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Electron-impact excitation of argon: II. The lowest resonance $4s\left[\frac{3}{2}\right]_1$ and metastable $4s\left[\frac{3}{2}\right]_2$ and $4s'\left[\frac{1}{2}\right]_0$ states

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Abstract. Absolute normalized differential electron-impact excitation cross sections (DCSs) are presented for the lowest three electronic states of argon: the lowest resonance $4s\left[\frac{3}{2}\right]_1$ ($1s_4$ in Paschen's notation) state and two neighbouring metastable $4s\left[\frac{3}{2}\right]_2$ and $4s'\left[\frac{1}{2}\right]_0$ ($1s_5$ and $1s_3$ in Paschen's notation) states. Direct, i.e. free of a cascade contribution, excitation cross sections were obtained experimentally for the $4s\left[\frac{3}{2}\right]_1$ state at 20, 40, 50 and 80 eV and for the $4s\left[\frac{3}{2}\right]_2$ and $4s'\left[\frac{1}{2}\right]_0$ states at 20 and 40 eV impact energies. The measurements were performed at scattering angles between 5° and 150° with an angular resolution of better than 2°. The ratios: $\boldsymbol{r} = \mathrm{DCS}\left(4s\left[\frac{3}{2}\right]_2\right)/\mathrm{DCS}\left(4s'\left[\frac{1}{2}\right]_0\right)$ and $\boldsymbol{r}' = \mathrm{DCS}\left(4s\left[\frac{3}{2}\right]_1\right)/\mathrm{DCS}\left(4s'\left[\frac{1}{2}\right]_1\right)$ were determined and compared with other available experimental and theoretical results. The absolute DCS scale was established with respect to DCSs of the $4s'\left[\frac{1}{2}\right]_1$ state (Filipović *et al* 2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** 677) using the peak intensity ratios in the energy-loss spectra. The DCSs were extrapolated to 0° and 180° and numerically integrated to yield integral, momentum transfer and viscosity cross sections.

1. Introduction

This paper is the second in a series of articles dealing with electron-impact excitation of argon. In the first paper we measured the cross sections for excitation of the $4s'\left[\frac{1}{2}\right]_1$ resonance, $4p\left[\frac{1}{2}\right]_1$ and $4p'\left[\frac{1}{2}\right]_0$ states (Filipović *et al* 2000). Complementary $4s\left[\frac{3}{2}\right]_1$ (the lowest resonance, $1s_4$ in Paschen's notation) and two neighbouring metastable $4s\left[\frac{3}{2}\right]_2$ and $4s'\left[\frac{1}{2}\right]_0$ states ($1s_5$ and $1s_3$ in Paschen's notation) are reported here. The $4s\left[\frac{3}{2}\right]_1$ state is a component of a triplet ^3P_J ($J = 2, 1, 0$) state if the Russell–Saunders (LS) coupling scheme is applied, but it is, in fact, a resonance state, with a lifetime of 1.0×10^{-8} s (Edgell 1961). Excitation functions for resonance states are typically smooth with a broad maximum. Transitions from the $4s\left[\frac{3}{2}\right]_2$ and $4s'\left[\frac{1}{2}\right]_0$ states to the ground state ($J = 0$) are optically forbidden. If the lifetime is longer than 10^{-5} s, the state is a metastable one. According to this criterion the $4s\left[\frac{3}{2}\right]_2$ and $4s'\left[\frac{1}{2}\right]_0$ are 'very long-lived' metastable states because of their lifetimes of 55.9 and 44.9 s, respectively (Small-Warren and Lue-Yung 1975). In electron spectra recorded under the conditions of higher impact energies and small scattering angles (optical-like spectra), peaks corresponding to these states are of very low relative intensities.

There are two different experimental approaches for electron-impact excitation of noble gases. One is direct (free of a cascade contribution) electron scattering by a target atom, as

Table 1. Summary of electron–argon cross section measurements: DCS (differential), Q_i (integral), Q_m (momentum transfer), Q_v (viscosity), $\boldsymbol{r} = \mathrm{DCS}\left(4\mathrm{s}\left[\frac{3}{2}\right]_2\right)/\mathrm{DCS}\left(4\mathrm{s}'\left[\frac{1}{2}\right]_0\right)$ and $\boldsymbol{r}' = \mathrm{DCS}\left(4\mathrm{s}\left[\frac{3}{2}\right]_1\right)/\mathrm{DCS}\left(4\mathrm{s}'\left[\frac{1}{2}\right]_1\right)$.

Reference	E_0 (eV)	θ (deg)	Quantities
Tam and Brion (1973)	30	5–90	Relative DCS, $\boldsymbol{r}'$
McConkey and Donaldson (1973)	20–200		Q_i
Chutjian and Cartwright (1981)	16, 20, 30, 50, 100	5–138	DCS, Q_i, Q_m
Mason and Newell (1987)	Onset–140		Q_i
Khakoo *et al* (1992)	30	10–120	$\boldsymbol{r}$, $\boldsymbol{r}'$
Khakoo *et al* (1994)	30	0–50	$\boldsymbol{r}$, $\boldsymbol{r}'$
Mityureva and Smirnov (1994)	Onset–60		Q_i
Schappe *et al* (1994)	Onset–100		Q_i
Tsurubuchi *et al* (1996)	Onset–1000		Q_i
Present	20, 40, 50, 80	5–150	DCS, Q_i, Q_m, Q_v

used by Tam and Brion (1973). They measured DCSs for the $4\mathrm{s}\left[\frac{3}{2}\right]_2$ and $4\mathrm{s}\left[\frac{3}{2}\right]_1$ states as well as the $4\mathrm{s}'\left[\frac{1}{2}\right]_1$ and 4p states, limited to the forward hemisphere. The second approach is an optical method of apparent cross section measurement as performed by McConkey and Donaldson (1973). They obtained the integral (Q_i) cross section of the $4\mathrm{s}\left[\frac{3}{2}\right]_1$ state using measured absolute apparent excitation functions of both the $4\mathrm{s}\left[\frac{3}{2}\right]_1$ and $4\mathrm{s}'\left[\frac{1}{2}\right]_1$ states. The cascade contribution was estimated to be the difference between the $4\mathrm{s}\left[\frac{3}{2}\right]_1$ function and that of the $4\mathrm{s}'\left[\frac{1}{2}\right]_1$ state when normalizing to the $4\mathrm{s}\left[\frac{3}{2}\right]_1$ magnitude above an incident electron energy (E_0) of 200 eV.

