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K and L- shell X-ray Transition-Probability Ratios by Proton Bombardment

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Abstract— The present work aimed to study the energy dependence of K_α/K_β X-ray transition-probability ratios for the elements Fe, Cu, Se, Ag and L_α/L_β, L_α/L_γ and L_α/L_ι X-ray transition-probability ratios for the elements Ag, Pb and Bi. It has been measured by proton bombardment in the energy range of 1.5 to 3.0 MeV. Our results provide constant transition-probability ratios for K shell X-ray with varying proton energies, while for L X-ray, transition probability ratios show variation with incident proton energy. X-ray transition probability has been compared with the theoretical ratios, calculated by relativistic Hartee-Slater theory and using sub-shell direct ionization cross-sections.
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Keywords— PIXE, X-ray Transition Probability,Emmision rate, Fluorescence yield, ECPSSR theory,Inner-shell ionization cross section.

I. INTRODUCTION

Accurate atomic data such as X-ray transitions- probability ratios, fluorescence yields and Coster-Kronig transition probability, are important for applications of trace element analysis by Particle Induced X-ray Emission (PIXE), X-ray Fluorescence (XRF) and fundamental physical processes studies of ion-atom collisions. Several experimental as well as theoretical data for X-ray transition probability ratios has been reported [1-7], but our aim was to measure X-ray transition probability ratios through PIXE (Proton Induced X-ray emission) from 1.5-3.0 MeV. PIXE measurement is a non destructive trace elemental analysis being done in this energy range. Low Z elements are determined by K X-ray, where as high Z elements are determined by L X-ray, due to poor detection efficiency of Si(Li) detector at high energy K X-ray. For this we have selected low Z elements (Fe, Cu, Se, Ag) to measure K shell X-ray transition probability ratios and high Z elements (Ag, Pb, Bi) to measure L shell X-ray transition probability ratios with consecutive atomic number 82 and 83. Transition probability directly related with X-ray production cross section and X-ray cross section is directly related with concentration of element [8]. Experimental X-ray transition probability ratios are required for testing the validity of the theories on inner-shell ionization by proton. Measured K-shell X-ray transition probability ratios has been compared with theoretical ratios calculated by J.H. Scofield with the help of relativistic Hartee-Slater theory [9-10], while L-shell X-ray transition probability ratios are calculated by using the method of G.A.Bissinger [11] in which we have calculated direction ionization cross section by ECPSSR theory [12-14]. This theory is enable to calculate direct ionization cross section by Proton. ECPSSR theory is the modified form of the Plane wave Born approximation (PWBA) theory and it includes the effect of projectile energy loss (E), Coulomb deflection of the projectile (C), polarization effects of the target electron and relativistic effects (R).The accuracy of the measurement mainly depends on the detection efficiency of the detector for both K and L-shell X-ray. PIXE measurement sensitivity is up to the ppb level. Therefore choice of pure metallic targets become very important.

This paper has been organized into five distinct sections- Introduction, theoretical X-ray intensity calculation for L-shell, experimental details and data analysis, results, discussion and conclusion. The introduction section covers briefly about X-ray transition probability with PIXE and the importance of the method has been discussed. In theoretical X-ray intensity calculation section, we have mentioned formula for calculating X-ray intensity for L-shell. The experimental details and data analysis section provides technical details related to the measurements. Results section provides variation of K and L sub-shell X-ray tansition probability with varying energies of Proton. In the discussion and conclusion section, the results obtained are discussed and major conclusions drawn from the results are provided.

II. THEORETICAL INTENSITY FORMULA.

Theoretical intensity of L-shell X-ray can be calculated using the method of Ref. [11].

$$I_{L\alpha} = \{[n_1(f_{13}+f_{12}f_{23})+n_2\, f_{23} + n_3]\omega_3\}F_{3\alpha}$$

$$I_{L\beta} = n_1\, \omega_1\, F_{1\beta} + (n_1 f_{12}+n_2)\, \omega_2\, F_{2\beta} + [n_1(f_{13}+f_{12}f_{23})+n_2\, f_{23} + n_3]\omega_3 F_{3\beta}$$

$$I_{L\gamma} = n_1\, \omega_1\, F_{1\gamma} + (n_1 f_{12}+n_2)\, \omega_2\, F_{2\gamma}$$

$$I_{L\iota} = \{[n_1(f_{13}+f_{12}f_{23})+n_2\, f_{23} + n_3]\omega_3\}F_{3\iota}$$

