6th Conference on Nuclear and Particle Physics
17-21 Nov. 2007 Luxor, Egypt

EMPIRICAL K -SHELL IONIZATION CROSS SECTIONS OF ELEMENTS FROM $_4$Be TO $_{92}$U FOR PROTON IMPACT

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Abstract

Experimental ionization cross sections for K-shell by protons for elements from Be to U available in the literature from 1953 to 1999 are fitted to deduce the empirical K-shell ionization cross sections, 5400 values of cross sections have been collected from various references. The deduced empirical cross sections are compared to the experimental data and to the theoretical values obtained by our research group within the ECPSSR theory and also with the reference ionization cross sections obtained by Paul and Sacher [H. Paul and J. Sacher At. Data. Nucl. Data. Tables 42 (1989) 105].

Keywords : *K-shell ionization cross sections; fitting; empirical cross sections.*

1. INTRODUCTION

During the last decades, considerable efforts have been directed to the study of inner-shell ionization by charged particles. Many different theories have been developed in a attempt to explain the ionization cross sections obtained from experiments. Among them, the ECPSSR theory [1-3], is the one being tested by a largest body of experimental data now available for light ions. As a result, we have much more data available from K and L-shell ionization cross sections for proton impact compared with those of 30 years ago, when Johansson and Johansson [4] derived their analytical formulas fitted to the K and L-shell ionization cross sections. At that time, only total L shell ionization cross section data were available, so they only derived the fitting formulas for the total ionization cross section. After, several authors have tried to perform fittings of the available experimental data with analytical functions and thus some contributions have been reported essentially for the L-shell ionization cross sections. The first contribution which modelled the ionization cross sections of the three L subshells is that of Miyagawa et al. [5]. Later, Sow et al. [6] reported new parameters for the calculation of the L subshell ionisation cross sections. Further, Orlic et al. [7] from the same research group reported empirical formulas for the calculation of empirical ionization cross sections for protons. An other major contribution is the one reported by Reis and Jesus [8] who tried to calculate semi-empirical L X-ray production cross sections by the normalization of experimental cross sections with the corresponding theoretical values calculated within the ECPSSR theory. After, Strivay and Weber [9] have reported empirical formulas based on the direct fitting of experimental L X-ray production cross sections. Later, our research group has reported "reference" Li subshell ionization cross sections for elements with $71 \leq Z \leq 80$ for protons of 0.5 to 3.0 MeV [10] and semi-empirical and empirical L X-ray production cross sections for elements with $50 \le Z \le 92$ for protons of 0.5 to 3.0 MeV [11]. For the K shell ionization cross section, Paul and Sacher [12] presented the so-called "reference" K shell ionization cross sections by proton impact for most of the elements and in a board projectile energy range. These reference cross sections are based on the fitting of the experimental data normalized to the theoretical values calculated within the ECPSSR theory. In their work on 1989, Paul and Sacher used 4000 experimental data points for fitting. Since than a large number of experimental data is become available calling for a new fitting. Then, we used in this work 5402 experimental data points collected from various references. All targets from $_4$Be to $_{92}$U are covered for all the proton energy ranges available. The results are compared, for each Z-group, with the theoretical ones from the ECPSSR theory and with those reported by Paul and Sacher [12].

2. DATA ANALYSIS

Experimental K-shell ionization cross sections were tabulated by Rutledge and Watson [13] as early as 1973. Gardner and Gray [14] tabulated all experimental Auger, X-ray production cross sections from 1973 to 1977. They chose to tabulate only directly experimentally determined quantities to facilitate the comparison of data with the theory. Since 1977, Paul and co-workers are the main research group which has interested to the tabulating of the K-shell ionization cross sections. The bulk of their work is summarized in an extensive report by Paul and Muhr [15] in 1986. In the present work, the database used for the calculation of empirical K-shell ionization cross sections for protons relies on different compilations. The first compilation is that of Heitz et al. [16] of experimental data published before 1981. The second compilation is that of Lapicki [17] regrouping the experimental K X-ray production cross sections for protons and helium ions on target atoms from beryllium to uranium published up to January 1988. The third compilation is due to Paul et al. [12,18] consisting on two tables regrouping the experimental data published before 1991. The first table [12] comprises 4300 values for protons and the second about 184 values. In addition to these compilations, we extracted an important number of data from other works published from 1992 to 1999 [19-23]. So, we have to our disposal a database consisting of a total of 5402 experimental data. The distribution of these data according to the target atomic number is presented in Figure 1.