Experimental techniques and theoretical methods applied in electron-impact excitation of the lowest two metastable states of argon are discussed by Fabrikant *et al* (1988). Only measurements by Chutjian and Cartwright (1981) cover scattering in both forward and backward hemispheres. They performed an experiment where the 4s and 4s′ states were resolved. Mason and Newell (1987) utilized a time-of-flight technique for measuring the total metastable integral cross sections for the $4\mathrm{s}\left[\frac{3}{2}\right]_2 + 4\mathrm{s}'\left[\frac{1}{2}\right]_0$ states. Khakoo *et al* (1994) re-analysed their measurements at $E_0 = 30$ eV (Khakoo *et al* 1992) and found that the experimental ratio $\boldsymbol{r} = \mathrm{DCS}\left(4\mathrm{s}\left[\frac{3}{2}\right]_2\right)/\mathrm{DCS}\left(4\mathrm{s}'\left[\frac{1}{2}\right]_0\right)$ corresponds closely to the statistical weight ratio of 5 at all scattering angles covered. Also, they found a significant deviation of both the experimentally and theoretically obtained ratio $\boldsymbol{r}' = \mathrm{DCS}\left(4\mathrm{s}\left[\frac{3}{2}\right]_1\right)/\mathrm{DCS}\left(4\mathrm{s}'\left[\frac{1}{2}\right]_1\right)$ from the value of approximately 0.3, obtained by Tam and Brion (1973) for scattering angles between 0° and 90° at the same impact energy.

Tsurubuchi *et al* (1996) utilized an optical polarized-free method in measurements of electron-impact emission cross sections. To obtain direct electron-impact excitation cross sections for the $4\mathrm{s}\left[\frac{3}{2}\right]_1$ and $4\mathrm{s}'\left[\frac{1}{2}\right]_1$ resonance states they subtracted the total cascade contribution to these states. Mityureva and Smirnov (1994) measured Q_i for metastable $4\mathrm{s}\left[\frac{3}{2}\right]_2$ and $4\mathrm{s}'\left[\frac{1}{2}\right]_0$ states using an optical absorption method. Using a laser-induced fluorescence technique, Schappe *et al* (1994) obtained electron-impact excitation cross sections for argon metastable states separately, subtracting the cascade contribution from the apparent cross section values. A survey of experimental work is presented in table 1.

Padial *et al* (1981) calculated cross sections for the lowest four electronic states of argon using first-order many-body theory (FOMBT). Utilizing the relativistic distorted-wave Born approximation (RDWBA), Bartschat and Madison (1987) considered the importance of relativistic effects in the description of the target states as well as in the wavefunction

Table 2. Summary of electron–argon cross section calculations: FOMBT (first-order many-body theory), DWBA (distorted-wave Born approximation), RDWBA (relativistic DWBA), CRDWA (complete relativistic distorted-wave approximation), SRDWBA (semi-relativistic DWBA), SRRM (semi-relativistic R-matrix calculation).

Reference	E_0 (eV)	Theory	Quantities
Padial *et al* (1981)	16, 20, 30, 50, 80.4	FOMBT	DCS, Q_i, Q_m
Bartschat and Madison (1987)	16, 20, 30, 50, 80, 100	RDWBA	DCS
Bartschat and Madison (1992)	30	RDWBA	DCS, $\boldsymbol{r}$, $\boldsymbol{r}'$
Zuo *et al* (1992)	15–500	CRDWA	DCS
Griffin *et al* (1995)	Threshold–20	R-matrix	Q_i
Madison *et al* (1998)	Threshold–150	SRDWBA	DCS, Q_i
Zeman *et al* (1998)	15, 20, 30	SRRM	DCS, Q_i

for the continuum electron for the lowest two resonance states. Bartschat and Madison (1992) extended their RDWBA calculations to the lowest two metastable states, emphasizing the results of $\boldsymbol{r}$ and $\boldsymbol{r}'$. Zuo *et al* (1992) exploited a complete relativistic distorted-wave approximation (CRDWA) in a calculation of DCSs of the lowest four excited states of argon. Griffin *et al* (1995) applied the R-matrix method in which the effects of dipole polarizability are included for the 4s $\left[\frac{3}{2}\right]_2 + 4\mathrm{s}' \left[\frac{1}{2}\right]_0$ metastable states. Zeman *et al* (1998) used the R-matrix method in their semi-relativistic approach (SRRM) in the consideration of the lowest four excited states of argon. Madison *et al* (1998) utilized a semi-relativistic first-order distorted-wave Born approximation (SRDWBA) for the lowest 12 states of argon. A survey of theoretical work is presented in table 2. As one can see from tables 1 and 2, direct measurements following the new calculations are very scarce. Actually, direct measurements of electron-impact excitation cross sections of the lowest three individual (experimentally resolved) electronic states of argon are limited to the measurements by Chutjian and Cartwright (1981) and Khakoo *et al* (1992, 1994). As Bartschat and Madison (1992) pointed out, the experimental data exhibit significant disagreement between each other. Thus, we measured the direct electron-impact excitation cross sections within a large angular range using an electron spectrometer with high enough energy and angular resolution to resolve the lowest electronic states of argon.

In this paper we report DCSs, $\boldsymbol{r}$ and $\boldsymbol{r}'$, in order to compare our data with both experimental and theoretical results. The large angular range enabled a more accurate extrapolation of DCSs to 0° and 180° with the aim of performing numerical integration to give integrated (Q_i), momentum transfer (Q_m) and viscosity (Q_v) cross sections. These cross sections, as an addition to those reported in Filipović *et al* (2000), complete a reliable set of cross sections for individual states of argon.

2. Apparatus and experimental procedure

The experimental set-up used in these measurements was an electron-impact spectrometer previously described in Filipović *et al* (1988). Briefly, an atomic beam effusing through the platinum–iridium needle in a vertical direction was cross-fired perpendicularly by a monoenergetic electron beam. Electrons scattered at a given angle, defined with respect to the primary electron beam direction, were accepted by the analyser, which was movable around the atomic beam. Both the monoenergizer and the analyser consist of input and output gold-plated cylindrical electrostatic lenses and a hemispherical molybdenum electrostatic energy selector. A hairpin thoriated tungsten cathode was used as the electron source. A channel electron multiplier in single-electron counting mode was employed as a detector. The spectrometer operates in energy-loss (ΔE) mode.

