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Electron-impact ionization cross-section of argon (σ_{n+}, $n = 7,8$)

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Abstract

Multiple ionization cross-sections of argon (Ar–Ar^{n+}, with $n = 7,8$) by electron impact are measured for energies ranging from threshold up to 3000 eV. The data were obtained by a time-of-flight mass-to-charge spectrometric technique. A comparison with other experimental data and with semi-empirical calculations is presented. The integrated oscillator strength (M^2_{7+}) for the production of Ar^{7+} was determined. © 1997 Elsevier Science B.V.

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1. Introduction

Multiple ionization cross-sections (MICS) of atoms by electronic collision are needed in many fundamental applications in different technological and scientific areas such as, for instance, plasma physics and astrophysics. Nevertheless, accurate argon multiple ionization measurements are still scarce and fragmentary. Moreover the discrepancies between different measurements very often exceed the combined uncertainties. Obtaining MICS presents a serious experimental challenge [1–3]. Efforts have been made to establish more precise data on multiple ionization cross-sections, mainly for noble gases. Even so, there are large differences between the data from different experimental groups both in magnitude and in shape. One of the best reviews up to the 60s of the several experiments on ionization cross-sections was given by Kieffer and Dunn [4]. Systematic discrepancies can be found in measurements performed by groups using different experimental approaches, such as effusive gas beams and static gas targets. Bruce and Bonham [2] and Tarnovsky and Becker [3] discussed the possible sources of the differences in the magnitude of cross-sections reported by laboratories using various techniques. Some of these disagreements have also been attributed to autoionization states present in the target final state [5,6].

From a theoretical point of view, the picture is also unsatisfactory. In the last two decades several efforts have been made to derive calculation schemes for the ionization cross-sections [7–11]. Recently, Fisher et al. [12] have derived an empirical expression that was found to agree well with a variety of experimental ionization

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cross-sections. Recent alternative approaches have been published by Shevelko and Tawara [13] and Deutsch et al. [14,15].

In the low-momentum transfer regime, the generalized oscillator strength (GOS) can be related to the optical oscillator strength (OOS) by:

$$M_{n+}^2 = \int \left(\frac{R}{\epsilon}\right) \frac{df}{d\epsilon} d\epsilon \qquad (1)$$

where R is the Rydberg energy and $df/d\epsilon$ is the generalized oscillator strength.

Multiple ionization can be produced mainly by two processes: inner-shell ionization with subsequent secondary electron ejection and outer-shell ionization with simultaneous electron ejection. The reaction channel can be investigated using photo-ionization data [16–19]. The matrix-element squared M_{n+}^2 is obtained by integrating the generalized oscillator strength for the processes considered [16].

Commonly, the most accurate information is available on the lower degrees of ionization (i.e., n = 1 to 3) of argon from the neutral atom by electron impact. In a previous study [5], a set of partial cross-sections for ionization of Ar has been presented for n = 3 to 6. In the present article this work is extended to n = 7 and 8. Straub et al. [20] and Rapp and Englander-Golden [21] have measured the total ionization cross-sections for argon and their values differ by about 10%, which is within the combined uncertainties.

2. Experimental apparatus

A detailed description of the apparatus, its operation and performance has been given elsewhere [5] (see also Fig. 1). Briefly, the electron gun is designed for electron energies from 90 to 3000 eV with a typical incident current of 0.01–0.1 nA (DC equivalent) and pulsed each 20 μs at 0.4% duty cycle. Pulses of electrons are directed toward a 3-cm-long gas cell through a pair of diaphragms (1.5 and 2 mm diameter).