Ionization cross-section ratio,

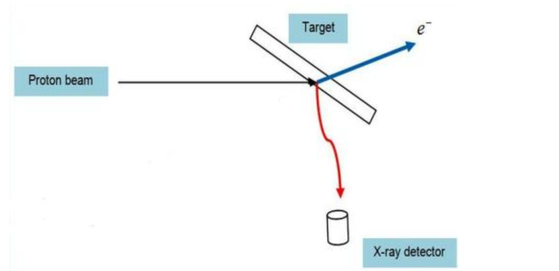
$$n_1 = \sigma_{L1} / \sigma_{L3}$$
$$n_2 = \sigma_{L1} / \sigma_{L3}$$
$$n_3 = 1$$

Symbol, $F_{3\alpha}$, $F_{1\gamma}$, $F_{3\iota}$,$F_{2\gamma}$, $F_{2\beta}$ represent fraction of radiative transition for L sub-shell [15].

$f_{12,}$ f_{13} f_{23} and $\omega_{1,}$ $\omega_{2,}$ ω_3 are Coster-Kronig yields, fluorescence yields respectively taken from Campbell work [16]. σ_{L1} , σ_{L2} , σ_{L3} are sub-shell direct ionization cross sections, calculated using ECPSSR theory Ref.[9-11].

III. EXPERIMENTAL DETAILS AND DATA ANALYSIS

Thin metallic targets were prepared by thermal evaporation method on 10μg/cm^2 Mylar backing. The thicknesses of targets were of the order of few hundred μg/cm^2. The targets were mounted on ladder in vacuum chamber, which is designed for PIXE measurement at Ion Beam Laboratory (IBL), IOP Bhubaneshwar, India. Proton beam of variable energy ranges from 1.5 MeV to 3.0 MeV having beam size 2mm and beam current 5-9 nA, was bombarded by 3.0 MV Pelletron accelerator. The X-ray detector was a Si(Li) detector having a 1.28 μm Be window and energy resolution of 165 eV at 5.9 keV. The angle between X-ray detector and beam direction was 90^o. Targets were positioned on ladder in such a way that the target makes 45^o w.r.t. proton beam direction. In reaching to the detector; X-ray traversed a 20μm Mylar window and 5.0 cm in air. The energy calibrations of spectrum has been done by using standard X-ray sources ^{56}Fe and ^{241}Am. X-ray detection efficiency as a function of energy has been taken from Ortech company[18], for detector Si(Li) Model No. SLP-06 165 OPT- 0.5. K X-ray were analyzed by Origin fitting software and L subgroup X-ray by Candle software which is freely available on official site of IUAC, New Delhi. Targets thicknesses were arranged in such a way that there was no significant absorption of K X-Ray and but absorption correction has been done for L X-ray lines. Theoretical intensity ratio of L-shell X-ray has been calculated using the method used in Ref. [11].



. Figure 1: Schematic diagram of the experimental set-up.

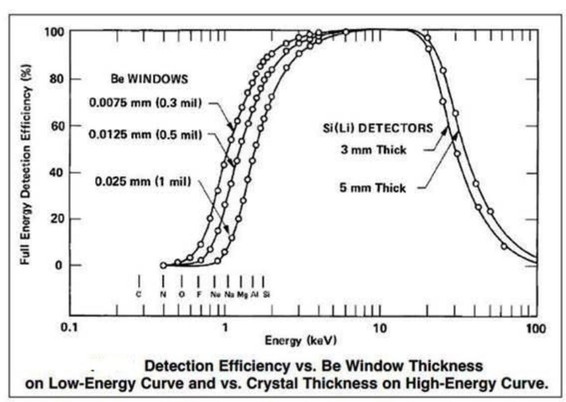


Figure 2: X-ray detection efficiency variation with photon energy **Ortech Company**, for detector Si(Li) Model No.SLP-06 165 OPT- 0.5.

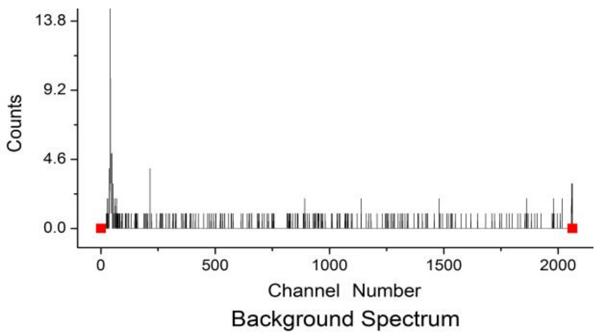


Figure 3: Background spectrum of target holder.

