

A measurement of the cross section for electron impact ionization of Ar^{2+} , Kr^{2+} and Xe^{2+}

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Abstract. The crossed electron-ion beams technique was used to measure absolute cross sections for single ionization of Ar^{2+} , Kr^{2+} and Xe^{2+} ions at electron energies ranging from threshold to 2000 eV. The total error in the cross sections at a 90% confidence level is estimated to be ± 4.8 , ± 4.9 and $\pm 7\%$ or less for Ar^{2+} , Kr^{2+} and Xe^{2+} respectively, at energies more than a few electron volts above threshold. In contrast to some previous measurements, the metastable contents of the ion beams were small even in the case of Xe^{2+} . All measured cross section curves show significant contributions from excitation-autoionization and possibly direct ionization of inner-shell electrons. There is evidence for resonance-excitation-double-autoionization in the case of Xe^{2+} . Comparisons are made with other available experimental and theoretical data. The present results for Ar^{2+} are in excellent agreement with those previously measured in our laboratory and elsewhere. For Kr^{2+} and Xe^{2+} generally good agreement with other experimental results is also observed. However, the clear structures previously noted by Danjo *et al* for all three ions between 100 and 200 eV are not apparent in our cross sections.

1. Introduction

The process of electron impact ionization of rare gas ions is fundamental to the understanding of collision mechanisms and has direct applications in astrophysics, atmospheric physics and laboratory plasma physics. The need for a more detailed understanding of the underlying processes within plasmas intended for controlled thermonuclear fusion has accelerated the study of ionization processes, both theoretically and experimentally. Accurate modelling of plasmas requires reliable cross section data for electron impact ionization of the impurity ions which are likely to occur in the plasma, including the ions of neon and argon which are introduced for controlling the temperature in the boundary region. Krypton gas may also be used in the next generation of Tokamak machines for the same purpose. Therefore, a reliable set of cross section measurements for rare gas ions of various charge states is required. The present series of measurements lie within a general programme of acquiring atomic collision data for fusion research.

Our previous ionization measurements on metal ions (Man *et al* 1987a, b, c) and on rare gas ions (Man *et al* 1987d) have revealed significant contributions to the total ionization from excitation-autoionization (in which an inner shell target electron is

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excited into an upper level with energy greater than the ionization threshold, and this excited ion then autoionizes) and from inner-shell ionization (in which an electron from an inner shell is removed directly). Such processes are known to dominate increasingly over outer-shell ionization when the charge state of the ions increases along an isoelectronic or isonuclear sequence (Müller *et al* 1980, Falk *et al* 1983, Griffin *et al* 1984, Achenbach *et al* 1984, Gregory *et al* 1983, 1986, 1987). In some cases of highly charged ions of high Z , resonance-excitation-double-autoionization (REDA, in which a temporary capture of an incident electron with simultaneous excitation of an inner-shell electron is followed by a decay of the resonance via double autoionization) could exceed direct ionization by at least an order of magnitude over a limited energy range (LaGattuta and Hahn 1981). For doubly-charged ions the process is not expected to be important, but our result for Xe^{2+} may suggest otherwise (section 3.4). The present measurements for Ar^{2+} , Kr^{2+} and Xe^{2+} constitute a systematic and thorough set of cross section data made at small energy intervals in the range from threshold to 2000 eV. This permits a detailed search for structures in the cross section curves from which contributions from the aforementioned indirect processes may be identified.

Electron impact ionization cross sections of doubly-charged rare gas ions reported in this paper have been measured before, but usually as part of an isoelectronic, isoionic or isonuclear sequence or other grouping (Müller *et al* 1980, Gregory *et al* 1985, Achenbach *et al* 1984, Griffin *et al* 1984, Gregory 1985, Diserens *et al* 1988, Mueller *et al* 1985). Relative cross sections for Ar^{2+} , Kr^{2+} and Xe^{2+} as a function of different populations of excited ions in the source have been reported by Latypov *et al* (1964), but relevant graphs, data and operational conditions necessary for comparisons are not given. The only whole set of absolute measurements for doubly-charged rare gas ions is by Danjo *et al* (1984). Their results show a large enhancement for the Ar^{2+} and Kr^{2+} curves at 160 and 140 eV respectively. Their Xe^{2+} curve, on the other hand, shows enhancements at several energies, including 160 eV. Their Ar^{2+} , Kr^{2+} and Xe^{2+} curves all show significant signals below the threshold energy, which indicates that there were substantial fractions of metastable or highly excited ions in their incident ion beams. It is important to examine the origin of the enhancements in the 150 eV region to establish whether they are due to excited ions in the target beam.

