Measurement of $K_\beta/K_\alpha$ values using proton beam

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Abstract. The $K_\beta/K_\alpha$ intensity ratios are measured in some 3$d$ shell elements by using a 2 MeV proton beam along with a high resolution Si(Li) detector. The present $K_\beta/K_\alpha$ intensity ratios are in good agreement with Scofield modified theoretical values, thus supporting the basic assumptions in that theory. From the present $K_\beta/K_\alpha$ intensity ratios, it is evident that due to chemical effects, the experimental $K_\beta/K_\alpha$ intensity ratios will be increased while they will be decreased due to the presence of simultaneous $M$-shell vacancies which are produced due to proton excitation.

Keywords. $K_\beta/K_\alpha$ intensity ratios; 2 MeV proton beam; Si(Li) detector; Scofield modified theory; chemical effects; multiple ionization phenomenon.

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1. Introduction

Study of the $K$ x-ray intensity ratios is of importance to understand the atomic inner shell ionization process and to test the relevant existing theories. The availability of highly sophisticated x-ray detectors with good energy resolution permits accurate measurement of x-ray intensities. The $K$-ray intensity ratios have been measured by several authors [1–28].

The $K$ x-ray intensity ratios due to different authors with different excitation modes are summarized in table 1. In the same table, the theoretical $K_\beta/K_\alpha$ intensity ratios due to Scofield’s old [29] and modified [30] theories are also given. From the table, it is observed that the $K_\beta/K_\alpha$ intensity ratios due to some of the authors [1,5,8,21,34] for some of the elements agree with Scofield’s old theoretical values [29] while the $K_\beta/K_\alpha$ intensity ratios due to some other authors agree with Scofield’s modified [30] theoretical values. However, limited experimental data on $K_\beta/K_\alpha$ intensities due to the elements $62 \leq Z \leq 82$ with proton excitation is available. Kasagi et al [11] have used 3.5 MeV protons as exciting agents and measured the $K_\beta/K_\alpha$ intensity ratios in the region of the elements $62 \leq Z \leq 82$ and found reasonable agreement with Scofield modified [30] theoretical values. Richard et al [32] have used proton beam in
the energy range 6 to 10 MeV and studied the dependence of $K_{\beta}/K_{\alpha}$ intensity ratios with projectile energy. The $K_{\beta}/K_{\alpha}$ intensity ratios for Cu element obtained by them is lower than Scofield modified [30] theoretical values.

Benka [10] studied the energy dependence of $K_{\beta}/K_{\alpha}$ intensity ratios in Si element with different proton energies. They have observed that $K_{\beta}/K_{\alpha}$ intensity ratios will depend on proton energy. They have explained this energy dependence as due to the formation of simultaneous $3p$ ($M$-shell) vacancies in addition to a vacancy in the $1s$ ($K$-shell) shell (multiple ionization phenomenon). Li et al [15] have measured the $K_{\beta}/K_{\alpha}$ intensity ratios in some 3d shell elements using charged particle excitation. The $K_{\beta}/K_{\alpha}$ intensity ratios obtained by them due to carbon ions are also given in table 1. Their values due to carbon ions are much higher than Scofield’s [29,30] theoretical values and also the experimental values due to other excitation modes.

Brunner et al [41], Kucukonder et al [34], Tamaki et al [36], Mukoyama et al [7], Kataria et al [35] and Raghaviah et al [42] have studied the effect of chemical environment on $K_{\beta}/K_{\alpha}$ intensity ratios in some 3d shell elements. They have observed that the $K_{\beta}/K_{\alpha}$ intensity ratios are enhanced by about 4 to 5% due to the chemical effects.