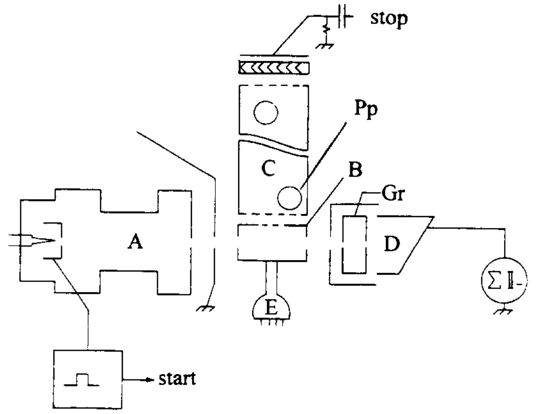

Fig. 1. Schematic view of the experimental set-up: A, electron gun; B, gas target; C, time-of-flight tube; D, Faraday cup; E, pressure gauge; pA, picoammeter; Gr, guard ring; Pp, pump-out ports.

The gas cell is filled with research grade gas (99.99% purity) at a pressure below 10^{-4} torr, which ensures single-collision conditions. After passing through the cell, the electron beam is collected in a large Faraday cup coupled to a picoammeter. The secondary electron emission was sufficiently reduced by a guard ring kept at −70 V.

After each incident electron bunch, the ionic states present in the cell are extracted by a 50-V cm^{-1} pulsed electric field (for 17 μs), transmitted through a 140-mm time-of-flight (TOF) Wiley–McLaren-type mass spectrometer [22] and detected by a set of micro-channel plates. The rise-time of the extraction pulse is coincident with the fall-time of the incident electron pulse.

The ions leave the interaction region through a 0.7-cm diameter hole in the top plate of the cell which is covered by an 82% transparent molybdenum grid. They are accelerated across a 0.5-cm gap before they enter a free-field region. Both ends of the TOF tube are closed by Mo mesh. The drift region is pumped through four ports of 0.5 cm diameter each. A large optical TOF tube was adopted to ensure maximum ion detection. To optimize the TOF focusing properties, the ionic trajectories were modelled using SIMION [23].

During all experiments the pressure inside the collision chamber was kept sufficiently low that the probability that an incident electron undergoes more than one collision is negligible. The single-collision regime was checked at each incident energy. The initial growth rate of the ratio I^{n+}/I^{-} is linear (I^{n+} is the ion current detected for the species n and I^{-} is the electron current). For a fixed incident energy, the ratio can be expressed as:

$$\frac{I^{n+}}{I^{-}} = \epsilon\pi(P)\sigma_{n+} \qquad (2)$$

where $\pi(P)$ is the number of target particles per square centimeter, which is proportional to the gas cell pressure P, and ϵ is the efficiency of ionic collection.

Special attention was paid to the incomplete collection and detection of the ions. The ionic transmission through the TOF spectrometer may depend on the charge-state focusing, particularly for highly charged ions. Simulations using SIMION showed that under our experimental conditions all extracted ions fly along trajectories parallel to the TOF tube axis. Therefore, the geometric loss fraction is independent of the charge state of the ion.

The most crucial problem is due to electron capture by the ion in collisions with the background gas. In our case, the ion loss during their flight to the detector was minimized by the low pressure inside the chamber. Along the ion trajectories (about 150 mm in total length) the ions move from a pressure $< 10^{-4}$ torr inside the gas cell to 10^{-7} torr in the detection region. The electronic capture cross-section at 50 eV (with n = 7 and 8) estimated by a Landau–Zener model [24] is of the order of 10^{-15} cm^2 for both Ar^{7+} and Ar^{8+} ions. Therefore, less than 8% of the ions are removed from their initial charge state and hence the effect of incomplete ion collection is smaller than the total uncertainty.

We have considered the experimental arguments of Bruce and Bonham [2] for all measurements carried out. For example, the accelerating potential at the entrance of the micro-channel plate was 5 kV. In our case, the determined ratios R_n for low charge states (n = 2 to 6) agree within the uncertainty with previously reported data [25–29] for 2 keV electron-impact energy. The same ratio analysis has been performed for neon [6] showing good agreement with the data in the literature. Hence we conclude that the dependence of ϵ on the ionic species is small for the geometry adopted, and can, therefore, be considered constant for all states studied here.