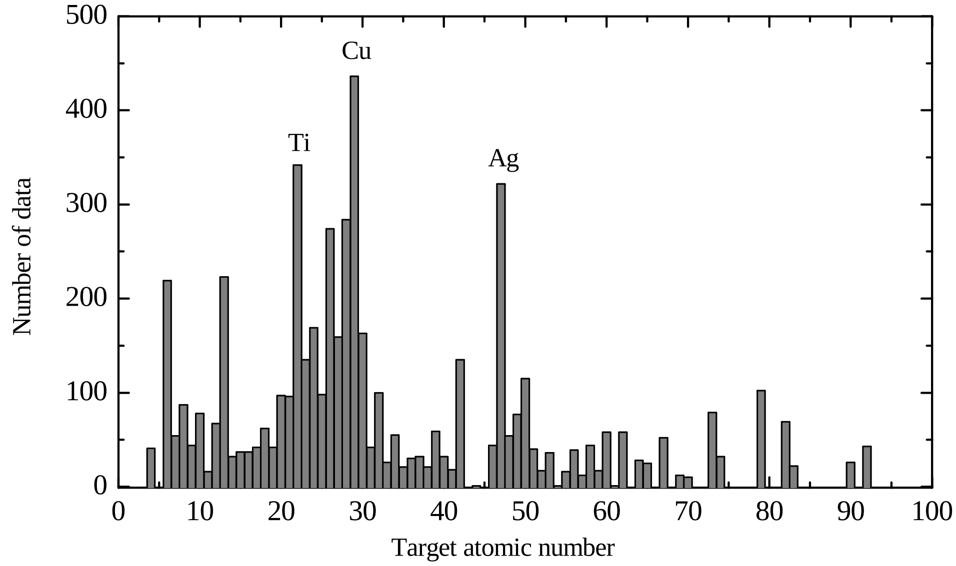

Figure 1 Distribution of experimental K-shell ionization cross sections by proton impact versus target atomic number

It can be seen from Figure 1 that:

(i) The most exploited targets for K-shell ionization cross section studies are metallic targets in the $20 \prec Z \prec 30$ region such as Ti, Fe, Ni and Cu and for Ag.
(ii) No experimental data are reported for elements with $84 \le Z \le 89$ due to the fact that they are difficult to handle and not readily available.
(iii) For the region $52 \le Z \le 92$, we have only a few number of experimental data because of the falling-down of the cross sections values (few millibarns or less).
For the rest of the target atomic numbers, it can be concluded that the experimental K-shell ionization cross section data are quite uniformly distributed.
The data are divided into nine Z-groups according to their target atomic numbers : $4 \le Z \le 10$, $11 \le Z \le 20$, $21 \le Z \le 30$, $31 \le Z \le 40$, $41 \le Z \le 50$, $51 \le Z \le 60$, $61 \le Z \le 70$, $71 \le Z \le 80$, and $81 \le Z \le 92$. It well be noted that each group will have a number of experimental data which should be sufficient to produce quite reliable fitting results within the group. For a given projectile, the following proportionality $\sigma U_K^2 \propto Z_1^2((m/M)(E/U_K))^4 = Z_1^2 f(E/\lambda U_K)$ is established, where λ is the ratio of proton mass to electron mass., m and M the mass of the electron and of the projectile, $U_K = Z_2^2$ Rydberg the binding energy of the target electron in the specific shell and E the projectile energy, Z_1 and Z_2 the atomic numbers of the projectile and of the target atom respectively, σ the ionization cross section of any atomic shell given by Merzbacher and Lewis [24]. Thus a plot of $\sigma U_K^2 / Z_1^2$ vs $E/\lambda U_K$ should yield the same results for all target atoms. This scaling law is in good agreement with results of Garcia et al. [25] who established that the K-shell cross section measurements for protons are well presented in terms of a “universal” curve. In this work, we calculated the empirical cross sections in the same manner used by Garcia et al. [25]. Having in mind that the experimental data were extracted from different references and hence measured under different experimental conditions, making the fitting of σU_K^2 vs $E/\lambda U_K$ in this situation would present large discrepancies between the empirical values and the experimental ones because of the spread of the various experimental data. So, in order to perform a suitable fitting and to deduce reliable empirical cross sections, the experimental data are normalized to their corresponding theoretical values by calculating the ratios $\sigma_{exp}/\sigma_{ECPSSR}$ where σ_{ECPSSR} refers to our theoretical cross sections calculated within the ECPSSR theory based on the approach of Cohen et al. [26]. Then, a criterion rejection is adopted by considering only the experimental data for which the ratio $\sigma_{exp}/\sigma_{ECPSSR}$ varies within the range of $0.5 - 1.5$. This criterion rejects 327 cross sections out of 5402 (about 6%). The analytical function used for the fitting is the following polynomial :