In the present work, the $K_{\beta}/K_{\alpha}$ intensity ratios have been measured in some 3d shell elements using 2 MeV proton beam. The motivation of the present work is (1) to verify

<table>
<thead>
<tr>
<th>Element</th>
<th>Present (proton)</th>
<th>Early authors</th>
<th>$K_{\beta}/K_{\alpha}$ (experimental)</th>
<th>$K_{\beta}/K_{\alpha}$ (theory)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca</td>
<td>0.131 ± 0.003</td>
<td>0.069 [21] (RD), 0.128 [20] (X), 0.128 [31] (SE), 0.175 [15] (C)</td>
<td>0.106</td>
<td>0.131</td>
</tr>
<tr>
<td>Ti</td>
<td>0.134 ± 0.003</td>
<td>0.064 [21] (RD), 0.123 [1] (RD), 0.103 [34] (P), 0.132 [20] (X), 0.133 [1] (X), 0.134 [31] (SE), 0.162 [15] (C)</td>
<td>0.114</td>
<td>0.125</td>
</tr>
<tr>
<td>V</td>
<td>0.135 ± 0.003</td>
<td>0.105 [21] (RD), 0.121 [1] (RD), 0.148 [4] (P), 0.134 [1] (X), 0.128 [8] (X), 0.156 [15] (C)</td>
<td>0.116</td>
<td>0.137</td>
</tr>
<tr>
<td>Cr</td>
<td>0.136 ± 0.003</td>
<td>0.113 [21] (RD), 0.127 [1] (RD), 0.131 [36] (RD), 0.112 [5] (P), 0.135 [20] (X), 0.134 [1] (X), 0.132 [8] (X), 0.135 [31] (SE), 0.152 [15] (C)</td>
<td>0.115</td>
<td>0.134</td>
</tr>
<tr>
<td>Mn</td>
<td>0.136 ± 0.003</td>
<td>0.122 [21] (RD), 0.129 [24] (P), 0.134 [25] (P), 0.115 [5] (P), 0.137 [35] (P), 0.138 [23] (P), 0.135 [7] (X), 0.137 [35] (X), 0.153 [15] (C)</td>
<td>0.119</td>
<td>0.138</td>
</tr>
<tr>
<td>Fe</td>
<td>0.135 ± 0.003</td>
<td>0.128 [21] (RD), 0.129 [1] (RD), 0.116 [34] (P), 0.136 [20] (X), 0.125 [1] (X), 0.133 [8] (X), 0.135 [31] (SE), 0.153 [15] (C)</td>
<td>0.121</td>
<td>0.139</td>
</tr>
<tr>
<td>Zn</td>
<td>0.139 ± 0.003</td>
<td>0.135 [21] (RD), 0.136 [1] (RD), 0.134 [24] (P), 0.139 [25] (P), 0.137 [1] (X), 0.151 [18] (X), 0.138 [8] (X), 0.133 [33] (E), 0.135 [13] (E), 0.138 [31] (SE), 0.164 [15] (C)</td>
<td>0.124</td>
<td>0.141</td>
</tr>
</tbody>
</table>

RD: radioactive decay; P: photons; X: x-rays; E: electrons; SE: semi-emperical (Salem); C: carbon ions.
Table 2. List of compounds used in the present work.

<table>
<thead>
<tr>
<th>Element</th>
<th>Compound</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca</td>
<td>CaCO₃</td>
</tr>
<tr>
<td>Ti</td>
<td>TiO₂</td>
</tr>
<tr>
<td>V</td>
<td>V₂O₅</td>
</tr>
<tr>
<td>Cr</td>
<td>K₂Cr₂O₇</td>
</tr>
<tr>
<td>Mn</td>
<td>MnSO₄</td>
</tr>
<tr>
<td>Fe</td>
<td>FeSO₄</td>
</tr>
<tr>
<td>Zn</td>
<td>ZnSO₄</td>
</tr>
</tbody>
</table>

which Scofield theoretical values will explain the $K_β/K_α$ intensity ratios due to proton excitation; (2) to observe the possible chemical effects; (3) whether multiple ionization takes place with proton excitation.