3. Results and discussion

All cross-sections were determined from the ratio of the ion abundances. The results are free from uncertainties due to absolute pressure measurements and fitting procedures. Subsequently, our data have been normalized to the values of Straub et al. [20]. We considered the total ionization cross-section as $\sigma_{tot} = \Sigma_n n\sigma_{n+}$ and $I^{tot} = \Sigma_n n I^{n+}$. Fig. 2 shows the a time-of-flight spectrum of the species detected. The ratios $R_n = \sigma_{n+}/\sigma_{+}$ for n = 7,8 measured at 2 keV impact-energy are presented with the available data from Schram et al. [28] in Table 1.

The total experimental uncertainties of the normalized ionization cross-section (NICS) result

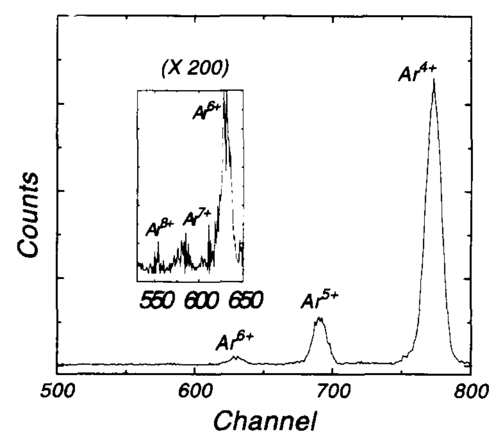

Fig. 2. Time-of-flight spectrum of Ar^{n+} ionized by 2 keV electrons.

Table 1
The ratios $R_n = \sigma_{n+}/\sigma_+$ for $n = 7, 8$ measured at 2 kev electron-impact energy

R_n	Present work	Ref. [28]
R_2	$(4.35 \pm 0.02) \times 10^{-2}$	$(4.4 \pm 0.2) \times 10^{-2}$
R_3	$(2.65 \pm 0.06) \times 10^{-3}$	–

from the statistical errors added in quadrature for the quantities used in the normalization procedure. The statistical counting uncertainties for I^{n+}/I^{tot} ($n = 7,8$) were approximately 12%, and 18%, respectively, at each electron impact energy after subtracting the background. The energy-spread of the incident electron beam was around 2 eV. The data from Straub et al. [20] are reliable within 3.5% (as quoted by the authors). Considering standard error reduction for independent uncertainties the overall uncertainty of the present NICS is about 14% and 19% for σ_{7+} and σ_{8+}, respectively. The results are presented in Table 2.

For σ_{7+} (Fig. 3) the only experimental result found in the literature in the present range of incident energy is from Schram et al [28]. The present data agree well with those of Ref. [28]. For σ_{8+}, (Fig. 4), to the best of our knowledge, the present data are the first results in this energy range.

The asymptotic expression for electron-collision cross-sections derived for energies higher than the maximum cross-section, was fitted to the Ar^{7+} data and is displayed in the form of a Fano–Bethe plot in Fig. 5. Our results for Ar^{7+} are shown to be proportional to $\ln E/E$ confirming that the process is strongly dominated by an optically allowed process. On the other hand, σ_{8+} has a $1/E$ dependence and the process is therefore dominated by ejection of eight electrons. For Ar^{8+} we did not have enough experimental points above the maximum region for a reasonably accurate plot of the asymptotic cross-section shape. No Auger features were observed in either spectrum in the energy-range studied.