Several theoretical calculations have been made of cross sections for rare-gas multiply-charged ions. The Oak Ridge Group (e.g. Griffin *et al* 1982, 1984, Pindzola *et al* 1986) have used a distorted-wave theory which includes direct and indirect processes. The excitation-autoionization process is included and an attempt is made to estimate contributions from REDA. A detailed calculation of the cross section for Xe^{2+} (and other multiply-charged Xe ions) has been made by Griffin *et al* (1984), but we are not aware of similar calculations for Ar^{2+} or Kr^{2+} . Comparison of our results with the calculated Xe^{2+} cross section is made in section 3.4. Younger (1981a, b) has evolved a parametrized version of the distorted-wave exchange theory. For Ar^{2+} , only direct ionization of 3p and 3s electrons is included and excitation-autoionization is excluded from the calculation. The difficulties of calculating total ionization cross sections are increased by the occurrence of inner-shell and indirect processes for complex ions. Disagreements between theory and experiment are usually greatest a little above the threshold energy, which is a particularly important region in plasma physics.

Since these methods require complicated calculations and do not guarantee reliable results, semi-empirical formulae, which are easy to apply, have been developed for

the use of the plasma physics and astrophysics communities and have met with some success. The formula of Lotz (1967) has been widely used for singly- and low-charged ions and gives a reasonably good result for direct ionization in many cases. However, when the cross section is strongly influenced by indirect or inner-shell processes, as is normally the case for multiply-charged ions, the Lotz formula tends to underestimate the cross section, especially near threshold where excitation-autoionization of inner-shell electrons is prevalent.

In order to take some account of excitation-autoionization, Burgess and Chidichimo (1983) have presented another formula for multiply-charged ions based on the Lotz formula. A good prior knowledge of the autoionizing energy levels of an ion is needed before this formula can be applied, but a much better agreement in the low-energy region can be expected. In sections 3.2 and 3.3 the results of the Burgess and Chidichimo formula are compared with our experimental results. The accumulation of high-quality experimental data is a valuable test of the adjustable parameters in the semi-empirical formula and also permits an evaluation of the general applicability of this and other such formulae.

2. Experimental technique

The experiments were performed at the Culham Laboratory using the fast crossed electron-ion beams technique. Both the apparatus and technique used for our measurements are as described briefly in recent papers (Man *et al* 1987a, b, c) for the measurements on Mo^+ , Cr^+ and Ta^+ , and in detail by Montague *et al* (1984) in their report on measurements for ground-state helium atoms. The sputter ion source (described by Man *et al* (1987c)), usually used for producing metal ions, was used in the present experiments, but no voltage was applied to the sputter electrode and an appropriate gas was fed into the arc chamber. The focusing electron gun described by Diserens *et al* (1988) was used.

Data were taken with ion beam energies of 4 and 8 keV, except for Xe^{2+} , which was limited to 4 keV because of the available magnetic field intensity of the mass selecting magnets. Ion currents ranged between 0.2 nA for Xe^{2+} at 4 keV and 1.5 nA for Ar^{2+} at 4 keV. It was not possible to obtain sufficiently intense and stable Ar^{3+} , Kr^{3+} or Xe^{3+} ion beams from the source, so the particle detector (Johnston type MM1) was calibrated using Ar^{2+} , Kr^{2+} and Xe^{2+} beams of the same velocities as their respective triply-charged ions. The detection efficiency of electron multipliers of the type used in this experiment is more dependent upon ion velocity than energy or charge state of an atomic species (Peart and Harrison 1981). A similar procedure has previously been used in this laboratory (Aitken and Harrison 1971, Woodruff *et al* 1978, Diserens *et al* 1988). The present measured detector efficiencies are 1.00 ± 0.03 for the Ar^{2+} data at 8 keV, 0.90 ± 0.04 for the Kr^{2+} data at 8 keV and 0.80 ± 0.04 for the Xe^{2+} data at 4 keV. The peak cross sections measured at the two ion energies of Ar^{2+} and Kr^{2+} differed by 2 and 7% respectively and these are within the combined error ranges of the measurements. The data for Ar^{2+} and Kr^{2+} at the higher ion energy were generally preferred because measurements of the detector efficiency were found to be more reliable and the ion beams were more stable.

The electron beam energy was varied from several electron volts below the threshold energy to 2000 eV, and the maximum electron current used was 400 μA (averaged over the duty cycle of the electron pulses). As examples of the signal and background count

rates, for Ar^{2+} at the peak of the cross section, an electron current of $285\ \mu\text{A}$ gave a mean signal count rate of typically 280 Hz at 4 keV, and the background count rate arising from interaction of the target beam with the background gas and slit edges was 35 Hz, giving a signal-to-background ratio of 8:1. For Xe^{2+} at 4 keV, an electron current of $275\ \mu\text{A}$ gave a mean signal count rate of 120 Hz and a background count rate of 1.5 Hz, giving a signal-to-background ratio of about 90:1. To minimize any effects due to short term changes of the product ion detection efficiency, measurements of the cross section were frequently repeated at a fixed reference energy (usually at 100 eV) and these were used to normalize the cross sections at other energies. Careful measurements were made at these reference energies immediately after measurements of the detection efficiency were made. To make the principal observed thresholds agree with the known ionization energies of Moore (1971), a correction of 0.5 eV was subtracted from the measured cathode-to-anode voltage to give the true mean electron energies. The FWHM of the electron beam energy distribution from the type of electron gun and cathode used here has been previously found to be 0.6 eV at the maximum current used for the present experiments (Diserens 1984). The ion beam velocity contributed negligible additions to the collision energies (0.1 eV in the worst case of Ar^{2+} at 8 keV), so the mean electron energy was therefore taken to be the collision energy.