2. Experimental details

In the present work, $K_β/K_α$ intensity ratios were measured for elements Ca, Ti, V, Cr, Mn, Fe and Zn using a 2 MeV proton beam. The elements used in the present work are taken in the form of their compounds. The chemical compounds of different elements that are used in the present work are shown in table 2. Targets of different elements are prepared using dipping technique. The thickness of different targets range from 2 mg/cm² to 3 mg/cm².

The present experiments were carried out using 3 mV tandem pelletron accelerator facility at the Institute of Physics, Bhubaneswar. The collimated beam of protons of diameter 1.5 mm is directed on to the target. The target is kept at an angle of 45° to the beam direction. The emitted $K_x$-x-rays are passed through a 3.5 mg/cm² mylar window, 5 cm air gap and 0.012 mm thick Be window and reach the Si(Li) detector. The Si(Li) detector is kept at an angle of 90° to the beam direction. The resolution of the Si(Li) detector is 160 eV (FWHM) at 5.9 keV photon energy.

The efficiency of the Si(Li) detector at different energies is calculated using $^{241}$Am, $^{55}$Fe, $^{57}$Co and $^{137}$Ba radioactive sources. The efficiency of the detector is also calculated theoretically as described by Padhi et al [37]. The following expression is used to calculate the intrinsic efficiency

$$e_δ = e^{-\mu_B X_{Be} + \mu_A X_{Au} + \mu_{Si} X_{Si}} (1 - e^{-\mu X_{Si}}),$$

where $\mu$ is the absorption coefficients due to Be window of the detector. The thickness of the Au layer on the Si(Li) crystal at the given energy is $X_{Au}$ and $\Delta X_{Si}$ is the thickness of the insensitive region of the Si(Li) crystal. The $X$'s are as per specification of the detector manufacturer and the absorption coefficients are taken from the tables of Hubbell et al [38]. The values of $X$ used in the present calculation are given in table 3. The detector efficiency is calibrated at different energies. The resulting efficiency curve as obtained is shown figure 1.

Each target is exposed to proton beam and $K$-x-rays spectrum is collected for sufficient long time. The time of collection is such that the uncertainty in counting statistic is less than 1%. The $K$-x-ray spectrum of Fe element recorded with proton beam is shown in figure 2.
Table 3. X-values used in the efficiency equation of the detector.

<table>
<thead>
<tr>
<th>Element</th>
<th>X-value (gm/cm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>X$_{He}$</td>
<td>0.002253</td>
</tr>
<tr>
<td>X$_{Au}$</td>
<td>0.000038622</td>
</tr>
<tr>
<td>$\Delta$ X$_{Si}$</td>
<td>0.000023212</td>
</tr>
<tr>
<td>X$_{Si}$</td>
<td>0.69636</td>
</tr>
</tbody>
</table>

Figure 1. The efficiency curve of the Si(Li) detector.

3. Data analysis

The areas under the $K_\alpha$ and $K_\beta$ x-ray components are calculated for each element by using the 'analytical x-ray analysis by iterative least square' (AXIL) software program. Using the detector efficiency values, the areas under the $K_\alpha$ and $K_\beta$ x-ray components are converted to the respective intensities using the relation

$$I_{K_\alpha} / I_{K_\beta} = (A_{K_\beta} / A_{K_\alpha}) / (\varepsilon_{K_\beta} / \varepsilon_{K_\alpha}),$$

where $I_{K_\alpha}$ and $I_{K_\beta}$ are the intensities of $K_\alpha$ and $K_\beta$ x-ray components. $A_{K_\alpha}$ and $A_{K_\beta}$ are the areas under $K_\alpha$ and $K_\beta$ x-ray components, $\varepsilon_{K_\alpha}$ and $\varepsilon_{K_\beta}$ represent the efficiency values corresponding to $K_\alpha$ and $K_\beta$ x-ray energies. The efficiency values are taken from figure 1. The $K_\alpha$ and $K_\beta$ intensity values thus obtained are without self-absorption correction. The intensities are corrected for self-absorption using the formula...
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![Graph showing the $K\alpha$ x-ray spectrum of Fe due to proton beam.](image)