Table 2
Partial ionization cross-section of argon by electron collision. The uncertainties are described in the text

Energy (eV)	σ_{7+} (10^{-22} cm^2)	σ_{8+} (10^{-22} cm^2)
1100	0.36	–
1300	1.05	–
1500	0.98	–
1700	1.50	0.30
1900	1.44	0.85
2000	1.67	0.87
2200	1.81	1.57
2400	1.98	1.26
2600	1.91	1.40
2800	2.01	1.61
3000	1.94	1.50

The integrated oscillator strength for Ar^{7+} (M^2_{7+}) was found to be $(1.50 \pm 0.161) \times 10^{-5}$. This value compares well with the value of 1.92×10^{-5} reported by Schram et al [28].

4. Comparison with calculations

In Figs. 3 and 4 we also show cross-section values obtained from three semi-empirical methods, the approach of Fisher et al. [12], the method of Shevelko and Tawara [13] and the DM formalism of Deutsch et al. [14,30]. The data based on the method of Fisher et al. [12] were previously used in a comparison with our earlier measured results for Ar^{n+} (n = 3–6) as discussed in the paper of Almeida et al. [5]. For n = 3 the calculated values were higher than the experimental data by about 50%, whereas good agreement was found for n = 4–6. In the present case of Ar^{7+} and Ar^{8+}, the calculated values based on the formalism of Fisher et al. [12] are lower than the measured cross-sections by factors of about 2.5 and 30, respectively. Shevelko and Tawara [13] have presented a semi-empirical formula for multiple ionization cross-sections for atoms and ions by electron impact deduced on the basis of the Bethe–Born dependence of σ_n on the incident energy. Their simple formula depends only on three atomic parameters (the minimal ionization potential, the atomic number of the target atom and the degree of ionization).

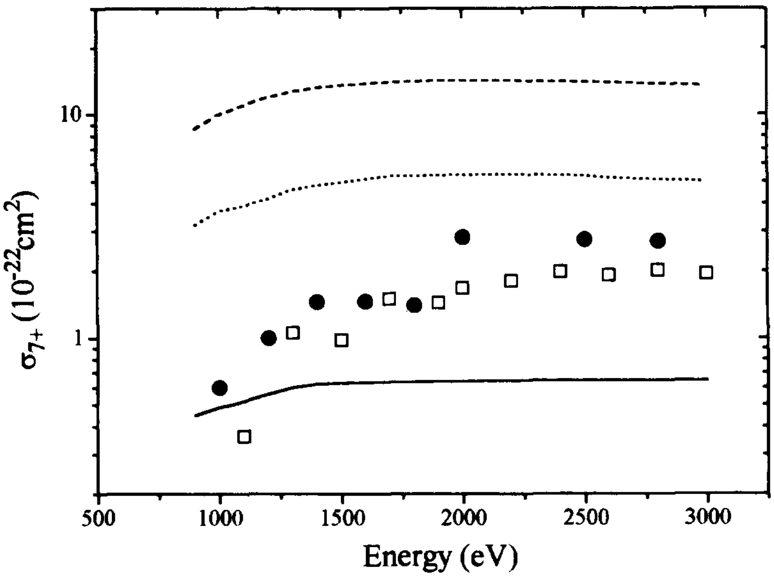

Fig. 3. Electron-impact ionization cross-section σ_{7+} as a function of the incident energy. □ present data; ● Schram et al. [28]; ——— Fisher et al. [12]; - - - Shevelko and Tawara [13]; · · · Deutsch et al. [15].

Their calculated cross-sections for Ar^{7+} and Ar^{8+} are higher than our experimental values by factors of 5 and 3, respectively.