The energies of the lowest four states of argon lie within 0.5 eV. The first three states are the subject of this work and the energy-loss feature of the fourth was used as a reference peak for the absolute DCS calibration. The overall energy resolution of 45 meV (FWHM) was sufficiently high to resolve all four peaks in the energy-loss spectra. The angular resolution was better than 2°. In the studied range of residual electron energies ($E_0 - \Delta E$), the transmission of the analyser was independent of which of the four peaks the optics was focused on.

Relative DCSs were obtained as follows. For a given E_0 and ΔE, the analyser position was changed from −30° to 150° and the angular distribution of inelastically scattered electrons was measured. Data between −5° and +5° are not reliable due to the influence of the primary electron beam. The real zero scattering angle was determined according to the symmetry of the scattering intensity around the instrumental zero. A correction of the intensity distribution was made due to the angular dependence of the effective interaction volume (Brinkmann and Trajmar 1981). Data points at each scattering angle were determined as a weighted arithmetic mean of from three to five intensity distributions. In the case of the metastable states at $E_0 = 40$ eV, only energy-loss spectra were utilized (measurements of relative DCSs were not carried out due to low signal intensities). The obtained relative DCSs were transferred to the absolute scale according to the intensity ratios of particular peaks and the reference peak. These ratios were determined from energy-loss spectra.

3. Results

Direct, i.e. free of cascade contribution, excitation cross sections were obtained experimentally for the 4s $\left[\frac{3}{2}\right]_1$ resonance and the 4s $\left[\frac{3}{2}\right]_2$ and 4s′ $\left[\frac{1}{2}\right]_0$ metastable states. Energy-loss spectra between 11.5 and 12 eV were taken at numbered energies and scattering angles (table 1). Representative energy-loss spectra at 40 eV, 10°, 'optical-like', and at 20 eV, 150° are shown in figure 1. The designation of peaks in the spectra is based on Moore's tables (Moore 1958). Relative DCSs were transferred to the absolute scale in accordance with energy-loss spectra and DCSs of the 4s′ $\left[\frac{1}{2}\right]_1$ reference state previously published by Filipović *et al* (2000). Because it is difficult to discuss errors related to each data point, we estimated the level of errors for DCSs rather than errors at a particular point.

3.1. The 4s$\left[\frac{3}{2}\right]_1$ resonance state

DCSs of the 4s $\left[\frac{3}{2}\right]_1$ state were obtained experimentally at 20, 40, 50 and 80 eV and between 5° and 150° scattering angles. These DCSs are presented in table 3, and shown in figure 2 together with other data. The level of error for DCSs is estimated to be 0.30 (30%). It is obtained as a square root of the sum of squares of particular contributing errors. Contributions to this value are: 0.28 in DCSs of the 4s′ $\left[\frac{1}{2}\right]_1$ reference state, 0.05 due to statistical error, 0.07 due to path-length correction and 0.08 due to absolute scale determination.

Integrated cross sections: Q_{i}, Q_{m} ($n = 1$) and Q_{v} ($n = 2$), defined as

$$Q_{\mathrm{i}}(E_0) = 2\pi \int_0^{\pi} \mathrm{DCS}(\theta) \sin\theta \,\mathrm{d}\theta \qquad (1)$$

$$Q_{\mathrm{m(v)}}(E_0) = 2\pi \int_0^{\pi} \mathrm{DCS}(\theta) \left[1 - \left(1 - \frac{\Delta E}{E_0}\right)^{n/2} \cos^n\theta\right] \sin\theta \,\mathrm{d}\theta \qquad (2)$$

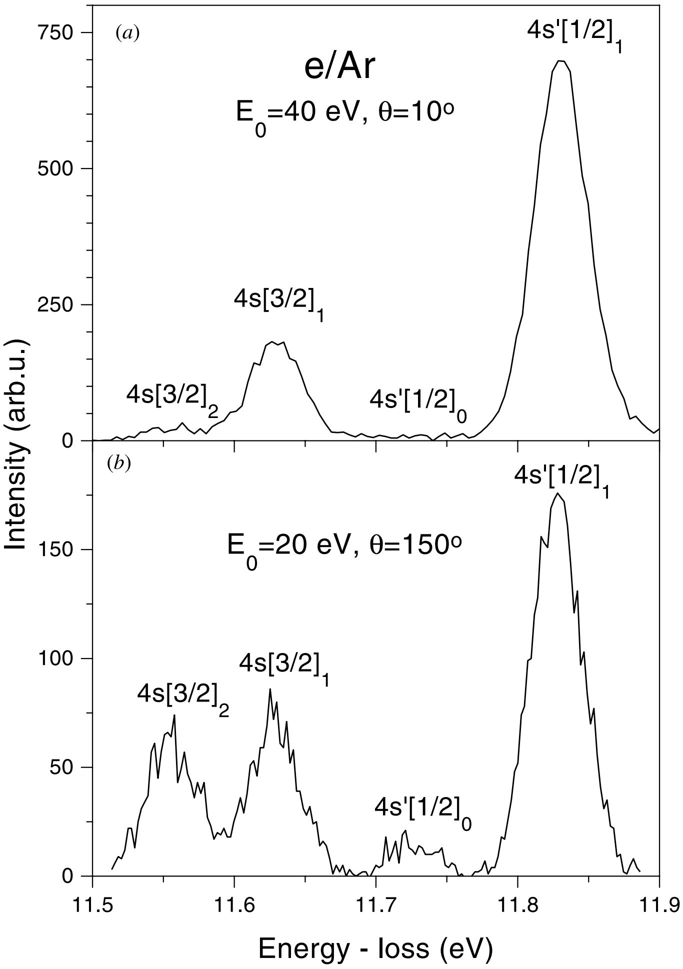

Figure 1. Energy-loss spectra of argon: (*a*) higher impact energy ($E_0 = 40$ eV) and small scattering angle ($\theta = 10°$); (*b*) lower impact energy ($E_0 = 20$ eV) and large scattering angle ($\theta = 150°$).

where ΔE is the excitation energy of the given state, were obtained by numerical integration of the DCSs presented in table 3 and extrapolated to 0° and 180°. The extrapolation procedure was based on the least-square method of fitting experimental data points by polynomial functions. The range of experimental points used in the fitting procedure was from the nearest structure in DCS towards a 0° or 180° scattering angle. The integrated cross section values are presented in table 4 together with other data for comparison. The level of error of these cross sections is estimated to be 0.33. Contributions to this value are 0.30 due to the error of the DCSs, and 0.15 due to extrapolation to 0° and 180° and numerical integration.