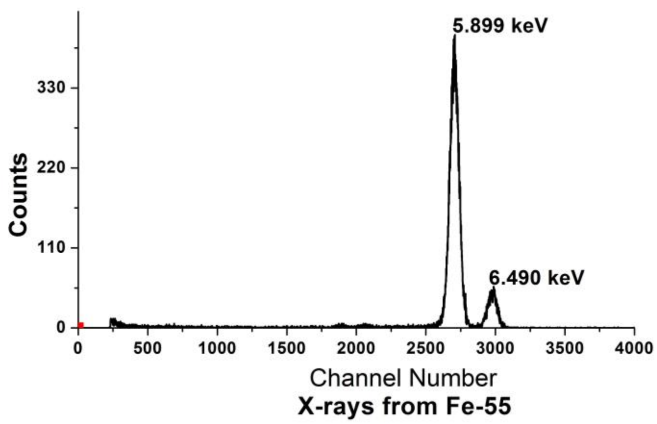


Figure 4. Spectrum of [55]Fe X-ray source for energy calibration.

IV. RESULTS

Characteristics X-ray spectra of Cu and Pb element are shown in figures (7) and (11), transition probability ratios of K_α/K_β L_α/L_β and L_α/L_γ are shown in tables.

Table 1. Experimental K_α/K_β X-ray transition-probability ratios for the elements Fe. Cu, Se and Ag.

Element	Present Work K_α/K_β	Scofield K_α/K_β
Fe	7.2419	7.1890
Cu	7.2727	7.2510
Se	6.3573	6.1576
Ag	4.7891	4.6948

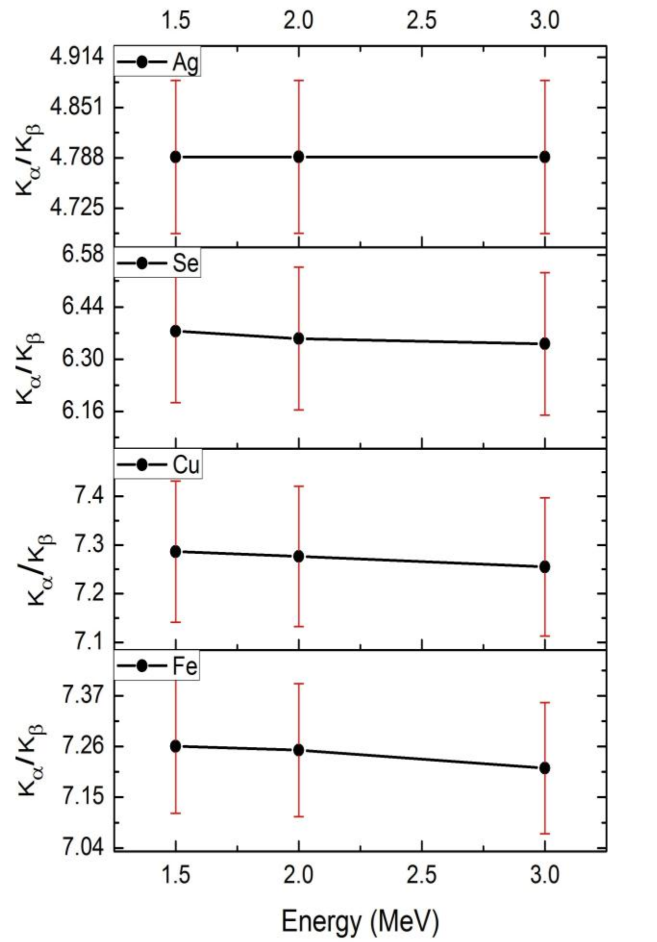


Figure 5: Variation of K_α/K_β ratios of Fe, Cu, Se and Ag with proton energy.

