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Optical Properties and Spectroscopic Study of Nd^{3+}, Eu^{3+}, Pr^{3+} Doped Aluminobismuthphosphate Glasses as Optical Amplifiers

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Abstract— **A new class of glasses with composition in mol. wt. % P_2O_5 (44.41), Bi_2O (23.51),Na_2O (19.38),Al_2O_3(12.70) for 0.5 mol. wt. % of Nd^{3+}, Eu^{3+}, Pr^{3+} were fabricated and characterized for Spectroscopic & Optical properties. Standard J-0 model predicted the three phenomenological (omega) parameters and the value of these parametric were employed to obtain decay rates, radiative lifetime, branching ratios of the principal intermainfold transitions. Omega parameter depicted the structural bonding of the host material with those of RE ions. ($^5D_0 \rightarrow {}^7F_4$) transition in case of Eu^{3+} :ABP in the red spectral region have a high fluroscence branching ratio and therefore can be fabricated as EL devices used in display technology. The figure of merit for gain($\sigma_p x \tau_R$)comes out to be 18.37 x $10^{-24}cm^2s$ for transition($^4F_{3/2} \rightarrow {}^4I_{13/2}$)at 3.38ms for Nd^{3+}:ABP sample which is quiet high for these alumino-bismuth phosphate glasses as compared to fluoride and silicate glasses. This makes it suitable for signal amplification in the third telecom window .High value of product of (FWHM xσ_p)is observed to be 795.3 x $10^{-28}cm^3$ in case of Pr^{3+}:ABP for transition ($^3P_0 \rightarrow {}^3H_5$)and 598.4 x 10^{-28} cm^3 for $Nd^{3+:}$ ABP($^4F_{3/2} \rightarrow {}^4I_{13/2}$) shows suitability as optical material to be used as broad band fiber amplifier.**

Keywords: **Phosphate glass; Rare earth doped glass; Optical Spectroscopy; Judd-Ofelt parameters**

I. INTRODUCTION

Glasses containing rare earth ions have attracted much attention as potential materials for various optical devices such as lasers, up conversion stimulated phosphor and high density optical storage. Knowledge of local structure surrounding rare-earth ions in glasses is important not only for interpreting their optical properties. It is also useful for designing laser glasses or phosphors [1]. In the present work we have fabricated trivalent lanthnides Nd^{3+},Eu^{3+}, Pr^{3+} as the active ions in aluminobismuth phosphate glasses as host matrix.

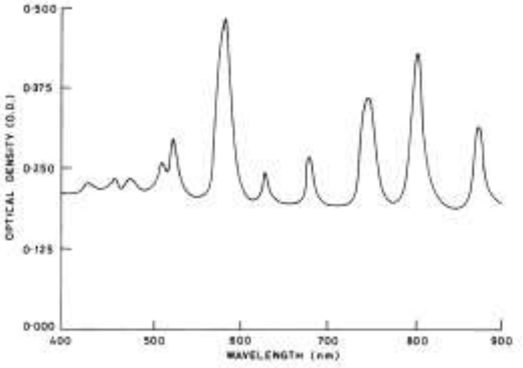

Fig. 1 Absorption Spectrum of 0.5 mol. wt% of ABPND glass specimen.

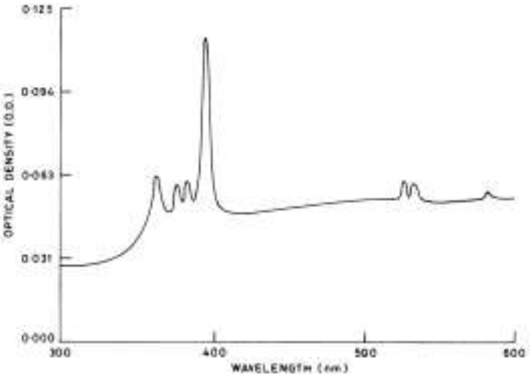

Fig. 2 Absorption Spectrum of 0.5 mol. wt% of ABPEU glass specimen.

