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Spectral and TransmittanceProperties of Nd^{3+} ions doped Zinc LithiumLead Vanadium Boromolybdate Glasses

S.L.Meena
CeramicLaboratory, Department of physics, Jai NarainVyas University, Jodhpur 342001(Raj.) India

Abstract
Glass of the system: (40-x) MoO_3:10ZnO:10Li_2O:10PbO:10V_2O_5:20B_2O_3: xNd_2O_3. (where x=1, 1.5,2 mol %) have been prepared by melt-quenching method.The amorphous nature of the glasses was confirmed by X-ray diffraction studies.Optical absorption,Excitation, fluorescence and Transmittance spectra were recorded at room temperature for all glass samples. Slater-Condon parameters F_k(k=2, 4, 6), Lande' parameter ξ_{4f} and Racah parameters E^k(k=1, 2, 3) have been computed. Using these parameters energies and intensities of these bands has been calculated. Judd-Ofelt intensity parameters Ω_λ (λ=2, 4, 6) are evaluated from the intensities of various absorption bands of optical absorption spectra. Using these intensity parameters various radiative properties like spontaneous emission probability (A), branching ratio(β_R), radiative life time(τ_R) and stimulated emission cross–section(σ_p)) of various emission lines have been evaluated.

Keywords: *ZLLVBM Glasses, Optical Properties, Judd-Ofelt Theory, Transmittance Properties.*

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I. Introduction

Glasses are receiving considerable attention due to their unique properties like highrefraction index, hardness, transparency, electrical, optical and physical properties[1-5].Among the numerous glass hosts, molybdate glass combines the attributes of wide transmission region, low phonon energy, high refractiveindex and excellent rare earth ions solubility.Molybdate glasses are potentially important host materials for developing rare earth doped optical devices. Molybdateglasses have excellent transparency, good mechanical and thermal stability [6-8].The additionof ZnO increases both the tendency of glass formation, refractiveindex while decreases the optical energy band gap [9].Molybdatebased glasses have been recently the subject of many types of research according to their promising mechanical, optical and magnetic properties as well their chemical durability, large refractive indices and good optical transmission [10-12].

The present work reports on the absorption and emission properties of Nd^{3+} doped zinc lithium lead vanadium boromolybdate. The intensities of the transitions for the rare earth ions have been estimated successfully using the Judd-Ofelt theory, The laser parameters such as radiative probabilities(A),branching ratio (β_R), radiative life time(τ_R) and stimulated emission cross section(σ_p) are evaluated using J.O.intensity parameters(Ω_λ, λ=2,4 and 6).

II. Experimental Techniques

Preparation of glasses

The following Nd^{3+} doped Boromolybdate glass samples (40-x) MoO_3:10ZnO:10Li_2O:10PbO:10V_2O_5:20B_2O_3:xNd_2O_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 MoO_3,ZnO, Li_2O,PbO,V_2O_5, $B_2O_3$and Nd_2O_3. They were thoroughly mixed by using an agate pestle mortar. Then melted at 1075˚C by an electrical muffle furnace for 2 hours. After complete melting, the melts were quickly poured in to a preheated stainless steel mould and annealed at temperature of 350˚C for 2 h 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.

Sample	Glass composition (mol %)
ZLLVBM (UD)	40MoO_3:10ZnO:10Li_2O:10PbO:10V_2O_5:20B_2O_3
ZLLVBM (ND1)	39MoO_3:10ZnO:10Li_2O:10PbO:10V_2O_5:20B_2O_3:1Nd_2O_3
ZLLVBM(ND1.5)	38.5MoO_3:10ZnO:10Li_2O:10PbO:10V_2O_5:20B_2O_3:1.5Nd_2O_3

ZLLVBM(ND2) $38MoO_3:10ZnO:10Li_2O:10PbO:10V_2O_5:20B_2O_3: 2Nd_2O_3$

ZLLVBM(UD) -Represents undopedZinc LithiumLead Vanadium BoroMolybdate glass specimens.
ZLLVBM(ND) -Represents Nd^{3+} doped Zinc LithiumLead Vanadium BoroMolybdate glass specimens.

III. Theory

3.1Oscillator Strength

The intensity of spectral lines is expressed in terms of oscillator strengths using the relation [13].

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

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 [14], using the modified relation:

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

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.

3.2. Judd-Ofelt Intensity Parameters

According to Judd[15] and Ofelt[16] 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:

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

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

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

$\lambda = 2, 4, 6$

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.