3.2. The $4s\left[\frac{3}{2}\right]_2$ *and* $4s'\left[\frac{1}{2}\right]_0$ *metastable states*

DCSs of $4s\left[\frac{3}{2}\right]_2$ and $4s'\left[\frac{1}{2}\right]_0$ were obtained experimentally at 20 and 40 eV and between scattering angles of 10° and 150°. These DCSs are presented in table 5, and shown in figures 3

Table 3. Electron–argon DCSs for the 4s $\left[\frac{3}{2}\right]_1$ excitation in units of 10^{-23} m^2 sr^{-1}.

	E_0 (eV)			
θ (deg)	20	40	50	80
5			529	470
10	40.2	182	165	155
15	27.0	107	70.3	37.5
20	16.8	46.1	24.3	7.80
30	8.16	17.4	7.59	2.64
40	6.14	8.98	4.13	0.917
50	5.71	4.71	2.13	0.378
60	5.27	2.22	0.877	0.202
70	4.24	1.27	0.449	0.124
75			0.390	0.107
80	3.19	1.06	0.409	0.137
90	2.71	1.18	0.732	0.314
95				0.356
100	2.37	1.39	0.940	0.310
110	2.94	1.49	1.18	0.197
120	3.6	1.29	0.985	0.101
130	4.19	0.860	0.441	0.066
140	5.17	0.648	0.338	0.111
150	6.32	0.827	0.445	0.336

Table 4. Integrated cross sections for the 4s $\left[\frac{3}{2}\right]_1$ state of argon in units of 10^{-23} m^2.

	E_0 (eV)			
	20	40	50	80
Integral				
Experiment				
McConkey and Donaldson (1973)	50	83	92	86
Chutjian and Cartwright (1981)	37.0 ± 10.7		57.8 ± 16.8	
Tsurubuchi *et al* (1996)	15 ± 2	68 ± 9	62 ± 8	50 ± 7
Present	69 ± 23	93 ± 31	74 ± 24	53 ± 17
Theory				
Padial *et al* (1981)	78.4		71.5	63.8
Bartschat and Madison (1987)	101		70	57
Zuo *et al* (1992)	103	51.9	51.1	44.4
Madison *et al* (1998)	167	99.6	97.4	85.7
Zeman *et al* (1998)	63			
Momentum transfer				
Experiment				
Chutjian and Cartwright (1981)	36.7 ± 11.4		9.4 ± 2.9	
Present	60 ± 20	31 ± 10	20 ± 7	10.2 ± 3.4
Theory				
Padial *et al* (1981)	64.7		18.8	9.36
Viscosity				
Present	54 ± 18	42 ± 14	26 ± 9	12 ± 4

and 4 together with other data. The level of error for the DCSs of the 4s $\left[\frac{3}{2}\right]_2$ state is 0.30, with the same consideration as in the case of the 4s $\left[\frac{3}{2}\right]_1$ state. Integrated cross sections for

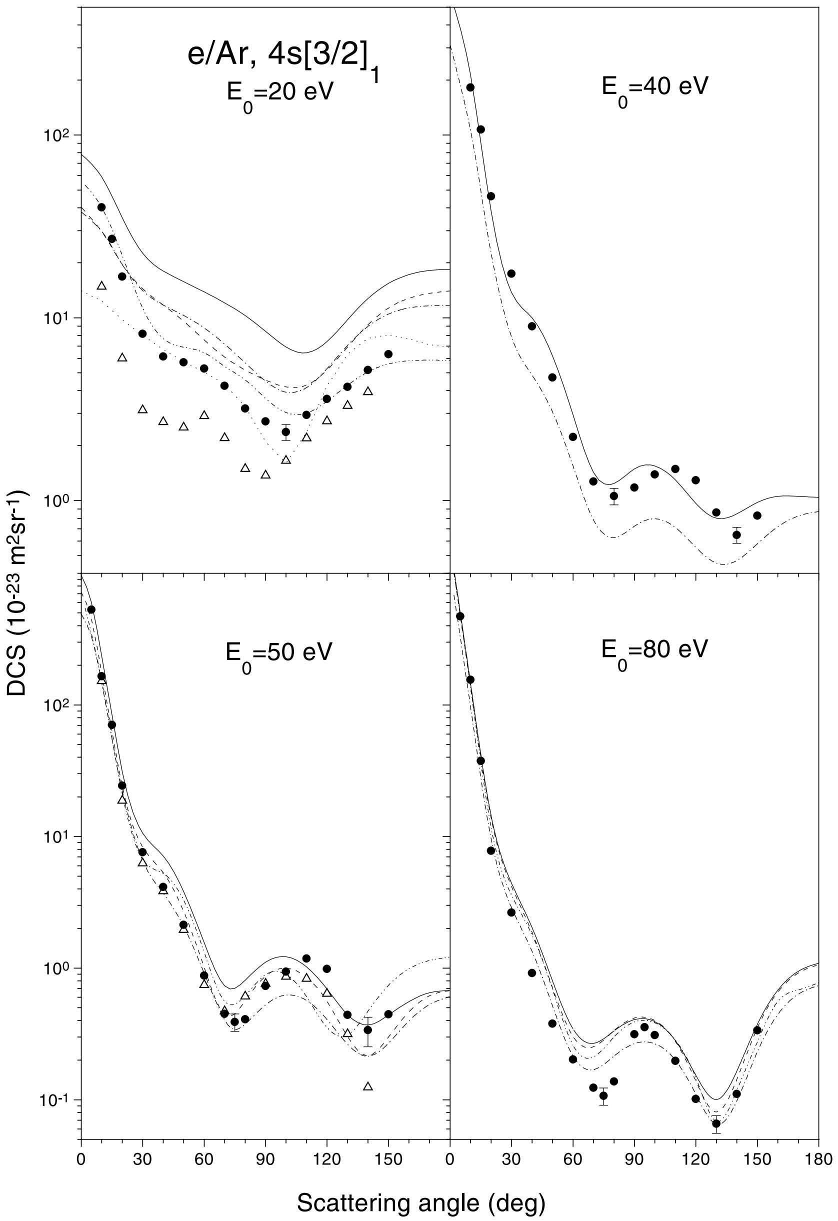

Figure 2. Differential cross sections for the excitation of the 4s $\left[\frac{3}{2}\right]_1$ resonance state at incident electron energies of 20, 40, 50 and 80 eV. Experiments: ●, present (statistical error bars are indicated); △, experiment of Chutjian and Cartwright (1981). Calculations: ——, SRDWBA Madison *et al* (1998); · · · · · ·, SRRM Zeman *et al* (1998); — · —, CRDWA Zuo *et al* (1992); – – –, DWBA Bartschat and Madison (1987); — · · —, FOMBT Padial *et al* (1981).

this state were determined by extrapolation of DCS values from table 5, to 0° and 180° and numerical integration. The level of error for this state is estimated to be 0.33.