3. Results and discussion

3.1. Results

The present results are shown as the plotted points in figure 1. The 90% confidence limits due to counting statistics at the peak ranged from $\pm 0.6\%$ for Ar^{2+} at 4 keV to $\pm 1.8\%$ for Kr^{2+} at 8 keV. The systematic error in the measurements is mainly due to the error in the measurement of the efficiency of the electron multiplier. Sources of systematic errors in measurements with this apparatus have been discussed by Montague *et al* (1984). The total systematic error in the present measurements is estimated to be about $\pm 4.7\%$ at a 90% confidence level. This is somewhat larger than for singly-charged rare gas ions because of greater errors in the detector efficiency measurements and because the smaller ion beam currents caused measurements of the effective beam height to be less accurate (both of which also caused greater statistical errors).

Tables 1–3 list values of the recommended cross section Q taken from a smooth curve drawn through the three sets of experimental data points of figure 1. No attempts have been made to fit the curves to small unidentified features in the experimental data. Where the structures are prominent and well resolved, as in the case of Xe^{2+} , the curve has been fitted to them. The total error in Q in each case is compounded from the overall systematic error and an error computed from the deviations of the experimental points from the smooth curve.

3.2. Discussion of Ar^{2+} results

The ionization cross section of Ar^{2+} is a smooth curve showing no structure. As discussed by Man *et al* (1987d) for the cross section measurements of singly-charged rare gas ions, the Ar atoms in the ion source are most probably in the ground state

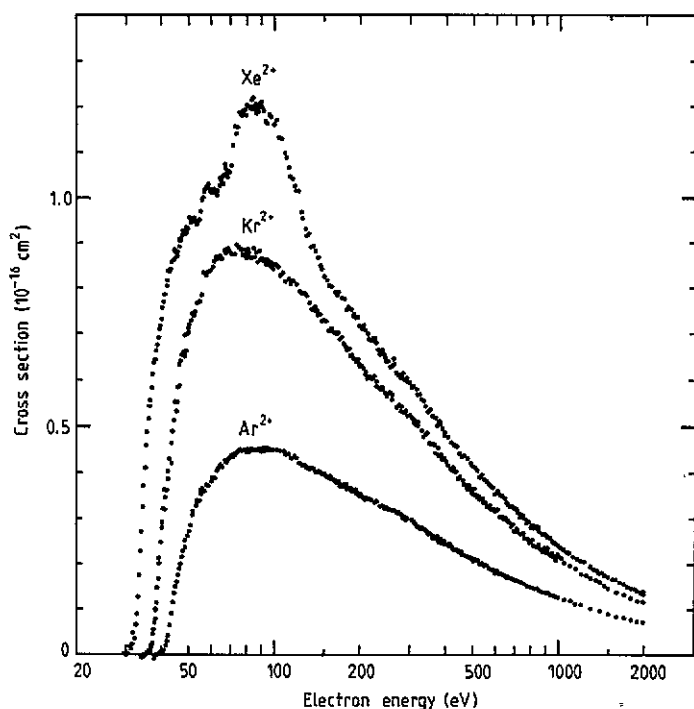


Figure 1. The ionization cross sections for Ar^{2+} , Kr^{2+} and Xe^{2+} as a function of electron energy. The full curves shown in figures 2, 3 and 4 are the best-fit lines through these experimental points. The cross section values given in tables 1-3 are obtained from these lines.

$3p^6$ (1S) and Ar^+ ions are likely to be $3p^5$ (2P) consisting of the fine-structure states $^2P_{3/2}$ and $^2P_{1/2}$, which are separated by less than 0.2 eV. The Ar^{2+} ions produced in the ion source should be predominantly in the ground state configuration $3p^4$ (3P) consisting of three states, 3P_2 , 3P_1 and 3P_0 separated by about 0.2 eV. All three states are likely to be present in the beam extracted from the source. In addition, the $3p^4$ configuration contains two metastable states, 1D_2 and 1S_0 , which lie 1.7 and 4.1 eV respectively above the ground state, and they may be expected to contribute to the ionization cross section. Indeed the signature of the presence of these metastable ions is the appearance of a small signal below the 40.9 eV threshold energy of the Ar^{2+} - Ar^{3+} ground-to-ground state transition, $3p^4$ (3P_2)- $3p^3$ ($^4S_{3/2}$). Transitions into excited states of Ar^{3+} are also likely since the $3p^3$ (2D) and $3p^3$ (2P) states lie only 2.6 and 4.3 eV respectively above the ground state. Ar^{2+} also possesses two higher metastable states, 5D_4 at 18.0 eV and 3F_4 at 23.1 eV, belonging to the $3p^3 3d$ configuration. However, it is most unlikely that an appreciable number of these ions would be excited in our low-temperature ion source.