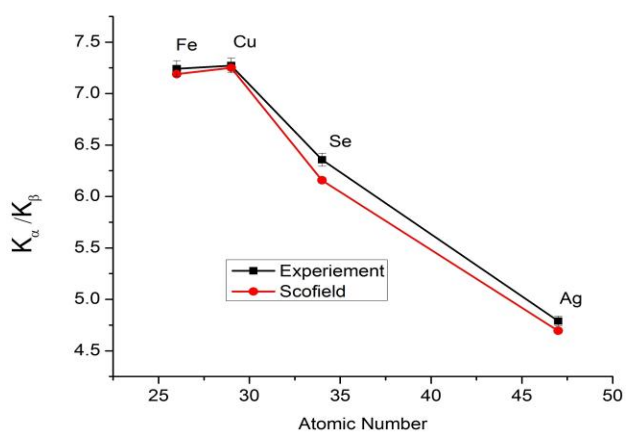


Figure 6: Variation of K_α/K_β ratio with Atomic number.

It is clear from our measurement that ratios K_α/K_β are independent from projectile energy and ratios decrease with increase of atomic number.

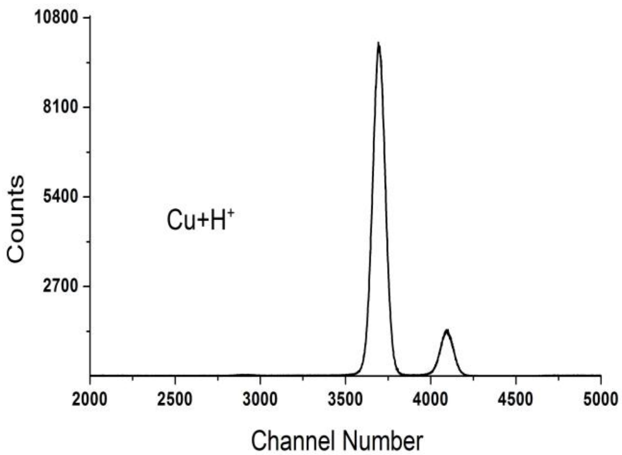


Figure 7: K-shell characteristics X-ray spectrum of Cu produced by 1.5 MeV Proton.

Table 2. Energy dependence of experimental X-ray transition-probability ratios (L_α/L_β) for the elements Ag, Pb, and Bi.

Element	L_α/L_β (1.5 MeV)	L_α/L_β (2.0 MeV)	L_α/L_β (3.0 MeV)
Ag	1.6846	1.6201	1.5118
Pb	2.0257	1.9140	1.8385
Bi	1.9888	1.9081	1.8301

Table3. Energy dependence of theoretical X-ray transitions-probability ratios (L_α/L_β) for the elements Ag, Pb, and Bi.

Element	L_α/L_β (1.5 MeV)	L_α/L_β (2.0 MeV)	L_α/L_β (3.0 MeV)
Ag	1.8793	1.8537	1.8264
Pb	1.8722	1.7868	1.6674
Bi	1.8778	1.7942	1.6747

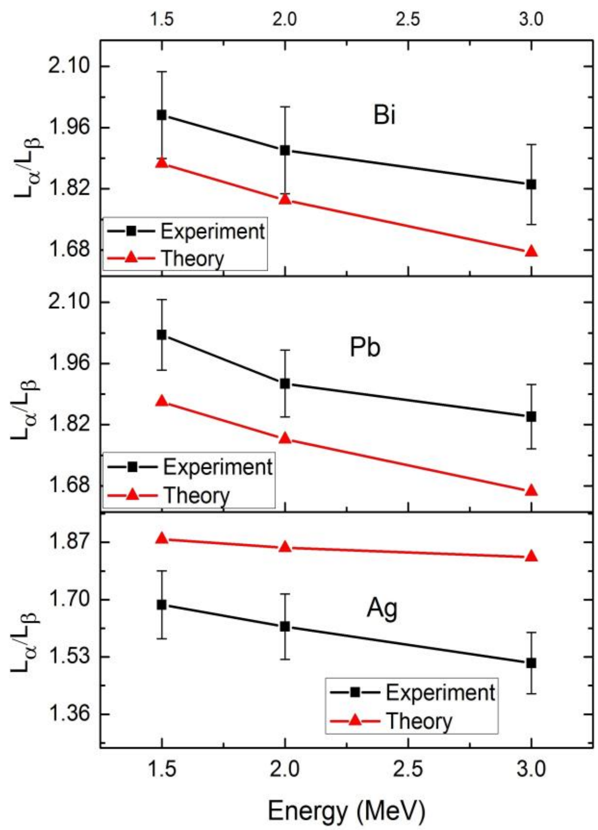


Figure 8: Variation of L_α/L_β ratio with energy for Ag, Pb and Bi elements.