3.3Radiative 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 many fold $|4f^N (S,L)J\rangle$ is given by

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

S L J

The radiative life time is given by

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

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'> to a final manifold
│4f^{N} (S,L)J >| is expressed as

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

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

IV. Result and Discussion

4.1. XRD Measurement

Figure 1presents the XRD pattern of the samples shows 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.

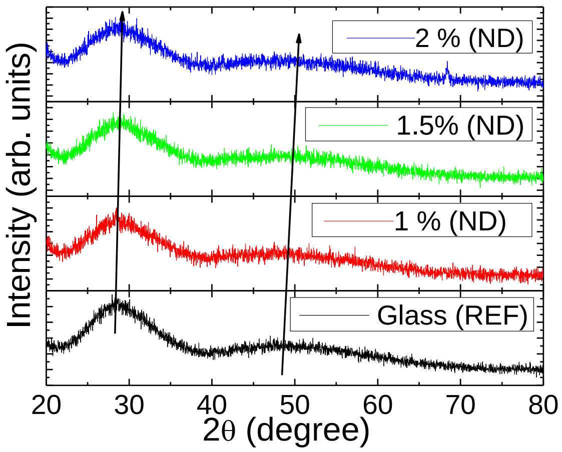

Fig.1:X-ray diffraction pattern of MoO_3:ZnO:Li_2O:PbO:V_2O_5:B_2O_3:Nd_2O_3 glasses.

4.2 Transmittance Spectrum

The Transmittance spectrum of Nd^{3+}doped in zinc lithium lead vanadium boromolybdate glass is shown in Figure 2.

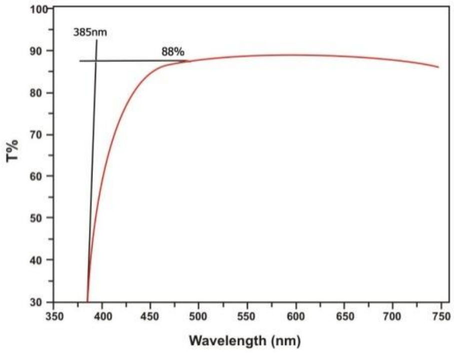

Fig. (2) Transmittance spectrum of Nd^{3+} doped ZLLVBM glasses.

4.3. Absorption spectra

The absorption spectra of ZLLVBM (ND01) glass, consists of absorption bands corresponding to the absorptions from the ground state $^4I_{9/2}$ of Nd^{3+} ions. Nine absorption bands have been observed from the ground state $^4I_{9/2}$ to excited states $^4F_{3/2}$, $^4F_{5/2}$, $^4F_{7/2}$, $^4F_{9/2}$, $^2H_{11/2}$, $^4G_{5/2}$, $^4G_{7/2}$, $^4G_{9/2}$, and $^2G_{9/2}$ for Nd^{3+} doped ZLLVBM(ND 01) glass.

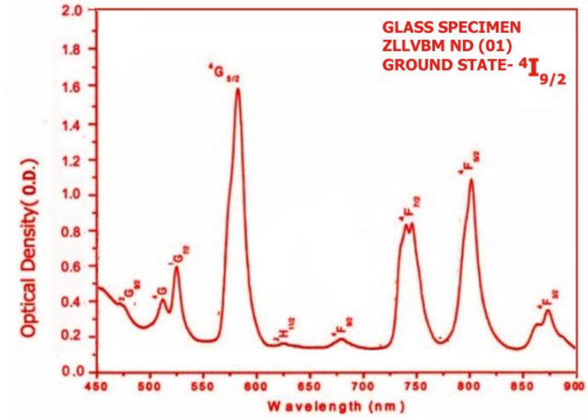

Fig.3: Absorption spectra of ZLLVBM (ND 01) glass.

The experimental and calculated oscillator strength for Nd^{3+}ions in ZLLVBM glasses are given in **Table 2.**

Table 2.Measured and calculated oscillator strength ($P^m \times 10^{+6}$) of Nd^{3+} ions in ZLLVBM glasses.