*Figure 2. K x-ray spectrum of Fe due to proton beam.*

$$I = I_0[1 - e^{-\mu t}]$$

Here $I_0$ represents the intensity of x-rays after absorption correction, $t$ represents the thickness of the target and $\mu$ represents the attenuation coefficient. These attenuation coefficients are calculated using the total cross sections given in the tables of Storm and Israel [39]. In the present work, since compounds are used, the attenuation coefficient corresponding to the respective compound is evaluated using 'sum rule' [40].

4. Results and discussion

The $K_\beta/K_\alpha$ values obtained in the present work with 2 MeV proton beam for different elements are given in table 1. The present experimental values are associated with an overall uncertainty of about 2.2% which is contributed cumulatively by individual uncertainty due to detector efficiency, counting statistics and self-absorption correction.

From table 1, it is observed that the $K_\beta/K_\alpha$ values obtained in the present work did not support Scofield theoretical calculations [29] while they support Scofield modified theoretical values [30]. In Scofield modified version [30], the radial wave functions of all the single particle states of a given sub-shell are assumed to be identical. He has included the exchange correction in the calculation of decay rates. In modified version, a separate relativistic HF calculation is considered for the initial state with a vacancy in the 1s sub-shell and for final state with a vacancy in a p sub-shell.

The effect of chemical environment on $K_\beta/K_\alpha$ has been studied by several authors [1,5,8,21,34] in 3d-shell elements. Their results indicated that the $K_\beta/K_\alpha$ intensity ratios of the elements when they are in compound form are 4–5% higher than those for pure elements as well as Scofield modified theoretical values [30]. In the present work, the elements are taken in the compound form and due to chemical effects, it is expected that the present $K_\beta/K_\alpha$ intensity ratios may be higher than Scofield theoretical values and other earlier data by about 4 to 5%. But, the present experimental values are in close agreement with Scofield theoretical values [30]. This may be due to the multiple ionization taking place in the target atoms while using protons as exciting agents.

Benka [10] has explained the variation of $K_\beta/K_\alpha$ intensity ratios with proton energy as due to the formation of simultaneous M-shell vacancies in addition to a K-shell vacancy.
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Li et al [15] calculated the functional dependence of $K_\beta/K_\alpha$ intensity ratios on the initial inner shell vacancy configuration. From their calculations, it is noticed that the $K_\beta/K_\alpha$ intensity ratio is a strongly increasing function of the simultaneous L-shell vacancies and a smoothly decreasing function of simultaneous M-shell vacancies. In the present work, since protons are used as projectiles, as mentioned by Benka [10] simultaneous M-shell vacancies may be produced and according to Li et al [15], these simultaneous M-shell vacancies will reduce the experimental $K_\beta/K_\alpha$ intensity ratios.

In the present work, since compounds are used, due to chemical effects, the experimental $K_\beta/K_\alpha$ intensity ratios may be higher than the normal values, while the experimental $K_\beta/K_\alpha$ intensity ratios may simultaneously be reduced due to multiple ionization effects. Hence, the present $K_\beta/K_\alpha$ intensity ratios may be expected to be in agreement with Scofield theoretical values as well as earlier experimental values which is evident from table 1.

5. Conclusion

In the present work, the $K_\beta/K_\alpha$ intensity ratios are measured in some 3d shell elements by using a 2 MeV proton beam. From the present results, the following are the conclusions:

1. The present $K_\beta/K_\alpha$ intensity ratios are in good agreement with Scofield modified theoretical values thus supporting the basic assumptions of that theory.

2. From the present $K_\beta/K_\alpha$ intensity ratios, it is evident that due to chemical effects, the experimental $K_\beta/K_\alpha$ intensity ratios may be increased while they may be decreased simultaneously due to the presence of simultaneous M-shell vacancies that are produced due to proton excitation.

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References


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