The 4s′ $\left[\frac{1}{2}\right]_0$ peak has the lowest intensity in the energy-loss spectra at all scattering angles. Thus statistical errors are generally higher and they are indicated in figure 4. Other contributing

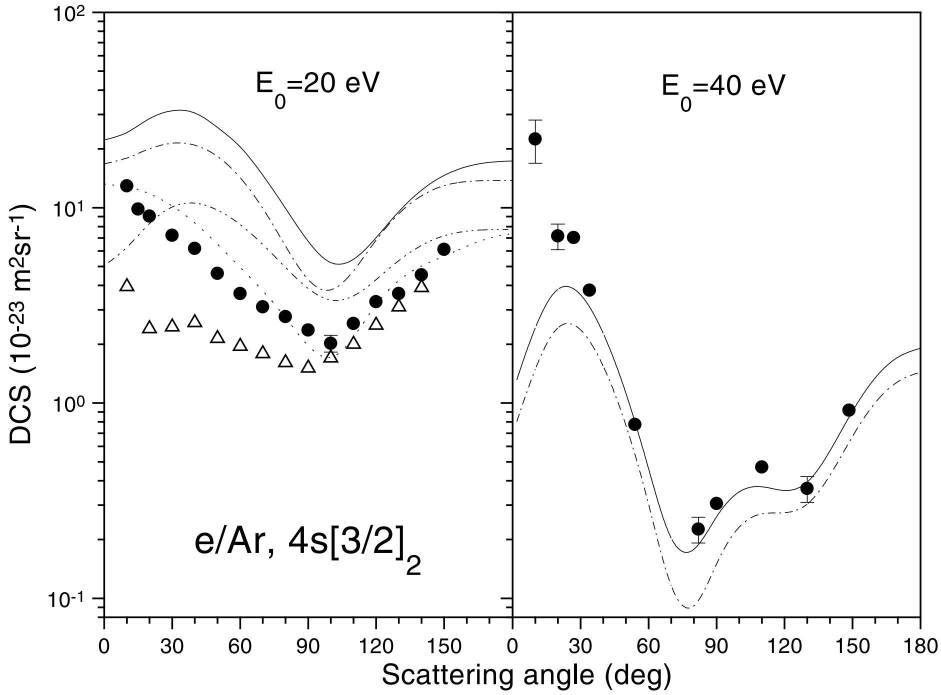

Figure 3. Differential cross sections for the excitation of the 4s $\left[\frac{3}{2}\right]_2$ metastable state at 20 and 40 eV. Experiments: ●, present (statistical error bars are indicated); △, Chutjian and Cartwright (1981). Calculations: ——, SRDWBA Madison *et al* (1998); · · · · · ·, SRRM Zeman *et al* (1998); — · —, CRDWA Zuo *et al* (1992); — · · —, FOMBT Padial *et al* (1981).

errors to the absolute DCS error are the same as for the 4s $\left[\frac{3}{2}\right]_1$ state. Integrated cross sections were calculated by numerical integration of DCSs previously extrapolated to 0° and 180°. The levels of error for the integrated cross sections are 0.35 at 20 eV and 0.45 at 40 eV.

The present integrated cross section data for both the 4s $\left[\frac{3}{2}\right]_2$ and 4s′ $\left[\frac{1}{2}\right]_0$ metastable states are presented in table 6. Other integrated cross section data available in the literature are given in the same table.

3.3. DCS ratios $\boldsymbol{r}$ and $\boldsymbol{r}'$

We have obtained the intensity ratio $\boldsymbol{r}'$, defined in section 1, at all measured energies and scattering angles according to the energy-loss spectra. As an example, the electron energy-loss spectrum at 40 eV, 148° is presented in figure 5. The result at large scattering angles for this impact energy is important because theory (Zuo *et al* 1992, Madison *et al* 1998) predicts an increase in the value of $\boldsymbol{r}'$. Our result is the only experimental one in this range. To unfold the four energy-loss features, Gaussian instrumental profiles were applied for a fitting of the raw experimental data. A synthetic profile is indicated by the full curve on the broken baseline, which is included as a linear function in the unfolding procedure. To show the quality of the fitting we present the difference between the raw experimental data and the synthetic profile at the bottom in the figure. The height ratio of the 4s $\left[\frac{3}{2}\right]_1$ to 4s′ $\left[\frac{1}{2}\right]_1$ Gaussian profiles was used as a good approximation of $\boldsymbol{r}'$. Our $\boldsymbol{r}'$ values are presented in figure 6 (bottom, open circles) for the energies of 20, 40, 50 and 80 eV. Fortunately, the errors of $\boldsymbol{r}'$ are much smaller than those of the DCSs themselves because only statistical errors (of 0.05) and the errors due

