

Total to K -shell photoelectric cross section ratios in Zr, Ag, Ta, and Th*

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Abstract. Total photon atomic cross sections in elements Zr, Ag, Ta and Th were determined around their K -edges in the energy region 6 to 400 keV with a good geometry set-up using proportional counter or Ge(Li) detector system. From these values the photoelectric cross sections were obtained by subtracting the theoretical values of coherent and incoherent scattering cross sections. The resulting photoelectric cross sections were fitted to curves above and below the K -edge for each element and extrapolated on either side and the total to K -shell photoelectric cross section ratios were determined with an error not exceeding 2%. The ratios were compared with the theoretical values obtained by Grodstein and others, as also from the empirical relation of Hubbell. The present values show good agreement with those of Scofield. A definite trend of decrease of the ratio as the energy increases is observed in the case of Th unlike in the previous experimental studies.

Keywords. Photoelectric cross sections; good-geometry; transmission experiment.

1. Introduction

Atomic photoeffect is the predominant process of interaction of photons with matter in the low energy region. The other two competing processes in this region are coherent and incoherent scattering. Although extensive investigations have been carried out on atomic photoeffect (Pratt *et al* 1973) there are still some uncertainties in the existing data on the relative magnitudes of the shell-wise cross sections. In the early investigations a 'five-fourth's law' was used for the total to K -shell cross section ratio (T/K). Based on Stobbe's (1930) formula, Grodstein (1957) reported both energy and atomic number dependences of the ratio. Later Rakavy and Ron (1965), Schmickley and Pratt (1967) and Scofield (1973) showed a definite energy and atomic number dependence of the ratio. From a direct determination of relative photoelectron intensities, using magnetic spectrometers, this ratio was experimentally determined by Hultberg (1959) and Grigorev and Zolotavin (1959). The sedata indicated no energy dependence of the ratio T/K . Davisson (1965) reported the values of the ratio at K -edges employing the data of Kirchner (1930). These values are smaller than those measured at higher energies, contrary to the recent theoretical

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predictions of Scofield (1973) and Storm and Israel (1970), where a decrease of the ratio with the increasing energy is expected. For these reasons we undertook accurate measurements of the ratio T/K for different elements near their K -edges.

2. Experimental

There are essentially three methods suitable for the measurement of total to K -shell (T/K) photoelectric cross section ratios near K -edges: (1) Measurement of the total photon cross sections and extrapolation of the extracted photoelectric cross sections towards K -edges. (2) Measurement of the K to total x-ray yields following the photoelectron emission. (3) Measurement of the emitted photoelectron intensities. The second method is a bit complicated and involves many errors due to various factors like self absorption, errors in the reported fluorescent yields and other corrections. Hence the resultant errors in the ratio are usually large. The third method is unsuitable as it involves the measurement of very low energy electron intensities in the low energy photon region. On the other hand, the first method is simple inasmuch as the total photon attenuation coefficients can be measured very accurately using suitable radioactive sources and high resolution detectors (Sivasankara Rao *et al* 1973, Lakshminarayana and Jnanananda 1961, Premachand *et al* 1976, Reddy *et al* 1976, McCrary *et al* 1967). Hence in the present investigations the ratios (T/K) are measured using the first method.

For the measurement of the total photon attenuation coefficients a good geometrical set-up is used, the suitability of which is already well discussed earlier (Davisson and Evans 1952; Lakshminarayana 1961; Premachand *et al* 1976; Reddy *et al* 1976). Photon attenuation coefficients were measured by conducting the transmission experiments on the good geometry set-up in the energy region 6 to 400 keV at suitable energies in elements Zr, Ag, Ta and Th adopting the usual procedure. Three detectors—an argon proportional counter (6–15 keV), a krypton proportional counter (15–40 keV) and a coaxial 35 cc Ge (Li) detector (40 to 400 keV), equipped with a Nuclear Data 512 Channel Analyser were used in the present investigations. A typical geometrical set-up is shown in figure 1. The graded structure of collimators and

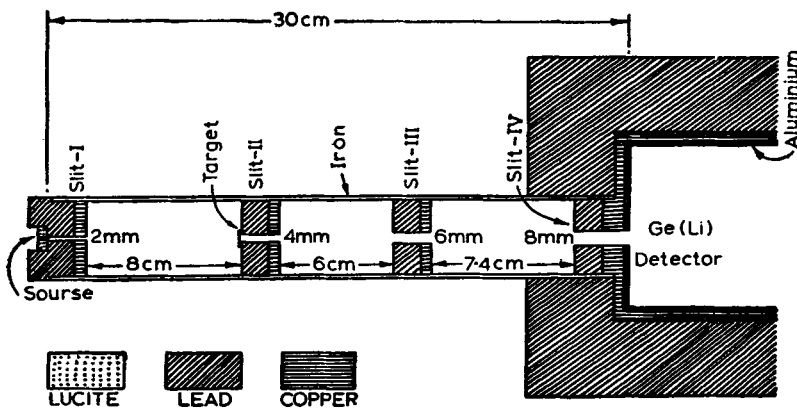


Figure 1. Geometrical set-up of Ge(Li) detector.

