

SINGLE AND DOUBLE IONIZATION CROSS SECTIONS OF THE ARGON ATOM BY ELECTRON IMPACT

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Single and double ionization of the argon atom by electron impact in the incident electron energy range from threshold to 100 eV were investigated. Partial ionization cross sections have been obtained by a normalization procedure to total ionization cross sections.

1. Introduction

Newest review papers and reports on values of ionization cross sections for atomic particles^{1,2,3,4)} show that the knowledge in the field of partial ionization cross sections of atoms, and specially of molecules, by electron collisions is scarce, as well as that among data of various authors big differences do exist. Data of this kind are needed for a better understanding of the interaction between electrons and the atomic particle, and for numerous fields of physics and related sciences for the interpretation of more complex processes and their modelling.

These were the reasons that we decided to reconstruct, change and improve an experimental apparatus⁵⁾ for the measurement of partial ionization cross sections. Detailed investigation of all parameters of the new experimental apparatus has been undertaken with argon as a target gas, since some earlier measurements of this atom are available⁶⁻¹²⁾ with which the results obtained can be compared, and a calibration procedure to total ionization cross sections¹³⁾ can be used.

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2. Experimental

The experimental apparatus used for present experiments contains the following main operational parts, shown in Fig. 1: a trochoidal electron monochromator (TEM) as a source of the incident electron beam, a parallel plate interaction chamber (IC) enclosed in a gas cell, a primary electron beam collector (PBC), a three-element cylindrical electrostatic lens system (TEL) for the acceleration

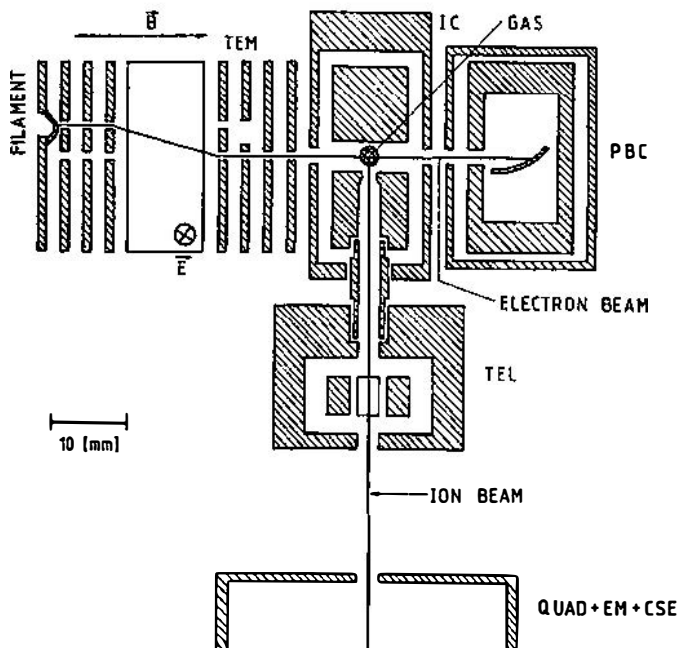


Fig. 1 Schematics of the experimental apparatus: TEM — trochoidal electron monochromator, IC — interaction chamber, PBC — primary electron beam collector, TEL — three-element cylindrical electrostatic lens system, QUAD — Quadrupole Mass Filter, IBD — ion beam detector.

and collimation of the ion beam, a quadrupole mass filter (QUAD) for the separation of the ion beam vs. m/q , and an ion beam detector (IBD). Ions formed in ionizing collisions are drawn out of the interaction chamber at an angle of 90° in respect to the electron beam axis.

2.1. Trochoidal electron monochromator

The primary electron beam is formed by a trochoidal electron monochromator^{1,4)}, a special combination of orthogonal electric and magnetic fields. The electron beam current was in the range of 30 nA, with an energy width of around 200 meV. For electron beam collimation and the TEM operation a homogeneous magnetic field of around 10^{-2} T parallel to the electron beam axis was used, generated by a Helmholtz coil pair.