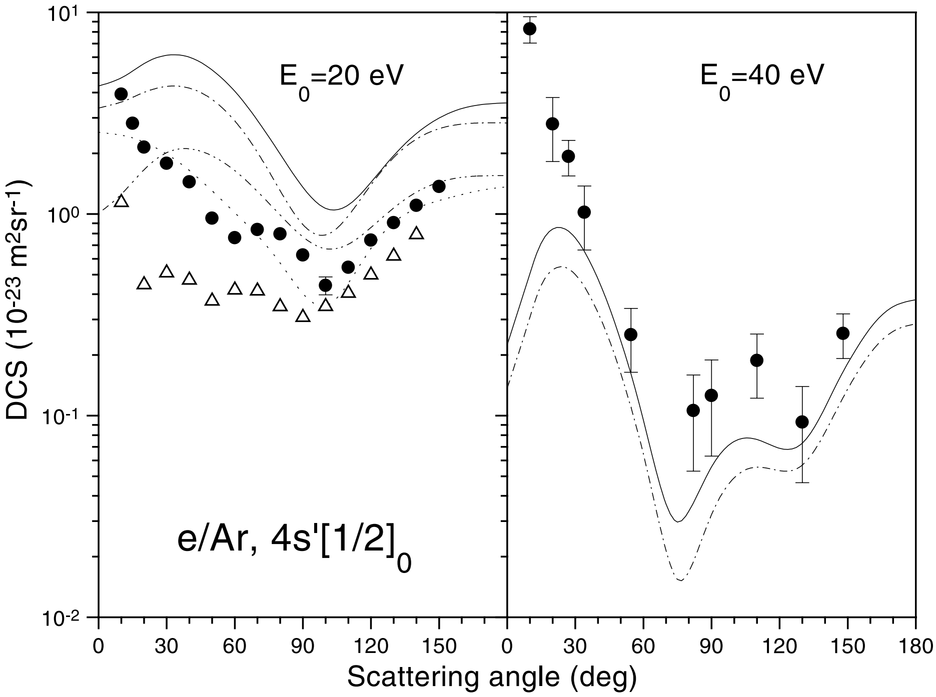

Figure 4. Differential cross sections for the excitation of the 4s′ $\left[\frac{1}{2}\right]_0$ metastable state at 20 and 40 eV. Experiments: ●, present (statistical error bars are indicated); △, Chutjian and Cartwright (1981). Calculations: ——, SRDWBA Madison *et al* (1998); · · · · · ·, SRRM Zeman *et al* (1998); — · —, CRDWA Zuo *et al* (1992); — · · —, FOMBT Padial *et al* (1981).

to energy and angular uncertainty are included in the determination of the total error. In this way we have established the level of error for r' to be 0.13. In positions of DCS minima the errors are larger. The error bars are indicated in the figure. The intensity ratio r', used for the absolute scale determination, is also important to test theoretically predicted branching ratios, as we will discuss in section 4.

Based on the absolute DCS values of the lowest two 4s $\left[\frac{3}{2}\right]_2$ and 4s′ $\left[\frac{1}{2}\right]_0$ metastable states we have considered their statistical branching ratios, r, at 20 and 40 eV, between 10° and 150°. We found that values of r are relatively close but smaller than the suspected value of 5. Our results of r are presented in figure 6 (top, full circles). Due to different statistical errors, the error bars are indicated for each data point.

4. Discussion and conclusion

The lowest three excited states of argon, for which the DCSs are presented in this paper, are designated in Racah notation as the 4s $\left[\frac{3}{2}\right]_2$, 4s $\left[\frac{3}{2}\right]_1$ and 4s′ $\left[\frac{1}{2}\right]_0$ states. The structure of excited states is given by j_h–l type coupling, where j_h is the ion–core angular momentum, and l is the orbital angular momentum of the excited electron. If the LS coupling scheme is applied, the 4s $\left[\frac{3}{2}\right]_1$ state is a component of a triplet state where generally nearly isotropic DCSs are expected. However, measured DCSs for this state are sharply forward peaked at each impact energy, typical for resonance states. Our results show a shoulder around 50° at 20 eV which is consistent with all available calculations, except the measurement by Chutjian

Table 5. Electron–argon DCSs for the $4s\left[\frac{3}{2}\right]_2$ and $4s'\left[\frac{1}{2}\right]_0$ excitation in units of 10^{-23} m^2 sr^{-1}.

	$4s\left[\frac{3}{2}\right]_2$		$4s'\left[\frac{1}{2}\right]_0$	
θ (deg)	20 eV	40 eV	20 eV	40 eV
10	12.9	22.5	3.93	8.28
15	9.84		2.82	
20	9.06	7.17	2.15	2.80
27		7.03		1.93
30	7.22		1.78	
34		3.78		1.02
40	6.19		1.44	
50	4.61		0.953	
54		0.777		0.252
60	3.64		0.763	
70	3.11		0.838	
80	2.77		0.797	
82		0.226		0.106
90	2.36	0.306	0.627	0.126
100	2.02		0.443	
110	2.55	0.470	0.544	0.188
120	3.30		0.742	
130	3.64	0.365	0.906	0.093
140	4.53		1.10	
148		0.919		0.256
150	6.12		1.37	

Table 6. Integrated cross sections for electron-impact excitation of $4s\left[\frac{3}{2}\right]_2$ and $4s'\left[\frac{1}{2}\right]_0$ states of argon in units of 10^{-23} m^2.

	$4s\left[\frac{3}{2}\right]_2$		$4s'\left[\frac{1}{2}\right]_0$	
	20 eV	40 eV	20 eV	40 eV
Integral				
Chutjian and Cartwright (1981)	31.9 ± 9.3		6.44 ± 2.13	
Padial *et al* (1981)	78.7		15.7	
Zuo *et al* (1992)	250	7.5	28	1.5
Mityureva and Smirnov (1994)	300	145	75	
Schappe *et al* (1994)	40 ± 12	7.6 ± 2.3	13.5 ± 3.4	3.4 ± 0.9
Madison *et al* (1998)	181	11	36	2.2
Zeman *et al* (1998)	59		12	
Present	53 ± 17	18 ± 6	12 ± 4	6.3 ± 2.9
Momentum transfer				
Chutjian and Cartwright (1981)	36.5 ± 12.5		7.27 ± 2.54	
Padial *et al* (1981)	72.6		14.5	
Present	50 ± 16	10.2 ± 3.4	12 ± 4	3.6 ± 1.6
Viscosity				
Present	42 ± 14	9.2 ± 3.0	10.1 ± 3.5	3.2 ± 1.4

and Cartwright (1981). All theories and both experiments show a higher angle minimum even though the minimum obtained by Chutjian and Cartwright (1981) is shifted towards a smaller angle. There are two DCS minima at 40, 50 and 80 eV in all calculations and both measurements. Very good agreement between the present data and the SRDWBA calculation