Table 1. List of radioactive isotopes used in the present work

| S. No. | Isotope | Half-life | Energies of photons selected in keV | |
|--------|-------------------|-----------|--------------------------------------|---|
| | | | X-rays* | Gamma Rays ^c |
| 1. | ⁵⁵ Fe | 2.6 Y | 5.959(K _{αβ}) ^a | — |
| 2. | ⁶⁵ Zn | 245 D | 8.136(K _{αβ}) ^a | — |
| 3. | ⁷⁵ Se | 120 D | 10.532(K _α) ^a | 121.11, 135.97, 264.67, 279.58 and 400.7†† |
| 4. | ⁸⁵ Sr | 64.5 D | 13.375(K _α) ^a | — |
| 5. | ⁹⁹ Mo | 67 H | 18.327(K _α) ^b | — |
| 6. | ¹¹³ Sn | 115 D | 24.138(K _α) ^b | — |
| 7. | ¹³³ Ba | 7.2 Y | 30.851(K _α) ^b | 81.00** |
| 8. | ¹⁶⁰ Tb | 72.1 D | 45.714(K _α) ^c | 86.79, 197.02, 215.64** |

(a) Argon counter detection, (b) Krypton counter detection, (c) Ge(Li) detection energies taken from:

*Storm and Israel (1970).

**Lederer *et al* (1967).

††Martin and Blichert-Toft (1970).

detector shielding are necessary to reduce the low energy background. The radioactive sources were obtained from Bhabha Atomic Research Centre, Bombay. These are listed in table 1, together with some important features. Circular discs of known diameter were cut for the foils of elements. These were weighed on a microbalance, and the weights were divided by the area of the foils to get the target thickness in gm/cm². The total photon atomic cross sections (μ_a) were estimated using the following expression developed by least square fitting (Lakshminarayana and Jnanananda 1961).

$$\mu_a = \frac{2.303 \sum_{i=1}^r \{\log(I_0/I_i)\} x_i}{\sum_{i=1}^r x_i^2} \cdot \frac{A}{N\rho}$$

where I_0 and I_i are the intensities without and with a foil of thickness x_i , A and ρ are the atomic weight and density of the absorber foil respectively. N is Avogadro's number and r refers to the number of different absorber foils employed for each element. For the small amount of coherent and incoherent scattering present in the observations, corrections were made using the tables of Brown (1966) and Storm and Israel (1970) employing the standard procedures.

Errors in the present experiment are due to (a) the statistics of counting (b) the interference of the fluorescent x-rays produced in the absorber foil (especially for thicker absorbers) due to the higher gamma energy photons present in the source when the cross sections for lower photon energies are measured, (c) the effect of K_β x-rays when they cannot be well resolved from the K_α x-rays due to poor resolution of the set up especially at lower transmissions (i.e. for thick absorbers), (d) uncertainties

Table 2. Total photon cross sections (Barns/atom) (error $\sim 1\%$)

| Energy (keV) | Zr | Ag | Ta | Th |
|--------------|-------|-------|-------|-------|
| 5.959 | 44760 | — | — | — |
| 8.136 | 19230 | 36910 | — | — |
| 10.532 | 9793 | 18630 | — | — |
| 13.375 | 5036 | 9700 | 53580 | — |
| 18.327 | 13520 | 4161 | 23810 | — |
| 24.138 | 6735 | 1970 | 11750 | 25710 |
| 30.851 | 3522 | 6114 | 6069 | 13780 |
| 45.714 | 1174 | 2162 | 2195 | 5105 |
| 81.0 | 252.8 | 460.3 | 2172 | 1178 |
| 86.79 | 210.1 | 375.6 | 1847 | 986.4 |
| 121.11 | — | 159.5 | 786.1 | 1603 |
| 135.97 | — | 120.0 | 586.0 | 1186 |
| 197.02 | — | — | 232.5 | 483.3 |
| 215.64 | — | — | 192.8 | 401.9 |
| 264.67 | — | — | — | 247.8 |
| 279.58 | — | — | — | 219.3 |
| 400.7 | — | — | — | 104.5 |

in the background and gaussian fitting of the photopeaks of the transmitted photons and (e) impurities in the foils. Of all, the errors arising out of (a) and (c) are considerable. The remaining three sources of errors are found to be negligibly small. The error contributions due to (a) and (c) are found to be of the order of 0.7% and 0.5% respectively. Hence the root mean squared total error is of the order of 0.8%. In no case it exceeded 1%. The total photon cross sections are given in table 2.