Using a different concept, Deutsch et al. [14,15,30] have recently extended the DM formalism [11], which was originally derived for the single ionization of an atom by electron impact, to atomic multiple ionization. In the case of the formation of highly charged rare-gas ions the semi-empirical fitting procedure used in the

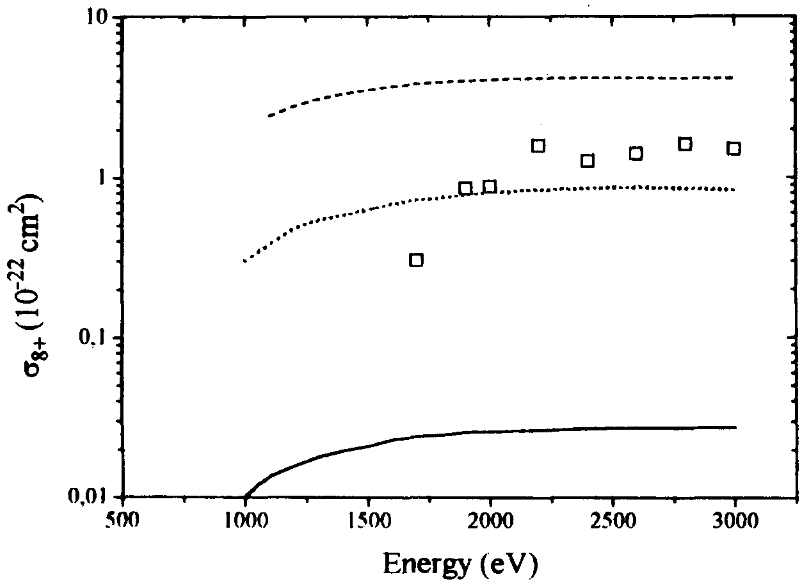

Fig. 4. Electron-impact ionization cross-section σ_{8+} as a function of the incident energy. □ present data; ——— Fisher et al. [12]; - - - Shevelko and Tawara [13]; · · · Deutsch et al. [15].

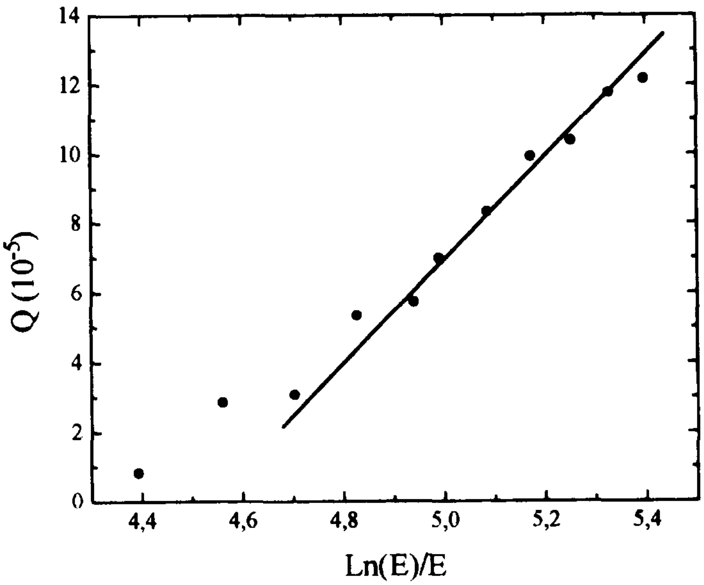

Fig. 5. Fano–Bethe plot for the process e+Ar ⇒ Ar^{7+}+8e. The quantity Q is defined as $\sigma_{n+}E/(R4\pi a_0^2)$.

DM formalism [14,30] has to be slightly modified as will be discussed in a forthcoming publication [15]. Using this modified fitting procedure, we were able to calculate cross-sections for the formation of Ar^{7+} in the framework of the DM formalism. As can be seen in Figs 3 and 4, the calculated Ar^{7+} cross-section is about 3 times higher than the present measured values, whereas there is good agreement between the DM calculations and the present measurements for Ar^{8+}.

5. Conclusions

The present result for σ_{7+} for argon are in good agreement with the only other measurement in the literature. The ionization cross-section for formation of Ar^{8+} is reported here for the first time. Recent efforts have been directed toward the study of multiple ionization cross-sections such as Ar^{n+} with $n > 4$ and despite the obvious difficulty of the measurements, the results have provided a better understanding of the ionization process.

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