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HIGH ENERGY PIXE: NEW EXPERIMENTAL K-SHELL IONIZATION CROSS SECTIONS FOR SILVER AND GOLD AND COMPARISON WITH THEORETICAL VALUES FROM ECPSSR/RECPSSR MODELS

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In high energy PIXE, a high energy ion beam, up to several tens of MeV, is used to analyze heavy elements in a thick sample. The knowledge of the interaction cross-section with the atoms is essential to perform quantitative analysis. As measured data in this range of energy are scarce, an experimental campaign was conducted at the C70 cyclotron of the GIP ARRONAX to measure the K-X-ray production cross-sections for silver and gold with proton energies 30, 42, 54 and 68 MeV. The detector efficiency, the beam intensity, and the target thickness were measured accurately to produce precise data. To convert the K-X-ray production cross-sections into the K-shell ionization cross-sections, an average value of the K-shell fluorescence yield, calculated from a compilation of existing data, was used. The outcomes of the campaign are in good agreement with the few existing data and the theoretical prediction of the RECPSSR model.

1. INTRODUCTION

ARRONAX, acronym for «Accelerator for Research in Radiochemistry and Oncology at Nantes Atlantique», is a high energy cyclotron. It allows the acceleration of several types of particle beams: 68 MeV alpha particles, 15-35 MeV deuterons and 30-68 MeV protons [1]. An experimental platform was implemented on ARRONAX to perform non-destructive material analysis with X-ray emission induced by high energy beams (HEPIXE) [2, 3, 4]. The HEPiXE has already been used to analyze geological samples with 66 and 85 MeV protons [5] and art objects (old paintings and coins) with 68 MeV protons [6]. One of the benefits of HEPiXE is the significant increase of the K-lines X-ray production cross-sections for medium and heavy elements with a high energy proton beam. Then, the identification of these elements with their K-X rays is possible allowing the analysis in depth of thick samples particularly if the bulk composition is different from the composition of the surface [6]. The knowledge of K-shell ionization cross sections is necessary to perform quantitative analysis. Currently, experimental data available at high energy are scarce. Therefore, we started to measure the K-X-ray production cross-sections for Ag and Au elements like to complement databases and to validate theoretical models. The experiments were carried out using high energy proton beams varying between 30 MeV and 68 MeV. Our results were compared with the existing data and with theoretical models.

1.1. Modeling and experimental cross section data

The inner shell ionization process by incident charged particles is modeled by different theoretical approaches, namely the Plane Wave Born Approximation PWBA [7] and the ECPSSR [8]. The last one is an upgrade of the PWBA model by the incorporation of corrective effects. The ECPSSR accounts for the energy loss (E) of the incident particle, the Coulomb deflection (C) of the incident particle, the perturbed-stationary state (PSS) and the relativistic (R) effect of the inner shell electrons. In addition, the relativistic effect of high energy projectile can be included, the model becoming RECPSSR [9]. The ECPSSR/RECPSSR model calculations, presented below, were performed using the ISICS software [10]. Many experiments have been carried out for the K-X-ray production and K-shell ionization cross sections induced by low energy protons [11-20]. Several comparisons with the experimental data have shown that the ECPSSR is a successful model for the determination of the cross section by light ions in the MeV energy range [16-18].

The database used in the present work, for Ag and Au, relies on different compilations available at high energy in the literature [21-28]. In the case of silver (Fig. 1), the scattering of the data, associated with large error bars (above 15 MeV), makes the comparison with the ECPSSR and RECPSSR models unrealistic. In the case of gold (Fig. 2), the data have large error bars for energy above than 20MeV. Thus, it is not possible to

distinguish the best model describing the data between ECPSSR and RECPSSR.

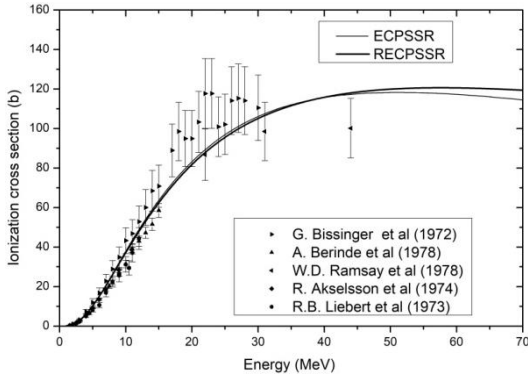


Figure 1 Experimental K-shell ionization cross section for silver with proton beam, compared to the ECPSSR/RECPSSR models.

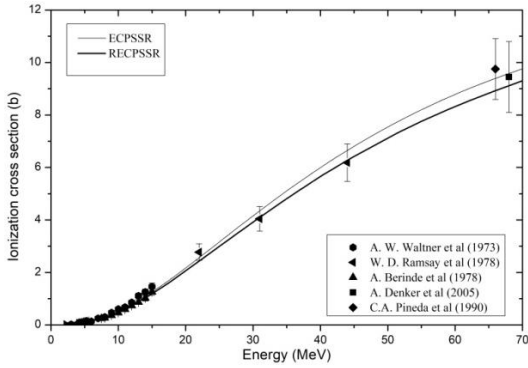


Figure 2 Experimental K-shell ionization cross section for gold with proton beam, compared to the ECPSSR/RECPSSR models.