Table 4. Energy dependence of experimental X-ray transition-probability ratios (L_α/L_γ) for the elements Ag, Pb, and Bi.

Element	L_α/L_γ (1.5 MeV)	L_α/L_γ (2.0 MeV)	L_α/L_γ (3.0 MeV)
Ag	16.1496	15.9305	14.6516
Pb	13.8441	12.6219	10.4588
Bi	13.9007	12.6556	11.0300

Table5. Energy dependence of theoretical X-ray transitions-probability ratios (L_α/L_γ) for the elements Ag, Pb, and Bi.

Element	L_α/L_γ (1.5 MeV)	L_α/L_γ (2.0 MeV)	L_α/L_γ (3.0 MeV)
Ag	19.6637	19.1314	18.6722
Pb	14.2635	13.0244	11.1637
Bi	14.2994	13.0997	11.2299

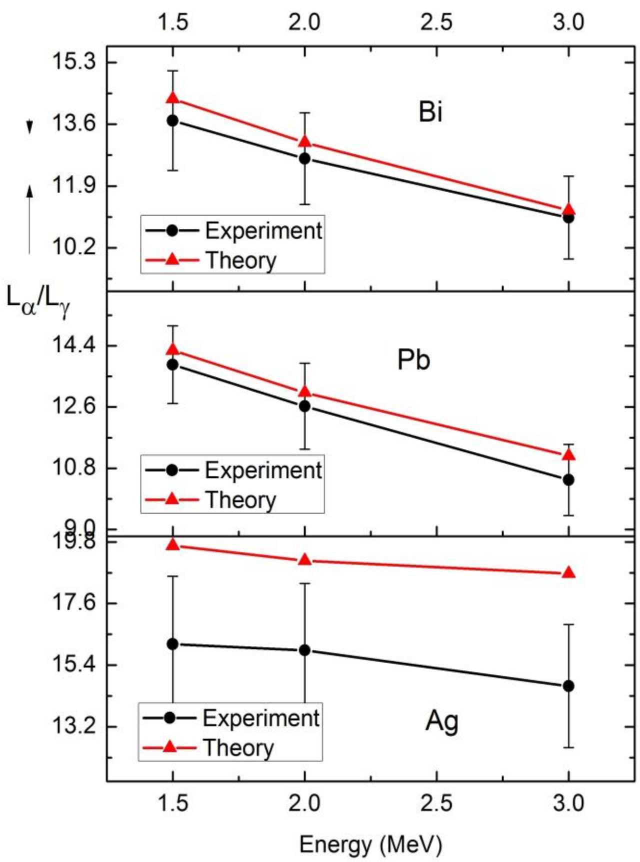


Figure 9: Variation of L_α/L_γ ratio with energy for Ag, Pb and Bi elements.

It is clear from our experimental and theoretical study that ratios L_α/L_β are dependent on projectile energy and ratios decreases with increase of projectile energy. Transition probability ratios (L_α/L_β) increases with the atomic number. Transition probability ratios (L_α/L_γ) also decreases with increase of projectile energy and atomic number.