The ionization cross section of Ar^{2+} has previously been measured in this laboratory (Diserens *et al* 1988), using the same crossed-beams apparatus, and elsewhere (Müller *et al* 1980, Danjo *et al* 1984, Mueller *et al* 1985), also using crossed-beams techniques. Figure 2 shows a comparison between the present data and the other experimental results and theoretical calculations. There is excellent agreement both in shape and in magnitude throughout the overlapping energy range between the present data and those of Diserens *et al*, thus confirming the consistency of measurements made with

Table 1. Recommended cross section for electron impact ionization of Ar^{2+} ions taken from a smooth curve drawn through the experimental points.

Mean electron energy E (eV) [†]	Recommended cross section $Q(E)$ (10^{-18} cm^2)	Total error in Q ($\pm\%$) [‡]	Mean electron energy E (eV) [†]	Recommended cross section $Q(E)$ (10^{-18} cm^2)	Total error in Q ($\pm\%$) [‡]
38.5	0.3	60	140	40.5	4.8
39	0.5	40	170	37.7	4.8
40	1.1	20	200	35.3	4.8
41	2.6	8.0	230	33.6	4.8
42	5.9	4.7	260	32.0	4.8
44	15.3	4.7	300	29.6	4.8
46	21.4	4.7	330	28.0	4.8
48	25.0	4.7	360	26.5	4.8
50	28.7	4.7	400	24.7	4.8
55	35.1	4.8	450	22.8	4.8
60	38.8	4.8	500	21.2	4.8
65	41.8	4.8	600	18.7	4.8
70	43.5	4.8	700	16.8	4.7
75	44.5	4.8	800	15.3	4.7
80	44.9	4.8	1000	12.9	4.7
85	45.2	4.8	1200	11.1	4.7
90	45.22	4.8	1400	9.7	4.7
100	45.0	4.8	1700	8.1	4.7
110	44.3	4.8	2000	7.2	4.7
120	43.2	4.8			

[†] ± 1.0 eV.[‡] 90% confidence limits. This error is a combination of systematic and random errors.

this apparatus. The present results also exhibit a similar fraction of metastable ions in the ion beam, as seen from the similar shape of the curve in the threshold region, reaffirming the reproducibility of the conditions of operation of the apparatus.

There is good agreement between all the experimental data in the sharply rising part of the Ar^{2+} curve from several electron volts above threshold to about 60 eV. Above the peak, the measurements of Müller *et al* (1980) are consistently smaller than the present results. As discussed by Man *et al* (1987d), their later-repeated results on the same ions are always 20–30% greater than their earlier data, probably as a result of improved experimental apparatus and technique. The measurements of Mueller *et al* (1985), on the other hand, are consistently greater than the present results especially near the peak of the curve where they are 9% greater. Their data are somewhat scattered and no structure can be identified on the curve. The measurements of Danjo *et al* (1984) show a prominent enhancement at around 160 eV. They attribute this hump to excitation-autoionization of the inner 2p electrons, but this seems unlikely since the lowest threshold is estimated to be approximately 240 eV. An alternative suggestion (Suzuki, private communication) is that the hump is due to a peak in the cross section for ionization of the 3s electrons. The enhancement is not reproduced in our results. Their cross section is consistently greater than ours but agrees quite well with that of Mueller *et al*. The fact that they agree with each other and that their curves are generally greater than ours is not too surprising since in both experiments a much greater fraction of metastable ions seem to be present in the beams than in ours, as witnessed by large

Table 2. Recommended cross section for electron impact ionization of Kr^{2+} ions taken from a smooth curve drawn through the experimental points.

Mean electron energy E (eV) [†]	Recommended cross section Q (E) (10^{-18} cm ²)	Total error in Q ($\pm\%$) [‡]	Mean electron energy E (eV) [†]	Recommended cross section Q (E) (10^{-18} cm ²)	Total error in Q ($\pm\%$) [‡]
36	0.5	60	140	75.0	4.8
37	1.5	50	170	69.0	4.8
38	6.0	20	200	63.0	4.8
39	17.0	4.8	230	59.0	4.8
40	25.5	4.8	260	56.0	4.8
42	39.0	4.8	300	52.0	4.8
44	50.0	4.8	330	49.0	4.8
46	57.5	4.8	360	46.5	4.8
48	65.0	4.8	400	43.0	4.8
50	71.0	4.8	450	39.5	4.8
53	77.0	4.8	500	36.5	4.8
56	80.5	4.9	550	34.0	4.8
60	84.5	4.9	600	32.2	4.8
65	87.0	4.9	700	28.2	4.8
70	88.0	4.9	800	25.5	4.8
75	88.5	4.9	1000	21.0	4.8
80	88.0	4.9	1200	18.0	4.7
90	87.0	4.9	1400	16.0	4.7
100	85.0	4.9	1700	13.3	4.7
110	82.5	4.9	2000	11.6	4.7
120	80.0	4.9			

[†] ± 1.0 eV.[‡] 90% confidence limits. This error is a combination of systematic and random errors.

signals below the ground-state ionization threshold. It is possible that the hump at around 160 eV in the curve of Danjo *et al* may also be the result of the large fraction of metastable and highly-excited ions in their beam, which would explain why it is not reproduced in our curve. Unfortunately the data of Mueller *et al* are not taken at sufficiently small energy intervals to resolve this issue.