2. MATERIALS AND METHODS

2.1. Experimental setup

The experimental setup is presented in the Fig. 3. After crossing a 75 μm Kapton window, the proton beam emerges in the air and passes through two Al collimator of 5 mm diameter. The target is located 30 cm downstream from the Kapton window. The beam energy loss in this air path is considered negligible. The X-rays, emitted from irradiated foils (Ag or Au), are detected normally to the target surface, placed at 45° with respect to the beam, by a shielded and cooled high purity germanium detector, HPGe (from Canberra, 50 mm^2 active area and 5 mm thick). The beam intensity is measured using a Faraday cup with a suitable electronics. The accuracy of the measured beam intensity is estimated around 1%. The beam intensity

was adjusted around 100 pA to keep a reasonable dead time, below 4%, in the detector. The typical irradiation time was several tens of minutes to insure good statistics. The different foils, dimensions 25 \times 25 mm^2 , were of high purity (99.9%) and supplied by GoodFellow. The different targets were 10.1 \pm 0.15 micrometer thick. Accurate values were obtained for each foil measuring their surface using a scanner (150 dpi) and their weight with a high precision scale (10^{-4} g).

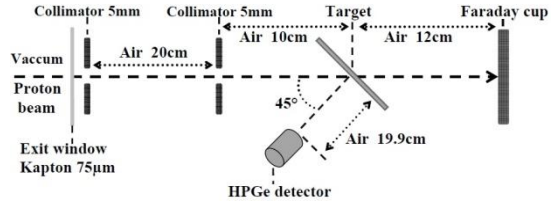


Figure 3 Schematic view of the experimental setup for cross section measurements

An efficiency model [29], optimized by the use of calibrated standard sources (^{241}Am , ^{109}Cd and ^{55}Fe), has been used to determine the detection efficiency for silver and gold K-X-rays. This model includes the intrinsic and the geometric efficiency of the detector. The errors on these efficiencies are below 3%.

2.2. X-ray spectrum

The table 1 presents the different allowed K_{α} and K_{β} transitions.

K_{α}	$K_{\alpha_1} = L_3 \rightarrow K$	
	$K_{\alpha_2} = L_2 \rightarrow K$	
K_{β}	K_{β_1}'	$K_{\beta_1} = M_3 \rightarrow K$
		$K_{\beta_3} = M_2 \rightarrow K$
	K_{β_2}'	$K_{\beta_5} = M_{4,5} \rightarrow K$
		$K_{\beta_2} = N_{2,3} \rightarrow K$
		$K_{\beta_4} = N_{4,5} \rightarrow K$

Tableau 1 K_{α} and K_{β} transitions

The graph (Fig. 4) shows two X-ray spectra obtained from irradiation of respectively Ag and Au targets with a 30 MeV proton beam. In the case of silver, the detected transitions are K_{α} , K_{β_1}' and K_{β_2}' and in case of gold, the detected transitions are K_{α_1} , K_{α_2} , K_{β_1}' and K_{β_2}' (Fig. 5). The different transitions listed in table 1 cannot be distinguished in the detected spectra due to the detector energy resolution.

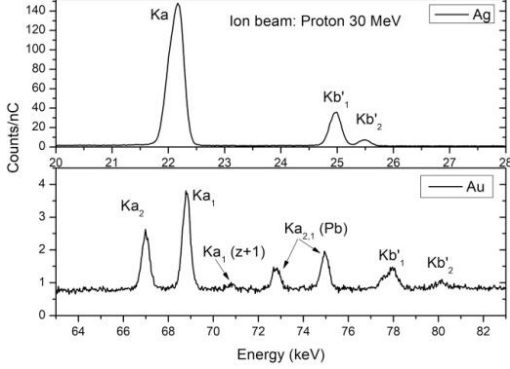


Figure 4: X-ray spectra obtained from the bombardment, with 30 MeV protons, of a silver target (top) and a gold target (bottom). Subscript a and b means alpha and beta respectively.

In the gold spectrum, the small peak, $K_{\alpha 1}(Z+1)$, comes from the internal conversion of the residual atoms called "nuclear satellites" produced by the (p, xn) nuclear reactions [22]. The position of the $K_{\alpha 2}(Z+1)$ peak is the same as the gold $K_{\alpha 1}$ peak. The $K_{\alpha 2}(Z+1)$ is at the same or lower level than the background, and its contribution to the area of the gold $K_{\alpha 1}$ peak is negligible. In the silver spectra, the satellites were not visible. The 72.8 keV and 74.97 keV peaks correspond to the K-X-rays of lead atoms contained in the detector shield. The peak area of a given transition was calculated by the subtraction of a linear background from the peak integral.