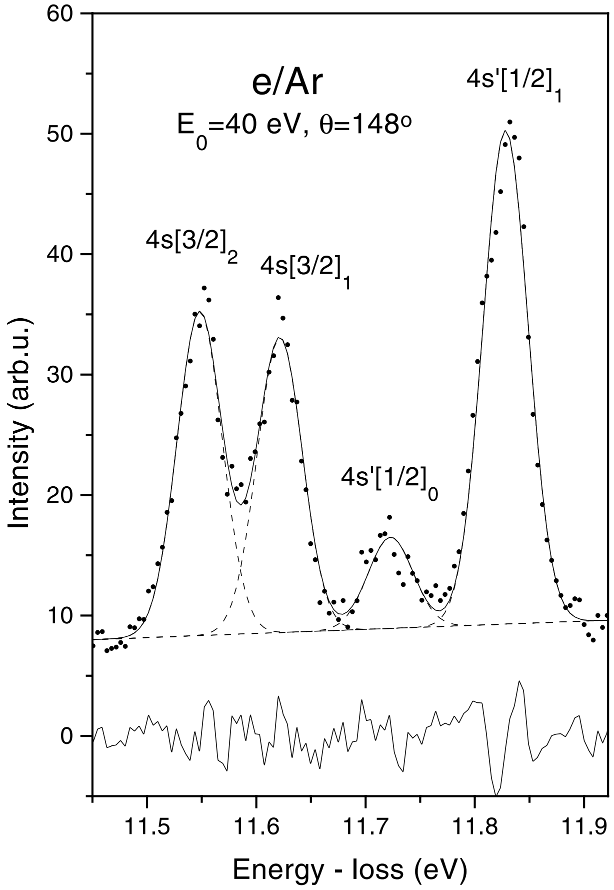

Figure 5. Electron energy-loss spectrum at $E_0 = 40$ eV and $\theta = 148°$: ●, raw data; – – –, unfolded Gaussian profiles on the baseline; ——, synthetic spectrum. The difference between raw data and synthetic spectrum is presented at the bottom.

(Madison *et al* 1998) is evident at 40 eV. Agreement between different calculations and both experimental results is very good at 50 eV. At 80 eV the agreement of our DCS result is very good with all calculations but compares best with the CRDWA calculation by Zuo *et al* (1992). Our absolute DCS values at all measured impact energies are in accordance with different calculations. Comparing our absolute DCS values with measurements by Chutjian and Cartwright (1981) our results are higher by approximately a factor of 1.5–2.5 at 20 eV. This difference arises from higher values of the reference state $4s'\left[\frac{1}{2}\right]_1$ used for normalization. At 40 eV, excellent agreement is achieved at all scattering angles except at the higher angle minimum. Integrated cross sections of the $4s\left[\frac{3}{2}\right]_1$ resonance state are in good agreement with other available results. There are only a few measurements of integral cross sections and they all have reasonably large errors due to the complexity of the experimental procedure. The agreement is satisfactory if one takes into account large error bars.

The DCS for the lowest $4s\left[\frac{3}{2}\right]_2$ metastable state at 20 eV is forward peaked according to both experiments and the SRRM calculation by Zeman *et al* (1998), while all DWBA calculations fall down at small scattering angles. There is one minimum in the present DCS at 100° that is consistent with all available calculations. In the measurements of Chutjian

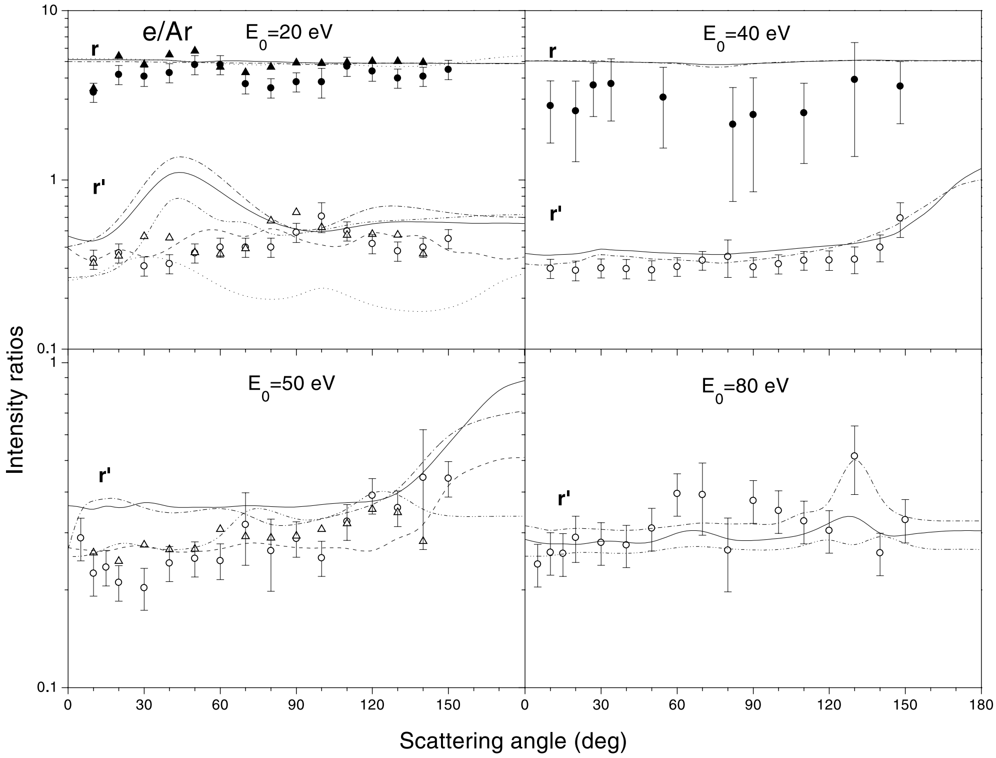

Figure 6. The ratios $r = \mathrm{DCS}\left(4s\left[\frac{3}{2}\right]_2\right)/\mathrm{DCS}\left(4s'\left[\frac{1}{2}\right]_0\right)$ and $r' = \mathrm{DCS}\left(4s\left[\frac{3}{2}\right]_1\right)/\mathrm{DCS}\left(4s'\left[\frac{1}{2}\right]_1\right)$ versus scattering angle at 20, 40, 50 and 80 eV. Experiments: ○ and ●, present r and r'; ▲ and △, Chutjian and Cartwright (1981). Calculations: ——, SRDWBA Madison *et al* (1998); · · · · · ·, SRRM Zeman *et al* (1998); — · —, CRDWA Zuo *et al* (1992); – – –, DWBA Bartschat and Madison (1987); — · · —, FOMBT Padial *et al* (1981).

and Cartwright (1981) the position of this minimum is shifted to 90° and the existence of the minimum at 20° is not confirmed by our experiment. The agreement in absolute DCS values is best with the SRRM calculations by Zeman *et al* (1998) over the whole angular range, but the experimentally obtained structure of DCS is richer than this theory predicts. In particular, at the scattering angle of 20° where we obtained an inflection point, there is no evidence of the structure in this calculation. The agreement exists with the absolute DCS measured by Chutjian and Cartwright (1981) at large scattering angles, while at lower scattering angles our results are systematically larger, mainly due to larger DCS values of the reference $4s'\left[\frac{1}{2}\right]_1$ state. At 40 eV a very good agreement between our DCS and the result of the SRDWB approximation by Madison *et al* (1998) is evident both in shape and absolute values except below 30° where our result is forward peaked.