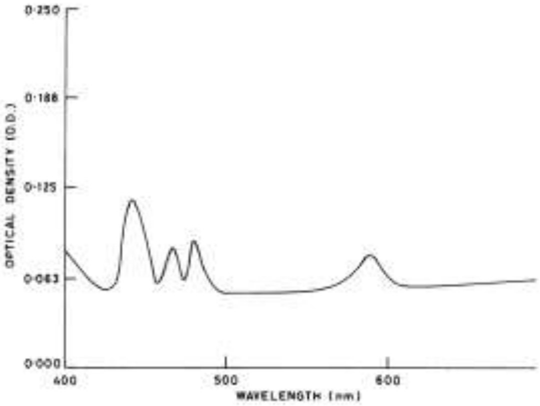

Fig. 3 Absorption Spectrum of 0.5 mol. wt% of ABPPR glass specimen.

II. EXPERIMENTAL

The non crystalline oxide glasses studied in this work were prepared with following compositions in mol wt% P_2O_5 (44.41), Bi_2O (23.51), Na_2O (19.38), Al_2O_3(12.70) for 0.5 mol. wt. % of Nd^{3+}, Eu^{3+}, Pr^{3+} rare earth ions. The three different glass specimens are termed as ABPND, ABPEU, ABPPR, respectively and were prepared using melt quenching technique. These rare earth doped aluminobismuthphosphate (ABP) glass specimens were melted in platinum crucible at 1000^{o} C for seven hours to have maximum optical purity and homogeneity. Silicon carbide Muffle furnace consisting of silicon carbide heating element rods fitted with automatic temperature controller (± 4^{o} C) was used. The glasses were cast onto preheated (up to 200^{o} C) rectangular heavy copper mould and annealed at 300^{o} C to reveal the strain and finally samples are grounded and polished. The refractive index for sample ABPND, ABPEU and ABPPR varies as 1.4987, 1.4993 and 1.4982 respectively. Thickness of the sample varies between 0.346 – 0.349 cm.

Optical spectrum in visible region at room temperature was recorded using Spctro Scan 80D/80DV in UV-VIS Spectrophotometer in the range 300-900 nm. The fluroscence Spectrum was recorded using Shimadzu RF-530 IPC instrument. The fluroscence Spectrophotometer irradiates the sample with excitation light and measures the fluroscence emitted from the irradiated sample to perform quantitative and qualitative analysis.

Glass specimen	Band	λ_{max} (nm)	υ (cm^{-1})	S_m ($x10^{-20}$)	S'_{cal} ($x10^{-20}$)
ABPND	$^4F_{3/2}$	875	11267	0.682	0.008
	$^4F_{5/2}$	806.2	12345	0.797	0.125
	$^4F_{7/2}$	748	13513	0.449	0.065
	$^4F_{9/2}$	674.9	14598	1.36	0.006
	$^2H_{11/2}$	627	16949	0.392	0.001
	$^4G_{5/2}$	585.4	17316	0.417	0.109
	$^4G_{7/2}$	524.9	18867	0.400	0.043
	$^4G_{9/2}$	512.4	19417	0.282	0.027
	$^2K_{15/2}$	476	21052	0.394	0.013
	$^4G_{11/2}$	460	21739	0.222	0.016
	$^2P_{1/2}$	428	23121	0.587	0.002
$\sigma_{r.m.s} = \pm 74.254\ x10^{-6}$					
Glass specimen	Band	λ_{max} (nm)	υ (cm^{-1})	S_m ($x10^{-20}$)	S'_{cal} ($x10^{-20}$)
ABPEU	$^5D_0$	361	17152	0.89	0.61
	$^5D_1$	378	19083	0.69	0.79
	$^5D_2$	383	21551	0.54	0.17
	$^5L_6$	395	25316	0.13	0.11
	$^5G_2$	464	26109	0.10	0.71