2.3. K-X-rays production cross section

The K_i -X-rays production cross-section formula is independent of the fluorescence yield ω_z and the intensity of a given transition. This K_i -X-rays production cross section is given by

$$\sigma_p(K_i) = \frac{N_X(K_i) \times \frac{\mu}{\rho}(K_i) \times M \times \cos(45)}{N_p \times N_A \times \varepsilon_{tot}(K_i) \times \left(1 - e^{-\frac{\mu}{\rho}(K_i) \times \rho \times d}\right)} \quad (1)$$

Where N_p and $N_X(K_i)$ are the number of incident protons and the detected number of K_i -X-rays photons, respectively, N_A is the Avogadro constant, M is the mass number of the target, μ/ρ is the photon mass attenuation coefficient at the energy of interest, ρ is the target density and ε is the detector efficiency at this energy. The error associated to the cross section measurement contains the errors on the particles beam number, the detector efficiency, the target thickness and the uncertainty on the number of X-rays. The total K X-ray production cross section $\sigma_p^T(K)$ is given by the sum of all K_i cross sections:

$$\sigma_p^T(K) = \sum_i \sigma_p(K_i) \quad (2)$$

2.4. K-shell ionization cross section

The K-shell ionization cross section $\sigma_I(K)$ is equal to the K-X-rays $\sigma_p^T(K)$ production cross section divided by the fluorescence yield ω_K :

$$\sigma_I(K) = \frac{\sigma_p^T(K)}{\omega_K} \quad (3)$$

This cross section is used to compare with the theoretical one given by the models.

The fluorescence yields were calculated by the weighted average [30] value of experimental data and are summarized in Table 2. The weighted average value and its associated standard deviation are given respectively by the equation (4) and (5):

$$\bar{\omega}_{Kw} = \left(\sum_{i=1}^N \left(\frac{1}{\Delta\omega_i} \right)^2 \right)^{-1} \sum_{i=1}^N \left(\frac{\omega_{Ki}}{(\Delta\omega_i)^2} \right) \quad (4)$$

$$\Delta\bar{\omega}_{Kw} = \sqrt{\left(\sum_{i=1}^N \left(\frac{1}{\Delta\omega_i} \right)^2 \right)^{-1} \sum_{i=1}^N \left(\frac{(\omega_{Ki} - \bar{\omega}_K)^2}{(\Delta\omega_i)^2} \right)} \quad (5)$$

Target	ω_{Ki}	Ref	$\bar{\omega}_K$	$\bar{\omega}_{Kw}$
Ag	0.843 ± 0.046	[32]	0.835	0.833 ± 0.011
	0.856 ± 0.025	[33]		
	0.857 ± 0.034	[34]		
	0.861 ± 0.072	[35]		
	0.826 ± 0.005	[36]		
	0.836 ± 0.04	[37]		
	0.829 ± 0.038	[38]		
	0.843 ± 0.047	[39]		
	0.818 ± 0.018	[40]		
	0.815 ± 0.041	[41]		
	0.827 ± 0.066	[42]		
	0.830 ± 0.026	[43]		
	0.821 ± 0.032	[44]		
0.834 ± 0.01	[45]			
Au	0.959 ± 0.014	[43]	0.963	0.959 ± 0.004
	0.967 ± 0.083	[46]		

Table 2 Summary of the experimental fluorescence yield ω_{Ki} , average value $\bar{\omega}_K$ and weighted average value $\bar{\omega}_{Kw}$ for Silver and gold.

3. RESULTS

3.1. K-X-ray production cross section

The K_{α} , $K_{\beta 1}$, and $K_{\beta 2}$ X-ray production cross sections for silver, obtained for four different proton beam energies, are given in Table 3.

Energy (MeV)	$\sigma_p(K_{\alpha})$ (barn)	$\sigma_p(K_{\beta'1})$ (barn)	$\sigma_p(K_{\beta'2})$ (barn)
30	74.8 ± 2.9	13.0 ± 0.8	2.3 ± 0.15
42	81.7 ± 3.0	13.8 ± 0.7	2.5 ± 0.14
54	83.5 ± 3.2	14.2 ± 0.8	2.5 ± 0.15

68	82.6 ± 3.2	14.3 ± 0.8	2.5 ± 0.15
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Table 3: The K_{α} , $K_{\beta 1}$, and $K_{\beta 2}$ X-ray production cross sections for silver for different beam energies.

The error associated to the X-ray production cross section for the main peak, K_{α} " $\sigma_p(K_{\alpha})$ ", is less than 4%, whereas it is less than 6 % for the other peaks, $K_{\beta 1}$ " $\sigma_p(K_{\beta 1})$ " and $K_{\beta 2}$ " $\sigma_p(K_{\beta 2})$ ".