Energy level from $^4I_{9/2}$	Glass ZLLVBM (ND01)		Glass ZLLVBM (ND1.5)		Glass ZLLVBM (ND02)	
	P_{exp}.	P_{cal}.	P_{exp}.	P_{cal}.	P_{exp}.	P_{cal}.
$^4F_{3/2}$	4.13	4.24	3.25	3.46	3.12	3.32
$^4F_{5/2}$	9.46	9.26	8.55	8.14	8.14	7.88
$^4F_{7/2}$	9.63	10.11	8.75	9.30	8.46	9.05
$^4F_{9/2}$	0.765	0.558	0.725	0.501	0.645	0.486
$^2H_{11/2}$	0.325	0.159	0.312	0.142	0.265	0.138
$^4G_{5/2}$	27.35	27.65	26.48	26.72	25.32	25.67
$^4G_{7/2}$	4.88	5.81	3.80	5.09	3.428	4.90
$^4G_{9/2}$	2.86	2.55	2.68	2.16	2.172	2.08
$^2G_{9/2}$	0.96	3.30	0.88	2.75	0.78	2.64
r.m.s.deviation	0.8754		0.8204		0.8336	

Table3.Computed values of Slater-Condon,Lande′, Racah, nephelauexetic ratio and bonding parameter for Nd^{3+} doped ZLLVBMglass specimens.

Parameter	Free ion	ZLLVBM(ND01)	ZLLVBM(ND1.5)	ZLLVBM (ND02)
$F_2(cm^{-1})$	331.16	324.552	324.601	324.5035
$F_4(cm^{-1})$	50.71	50.733	50.729	50.358
$F_6(cm^{-1})$	5.154	5.0368	5.0394	5.0353
$\xi_{4f}(cm^{-1})$	884.0	882.6421	882.633	882.7769
$E^1(cm^{-1})$	5024.0	4946.8370	4947.010	4946.2019
$E^2(cm^{-1})$	23.90	23.067999	23.0767	23.060344
$E^3(cm^{-1})$	497.0	489.60265	489.5979	489.5744
F_4/F_2	0.1531	0.15632	0.15628	0.15634891
F_6/F_2	0.0155	0.15519	0.01552488	0.015516825
E^1/E^3	10.1086	10.1038	10.1056	10.10306
E^2/E^3	0.0481	0.047116	0.047134	0.0471028
β'		0.9958	0.9959	0.9956
$b^{1/2}$		0.04583	0.04506	0.04690

Judd-Ofelt intensity parameters $\Omega_\lambda(\lambda = 2, 4$ and $6)$ were calculated by using the fitting approximationof the experimental oscillator strengths to the calculated oscillator strengths with respect to their electric dipole contributions.

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

Table 4. Judd-Ofelt intensity parameters for Nd^{3+} doped ZLLVBMglass specimens.

Glass Specimen	$\Omega_2(pm^2)$	$\Omega_4(pm^2)$	$\Omega_6(pm^2)$	Ω_4/Ω_6	Ref.
ZLLVBM (ND01)	2.239	9.122	3.558	2.5638	[P.W.]
ZLLVBM(ND1.5)	3.259	7.335	3.352	2.1882	[P.W.]
ZLLVBM(ND02)	3.129	7.032	3.268	2.1518	[P.W.]
LTTNd10	4.54	5.79	5.69	0.75	[17]
YZLBB(ND)	3.574	5.511	4.464	1.2345	[18]

4.4 Excitation Spectrum

The Excitation spectra of Nd^{3+}doped ZLLVBMglass specimens have been presented in Figure 4 in terms of Excitation Intensity versus wavelength. The excitation spectrum was recorded in the spectral region 700–1000 nm fluorescence at 1065nm having different excitation band centered at 808 nm and 888 nmare attributed to the ($^4I_{9/2}\rightarrow{}^4F_{5/2}$) and($^4I_{9/2}\rightarrow{}^4F_{3/2}$) transitions, respectively. The highest absorption level is $^4F_{5/2}$ and is at 808 nm.So this is to be chosen for excitation wavelength.

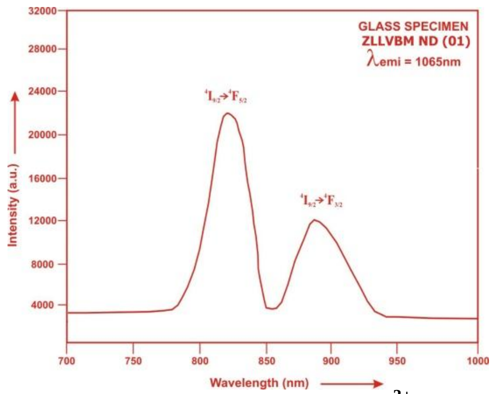

Fig. (4) Excitation spectrum of doped with Nd^{3+} ZLLVBM glasses.