	$^5G_4$	524	26455	0.70	0.64
	$^5D_4$	583	27700	0.62	0.22
$\sigma_{r.m.s} = \pm 0.45 \text{ x}10^{-6}$					
Glass specimen	Band	λ_{max} (nm)	υ (cm^{-1})	S_m ($x10^{-20}$)	S'_{cal} ($x10^{-20}$)
ABPPR	$^3F_2$	*2061.8	4850	3.150	3.640
	$^3F_3$	*1541	6489	6.467	6.535
	$^3F_4$	*1474.9	6780	2.347	3.497
	$^1G_4$	*1016.15	9841	0.298	0.215
	$^1D_2$	591.6	16903	0.28	1.20
	$^3P_0$	481	20790	2.62	1.96
	$^3P_1$	467	21413	0.38	0.12
	$^3P_2$	442	22624	0.35	0.19
$\sigma_{r.m.s} = \pm 144.92 \text{ x}10^{-6}$					

*Reported values(Ref. [23])

Table-I: Experimental **(**S_m) and Calculated S'_{cal} line strength of different transitions and their assignments for 0.5 mol. wt.% ABPND, ABPEU, ABPPR glass specimens.

III. RESULT

A. Spectroscopic Properties

Absorption spectra is reported for the three glass specimens in the UV/VIS/NIR region. Total eleven bands were recorded for ABPND in the range 400-900nm. We report seven bands in ABPEU sample in the range 300-600nm and total eight bands for ABPPR in the range 400-700nm. Transitions assigned to them are reported in Table-I along with the energy parameter (cm^{-1}) and observed and calculated line strength. The variation of wavelength with optical density in each case is depicted in Fig.1, Fig.2, Fig.3 respectively. These values serves as basis for calculation of line strength and omega parameters. Since intensity of various absorption bands in the glasses is expressed generally in terms of line strength. This experimental line strength S_m and theoritical line strength S'_{cal} can be given by $Eq^n(1)$

$$S'_{cal} = \sum_{\lambda=2,4,6} \Omega_\lambda \ | 4f^n (S,L)J|| U^\lambda || 4f^n (S' L')J' |^2 \text{ - } Eq^n(1)$$

$$= \frac{s'_{cal}}{e^2}$$

In the above equation Ω_λ is the J-O intensity parameter determined by least square fit method and reported in Table-II . $\| U^\lambda \|$ are doubly reduced squared matrix elements of the tensor operator of rank λ=2, 4, 6 calculated from the intermediate coupling approximation of the transition [ψ J → ψ'J'] where ψ J is the wave function of the ground state represented by | $4f^n$ (S,L)J> and ψ'J' is the wave function of the corresponding excited state [2] represented by $4f^n$ (S' L')J' >

λ	**ABPND**	**ABPEU**	**ABPPR**
2	4.09±1.37	1.98±1.57	1.22±1.84
4	5.64±2.36	2.03±1.86	3.08±2.43
6	5.71±2.36	2.53±2.23	3.94±1.8

Table-II : Judd- Ofelt intensity parameter for 0.5 mol.wt.% ABPND, ABPEU, ABPPR glass specimens
Ω_λ x10^{-20} cm^2

B. J-O Intensity Parameters

In all the three glass specimen studied by us maximum variation is seen in Ω_6 parameter and least variation is seen in Ω_2 parameter (Table-II). The root mean square deviations having quit low values (Table-I) are in usual range for such kind of analysis. In case of ABPND and ABPPR omega values varies as ($\Omega_6 \sim \Omega_4 > \Omega_2$) but for ABPEU omega varies as($\Omega_6 \sim \Omega_4 \sim \Omega_2$) Table-II. Ω_2 Parameter strongly depends on covalent bonding and on structural changes in the vicinitfy of the RE ions while Ω_6 parameter is related to rigidity and stability of the medium in which ions are situated. The spectroscopic quality factor(x= Ω_4/ Ω_6) is found to be 0.98 for ABPND, 0.80 for ABPEU and 0.78 for ABPPR. Thus quality factor is maximum for Nd $^{3+}$ doped covalent ABP glasses.