Table 4 presents the $K_{\alpha 1}$, $K_{\alpha 2}$, $K_{\beta 1}$, and $K_{\beta 2}$ X-ray production cross sections for gold obtained for four different proton beam energies.

Energy (MeV)	$\sigma_p(K_{\alpha 1})$ (barn)	$\sigma_p(K_{\alpha 2})$ (barn)	$\sigma_p(K_{\beta 1})$ (barn)	$\sigma_p(K_{\beta 2})$ (barn)
30	1.76 ± 0.05	1.04 ± 0.03	0.57 ± 0.03	0.17 ± 0.02
42	2.74 ± 0.07	1.63 ± 0.05	0.87 ± 0.04	0.27 ± 0.03
54	3.57 ± 0.10	2.13 ± 0.07	1.21 ± 0.06	0.31 ± 0.04
68	4.26 ± 0.13	2.53 ± 0.08	1.47 ± 0.07	0.34 ± 0.04

Table 4: The $K_{\alpha 1}$, $K_{\alpha 2}$, $K_{\beta 1}$, and $K_{\beta 2}$ X-ray production cross sections for gold for the different beam energies.

The experimental errors are less than 3% for $K_{\alpha 1}$ and $K_{\alpha 2}$, less than 6% for $K_{\beta 1}$ and close to 12% for $K_{\beta 2}$.

Table 5 presents the total K-X-ray production cross sections $\sigma_p^T(K)$ for silver and gold obtained for the four different proton beam energies (calculated according to equation 2). The error on the K-X-ray production cross section varies between 3.7 and 4.4 %.

Energy (MeV)	$\sigma_p^T(K)$ (barn)	
	Ag	Au
30	90.1 ± 3.9	3.53 ± 0.14
42	98 ± 3.8	5.52 ± 0.18
54	100.2 ± 4.1	7.22 ± 0.27
68	99.5 ± 4.1	8.60 ± 0.32

Table 5: K-X-ray production cross sections for silver and gold for the different beam energies.

3.2. K-shell ionization cross section

To compare the results with values extracted from theoretical models, the K-shell ionization cross sections have to be determined according to the equation (3).

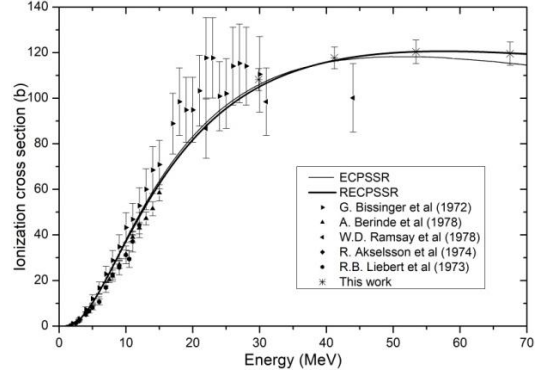


Figure 5 Experimental ionization cross sections as a function of the proton beam energy for silver compared with the ECPSSR/RECPSSR models. The stars correspond to the results presented in this work.

Fig. 5 presents our new experimental values for K-shell ionization cross sections for silver (stars) as a function of the proton beam energy, compared to the existing data and the ECPSSR/RECPSSR models (lines).

These points cover a common region (30 MeV-42 MeV) with those of W. D. Ramsay [23] and G. Bissinger [21]. The difference with our points varies between 2% and 12%. Our new data point at 30 MeV is in agreement with the existing one [21]. Our experimental data are in better agreement with the theoretical models than other experimental ones above 30 MeV. The two model calculations are in agreement with our data points considering the associated error bars.

Fig. 6 presents our new data points for K-shell ionization cross section for gold (stars) as a function of the proton beam energy, compared to the existing data (between 1973 and 2005) and the ECPSSR/RECPSSR models.

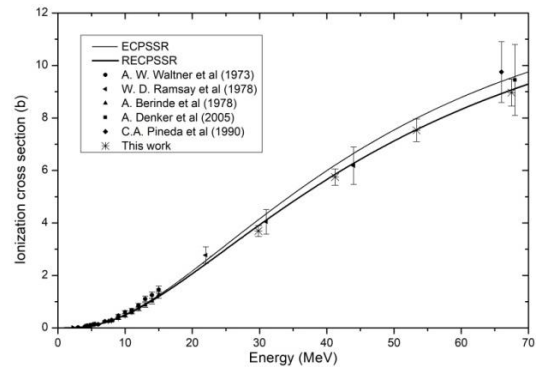


Figure 6 Experimental ionization cross sections as a function of the proton beam energy for gold compared with the ECPSSR/RECPSSR models. The stars correspond to the results presented in this work.

We found a 5% deviation with the data points published by A. Denker [28] at 68 MeV and a 9% difference from the data point of C.A. Pineda [27] at 66 MeV. These values are compatible with each other within the error bars. The new points, at 30 and 40 MeV, are in good agreement with the trend of the points given by W. D. Ramsay [23].