$$\ln\left(\sigma_{exp} U_K^2\right) = \sum_{j=0}^{n} A_j X^j \qquad (1)$$

where $X = \ln(E/\lambda U_K)$.

Each group was fitted with an analytical function to obtain a set of parameters.

The deviation of the cross sections from the corresponding fitted values is expressed in terms of the root-mean-square error (ε_{RMS}) calculated by using the following expression :

$$\varepsilon_{RMS} = \left[\sum_{j=1}^{N} \frac{1}{N} \left(\frac{\sigma_j(\text{data}) - \sigma_j(\text{fif})}{\sigma_j(\text{fif})} \right)^2 \right]^{\frac{1}{2}} \qquad (2)$$

where N is the number of data.

3. RESULTS AND DISCUSSION

The results of fitting for each Z-group are shown in Figures 2 (a) – (i). The dots are the experimental values and the curves are the fittings defined by Eq. (1). The parameters of these fittings are resumed in Table 1 where the fitting errors (ε_{RMS}) are also reported. We presented a figure for each Z-group in order to point out the different spread of the data in each case.

The examination of Figure 2 and Table 1 requires some comments:

- The scatter of the data in Figure 2 is partly due to the fact that the data taken from various references and consequently measured under different experimental conditions. However, though the figure is plotted in the "universal" scale, the scatter of the data is also due to some Z-dependence not taken into account in this work.
- It must be emphasized that the fittings given by Eq. (1) are not universal i.e. the coefficients reported in Table 1 are only valid in the energy ranges specified in Table 1. Any extension of the fittings outside the corresponding ranges might take unpredictable course and consequently erroneous cross sections.
- The Z-groups $4 \le Z \le 10$, $71 \le Z_2 \le 80$ and $81 \le Z_2 \le 92$ are considered as unfavourable cases as regards the numbers of rejected data which are 14%, 18% and 25% respectively as a consequence of the large spread of the data. The light targets $(4 \le Z_2 \le 10)$ deserve special consideration since here the L-shell is the valence shell, and chemical effects could occur. While these effects are note normally expected to influence ionization cross sections noticeably, they may well influence the fluorescence yields and hence the X-ray cross sections. For this Z-group, the data are generally X-ray production cross sections which we convert to K-shell ionization cross sections by using the fluorescence yields. The choice of the fluorescence yield does not have any influence on the final result because the ionization cross sections are solely based on Auger electron production measurements. On the other hand, it should be noted that the universal curve fitting for this Z-group is mostly determined by the experimental data for the carbon which represent 41% form the total number of data.
- The other Z-groups essentially $11 \le Z_2 \le 20$ and $21 \le Z_2 \le 30$ are favourable cases because they comprise the largest numbers of data which data are the most clustered. The Z-group $21 \le Z_2 \le 30$ comprises about 40% of the total number of data (2156 among 5402) and the universal curve depends strongly on the Cu data (436 among 2156).