3. Results

In the present investigations, the photoelectric cross sections were obtained by subtracting from the observed values of total cross sections, the theoretical values of coherent and incoherent scattering cross sections reported by Storm and Israel (1970) at various energies for the different elements. The subtracted contribution in no case exceeded 20% of the total except at 400 keV. The resultant experimental photoelectric cross sections are given in table 3. The theoretical values are obtained by interpolation, at the required energies and elements, using an IBM 1130 Computer. The error in the interpolations is of the order of 0.05%. Errors in the theoretical coherent and incoherent scattering cross sections are not specified by Storm and Israel (1970) in their reported data. However, as already stated, since the subtracted contribution is less than 20%, this error affects the final cross section in a secondary way. Hence the error arising from subtraction is neglected. The error in the photoelectric cross sections reported in table 3 in no case exceeds 1% and represents the experimental error in the present study.

Using the data in table 3, the total to *K*-shell photoelectric cross section ratios are estimated as follows: The data above and below *K*-edge of each element were fitted to log-log plots of first or second degree polynomials by the method of least squares using an IBM 1130 computer. A typical plot in the case of Ag is shown in figure 2.

Table 3. Photoelectric cross sections (Barns/Atom) (error~1%)

| Energy (keV) | Zr | Ag | Ta | Th |
|--------------|-------|-------|-------|-------|
| 5.959 | 44246 | — | — | — |
| 8.136 | 18826 | 36346 | — | — |
| 10.532 | 9474 | 18181 | — | — |
| 13.275 | 4787 | 9343 | 52590 | — |
| 18.327 | 13344 | 3911 | 23120 | — |
| 24.138 | 6611 | 1791 | 11250 | 24890 |
| 30.851 | 3429 | 5983 | 5702 | 13178 |
| 45.714 | 1115 | 2079 | 1977 | 4747 |
| 81.0 | 219.1 | 415.8 | 2068 | 1016 |
| 86.79 | 178.3 | 333.9 | 1751 | 838.4 |
| 121.11 | — | 128.0 | 720.9 | 1506 |
| 135.97 | — | 91.0 | 527.8 | 1101 |
| 197.02 | — | — | 190.2 | 424.7 |
| 215.64 | — | — | 153.4 | 350.8 |
| 264.67 | — | — | — | 202.3 |
| 279.58 | — | — | — | 175.8 |
| 400.7 | — | — | — | 70.8 |

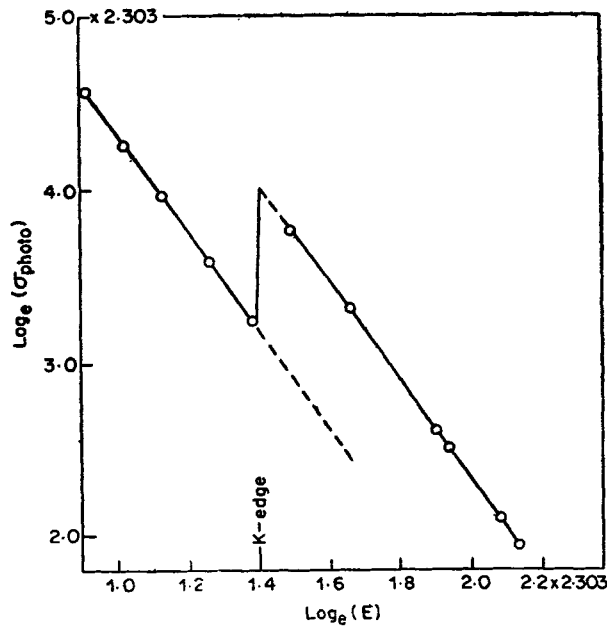


Figure 2. Least square fitted curves using below and above *K*-edge data of silver.

The plots below and above the *K*-edge were extrapolated towards *K*-edge (represented by broken line). The difference of the two plots at any energy, above the *K*-shell binding energy therefore directly gives the *K*-shell cross section at that energy. Because of the involved extrapolations, great care was taken to see that sufficient number of experimental points (5 to 7) were included on both sides of the *K*-edge covering adequate energy range. A simple test for the justification of the extrapolation was made as follows:

The experimental points of Th in the energy range 121 to 215 keV were fitted to a smooth curve and the values at 264.67 keV, 279.58 keV and 400.7 keV were obtained by extrapolation. These are found to be in satisfactory agreement with the experimental values within the experimental errors (i.e. $\sim 1\%$). The energy range used for curve fitting is only 100 keV and the extrapolations are made beyond the experimental range. The extrapolations up to two times the energy range, used for curve fitting are found to be accurate within 1% . Hence in the present investigation the extrapolation was made up to two times the experimental energy range. T/K ratios were obtained by dividing the total photoelectric cross section with the corresponding K -shell cross section. As the cross sections were fitted to curves above and below the K -edge and using the difference of values at each energy the T/K ratios were deduced, the error in the final ratios is of the order of $1.5 (\sqrt{1^2+1^2}\%)$. Hence an error of 2% is considered, for discussion.