Our points have error bars smaller than other existing data, which helps to compare with the two close models ECPSSR and RECPSSR: they are in the same trend as the ECPSSR model, but in better agreement with the RECPSSR model.

4. CONCLUSION

A new set of data for proton induced K-X-ray production cross sections have been obtained at the high energy cyclotron ARRONAX in Nantes (France). These new measurements were performed at 30, 42, 54 and 68 MeV for silver and gold with errors ranging between 4 and 7%. The weighted average values of the experimental fluorescence yields have been used to convert K-X-ray cross sections into K-Shell ionization cross sections. These latter are in good agreement with the trend of low energy measurements and within the error bars with the scarce existing high energy experimental data. As expected, the RECPSSR model which includes relativistic effects for the projectile seems better reproducing these new experimental data than the ECPSSR model.

The K-shell ionization cross section measurements now need to be extended to other medium and heavy elements, which is necessary to complete the database and to validate the theoretical models that can be used for HEPIXE quantitative analysis.

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Table 1

K_α	$K_{\alpha_1} = L_3 \rightarrow K$ $K_{\alpha_2} = L_2 \rightarrow K$	
K_β	K_{β_1}'	$K_{\beta_1} = M_3 \rightarrow K$ $K_{\beta_3} = M_2 \rightarrow K$ $K_{\beta_5} = M_{4,5} \rightarrow K$
	K_{β_2}'	$K_{\beta_2} = N_{2,3} \rightarrow K$ $K_{\beta_4} = N_{4,5} \rightarrow K$

Table 2

Target	ω_{Ki}	Ref	$\bar{\omega}_K$	$\bar{\omega}_{Kw}$
Ag	0.843 ± 0.046	[32]	0.835	0.833 ± 0.011
	0.856 ± 0.025	[33]		
	0.857 ± 0.034	[34]		
	0.861 ± 0.072	[35]		
	0.826 ± 0.005	[36]		
	0.836 ± 0.04	[37]		
	0.829 ± 0.038	[38]		
	0.843 ± 0.047	[39]		
	0.818 ± 0.018	[40]		
	0.815 ± 0.041	[41]		
	0.827 ± 0.066	[42]		
	0.830 ± 0.026	[43]		
0.821 ± 0.032	[44]			
0.834 ± 0.01	[45]			
Au	0.959 ± 0.014	[43]	0.963	0.959 ± 0.004
	0.967 ± 0.083	[46]		

Table 3

Energy (MeV)	$\sigma_P(K_{\alpha})$ (barn)	$\sigma_P(K_{\beta'1})$ (barn)	$\sigma_P(K_{\beta'2})$ (barn)
30	74.8 ± 2.9	13.0 ± 0.8	2.3 ± 0.15
42	81.7 ± 3.0	13.8 ± 0.7	2.5 ± 0.14
54	83.5 ± 3.2	14.2 ± 0.8	2.5 ± 0.15
68	82.6 ± 3.2	14.3 ± 0.8	2.5 ± 0.15

Table 4

Energy (MeV)	$\sigma_P(K_{\alpha 1})$ (barn)	$\sigma_P(K_{\alpha 2})$ (barn)	$\sigma_P(K_{\beta' 1})$ (barn)	$\sigma_P(K_{\beta' 2})$ (barn)
30	1.76 ± 0.05	1.04 ± 0.03	0.57 ± 0.03	0.17 ± 0.02
42	2.74 ± 0.07	1.63 ± 0.05	0.87 ± 0.04	0.27 ± 0.03
54	3.57 ± 0.10	2.13 ± 0.07	1.21 ± 0.06	0.31 ± 0.04
68	4.26 ± 0.13	2.53 ± 0.08	1.47 ± 0.07	0.34 ± 0.04

Table 5

Energy (MeV)	$\sigma_p^T(K)$ (barn)	
	Ag	Au
30	90.1 ± 3.9	3.53 ± 0.14
42	98 ± 3.8	5.52 ± 0.18
54	100.2 ± 4.1	7.22 ± 0.27
68	99.5 ± 4.1	8.60 ± 0.32