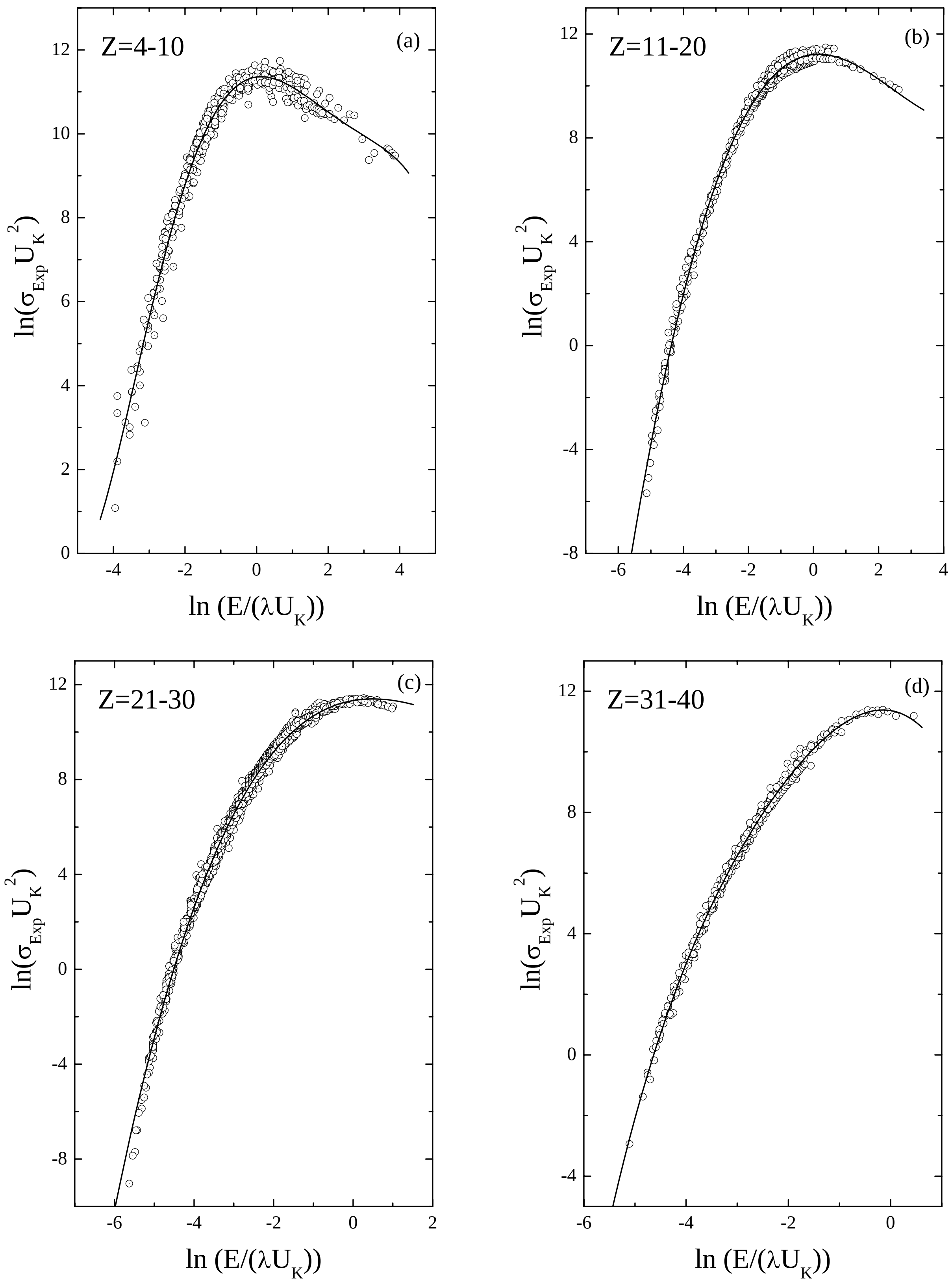

Figure 2 Plots of $\ln\left(\sigma_{Exp} U_K^2\right)$ as a function of the reduced proton energy $\ln\left(E/\lambda U_K\right)$ for each Z-group (a) – (i). The dots are experimental data. The curves are the fittings defined by Eq (1) and the coefficients listed in Table 1.