The DCS for the $4s\left[\frac{3}{2}\right]_0$ metastable state is forward peaked according to both experiments and the SRRM calculation by Zeman *et al* (1998), while all DWBA calculations, similarly as for the $4s\left[\frac{3}{2}\right]_2$ state, turn down at small scattering angles. At 20 eV our DCS has two minima, at 60° and 100°, and a shoulder at 30°. With respect to these angles, the three DCS minima obtained by Chutjian and Cartwright (1981) are shifted by 10° towards smaller angles. The agreement in absolute DCS values is best with the SRRM calculations by Zeman *et al* (1998) at 20 eV and with the SRDWBA result by Madison *et al* (1998) at 40 eV. At this energy our data points have larger error bars because of the lower intensity of the peak in energy-loss spectra used for DCS determination.

Integrated cross sections for the metastable states are important quantities in many applications and that is why we calculated them from our DCSs despite the relatively large errors associated with these kinds of measurements. Agreement with the measurements of Chutjian and Cartwright (1981) and those of Schappe *et al* (1994) at 20 eV is within the experimental error bars, although it is better with the latter. At this energy the best agreement is obtained with the calculations of Zeman *et al* (1998) for both metastable states. At 40 eV our integral cross sections are larger than other results except for those of Mityureva and Smirnov (1994) who have values which are larger by an order of magnitude. In the evaluation of integrated cross sections one must bear in mind that DCSs are much more sensitive to imperfections in both experiments and theories.

In conclusion, calculated DCSs satisfactorily reproduce experimental data of the $4s\left[\frac{3}{2}\right]_1$ resonance state, as opposed to the $4s\left[\frac{3}{2}\right]_2$ and $4s'\left[\frac{1}{2}\right]_0$ metastable states at small scattering angles. It seems that new ideas related to long-range interactions must be included in order to reproduce the measured small-angle DCS shapes of metastable states.

As we mentioned in section 3, DCS ratios are needed not only for normalization purposes. For the excitation from the ground state the cross sections for two fine-structure states with total electronic angular momenta J_1 and J_2 are related by the statistical branching ratio

$$\frac{\mathrm{DCS}(J_1)}{\mathrm{DCS}(J_2)} = \frac{2J_1 + 1}{2J_2 + 1}.$$

If this relation were not valid, one would suspect that relativistic effects in projectile–target interactions and the energy dependence of the scattering operator due to different excitation energies of fine-structure levels must be considered (Bartschat and Madison 1992). Our results of r and r' at 20 eV are compared with the only available measurements by Chutjian and Cartwright (1981). From figure 6 it is clear that there is a good agreement between both experiments. The r values at this energy show a richer structure in experiments than in calculations, although the results of Chutjian and Cartwright (1981) are close to 5 (mean value of 4.87), while our value is 4.1 ± 0.6 averaged for all measured angles. Our r' results show good agreement with the experiments of Chutjian and Cartwright (1981) and with the calculations of Bartschat and Madison (1987), while there is a remarkable spread of results among other calculations. At 40 eV our results of r, obtained with relatively higher statistical errors, are systematically lower than theories predict. It is interesting to note that r' is isotropic up to 130° at 40 eV according to the experiment and the calculations. The theoretically predicted increase of r' at large scattering angles (Zuo *et al* 1992, Madison *et al* 1998) is confirmed by our measurements. There are no other available experimental results to compare with at this energy. At 50 eV we determined only r' values and the behaviour is similar to that at 40 eV. At 80 eV theoretical results for r' indicate two maxima around 70° and 130° that are observed experimentally for the first time in this work.

In conclusion, r values obtained in this experiment are close to but smaller than theoretically predicted values. Present r' values at 40, 50 and 80 eV are well reproduced by calculations, while at the lowest measured energy of 20 eV, there are certain differences among calculations, indicating that further sophisticated predictions must be undertaken.

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References

Bartschat K and Madison D H 1987 *J. Phys. B: At. Mol. Phys.* **20** 5839
——1992 *J. Phys. B: At. Mol. Opt. Phys.* **25** 4619
Brinkmann R T and Trajmar S 1981 *J. Phys. E: Sci. Instrum.* **14** 245
Chutjian A and Cartwright DC 1981 *Phys. Rev.* A **23** 2178
Edgell W F 1961 *Argon, Helium and the Rare Gases* vol I, ed G A Cook (New York: Interservice) p 97
Fabrikant I I, Shpenik O B, Snegursky A V and Zavilopulo A N 1988 *Phys. Rep.* **159** 1
Filipović D, Marinković B, Pejčev V and Vušković L 1988 *Phys. Rev.* A **37** 356
——2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** 677
Griffin D C, Pindzola M S, Gorczyca T W and Badnell N R 1995 *Phys. Rev.* A **51** 2265
Khakoo M A, Beckmann C E, Trajmar S and Csanak G 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 3159
Khakoo M A, Tran T D, Bordelon D and Csanak G 1992 *Phys. Rev.* A **45** 219
Madison D H, Maloney C M and Wang J B 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** 873
Mason N J and Newell W R 1987 *J. Phys. B: At. Mol. Phys.* **20** 1357
McConkey J W and Donaldson F G 1973 *Can. J. Phys.* **51** 914
Mityureva A A and Smirnov V V 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 1869
Moore C E 1958 *Atomic Energy Levels* vol I–III *(NBS Circular no 467)* (Washington, DC: US Govt Printing Office)
Padial N T, Meneses G D, da Paixao F J, Csanak G and Cartwright D C 1981 *Phys. Rev.* A **23** 244
Schappe R S, Schulman M B, Anderson L W and Lin C C 1994 *Phys. Rev.* A **50** 444
Small-Warren N E and Lue-Yung C C 1975 *Phys. Rev.* A **11** 1777
Tam W-C and Brion C E 1973 *J. Electron Spectrosc. Relat. Phenom.* **2** 111
Tsurubuchi S, Miyazaki T and Motohashi K 1996 *J. Phys. B: At. Mol. Opt. Phys.* **29** 1785
Zeman V, Bartschat K, Noren C and McConkey J W 1998 *Phys. Rev.* A **58** 1275 and Private communication
Zuo T, McEachran R P and Stauffer A D 1992 *J. Phys. B: At. Mol. Opt. Phys.* **25** 3393