Figure1

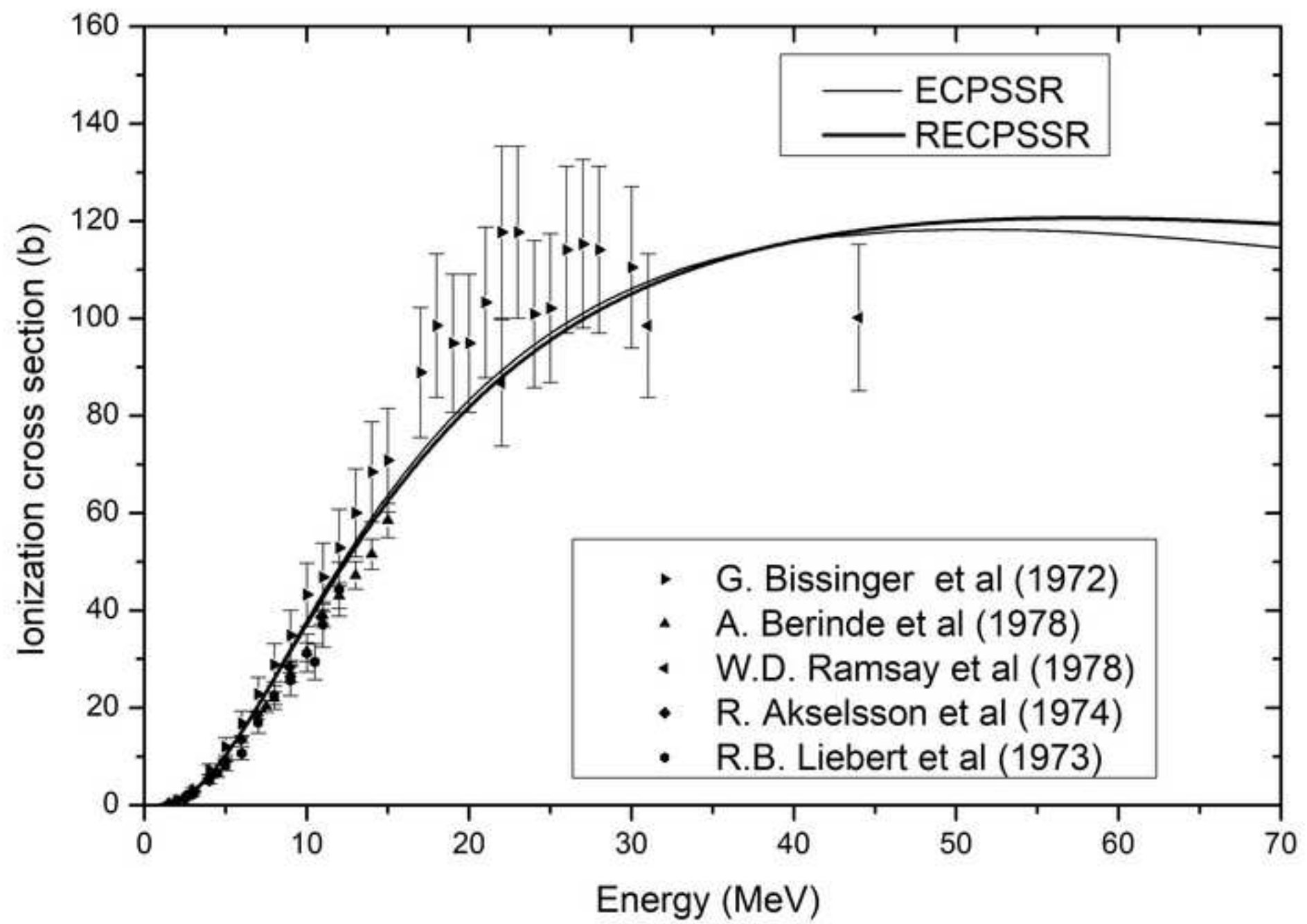


Figure2

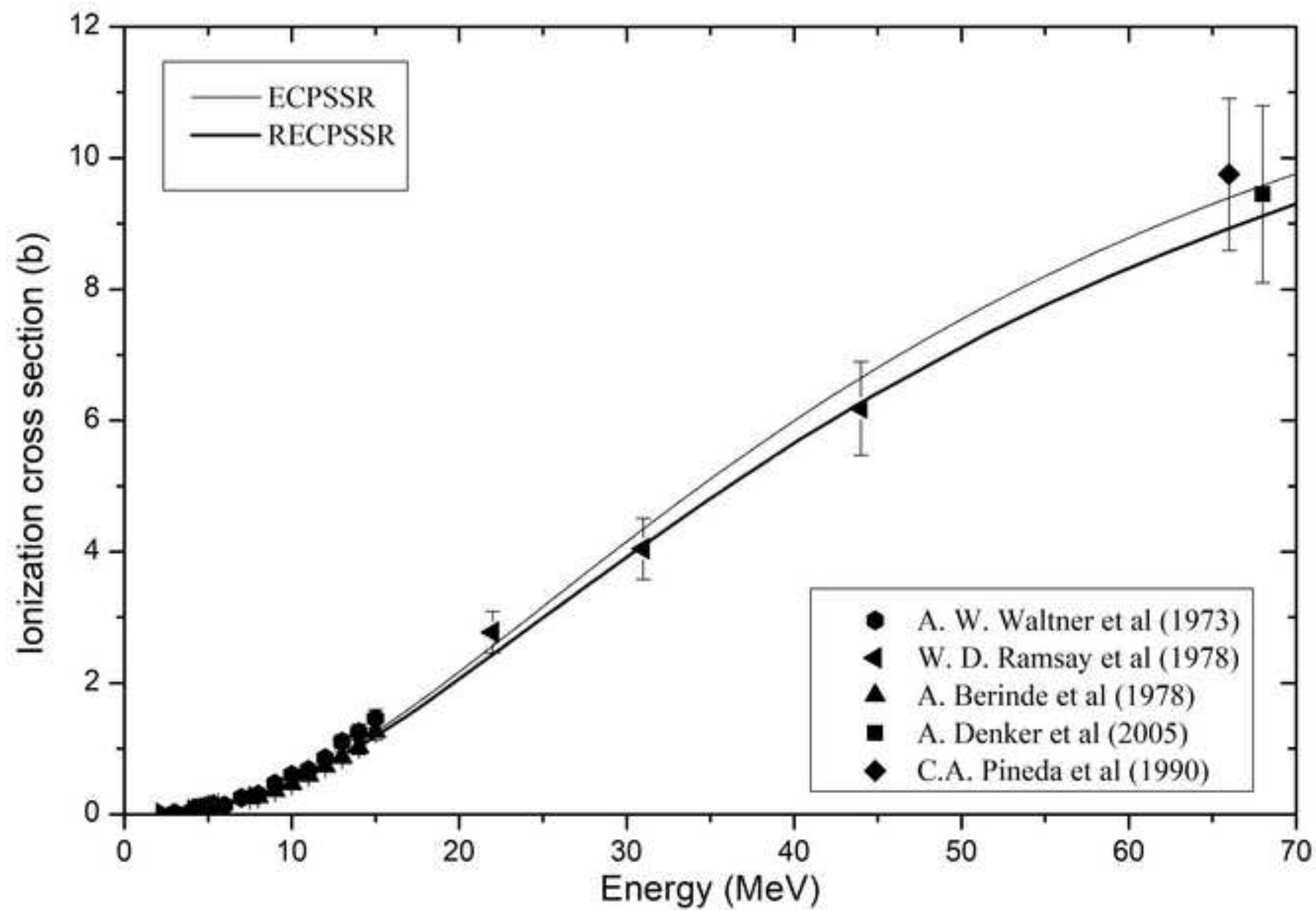