2.2. Interaction chamber

The monoenergetic electron beam of given energy enters through the entrance aperture a small closed gas cell which represents the interaction chamber. It is made of two parallel electrodes, allowing the application of a homogeneous electric field for the extraction of ions formed in ionizing collisions. One of the electrodes has a small aperture through which ions can leave the interaction chamber space towards the mass-dispersive element.

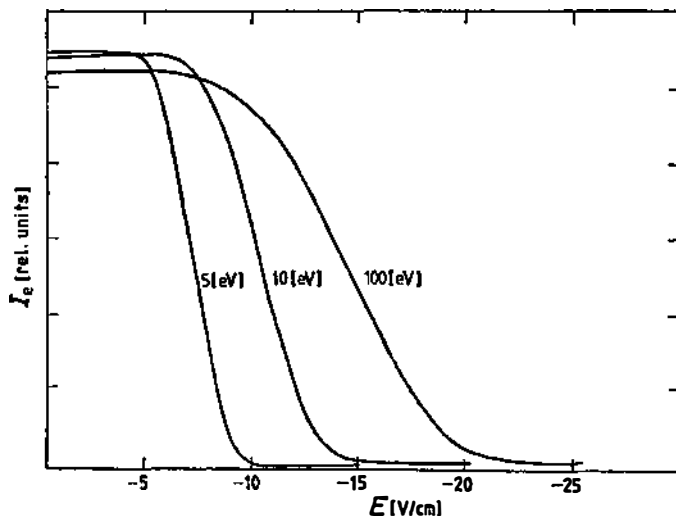


Fig. 2. Influence of the ion extraction electric field to the efficiency of the primary electron beam collection.

The intensity of the ion beam drawn out from the interaction chamber increases with the electric field strength, but simultaneously the path of the electron beam is changed. In the crossed electric and magnetic fields electrons drift in the same way as in the TEM. Only field strengths which keep the electron beam within the exit aperture of the interaction chamber gas cell can be used, since it must be detected and monitored by the electron beam collector. In Fig. 2 dependence of the primary electron beam intensities at the beam collector are given is the interaction chamber electric field strength for various incident electron energies. It can be seen that only field strengths lower than 2 V/cm satisfy the condition for total electron beam collection.

2.3. Three-element electrostatic lens system

The ion beam formed by drawing out ions from the interaction chamber is focused and adapted in energy to the mean path of the mass dispersive element by a three-element cylindrical electrostatic lens system^{15,16}. Two potentials of the three are fixed: the first element of the lens system is at the potential of the interaction chamber negative electrode, the third at the potential of the mass dispersive element entrance electrode. Care was taken that the filling factor of the

electrostatic lenses is kept below 0.3. Change of the second electrode potential allows the ion beam to be focused at the entrance aperture of the mass dispersive element (Fig. 3).

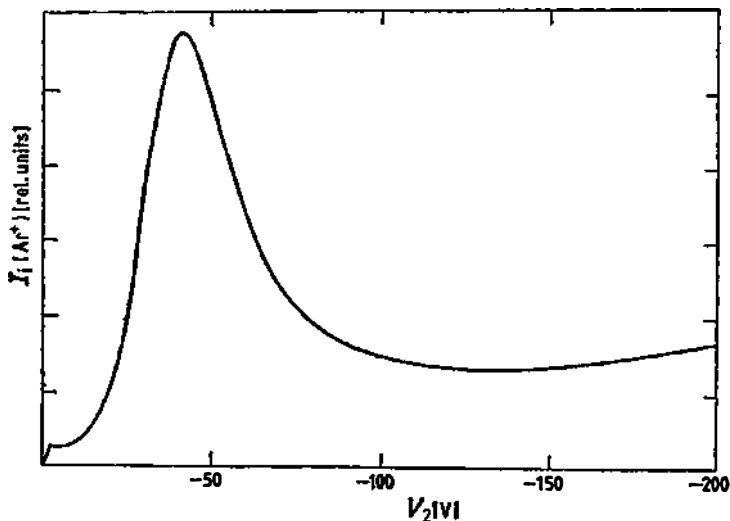


Fig. 3. Focusing of the ion beam to the entrance aperture of the mass spectrometer by the three-element cylindrical electrostatic lens system.