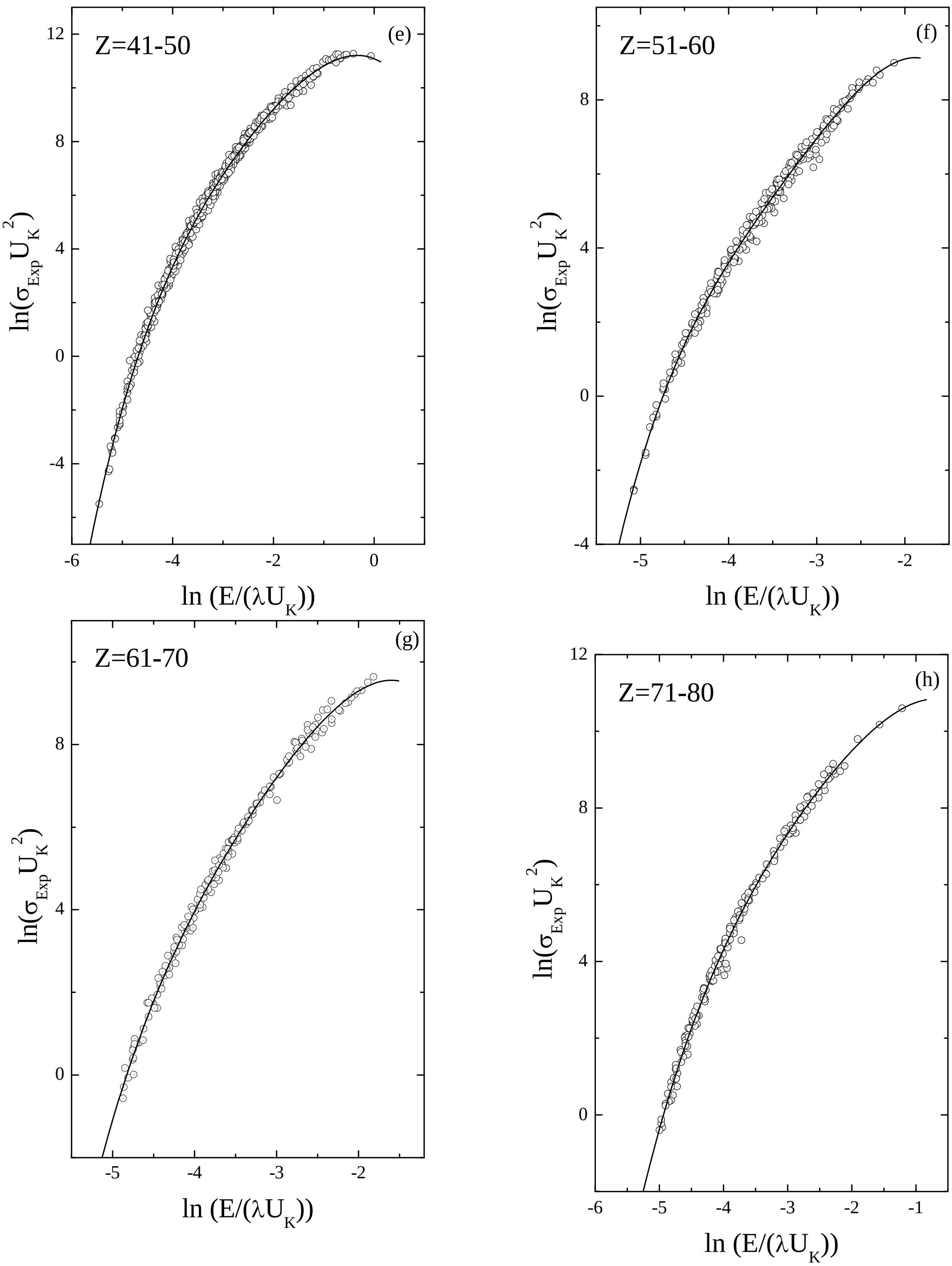

Figure 2 *(continued)*

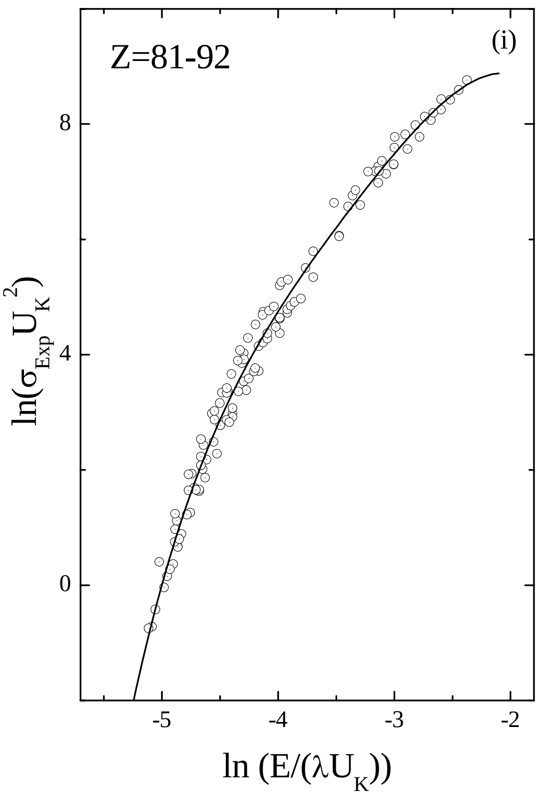

Figure 2 *(continued)*

Table 1: Fitting coefficients for the calculation of the empirical K-shell ionization cross sections for protons according to the Z-groups.

	K-Shell ionization cross sections				
Z- range	**4-10**	**11-20**	**21-30**	**31-40**	**41-50**
A_0	11.35397	11.20958	11.32958	11.36258	11.08617
A_1	0.11096	0.11958	0.28950	-0.27045	-0.75841
A_2	-0.44782	-0.39517	-0.32229	-0.92878	-1.26256
A_3	0.09074	0.04521	0.03767	-0.15441	-0.26433
A_4	0.00565	0.00177	-	-0.01742	-0.02929
A_5	-0.00251	-	-	-	-
ε_{RMS} (%)	22.6	17.4	17.9	14.1	16.3
Range of X	-3.95-3.83	-5.01-2.62	-5.48-0.88	-5.10-0.10	-5.26-0.06
No of data	531	655	2156	418	775
Rejected data	72	07	78	10	55

Z-range	51-60	61-70	71-80	81-92
A_0	-11.19842	2.03929	10.25152	-15.43859
A_1	-28.44505	-11.58286	-1.87635	-32.07755
A_2	-13.72839	-5.89107	-1.69380	-14.87613
A_3	-2.69603	-1.13487	-0.35176	-2.88539
A_4	-0.20262	-0.08900	-0.03461	-0.21395
ε_{RMS} (%)	18.5	15.2	14.8	19.6
Range of X	-5.07-2.12	-4.87-1.81	-4.99-1.21	-5.11-2.37
No of data	280	195	232	160
Rejected data	19	02	42	42

- For the heavy targets $61 \le Z_2 \le 70$, $71 \le Z_2 \le 80$ and $81 \le Z_2 \le 92$, the universal curve varied with an errors as 15-19%. The data for heavy atoms are less numerous and harder to measure; they seem to show large deviations from ECPSSR (see figure 3b). This will be discussed below.