4. Discussion

The T/K ratios at K -threshold are compared with different theoretical estimates. Hubbell (1969) fitted all earlier experimental ratios of T/K to an expression given by

$$T/K = 1 + 0.01481 \ln^2 Z - 0.000788 \ln^3 Z.$$

The values estimated using this expression, are furnished in table 4. It can be seen that these values agree fairly well with the present experimental values for Zr and Ag, while the values are lower than the present experimental values in the case of Ta and Th. It can be seen that the values of Grodstein (1957) are consistently lower in all the cases. The values of Scofield (1973) and Storm and Israel (1970) are in satisfactory agreement with the present experimental values within the range of errors.

T/K ratios at higher energies together with the corresponding experimental values of Grigorev and Zolotavin (1959) are presented in the same table along with the theoretical values of Scofield (1973). It can be seen from the table that there is in general agreement, between theory and experiment within the range of errors. In the case of Th, there is some indication of the variation, as can be seen from the trend of the values after allowing for the experimental errors. The energy dependence of the T/K ratio is quite pronounced in Th.

5. Conclusion

Thus it is concluded that the T/K ratios at K -threshold of the four elements, reported for the first time in the present work, show disagreement with the reported values of Grodstein (1959) but confirm the theoretical predications of Scofield (1972) and Storm and Israel (1970). The dependence of the obtained T/K values on Z is found to obey the relation $T/K = 1 + 6.666 \times 10^{-3} Z - 8.068 \times 10^{-5} Z^2 + 4.816 \times 10^{-7} Z^3$. A trend of the decrease of the T/K ratio with energy in accordance with the theoretical predictions of Scofield (1973) is noticed, especially in Th unlike in the previous experimental studies (Hultberg 1957).

Table 4. Total to K-shell photoelectric cross section ratios (error ~ 2%)

| S. No. | Zirconium | | Silver | | Tantalum | | Thorium | |
|--------|--------------|------------|--------------|------------|--------------|------------|--------------|------------|
| | Energy (keV) | Experiment | Energy (keV) | Experiment | Energy (keV) | Experiment | Energy (keV) | Experiment |
| 1. | 17.96* | 1.171 | 25.49* | 1.181 | 67.588* | 1.247 | 110.25* | 1.296 |
| | | 1.174(S) | | 1.188(S) | | 1.188(S) | | 1.241(S) |
| | | 1.16 (H) | | 1.17 (H) | | 1.17 (H) | | 1.21 (H) |
| | | 1.17 (SI) | | 1.19 (SI) | | 1.24 (SI) | | 1.24 (SI) |
| 2. | 18.327 | 1.173 | 30.851 | 1.174 | 81.00 | 1.239 | 121.11 | 1.283 |
| | | 1.13 (G) | | 1.14 (G) | | 1.16 (G) | | 1.16 (G) |
| | | 1.15 (R) | | 1.18 (R) | | 1.23 (R) | | 1.23 (R) |
| | | 1.172(S) | | 1.177(S) | | 1.233(S) | | 1.233(S) |
| 3. | 24.138 | 1.158 | 45.714 | 1.157 | 86.79 | 1.231 | 135.97 | 1.279 |
| | | 1.160(S) | | 1.163(S) | | 1.229(S) | | 1.229(S) |
| 4. | 30.851 | 1.145 | 121 | 1.156(GZ) | 121.11 | 1.214 | 197.02 | 1.246 |
| | | 1.152(S) | | — | | 1.215(S) | | 1.246 |
| | | | | 136 | | | | 215.64 |
| | | | | 1.162(GZ) | | | | 411 |
| | | | | | | | 513 | |
| | | | | | | | 603 | |
| | | | | | | | 1.24(GZ) | |
| | | | | | | | 1.205(GZ) | |
| | | | | | | | 1.185(GZ) | |
| | | | | | | | — | |
| | | | | | | | — | |

*Experimental K-shell binding energy (Storm and Israel 1970).

(H) Hubbell's (1969) expression value.

(SI) Storm and Israel (1970) value.

(S) Scofield (1973) value.

(G) Grodstein (1959) value.

(R) Rakavy and Ron's (1965) value.

(GZ) Grigorev *et al* (1959) value (the error in the values of GZ is of the order of 3%. Hence the energy dependence is not quite clear).

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