Figure 2 also shows comparisons of the present data with the semi-empirical formulae of Lotz (1967) and of Burgess and Chidichimo (1983) (bc) and with the parametrized distorted-wave formula of Younger (1981a, b) (dw). In the Lotz three-parameter calculation, we have taken the contributions from all electrons in the outer three shells, 3p, 3s and 2p into consideration. The ionization energies were taken as 40.9, 55 and 277 eV respectively. The three-parameter curve underestimates the cross section at lower energies and overestimates it at higher energies. The Lotz curve with one parameter (only the 3p electrons are included) shows good agreement with the present result at higher energies, but greatly underestimates the cross section at energies below the peak. The peak for both curves shifts to the higher energy by some 40 eV. The neglect of excitation-autoionization in the formula is probably the major source of discrepancy with the measured cross sections at low energies. The bc curve, which includes an allowance for excitation-autoionization, fits the experimental curve much better, especially in the low energy and peak regions, but at higher energies it progressively overestimates the cross section.

Table 3. Recommended cross section for electron impact ionization of Xe^{2+} ions taken from a smooth curve drawn through the experimental points.

Mean electron energy E (eV) [†]	Recommended cross section $Q(E)$ (10^{-18} cm ²)	Total error in Q ($\pm\%$) [‡]	Mean electron energy E (eV) [†]	Recommended cross section $Q(E)$ (10^{-18} cm ²)	Total error in Q ($\pm\%$) [‡]
30.5	0.5	100	72	112.5	5.8
31	1.1	50	74	116.1	5.8
32	3.5	20	76	118.0	5.8
33	10.0	4.8	78	119.0	5.8
34	25.5	4.8	80	119.7	5.8
35	39.5	4.8	82	120.2	5.8
36	48.5	4.8	86	120.5	5.8
37	57.0	4.8	90	120.0	5.8
38	63.5	4.8	95	118.5	5.8
40	72.5	4.8	100	115.7	5.8
42	81.0	4.8	110	109.0	4.8
43	85.0	5.0	120	100.5	4.8
44	86.3	5.0	130	92.5	4.8
45	87.0	5.0	140	87.0	4.8
46	88.7	5.0	160	80.5	4.8
47	90.5	5.0	180	76.1	4.8
48	92.3	5.0	200	72.5	4.8
50	94.5	5.8	230	68.0	4.8
51	95.3	5.8	260	64.0	4.8
52	95.3	5.8	300	59.4	4.8
53	94.6	5.8	330	56.0	4.8
54	95.0	5.8	360	52.7	4.8
55	97.5	5.8	400	48.6	4.8
56	100.5	5.8	450	45.0	4.8
57	102.1	5.8	500	42.2	4.8
58	102.6	5.8	550	39.0	4.8
59	102.4	5.8	600	36.6	4.8
60	101.8	5.8	700	32.5	4.8
61	101.4	5.8	800	29.2	4.8
62	101.5	5.8	1000	24.4	4.7
64	102.7	7.0	1200	21.0	4.7
66	104.6	7.0	1400	18.5	4.7
68	105.5	7.0	1700	15.8	4.7
70	106.6	7.0	2000	13.7	4.7

[†] ± 1.0 eV.[‡] 90% confidence limits. This error is a combination of systematic and random errors.

The DW curve underestimates the cross section at all energies, especially at energies below the peak, where it is in good agreement with the one parameter Lotz curve. The underestimation is likely to originate from the same neglect of excitation-autoionization. The peak for this curve is shifted to a higher energy by some 30 eV.

3.3. Discussion of Kr^{2+} results

The ground-state configuration of Kr^{2+} is $4p^4$ (3P) and the fine-structure states 3P_2 , 3P_1 and 3P_0 are separated by 0.7 eV. In addition, the $4p^4$ configuration has two low-lying metastable states 1D and 1S , which are 1.8 and 4.1 eV above the ground state, and