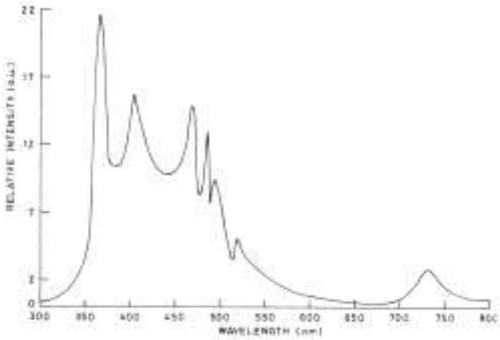

Fig. 4 Fluroscence Spectrum of 0.5 mol. wt% of ABPND λ_{ex}=254 nm

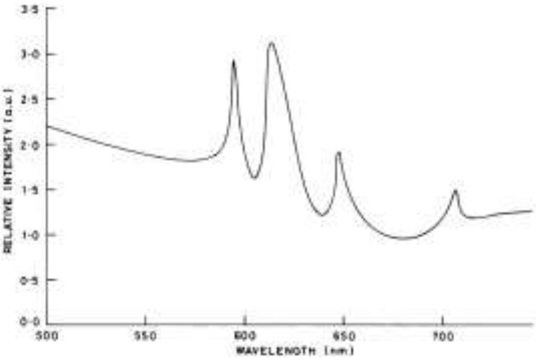

Fig. 5 Fluroscence Spectrum of 0.5 mol. wt% of ABPEU λ_{ex}=470 nm

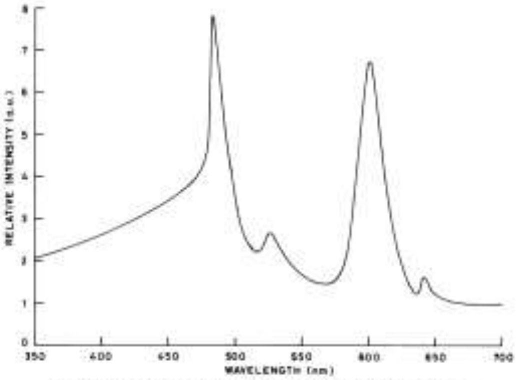

Fig. 6 Fluroscence Spectrum of 0.5 mol. wt% of ABPPR λ_{ex}=441 nm

C. FLUROSCENCE PROPERTIES

The fluroscence spectra for Nd^{3+} : ABP at excitation wavelength 254nm($^2F_{7/2} \rightarrow {}^4I_{9/2}$) and for Eu^{3+} : ABP at 470nm ($^5D_2 \rightarrow {}^7F_1$)and for Pr $^{3+}$: ABP at 441nm($^1I_6 \rightarrow {}^3H_4$) is shown in Fig.4,Fig.5,Fig.6 respectively. The transitions, along with their peak position and laser parameters are recorded in Table-III. Out of seven bands of ABPND sample Fig.4, one peak at 486nm ($^2K_{15/2} \rightarrow {}^4J_{9/2}$) is a magnetic dipole allowed transition and hence is not included for laser calculation.

Four fluroscence peaks appear in case of Eu^{3+}: ABP specimen Fig.5. Two of the transition namely($^5D_0 \rightarrow {}^7F_3$) is a weak transition therefore unable to calculate $\Delta\lambda_{eff}$ precisely and transition($^5D_0 \rightarrow {}^7F_1$) is a magnetic dipole allowed transition and therefore not included for calculation of laser parameter.

In case of Pr^{3+}: ABP glassy matrix four peaks are recorded and shown in Fig.6. Standard J-O theory and fluroscence parameters were used to calculate the spontaneous emission probability (A) fluroscence branching ratio (ß), radiative life time and stimulated emission cross section , measured fluroscence life time,radiative quantum effeciency are represented in Table- III.