We present, in Table 2, a comparison of the K-shell ionization cross sections from our ECPSSR calculations, our empirical values and the reference ones of Paul and Sacher [12] for one representative element from some selected Z-groups and for selected proton energies. In order to present the deviations between the results graphically, Figure 3 shows the evolution of the empirical K-shell ionization cross sections compared to the “reference” values reported by Paul and Sacher [12] as a function of the proton energy. All cross sections are normalized to their corresponding values from the ECPSSR theory. We note that the two methods used for the calculation of empirical (This work) and "reference" cross section [12] are different. The first method is based only of the experimental data while the reference cross section is based on both theoretical and experimental values. In spite of the relatively high number of experimental data used in this work (5402 data) by comparison with that used by Paul and Sacher [12] (4000 data), the results reported by the two approaches agree generally for proton energies above 3.0 MeV for all selected elements. At low proton energies, large discrepancies are observed between the two sets of results; the ratio σ/σ_{ECPSSR} tends less and less towards unity when going from high to low proton energy. For Au target at low proton energy, the ECPSSR prediction exceeds either the empirical value or the reference ones. For 0.7 MeV proton energy, the ratio is 0.53 for the empirical value and 0.19 for the reference value. We believe that the deviation is due essentially to a deficiency of the Coulomb correction [3]. Also, the measured K shell ionization cross section at 0.5MeV is only ~10^{-8} b for $_{79}$Au and at this energy on $_{29}$Cu the K-shell ionization cross section is nearly 1.6 b, some 10^{8} order of magnitude larger, so experimental uncertainties are highest for the $_{79}$Au and this is reflected in the huge spread of experimental data.

Table 2: Comparison of ECPSSR calculations, empirical K-shell ionization cross sections from this work, with the "reference" ones of Paul and Sacher [12] for selected elements.
First row : ECPSSR, second row : empirical calculation deduced from formula (1) and third row : Paul and Sacher [12].

K-Shell ionization cross section (barns)				
E (MeV)	Elements			
	$_{6}$C	$_{29}$Cu	$_{47}$Ag	$_{79}$Au
0.1	2.013 E+5	4.045 E-4	-	-
	1.883 E+5	3.528 E-4	-	-
	1.745 E+5	2.906 E-4	-	-
0.2	5.862 E+5	3.286 E-2	6.734 E-6	-
	5.925 E+5	2.138 E-2	7.049 E-6	-
	5.547 E+5	3.326 E-2	-	-
0.5	9.645 E+5	1.659 E+0	4.154 E-3	7.452 E-7
	1.059 E+6	1.463 E+0	3.415 E-3	6.910 E-7
	1.024 E+6	1.618 E+0	3.678 E-3	-
1.0	9.096 E+5	1.643 E+1	8.228 E-2	2.007 E-4
	9.704 E+5	1.592 E+1	8.012 E-2	1.065 E-4
	1.039 E+6	1.584 E+1	8.383 E-2	1.013 E-4
2.0	6.779 E+5	9.865 E+1	8.896 E-1	4.430 E-3
	6.898 E+5	9.384 E+1	8.494 E-1	3.453 E-3
	7.489 E+5	9.666 E+1	9.178 E-1	4.041 E-3
3.0	5.327 E+5	2.188 E+2	2.913 E+0	1.786 E-2
	5.389 E+5	2.053 E+2	2.645 E+0	1.631 E-2
	5.307 E+5	2.170 E+2	2.917 E+0	1.848 E-2
4.0	4.401 E+5	3.435 E+2	6.156 E+0	4.356 E-2
	4.514 E+5	3.226 E+2	5.445 E+0	4.213 E-2
	-	3.434 E+2	5.929 E+0	4.653 E-2
5.0	3.764 E+5	4.576 E+2	1.039 E+1	8.341 E-2
	3.949 E+5	4.334 E+2	9.129 E+0	8.213 E-2
	-	4.635 E+2	9.667 E+0	8.858 E-2
8.0	2.663 E+5	7.074 E+2	2.640 E+1	2.943 E-1
	3.028 E+5	6.976 E+2	2.401 E+1	2.865 E-1
	-	7.320 E+2	2.317 E+1	2.962 E-1
10	2.246 E+5	8.081 E+2	3.784 E+1	5.083 E-1
	2.687 E+5	8.204 E+2	3.571 E+1	4.874 E-1
	-	8.459 E+2	3.327 E+1	5.039 E-1