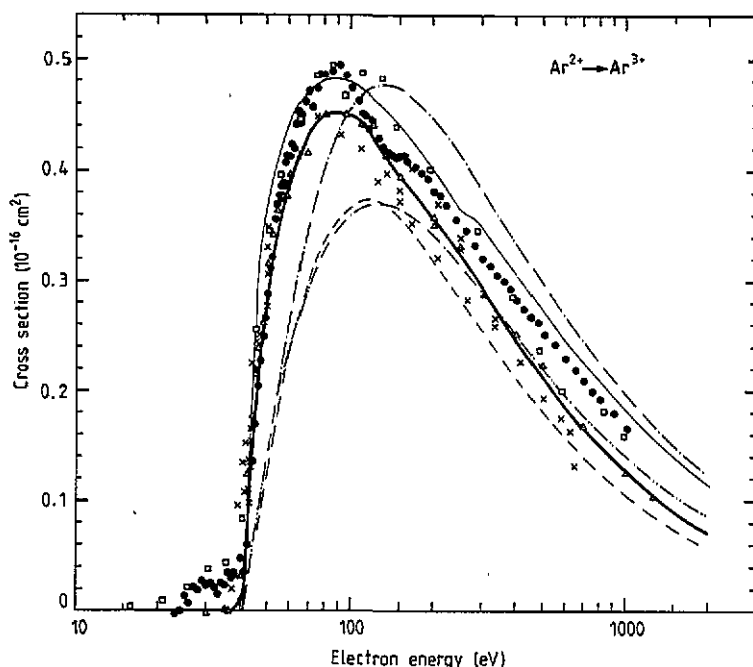


Figure 2. Electron impact ionization cross section of Ar^{2+} as a function of electron energy. The present results (—) are compared with the experimental measurements of Danjo *et al* (1984), ●; Müller *et al* (1980), ×; Diserens *et al* (1988), △; Mueller *et al* (1985), □; and theoretical calculations using the formula of Lotz (1967) with three parameters, — — —; with one parameter, — · — · —; formula of Burgess and Chidichimo (1983), — — —; and the formula of Younger (1981), — — — — —.

high-lying metastable states with energies between 17.2 and 21.7 eV. Since Kr^{2+} is more complicated than Ar^{2+} , with extra s and p subshells and the additional 3d subshell, the observed enhancement in the cross section curve is to be expected. The Kr^{2+} curve in figure 1 exhibits a small signal below the threshold energy, which is a sign of the presence of low-lying metastable ions in the beam, but the fraction of these ions in the beam is clearly very small. There are many partly-resolved structures along the curve and a small enhancement appears at about 300 eV. This is possibly caused by ionization of inner-shell electrons or excitation-autoionization of 3p or 3s electrons. Some of the unresolved structures around the peak are likely to be due to direct ionization and excitation-autoionization of 4s and 3d electrons.

Figure 3 shows a comparison between the present data and the experimental results of Danjo *et al* (1984), Gregory (1985) and Tinschert *et al* (1987). Our result is in good agreement with the first of these over most of the energy range except between 100 and 160 eV where their curve dips significantly and then is followed by an enhancement at 140 eV. This feature is similar to that observed in their Ar^{2+} data. Again their curve shows a large signal below the ground-state threshold energy indicating that their beam contained a larger fraction of metastable and highly-excited ions than ours. The threshold for this metastable contribution is at about 21 eV, indicating the presence of high-lying metastables with $4p^34d$ configuration. Gregory's curve is in much better agreement with ours in the near-threshold region and it approaches our curve at high energies. In the intermediate energy range, however, his measurements are consistently smaller than ours, about 14% smaller at the peak.

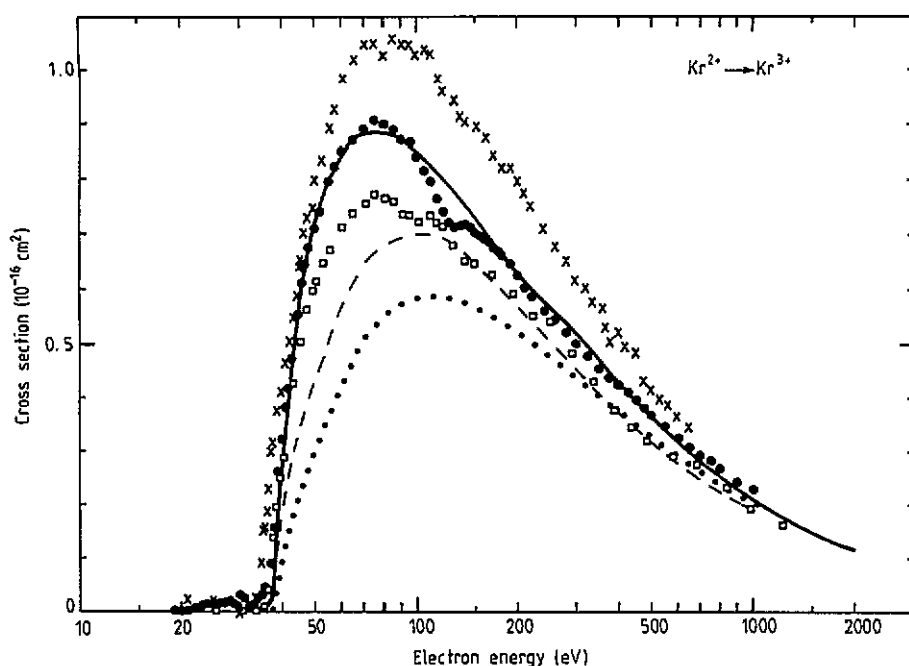


Figure 3. Electron impact ionization cross section of Kr^{2+} as a function of electron energy. The present result (—) is compared with the experimental measurements of Danjo *et al* (1984), ●; Gregory (1985), □; Tinschert *et al* (1987), ×; and calculations using the formula of Burgess and Chidichimo (1983), — — —; and distorted wave from Gregory (1985), · · · ·

The results of Tinschert *et al* are consistently much greater than ours and those of Danjo *et al*, by 19% at the peak. They state that this is caused by the presence of a large fraction of Kr^{2+} ions in low-lying $4p^4$ metastable states, although the reason why their beam should contain a large population of such states whereas the beam used by Danjo *et al* should contain only high-lying states is not discussed. In both experiments electron-cyclotron-resonance ion sources were used, in contrast to the low-temperature plasma sources used in Gregory's experiment and ours. Tinschert *et al* estimate that the influence of the low-lying metastable states, assumed to be principally $^1\text{S}_0$, would be to increase their measured cross section by about 14% at the peak, whereas the influence of the high-lying states on the results of Danjo *et al* would be to increase their measured cross section by 3%. Thus, assuming that our results represent the cross section for ground-state Kr^{2+} ions, the three sets of results are in agreement to well within the combined experimental errors. The results of Gregory appear to be anomalously low in the intermediate energy region.