The spontaneous emission probability A from an initial mainfold ψ'J' to a final mainfold ψ J is given by Eq^n (2).

$$A(\psi' J', \psi J) = \frac{64\pi^4 \upsilon^3}{3h(2J'+1)} \left[\frac{n(n^2+2)^2}{9} S_{ED} + S_{MD} n^2 \right] \tag{2}$$

Where n is the refractive index, h is the Planck's constant ,υ is the energy of transition, S_{ED} and S_{MD} are the calculated line strength and magnetic dipole transition respectively. The matrix elements of the doubly reduced unit tensor operator (3) have been used in the determination of Scale. Here $A_T(\psi' J')$ represents the total spontaneous emission probability as Sum of A (ψ' J', ψ J)

$$A_T(\psi J) = \sum_{\psi J} A(\psi' J', \psi J) \qquad (3)$$

The equations for laser parameters like fluroscence branching ratio Eq^n (4), measured fluroscence lifetime Eq^n (5) and stimulated emission cross section Eq^n (6) are mentioned below.

$$\beta_R(\psi' J', \psi J) = \frac{A(\psi' J', \psi J)}{A_T(\psi J)} \qquad (4)$$

$$\tau = \left[A(\Psi' J', \Psi J)\right]^{-1} \qquad (5)$$

$$\sigma_p = \frac{\lambda_p^4}{8\pi c n^2 \Delta\lambda_{eff}} A(\psi' J', \psi J) \qquad (6)$$

In Eq^n (6) λ_p is the fluroscence peak wavelength of the emission band and $\Delta\lambda_{eff}$ is the effective fluroscence line width. Review on the laser parameter listed in Table-III will through some light on how they are favorable for this material to become a potential candidate as an efficient laser system operating in visible region [4].

Transitions having spontaneous emission probability greater than 500 sec^{-1} , fluroscence branching ratio~0.5 and energy separation~ 6000 cm $^{-1}$ between the energy level under consideration and terminating level are considered to be good radiative transitions[11]. The last requirement is essential, in order to circumvent the non-radiative relaxation rates [5].

Since ($^4G_{7/2} \rightarrow {}^4I_{9/2}$) transitions in the green spectral region in ND $^{3+}$:ABP ,($^5D_0 \rightarrow {}^7F_4$) transition in red spectral region in Eu^{3+}:ABP and ($^1D_2 \rightarrow {}^3H_5$)in the orange spectral region in case of Pr^{3+}:ABP can be considered to be good laser transition in the visible region as per conditions mentioned above.Out of which Eu^{3+}:ABP have high value of fluroscence branching ratio therefore can be used in application such as fabrication of electroluminescent(EL)devices [6] for use in display technology.

The($^5D_0 \rightarrow {}^7F_1$) transition in ABPEU and ($^2K_{15/2} \rightarrow {}^4I_{9/2}$)in ABPND are magnetic dipole allowed transition (MD). The MD transitions contribute to sharp peak in emission spectra.

Stimulated emission cross section σ_p is the most important laser parameter as it signifies the rate of energy extraction from the laser material. The value of σ_p ranges from 1.73-7.23 pm^2 for ABPPR and are quiet high as compared to other borophosphate [24] and silicate glasses [2]. It signifies that the host material used along with the rare earth doping is quiet suitable for use as optical amplifier .The value of σ_p for ABPEU and ABPND in the present glass are comparable to σ_p values in case of other borophosphate glasses [7] and BPG glasses [26] with added advantage that the glasses presently prepared are chemically stable and Thermally durable .