Inside the third electrode two pairs of electrodes form orthogonal electronic fields for positioning the ion beam into the middle of the mass dispersive element entrance electrode. The optimal beam position is achieved with non-zero electric field strengths between detector plates, showing thus that ion paths are influenced by the guiding magnetic field of the electron monochromator.

2.4. Mass dispersive element

For the mass analysis of ions in our experiment a Balzers made, Type QMG 212, Quadrupole Mass Spectrometer was used, with an electron multiplier in the current amplification mode.

2.5. Vacuum conditions

The background pressure in the whole apparatus was around $5 \cdot 10^{-5}$ Pa, achieved by a mercury three-stage diffusion pump, backed by a mercury booster pump and a two-stage rotary pump. The vacuum system is fully automated against power and water supply features.

Argon gas of 99.99% purity, of VEB- Leipzig, was introduced through a needle valve into the gass cell until in the rest of the apparatus a pressure of $2 \cdot 10^{-4}$ Pa was reached. The gas pressure in the gas cell was calculated to be 150 times higher i. e. of the order of $3 \cdot 10^{-2}$ Pa, the binary collision conditions being fulfilled. The linearity of the ion signal vs. the gas pressure was proved in separate calibration procedures.

3. Experimental results

3.1. Singly and doubly charged argon ion signals

With all potentials on the interaction chamber electrodes, three-element electrostatic lens electrodes and X and Y detectors set to optimal values and a constant gas pressure, signal intensities of singly and doubly charged argon ions have been measured in the energy range from the threshold to 100 eV.

For each ionic species great number of ionization curves were taken for different gas pressures and ion beam detector amplifications. All measured curves were normalized at 100 eV to obtain a statistical mean value of the relative intensity curve. The individual curves differed from the average by less than 5%.

3.2. Single ionization cross sections

The averaged singly ionized argon atom signal was brought to an absolute scale by normalization to the total ionization cross section measured in our laboratory¹¹. Namely, for incident electron energies lower than 43.375 eV (threshold for the double ionization process), the measured singly ionized argon atom signal must be identical in shape with the total ionization cross section curve. By a mathematical method for comparison of numerical values in the whole energy range up to the double ionization threshold, absolute cross section values were obtained. They are given in Table 1 and compared with all other values available from the literature.

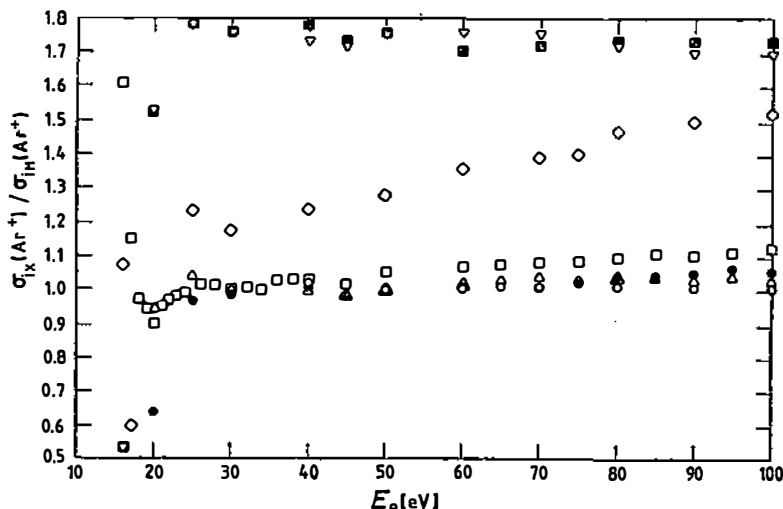


Fig. 4. Ratios of single ionization cross sections of the argon atom by different authors to our data: ∇ — Bleakney⁶, \diamond — Peterson⁷; \circ — Okudaira et al.⁸; \blacksquare — Crowe et al.⁹; \triangle — Stephan et al.¹⁰; \square — Wenzel et al.¹¹ and \bullet — Krishnakumar and Srivastava¹².