4.5. Fluorescence Spectrum

The fluorescence spectrum of Nd^{3+}doped in zinc lithium lead vanadium boromolybdate is shown in Figure 5. There are five broad bands($^4G_{7/2}\rightarrow{}^4I_{9/2}$), ($^4G_{7/2}\rightarrow{}^4I_{11/2}$) ($^4F_{3/2}\rightarrow{}^4I_{9/2}$),($^4F_{3/2}\rightarrow{}^4I_{11/2}$) and ($^4F_{3/2}\rightarrow{}^4I_{13/2}$) respectively for glass specimens.

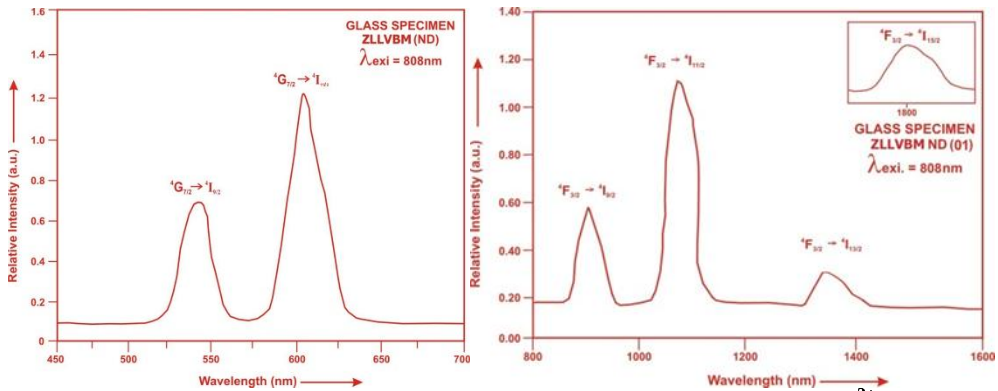

Fig.5:Fluorescence spectrum of ZLLVBM glasses doped with Nd^{3+}.

The wavelengths of these bands along with their assignments are given in **Table 5**.

Table 5.Emission peak wave lengths (λ_p), radiative transition probability (A_{rad}), branching ratio (β_R), stimulated emissioncrosssection (σ_p), and radiative life time (τ_R) for various transitions in Nd^{3+} doped ZLLVBMglasses.

Transition		ZLLVBM (ND01)				ZLLVBM (ND1.5)				ZLLVBM (ND 02)			
	λ_{max} (nm)	$A_{rad}(s^{-1})$	β	σ_p (10^{-20} cm^2)	τ_R(μs)	$A_{rad}(s^{-1})$	β	σ_p (10^{-20} cm^2)	τ_R (μs)	$A_{rad}(s^{-1})$	β	σ_p(10^{-20} cm^2)	τ_R(10^{-20} cm^2)
$^4G_{7/2}\rightarrow{}^4I_{9/2}$	532	3538.95	0.4361	0.528	123.23	3079.72	0.4092	0.473	132.86	2960.26	0.4086	0.475	138.03
$^4G_{7/2}\rightarrow{}^4I_{11/2}$	595	3108.81	0.3831	1.260		3208.02	0.4262	1.381		3088.62	0.4263	1.416	
$^4F_{3/2}\rightarrow{}^4I_{9/2}$	905	900.27	0.1109	0.905		736.289	0.0978	0.765		708.22	0.0977	0.761	
$^4F_{3/2}\rightarrow{}^4I_{11/2}$	1075	490.78	0.0605	2.405		430.615	0.05721	2.258		417.48	0.0576	2.356	
$^4F_{3/2}\rightarrow{}^4I_{13/2}$	1320	74.58	0.00919	0.431		70.406	0.00935	0.422		68.76	0.00949	0.430	
$^4F_{3/2}\rightarrow{}^4I_{15/2}$	1800	1.78	0.000219	0.0257		1.68	0.000224	0.0249		1.64	0.000226	0.0252	

V. Conclusion

In the present study, the glass samples of composition (40-x) MoO_3:10ZnO:10Li_2O:10PbO: 10V_2O_5:20 B_2O_3: xNd_2O_3.(wherex=1, 1.5, 2 mol %) have been prepared by melt-quenching method. The stimulated emission cross section (σ_p) has highest value for the transition ($^4F_{3/2}\rightarrow{}^4I_{11/2}$) in all the glass specimens doped with Nd^{3+} ion. This shows that ($^4F_{3/2}\rightarrow{}^4I_{11/2}$) transition is most probable transition.On the basis of spectrophotometric, transmittance reaches about 88% for all silicate glasses doped with Nd^{3+}ions.

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