Glass specimen	***Transition***	**λ_p (nm)**	**$\Delta\lambda$ (nm)**	***A (sec^{-1})***	**β_R**	**τ_R (μsec)**	**σ_p (pm^2)**
ABPND	$^4D_{3/2} \rightarrow {}^4I_{9/2}$	***365***	***21.4***	***283.79***	***0.4071***	***3523.73***	***0.0139***
	$^2D_{5/2} \rightarrow {}^4I_{9/2}$	***404***	***15.7***	***1257.59***	***0.1804***	***795.17***	***0.126***
	$^2P_{3/2} \rightarrow {}^4I_{9/2}$	***469***	***14.8***	***603.70***	***0.0866***	***1656.45***	***0.1173***
	$^2K_{15/2} \rightarrow {}^4I_{9/2}$	***486***	***11.2***	-	-	-	-
	$^4G_{9/2} \rightarrow {}^4I_{9/2}$	***494***	***9.3***	***1384.78***	***0.1986***	***722.136***	***0.5248***
	$^4G_{7/2} \rightarrow {}^4I_{9/2}$	***519***	***15.1***	***2977.61***	***0.4271***	***335.83***	***0.8488***

	$^2S_{3/2}\rightarrow{}^4I_{9/2}$	***728***	***12.7***	***463.528***	***0.0664***	***2157.36***	***0.6013***
	$^4F_{3/2}\rightarrow{}^4I_{13/2}$	****1335***	***11***	***296***	***0.04073***	***3378.38***	***5.44***
ABPEU	$^5D_0\rightarrow{}^7F_1$	***595***	***16***	-	-	-	-
	$^5D_0\rightarrow{}^7F_2$	***614***	***13***	***83.10***	***0.1239***	***12033.7***	***0.0538***
	$^5D_0\rightarrow{}^7F_3$	***648***	-	-	-	-	-
	$^5D_0\rightarrow{}^7F_4$	***707***	***14.6***	***587.52***	***0.8760***	***1702.07***	***0.5955***
ABPPR	$^3P_0\rightarrow{}^3H_4$	***483***	***15***	***10267.40***	***0.1670***	***97.39***	***2.25***
	$^3P_0\rightarrow{}^3H_5$	***528***	***11***	***17218.74***	***0.2801***	***58.07***	***7.23***
	$^1D_2\rightarrow{}^3H_4$	***602***	***12.2***	***5224.38***	***0.0850***	***191.42***	***3.34***
	$^1D_2\rightarrow{}^3H_5$	***644***	***16.9***	***2875.06***	***0.4677***	***347.8***	***1.73***

**Reported value(Ref. [25])*

TABLE-III: Wavelength of different fluroscence transitions and their spontaneous emission probability(A), fluroscence branching ratio (ß$_R$),measured radiative lifetime(τ_R),and stimulated emission cross section (σ_p) for ABPND, ABPEU, ABPPR glass specimens

DISCUSSION

Our aim is to study the radiative properties of fluroscence transitions, so that the samples prove to be a good host material for potential broad band amplifier.The glasses with lower bismuth content are easy to form compared to those with higher Bi content. Thermal stability of these glasses is also found to be in good command to form the glasses with ease[22] ,as is true in our case.

Peaks at 469nm ($^2P_{3/2} \rightarrow {}^4I_{9/2}$) and 494 nm ($^4G_{9/2} \rightarrow {}^4I_{9/2}$) in case of ND^{3+}:ABP glass specimen are rarely observed in case of oxide glasses but addition Al_2O_3 has increased their intensities considerably .This may be due to intermixing of electronic levels of Nd^{3+} with molecular orbitals of Al_2O_3 resulting in increase in the population of the excited states of Nd^{3+}. Such kind of effect has also been observed by Reisfeld et al.[8] in case of Sm^{3+} doped oxide glasses on addition of As_2O_3. In the present glass system high value of Ω_6 and relatively small value of Ω_2 parameter has been obtained. Nageno et al. has shown that $\Omega_2$parameter is affected [9] covalent chemical bonding between the central rare earth ion and the environment surrounding it and Ω_6 parameter is related to rigidity and stability of the medium in which the ions are situated. A look at Ω_2 values from Table- II for different RE ions, it may be said that the RE ions under study have the same co-ordination number. Eight coordination number for RE ion in the glasses may be assumed due to eight oxygen provided by both $[PO_4]^{3-}$ chains and the non-bridging oxygen bonded only to a single bismuth atom. With a slight decrease in covalency (Ω_2) parameter, Ω_6 parameter increase because of inverse relationship [10] between the two and as a consequence of it, we find a slight increase in stimulated emission cross section (σ_p) which is considerable in case of ABPPR.