The degree of agreement between our values and the other is more clearly shown in Fig. 4 as ratios of cross sections. One can see that the best agreement exists with recent measurements of Stephan et al.¹⁰, Wenzel et al.¹¹ and Krishna-

TABLE 1.

Electron energy (eV)	Authors							our results
	Bleakney (1930)	Peterson (1963)	Okudaira et al. (1970)	Crowe et al. (1972)	Stephan et al. (1980)	Wenzel et al. (1987)	Krishna-kumar and Srivastava (1988)	
16	0.01	0.02		0.01		0.03		0.02
17		0.078				0.15		0.13
18						0.29		0.30
19						0.44		0.47
20	0.98	0.60		0.98	0.61	0.58	0.41	0.64
21						0.71		0.75
22						0.85		0.88
23						0.96		0.98
24						1.10		1.11
25	2.14	1.48		2.14	1.26	1.25	1.16	1.20
26						1.34		1.32
28						1.57		1.55
30	3.06	2.04		3.06	1.76	1.74	1.72	1.74
32						1.92		1.90
34						2.07		2.07
36						2.24		2.19
38						2.39		2.31
40	4.13	2.91	2.39	4.25	2.39	2.47	2.39	2.39
45	4.30			4.35	2.49	2.56	2.48	2.51
50	4.36	3.11	2.60	4.36	2.49	2.62	2.50	2.49
55	4.25			4.22	2.49	2.62	2.50	
60	4.32	3.33	2.58	4.20	2.50	2.63	2.48	2.46
65					2.52	2.63	2.47	2.45
70	4.28	3.39	2.57	4.20	2.54	2.65	2.49	2.45
75		3.43			2.54	2.66	2.51	2.45
80	4.20	3.58	2.58	4.24	2.53	2.68	2.53	2.44
85					2.52	2.70	2.54	2.44
90	4.12	3.62	2.60	4.20	2.51	2.68	2.54	2.43
95					2.49	2.67	2.54	2.40
100	4.05	3.62	2.60	4.13	2.46	2.68	2.52	2.39

Single ionization cross section of the argon atom by electron impact; in units of 10^{-20} m².

kumar and Srivastava¹²⁾. The biggest discrepancies appear in the above-threshold region, presumably due to two reasons: firstly, to differences in the energy scale calibration, and secondly, to small signals of ions collected. In the energy range above 50 eV the biggest is the discrepancy with Wenzel et al.¹¹⁾ reaching almost 10%. With the other two sets of data the agreement is very good, differences not exceeding 5%.

3.3. Double ionization cross sections

For electron energies above the double ionization threshold the parallel plate method for the determination of the total ionization cross section directly measures a signal proportional to

$$\sigma_{i,tot} = \sigma_i(\text{Ar}^+) + 2 \cdot \sigma_i(\text{Ar}^{2+}),$$

since the total ionic charge is collected.

The quadrupole mass spectrometer has a variable transparency for ions of different masses, so that the signal measured for Ar^{2+} ions must be corrected for this behaviour. The way to do it is to compare the shape of the curve

$$I_{i,\text{tot}} = I_i(\text{Ar}^+) + 2 \cdot x \cdot I_i(\text{Ar}^{2+})$$

and that of the total ionization cross section. The comparison of numerical values in the whole measured incident electron energy range delivered a value for the normalization factor x , which allowed to bring the double ionization curve into an absolute scale. Numerical values obtained in this way are listed in Table 2 and compared with other data available from the literature.

TABLE 2.

