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 $K_e/K_\beta$ X-ray transition-probability ratios for the elements Fe, Cu, Se and L$_e$/L$_\beta$, L$_e$/L$_\gamma$, and L$_e$/L$_\delta$ 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.

Keywords— PIXE, X-ray Transition Probability, Emission 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 able 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 transition 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].

\[ I_{L\alpha} = \left[ n_1 (f_{13} + f_{12}f_{23}) + n_2 f_{23} + n_3 \right] \sigma_{L3} \]

\[ I_{L\beta} = n_1 \omega_1 F_{1\beta} + (n_1 f_{12} + n_2) \omega_2 F_{2\beta} + [n_1 (f_{13} + f_{12}f_{23}) + n_2 f_{23} + n_3] \omega_3 \sigma_{L3} \]

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

\[ I_{L\delta} = \left[ n_1 (f_{13} + f_{12}f_{23}) + n_2 f_{23} + n_3 \right] \omega_3 \sigma_{L3} \]

Ionization cross-section ratio,

\[ n_1 = \sigma_{L1} / \sigma_{L3} \]

\[ n_2 = \sigma_{L1} / \sigma_{L3} \]

\[ n_3 = 1 \]

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

\( f_{12}, f_{13}, f_{23} \) and \( \omega_1, \omega_2, \omega_3 \) are Coster-Kronig yields, fluorescence yields respectively taken from Campbell work [16]. \( \sigma_{L1}, \sigma_{L2}, \sigma_{L3} \) 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 10 \( \mu \)g/cm\(^2\) Mylar backing. The thicknesses of targets were of the order of few hundred \( \mu \)g/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 \( \mu \)m Be window and energy resolution of 165 eV at 5.9 keV. The angle between X-ray detector and beam direction was 90°. Targets were positioned on ladder in such a way that the target makes 45° w.r.t. proton beam direction. In reaching to the detector, X-ray traversed a 20\( \mu \)m Mylar window and 5.0 cm in air. The energy calibrations of spectrum has been done by using standard X-ray sources \( ^{38}\)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.](image)

![Figure 2: X-ray detection efficiency variation with photon energy.](image)
IV. RESULTS

Characteristics X-ray spectra of Cu and Pb element are shown in figures (7) and (11), transition probability ratios of $K_{\alpha}/K_{\beta}$, $L_{\alpha}/L_{\beta}$ and $L_{\alpha}/L_{\gamma}$ are shown in tables.

Table 1. Experimental $K_{\alpha}/K_{\beta}$ X-ray transition-probability ratios for the elements Fe, Cu, Se and Ag.

<table>
<thead>
<tr>
<th>Element</th>
<th>Present Work $K_{\alpha}/K_{\beta}$</th>
<th>Scofield $K_{\alpha}/K_{\beta}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>7.2419</td>
<td>7.1890</td>
</tr>
<tr>
<td>Cu</td>
<td>7.2727</td>
<td>7.2510</td>
</tr>
<tr>
<td>Se</td>
<td>6.3573</td>
<td>6.1576</td>
</tr>
<tr>
<td>Ag</td>
<td>4.7891</td>
<td>4.6948</td>
</tr>
</tbody>
</table>

Figure 3: Background spectrum of target holder.

Figure 4: Spectrum of $^{55}$Fe X-ray source for energy calibration.

Figure 5: Variation of $K_{\alpha}/K_{\beta}$ ratios of Fe, Cu, Se and Ag with proton energy.

Figure 6: Variation of $K_{\alpha}/K_{\beta}$ 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.

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

<table>
<thead>
<tr>
<th>Element</th>
<th>$L_α/L_β$ (1.5 MeV)</th>
<th>$L_α/L_β$ (2.0 MeV)</th>
<th>$L_α/L_β$ (3.0 MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag</td>
<td>1.6846</td>
<td>1.6201</td>
<td>1.5118</td>
</tr>
<tr>
<td>Pb</td>
<td>2.0257</td>
<td>1.9140</td>
<td>1.8385</td>
</tr>
<tr>
<td>Bi</td>
<td>1.9888</td>
<td>1.9081</td>
<td>1.8301</td>
</tr>
</tbody>
</table>

Table 3. Energy dependence of theoretical X-ray transitions-probability ratios ($L_α/L_γ$) for the elements Ag, Pb, and Bi.

<table>
<thead>
<tr>
<th>Element</th>
<th>$L_α/L_γ$ (1.5 MeV)</th>
<th>$L_α/L_γ$ (2.0 MeV)</th>
<th>$L_α/L_γ$ (3.0 MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag</td>
<td>16.1496</td>
<td>15.9305</td>
<td>14.6516</td>
</tr>
<tr>
<td>Pb</td>
<td>13.8441</td>
<td>12.6219</td>
<td>10.4588</td>
</tr>
<tr>
<td>Bi</td>
<td>13.9007</td>
<td>12.6556</td>
<td>11.0300</td>
</tr>
</tbody>
</table>

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

Figure 8: Variation of $L_α/L_β$ ratio with energy for Ag, Pb and Bi elements.
Element | $L_{\alpha}/L_{\gamma}$ (1.5 MeV) | $L_{\alpha}/L_{\gamma}$ (2.0 MeV) | $L_{\alpha}/L_{\gamma}$ (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 $L_{\alpha}/L_{\gamma}$ ratio with energy for Ag, Pb and Bi elements.

It is clear from our experimental and theoretical study that ratios $L_{\alpha}/L_{\beta}$ are dependent on projectile energy and ratios decreases with increase of projectile energy. Transition probability ratios $(L_{\alpha}/L_{\gamma})$ increases with the atomic number. Transition probability ratios $(L_{\alpha}/L_{\gamma})$ also decreases with increase of projectile energy and atomic number.

Figure 10: Variation of $L_{\alpha}/L_{\alpha}$ 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 Kα/Kβ 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 Kα/Kβ X-ray transitions-probability ratios are independent from nature of excitation mechanism. Figure 5 also shows that Kα/Kβ 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 Kα/Kβ 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 Lα/Lβ ratio of the elements Pb, Bi and Ag are 4-6%. The uncertainty for Lα/Lγ ratios are 10-15 % and in Lα/Lα ratios are 8-10% depending upon the counting statistics, incident particle energy and target thickness. For all the three elements L-X ray ratios Lα/Lβ, Lα/Lα and Lα/Lγ were measured and compared to ECPSSR theoretical intensity. Theoretical Lα/Lβ 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 Lα/Lβ is about 8% higher than those measured results [17]. For Lα/Lγ our results are 10% and 12% higher at 1.00 and 3.00 MeV, respectively. For the cases of Lα/Lβ and Lα/Lγ, 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 Lα/Lγ are independent from incident proton energy theoretically as well as experimentally for all three targets. In case of Bi, for Lα/Lβ 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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