It is only in the results of Danjo *et al* that structure in the region of 150 eV is clearly apparent. If the interpretation is correct that it was only in their experiment that the ion beam contained ions in high-lying metastable states, then it seems possible that the structure is caused by enhanced inner-shell ionization of such ions.

Figure 3 also shows a comparison between the present experimental data and the distorted-wave calculation from Gregory (1985) (attributed to M S Pindzola), and the semi-empirical formula of Burgess and Chidichimo (1983). The DW calculation, which includes only direct ionization of $3p$ and $3s$ electrons, grossly underestimates the cross section at low and intermediate energies. The semi-empirical formula of Burgess and

Chidichimo, which takes into account the excitation-autoionization process, improves the agreement with the experimental curve. This shows that inner-shell electrons in Kr^{2+} undergo ionization indirectly and must be taken into account in any theoretical calculations to accurately describe the ionization process in a plasma system.

3.4. Discussion of Xe^{2+} results

Similarly to the cases of Ar^{2+} and Kr^{2+} , the ground state configuration of Xe^{2+} is $5p^4$ (3P). The 3P state consists of the three fine structure states 3P_2 , 3P_1 and 3P_0 spanning 1.2 eV. The two low-lying metastable states 1D_2 and 1S_0 of the same configurations, occurring 2.1 and 4.5 eV respectively above the ground state, can be expected to contribute to the total ionization cross section. The increased number of subshells compared with Ar^{2+} and Kr^{2+} will enhance the cross section even further, particularly through excitation-autoionization. There are several well defined structures in the region around the maximum of the curve and at intermediate energies (figure 1). The most prominent feature is a hump centred at 85 eV. Other well resolved enhancements are centred at 52 and 58 eV. Close examination of the curve shown in figure 1 suggests that there are many other partly resolved features in the range from 150 to 400 eV.

The first discontinuity at 45 eV coincides with the threshold for removal of a 5s electron, but the other features up to 140 eV are probably caused by excitation of 5s and 4d electrons followed by autoionization. The large hump at 85 eV most probably arises from excitation-autoionization of the abundant 4d electrons. The features at the higher energies are probably due to excitation-autoionization or direct ionization of the 4p and 4s electrons. The two well resolved structures at 52 and 58 eV are different from the other observed structures and appear to be due to a type of resonance, possibly from the resonance excitation-double autoionization (REDA) process discussed by LaGattuta and Hahn (1981). In Xe^{2+} this would be of the form $5s^25p^4 + e \rightarrow 5s^15p^5nl \rightarrow 5s^25p^3 + 2e$. This feature is quite similar to that observed in Ti^{2+} by Mueller *et al* (1985) and Diserens *et al* (1988).

Three previous measurements of the cross section for Xe^{2+} , all using the crossed-beams technique, have been made by Achenbach *et al* (1984), Danjo *et al* (1984) and Griffin *et al* (1984). The second and third measurements agree quite well with each other, reproducing many of the features, but the data of Achenbach *et al* are grossly higher than the other two and have a very different main threshold.

Figure 4 shows a comparison between the present data and the other experimental measurements. Our results are in best agreement with those of Griffin *et al* (1984). Both sets of results show small signals below the main threshold. The magnitude of the hump at 85 eV is almost exactly the same. The structures between 43 and 70 eV are quite well reproduced although there are differences in the exact shape and magnitude of these structures. The two curves are also in excellent agreement at higher energies. In the energy range between 110 and 300 eV our curve tends to fall more sharply after the main hump, but the maximum difference is no more than 8%, which is within the combined errors of the measurements.

There is reasonable agreement between our data and those of Danjo *et al* (1984) throughout the energy range, although the latter are consistently higher than ours. These differences are within the combined errors of the experiments at most energies. Their maximum at 85 eV coincides exactly with ours and it is about 6% greater. Their curve also shows many of the structures seen on our curve, but generally not so clearly resolved and at slightly different energies. The most significant differences between the