Electron energy [eV]	Authors							
	Bleakney (1930)	Peterson (1963)	Okudaira et al. (1970)	Crowe et al. (1972)	Stephan et al. (1980)	Wenzel et al. (1987)	Krishna-kumar and Srivastava (1988)	our results
45					0.001			
50	0.036		0.087	0.036	0.016	0.018	0.012	0.010
55					0.045			
60	0.20	0.254	0.154	0.252	0.082	0.077	0.099	0.086
65					0.113	0.108	0.134	0.130
70	0.312		0.176	0.378	0.133	0.130	0.167	0.160
75					0.144	0.147	0.193	0.180
80	0.382	0.449	0.186	0.435	0.154	0.158	0.204	0.195
85					0.159	0.164	0.211	0.205
90	0.44		0.191	0.465	0.162	0.170	0.22	0.214
95					0.162	0.171	0.225	0.219
100	0.47	0.507	0.194	0.478	0.168	0.177	0.228	0.225

Double ionization cross sections of the argon atom by electron impact; in units of 10^{-20} m^2 .

One method to check the validity of the normalization procedure is to compare the single to double ionization cross sections ratio at an energy of 100 eV with corresponding values of other authors, as shown in Fig. 5. Our value of 0.0785 for this ratio is in good agreement with 0.0732 obtained as an average from experiments made after 1960 except one¹³⁾. The difference is within 12% of this average value, proving that the normalization procedure is correct.

Ratios of double ionization cross sections of other authors to our values are shown in Fig. 6. Here again, the best agreement is with the recently obtained data by Stephan et al.¹⁰⁾, Wenzel et al.¹¹⁾ and Krishnakumar and Srivastava¹²⁾.

3.4. Comparison of total ionization cross sections

The contribution of the cross section for triple ionization, with the threshold at 84.275 eV, to the total ionization cross section in the range up to 100 eV can be neglected, being smaller than 0.5%. Thus, the sum of $[\sigma_i(\text{Ar}^+) + 2 \cdot \sigma_i(\text{Ar}^{2+})]$

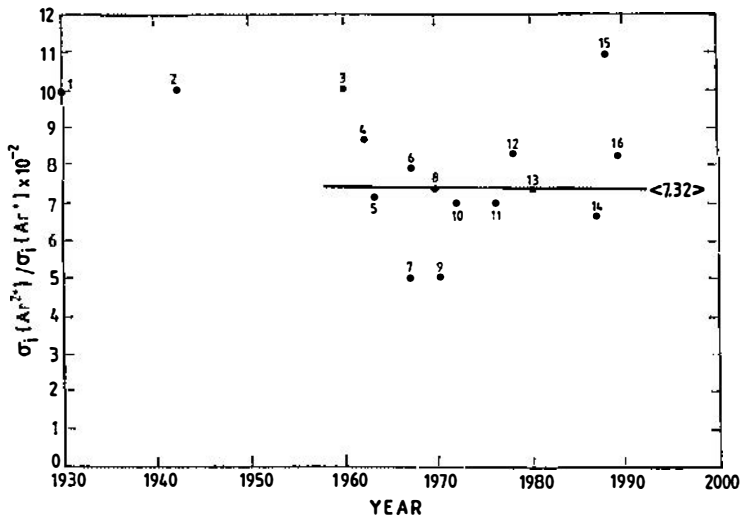


Fig. 5. Ratios of single to double ionization cross sections of various authors at an incident electron energy of 100 eV: 1 — Bleakney⁶⁾; 2 — Stevenson and Hipple¹⁷⁾; 3 — Fox¹⁸⁾; 4 — Fiquet-Fayard et Lahmani¹⁹⁾; 5 — Peterson⁷⁾; 6 — Melton and Rudolph²⁰⁾; 7 — Gaudin et Hageman²¹⁾; 8 — Morrison and Traeger²²⁾; 9 — Okudaira et al.⁸⁾; 10 — Crowe et al.⁹⁾; 11 — Drewitz²³⁾; 12 — Egger und Märk²⁴⁾; 13 — Stephan et al.¹⁰⁾; 14 — Wenzel et al.¹¹⁾; 15 — Krishnakumar and Srivastava¹²⁾.