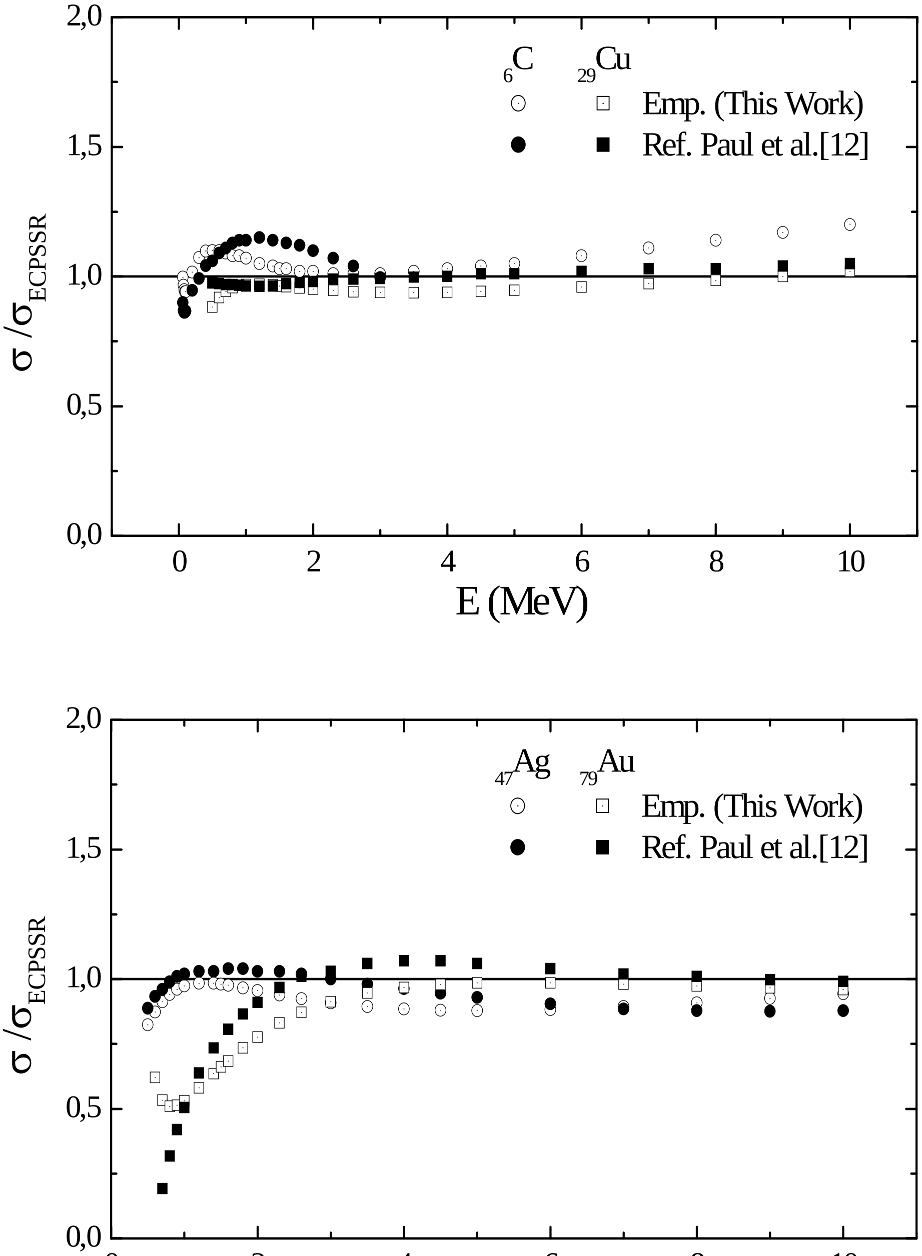

Figure 3 Empirical K-shell ionization cross sections from this work compared to those of Paul and Sacher [12] as a function of the proton energy for selected elements C, Cu, Ag and Au. All cross sections are normalized to their corresponding ECPSSR values (this work).

4. CONCLUSION

We reported in this contribution empirical ionization cross sections for elements with atomic number in the range 4 – 92 for proton impact. New fitting parameters are reported and are only valid in the region of the used experimental data. Because of the deviations observed between the theoretical values and the experimental data, the deduced empirical values should constitute a good compromise between the theory and the experiments and should be used in PIXE analysis. On the other hand, the comparison of our empirical K-shell ionization cross sections and the reference ones reported by Paul and Sacher shows a quite satisfactory agreement though the numbers of data used in the two approaches are different. However, some discrepancies are observed attesting that an analytical formula reproducing the experimental data is far to be achieved.

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