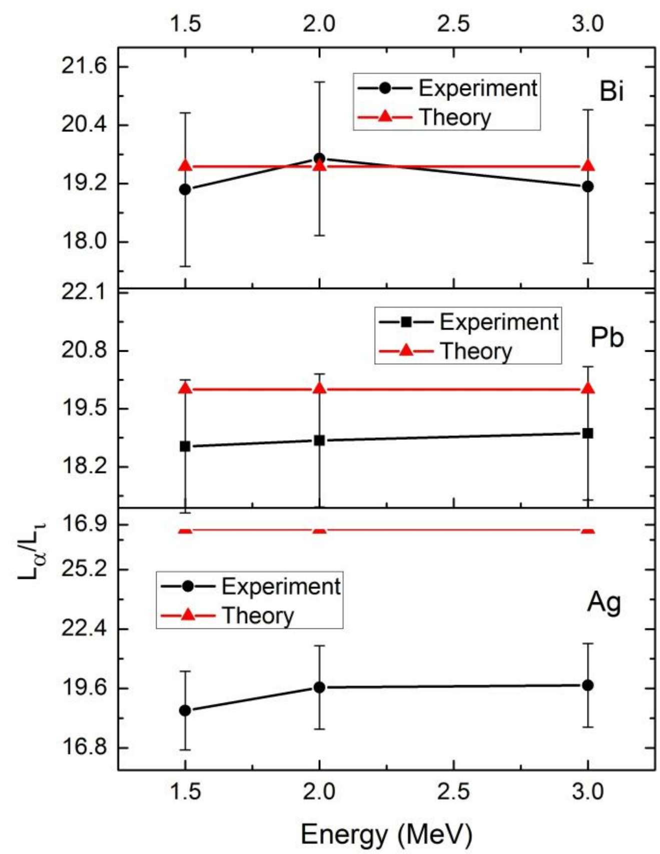


Figure 10: Variation of L_α/L_ι ratios with energy for Ag, Pb and Bi elements

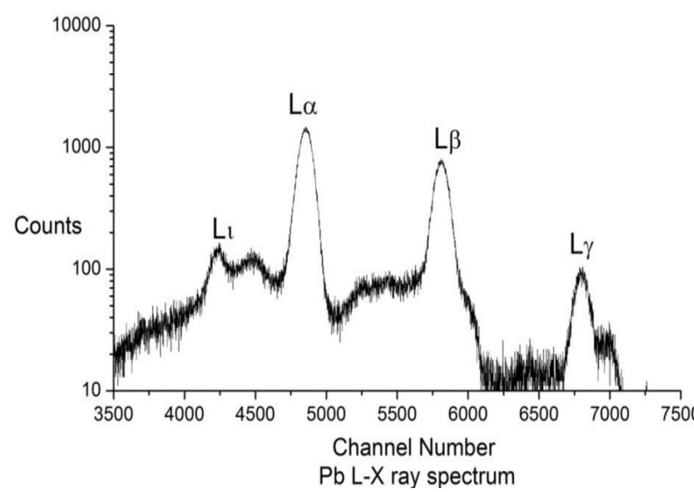


Figure 11: Typical characteristics L-shell X-ray spectrum of Pb target induced by 1.5 MeV Proton.

V. DISCUSSION AND CONCLUSION

In the measurement of L-shell intensity ratio, some common factors such as geometrical efficiency and attenuation of X-ray between target and X-ray detector are cancelled out. Thus X-ray intensity ratio can be determined accurately with the background corrections. The experimentally measured K_α/K_β X-ray transition-probability ratios are compared with the theoretical one calculated by J.H. Scofield [10]. The present data show good agreement with theory in comparison to other measured data [1,6]. It was found from the literature [6] that the K_α/K_β X-ray transitions-probability ratios are independent from nature of excitation mechanism. Figure 5 also shows that K_α/K_β X-ray transition-probability ratios are independent from incident proton energy. From present measurement it is also verified that the intensity ratio increases as Z increases. The uncertainty in the K_α/K_β X-ray transitions- probability ratio is found to be on average about 2%. The experimental values for the L-shell X-ray intensity ratios for Ag**,** Pb and Bi at three excitation energies are listed in tables 2-5. The uncertainty due to statistical error for L_α/L_β ratio of the elements Pb, Bi and Ag are 4-6%.The uncertainty for L_α/L_γ ratios are 10-15 % and in L_α/L_ι ratios are 8-10% depending upon the counting statistics, incident particle energy and target thickness..For all the three elements L-X ray ratios L_α/L_β, L_α/L_γ and L_α/L_ι were measured and compared to ECPSSR theoretical intensity. Theoretical L_α/L_β ratio for Ag predicts about 17% higher w.r.t. experimental measured ratio at 3.0 MeV, while at 1.5 MeV it is about 10%. Our result for Pb L_α/L_β is about 8 % higher than those measured results [17] . For L_α/L_γ our results are 10% and 12 % higher at 1.00 and 3.00 MeV, respectively. For the cases of L_α/L_β and L_α/L_γ, the ECPSSR predictions lower with the measured data over the proton energy range 1.5-3.0 MeV but it is higher when it compared with [7,17] for Pb and Bi at 3.0 MeV. Our study indicates that the ratios L_α/L_ι are independent from incident proton energy theoretically as well as experimentally for all three targets. In case of Bi, for L_α/L_β ratio theory predicts well within the experimental error at energy 1.5 and 2.0 MeV, while at 3.0 MeV energy experimental ratio is higher about 10% with respect to theoretical value calculated by G.E.Bessignger et al.[11] and experimental values measured by D.A.Close *et al.* [7].

VI. ACKNOWLEDGMENT

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