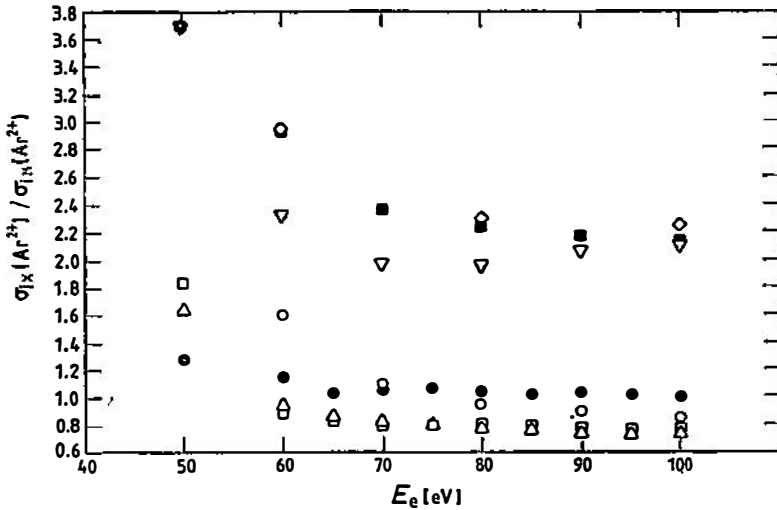


Fig. 6. Ratios of double ionization cross sections of the argon atom by different authors to our data: ∇ — Bleakney⁶⁾; \diamond — Peterson⁷⁾; \circ — Okudaira et al.⁸⁾; \blacksquare — Crowe et al.⁹⁾; \triangle — Stephan et al.¹⁰⁾; \square — Wenzel et al.¹¹⁾ and \bullet — Krishnakumar and Srivastava¹²⁾.

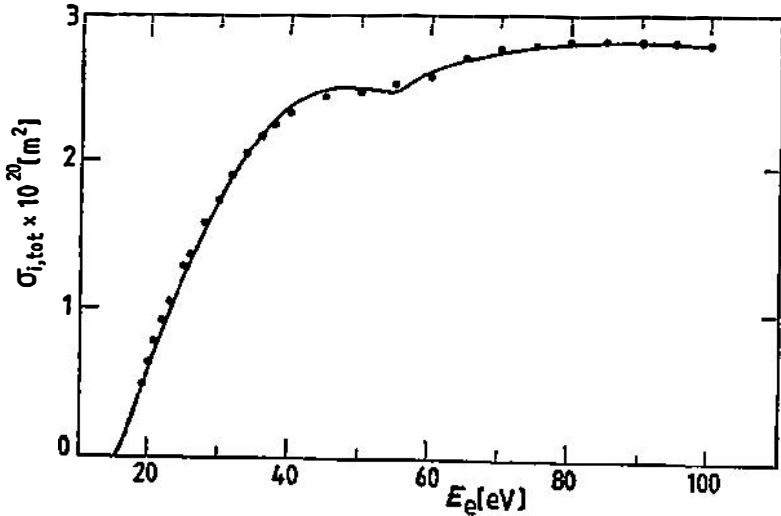


Fig. 7. Total ionization cross section of the argon atom: full curve by summation of the present single and double ionization cross sections; ● — values obtained by total ion charge collection in the parallel plate interaction chamber¹⁾.

determined in our measurements can be compared directly to the total ionization cross section measured by a parallel plate ionization chamber¹⁾. The two cross section curves are shown in Fig. 7. The agreement is very good in shape, which is, taking into account the procedure used for normalization, of importance.