Figure3

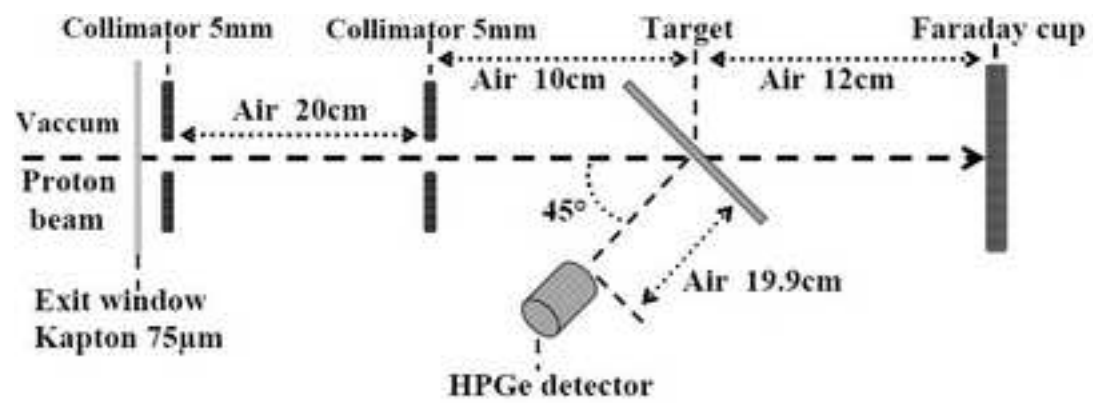


Figure4

