0.0703	0.0339	
$^4G_{5/2} \rightarrow ^6F_{7/2}$	1036	2.14	0.0084	0.0044		2.14	0.0084	0.0045		2.15	0.0085	0.0046	
$^4G_{5/2} \rightarrow ^6F_{9/2}$	1180	1.63	0.0064	0.0048		1.62	0.0064	0.0049		1.60	0.0063	0.0050	

**V. CONCLUSION**

In the present study, the glass samples of composition  $(45-x)MoO_3:10ZnO:10Li_2O:10Al_2O_3:25B_2O_3:xSm_2O_3$ . (where  $x=1, 1.5, 2$  mol %) have been prepared by melt-quenching method. The Judd-Ofelt theory has been applied to calculate the oscillator strength and intensity parameters  $\Omega_\lambda (\lambda=2, 4, 6)$ . The radiative transition probability, branching ratio are highest for ( $^4G_{5/2} \rightarrow ^6H_{7/2}$ ) transition and hence it is useful for laser action. The stimulated emission cross section ( $\sigma_p$ ) has highest value for the transition ( $^4G_{5/2} \rightarrow ^6H_{7/2}$ ) in all the glass specimens doped with  $Sm^{3+}$  ion. This shows that ( $^4G_{5/2} \rightarrow ^6H_{7/2}$ ) transition is most probable transition.

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