4. Conclusions

The thorough investigation of the experimental apparatus constructed and adapted for the measurement of partial cross sections was undertaken. The normalization procedure showed that reliable partial cross sections can be obtained for atomic species. For the time being the use of the apparatus to measure partial ionization cross sections for processes in which faster ions are formed was not investigated. In some cases, as in dissociative ionization processes, ions of considerable kinetic energies can be formed, changing the extraction efficiency from the interaction chamber, as well as the focusing properties of the three-element electrostatic lens system. Search for influences of these effects is our next goal.

With the available apparatus partial cross section determinations for a processes where the ions have thermal energy distributions, such as ions of inert gases and parent ions of molecules are next in our programme.

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References

- 1) H. Tawara and T. Kato, Atomic Data and Nuclear Data Tables **36** (1987) 167;
- 2) *Electron impact ionization*, Ed. T. D. Mark and G. H. Dunn, Springer Verlag, Berlin, 1985;
- 3) K. L. Bell, H. B. Gilbody, J. G. Hughes, A. E. Kingstone and F. J. Smit, J. Phys. Chem. Ref. Data **12** (1983) 891;
- 4) I. Shimamura, Sci. Papers, Inst. Physical and Chemical Research **82** (1989) 1 Rikagaku Kenkyusho, Japan;
- 5) N. Đurić and A. Stamatović, 7th SPIG, Rovinj, 1974; Contributed Papers, p. 55;
- 6) W. Bleakney, Phys. Rev. **36** (1930) 1303;
- 7) J. R. Peterson, Proc. 3rd International Conference on the Physics of Atomic and Electronic Collisions, London, 1963; Ed. M. R. C. McDowell, North Holland Publ. Co., Amsterdam, 1964, p. 465;
- 8) S. Okudaira, Y. Kaneko and I. Kanomata, J. Phys. Soc. Japan. **28** (1970) 1536;
- 9) A. Crowe, J. A. Preston and J. W. McConkey, J. Chem. Phys. **57** (1972) 1620;
- 10) K. Stephan, H. Helm and T. D. Märk, J. Chem. Phys. **73** (1980) 3763;
- 11) R. C. Wenzel, F. A. Baiocchi, T. R. Hayes and R. S. Freund, Phys. Rev. A **35** (1987) 559;
- 12) E. Krishnakumar and S. K. Srivastava, J. Phys. B, At. Mol. Opt. Phys. **21** (1988) 1055;
- 13) M. Kurepa, I. Čadež and V. Pejčev, Fizika **6** (1974) 185;
- 14) A. S. Stamatović and G. J. Schulz, Rev. Sci. Instrum. **39** (1968) 1752;
- 15) D. W. O. Heddle, J. Phys. E., Sci. Instrum. **2** (1969) 1046;
- 16) D. W. O. Heddle and M. V. Kurepa, J. Phys. E., Sci. Instrum. **3** (1970) 552;
- 17) D. P. Stevenson and J. A. Hipple, Phys. Rev. **62** (1942) 237;
- 18) R. E. Fox, J. Chem. Phys. **33** (1960) 200;
- 19) F. Fiquet-Fayard et M. Lahmani, J. Chem. Phys. **59** (1962) 1050;
- 20) C. E. Melton and P. S. Rudolph, J. Chem. Phys. **47** (1967) 1771;
- 21) A. Gaudin et R. Hageman, J. Chem. Phys. **64** (1967) 1209;
- 22) J. D. Morrison and J. C. Traeger, J. Chem. Phys. **53** (1970) 4053;
- 23) H. J. Drewitz, Int. J. Mass Spectrom. Ion Phys. **19** (1976) 313;
- 24) F. Egger und T. D. Märk, Z. Naturforsch. A **33** (1978) 1111.

PRESECI ZA JEDNOSTRUKU I DVOSTRUKU JONIZACIJU ATOMA
ARGONA UDAROM ELEKTRONA

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Ispitivana je jednostruka i dvostruka jonizacija atoma argona udarom elektrona upadne energije od praga do 100 eV. Parcijalni preseki za jonizaciju dobijeni su postupkom normalizacije na totalni presek za jonizaciju.

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