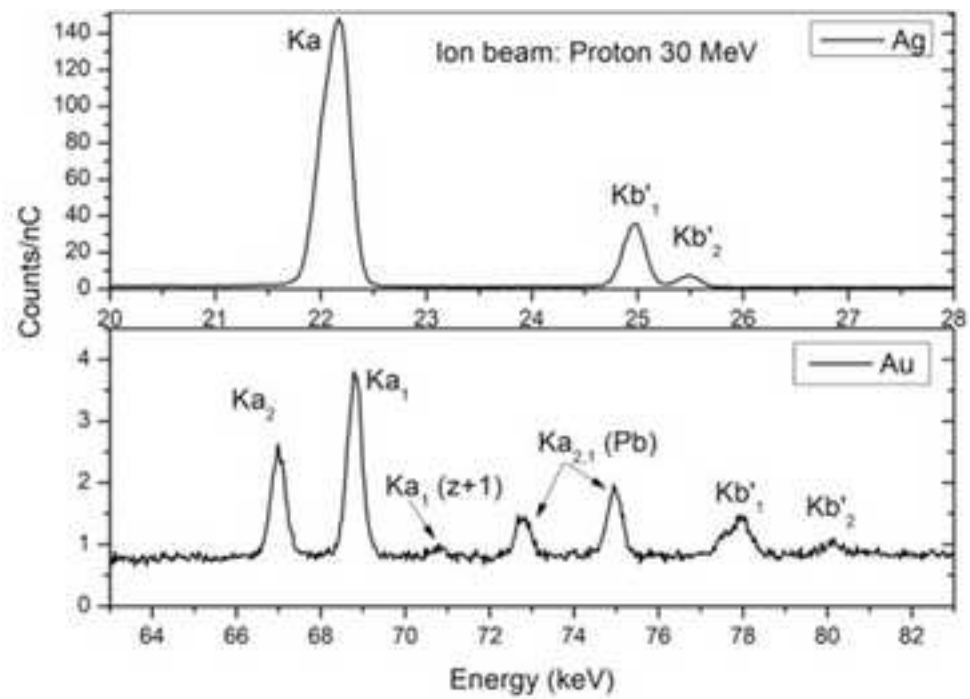


Figure 5

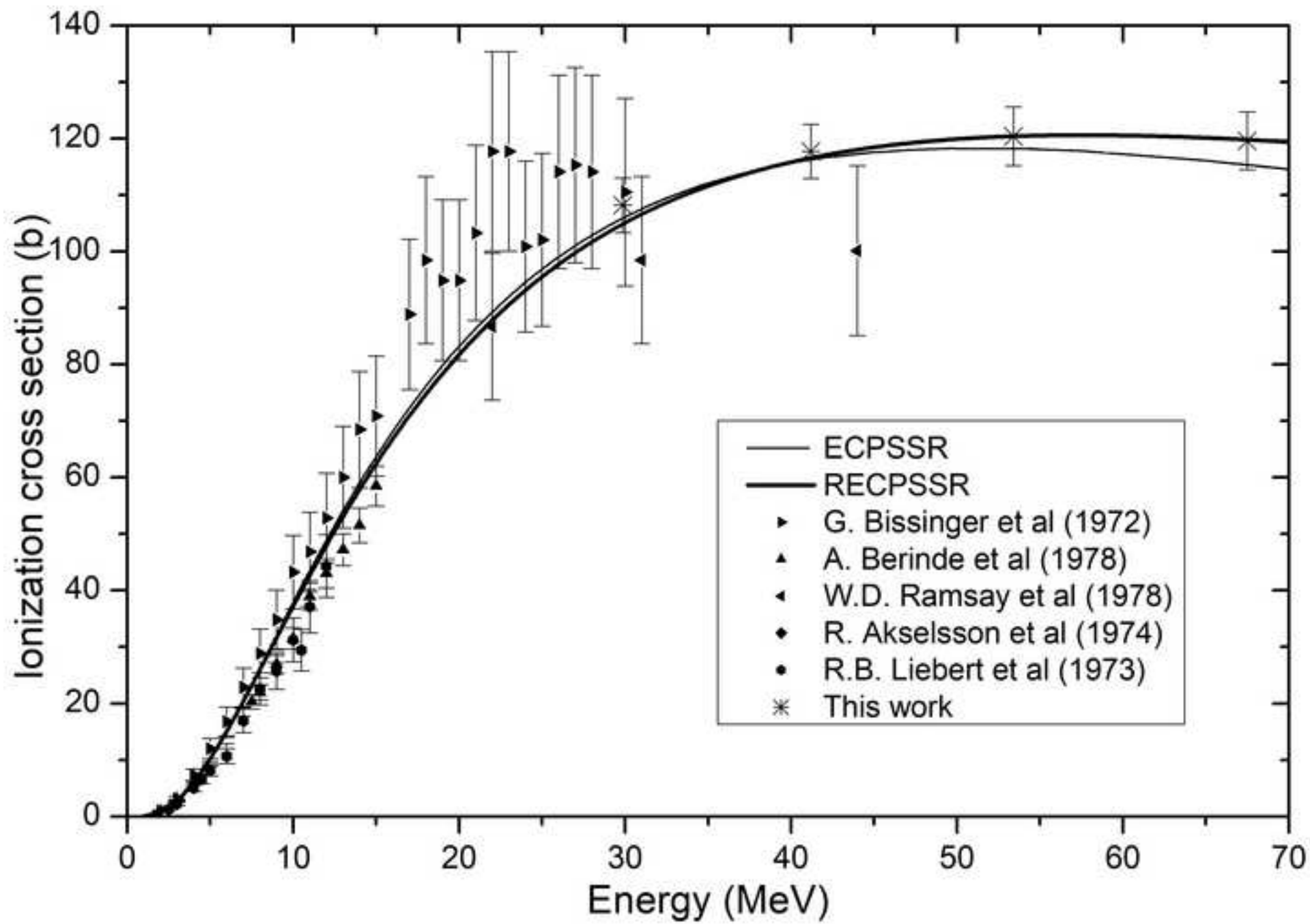


Figure6