The product of (FWHM X σ_p) is observed to be 795.3 x 10^{-28}cm^3 in case of Pr^{3+}:ABP for transition ($^3P_0 \rightarrow {}^3H_5$) and 598.4 x 10^{-28} cm^3 for Nd^{3+}:ABP($^4F_{3/2} \rightarrow {}^4I_{13/2}$) and is quiet high as compared to those observed in phosphate and silicate glasses [12, 13, 14]. Therefore, the present glass system suitable for optical materials to be used as broad band fiber amplifier. Such materials can be widely used [15, 16] in increasing the transmission capacity of wavelength division multiplex(WDM)syste .High fluroscence branching ratio for transition ($^5D_0 \rightarrow {}^7F_4$)for ABPEU comes out to be 0.8760and thus makes it sutiable for EL devices.

The calculated radiative lifetime of the ($^4F_{3/2} \rightarrow {}^4I_{13/2}$) level for ABPND glass system is 3.38ms and the product of(σ_p x τ_R) known as [17,18] the figure of merit for gain comes out to be 18.37 x 10^{-24}cm^{2}s which is quiet high for these alumino-bismuth phosphate glasses as compared to fluoride [12,19,20]and silicate [2] glasses. Considerable value of figure of merit for gain at 2.15ms for ABPND is 1.297 x 10^{-24} cm^2 s and for ABPEU at 1.702ms is 1.013 x 10^{-24} cm^{2}s are obtained. Therefore, this glass material properly drawn in the form of a fiber is quite suitable for signal amplification in the third telecom window [2]. Such optical amplifier easily compensate for the losses in the processing and distribution of optical signals while maintaining the high band width and low cross talk [21].

CONCLUSION

The spectroscopic analysis of Nd^{3+} , Eu^{3+} , Pr^{3+} rare earth ions in the ABP laser host has been performed following the standard J-O model that predicts the three phenomological parameters. Values of this parameters were then employed to obtain spontaneous emission probability, stimulated emission cross section , decay rates, radiative lifetimes and branching ratio of the principal intermainfold transitions to the lower lying mainfolds.

A comparision between the Ω_λ parameter shows that value of Ω_6 Parameter are higher than Ω_2 parameter which has attributed to decrease in covalency between the centeral RE ions and surrounding eight oxygens atoms provided by both $[PO_4]^{3-}$chains and the non-bridging oxygen bonded only to a single Bismuth atom.

Value of fluroscence branching ratio for transition ($^5D_0 \rightarrow {}^7F_4$) for Eu^{3+}:ABP makes it sutiable as electroluminescent devices. The figure of merit (σ_p x τ_R)for gain is 18.37x10^{-24} cm^{2}s for ($^4F_{3/2} \rightarrow {}^4I_{13/2}$) at 3.38ms for Nd^{3+} :ABP in the glass system which is quite high as compared to fluoride and silicate glasses. A high value of product (FWHM x $\boldsymbol{\sigma_p}$) is observed to be 795.3 x 10^{-28} cm^3 for Pr^{3+}: ABP for transition ($^3P_0 \rightarrow {}^3H_5$) and 598.4 x 10^{-28} cm^3 for $Nd^{3+:}$ABP ($^4F_{3/2} \rightarrow {}^4I_{13/2}$) shows suitability for these new class of aluminobismuthphosphate glasses to be used as optical amplifier.

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