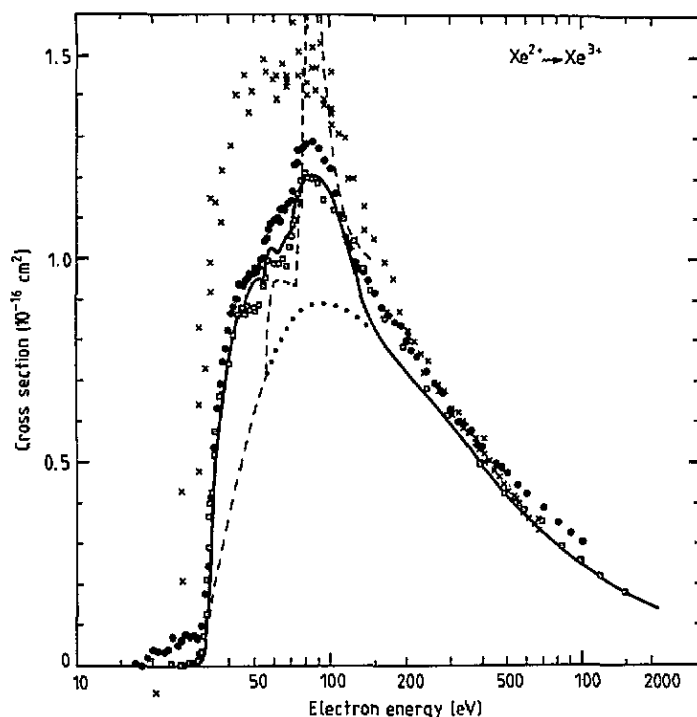


Figure 4. Electron impact ionization cross section of Xe^{2+} as a function of electron energy. The present result (—) is compared with the experimental measurements of Danjo *et al* (1984), ●; Griffin *et al* (1984), □; Achenbach *et al* (1984), ×; and theoretical calculations (distorted-wave) of Griffin *et al* (1984), direct ionization only ····; direct plus excitation-autoionization, — — —.

two curves are in the near-threshold region, where their main threshold energy is lower, and in the below-threshold region, where they observed large signals. Their lowest threshold is at 18 eV, some 14 eV lower than the threshold expected from the ground-ground state transition. This indicates that their ion beam contains not only low-lying metastable ions such as $5s^25p^4$ (1D_2) and $5s^25p^4$ (1S_0), but also higher metastables such as $5s^15p^5$ (3P). The presence of substantial fractions of metastable ions in their beam is probably also the cause of the generally larger cross section over the entire energy range.

The data of Achenbach *et al* (1984) are so scattered that it is not possible to identify any structure. The magnitude at the maximum and at lower energies is very much greater than the other three sets of data, although the disagreement decreases at higher energies. The main threshold energy deduced from the very limited data shows that it is some 10 eV lower than the ground-ground state transition. This indicates that their ion beam contained a considerable fraction of excited ions.

Figure 4 also shows a comparison between the present experimental data and the distorted-wave calculation of Griffin *et al* (1984). The calculation including only direct processes grossly underestimates the cross section at low and intermediate energies. This is to be expected since the abundant inner-shell electrons in Xe^{2+} undergo ionization indirectly. When the calculation takes into account the excitation-autoionization process for the 4d electrons, the agreement with the experimental curve improves.

However, the calculation is still too low at lower energies, suggesting that electrons other than those in the 4d subshell also contribute to the cross section. The peak, though it agrees well in energy with the experimental peak, grossly overestimates the cross section.

4. Summary and conclusion

Measurements on electron impact single-ionization of the doubly-charged rare gas ions Ar^{2+} , Kr^{2+} and Xe^{2+} (figure 1) show that the general magnitudes of the cross sections increase with increasing atomic number along the group, in agreement with the expectation from the decreasing ionization energy of the outer electrons. The cross sections for Kr^{2+} and Xe^{2+} are greatly enhanced because of contributions from the d-shell electrons. The progressively sharper rise in cross section just above the thresholds from Ar^{2+} to Xe^{2+} is probably due to greater contributions from excitation-autoionization of the increasing number of inner-shell electrons and thus the increasing number of autoionizing states. All the incident beams contained metastable and possibly long-lived excited states, as evidenced by the presence of ionization signals below the ionization energy of ground-state ions, but the populations are small even in the case of Xe^{2+} where it is estimated to be no more than a few percent†. Nevertheless the presence of the metastables could have a significant effect on the near-threshold region and could partly account for the sharp rises in the cross sections from the thresholds. However, since the ions are produced in a plasma source, the cross sections measured here with possibly small contributions from excited ions are of practical relevance in plasma physics. Structures on the curves are increasingly evident along the group, especially near the peaks and at intermediate energies. These range from no clearly resolved structure on the Ar^{2+} curve to several peaks and unresolved structures for Xe^{2+} . These structures are generally due to direct inner-shell ionization and indirect inner-shell excitation-autoionization. In Xe^{2+} , resonance-type structures are observed that may be due to the REDA process.

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† The metastable content of Ar^{2+} , Kr^{2+} and Xe^{2+} ion beams produced by direct electron impact with atoms has been measured by Nakamura *et al* (1985), Kobayashi *et al* (1983) and by Hamdan *et al* (1988), respectively, using the technique of translational energy spectroscopy. They find the fractions of metastable $^1\text{D}_2$ and $^1\text{S}_0$ states are approximately 0.3 and ≈ 0.05 , respectively, i.e. roughly in accordance with the expectation from statistical weight considerations. In our experiment, in which the ions are generated in a low-temperature plasma source, the metastable fractions appear to be much smaller.

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