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Absolute cross sections for multiple ionization of noble gases by swift proton impact

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Abstract
Absolute cross sections, σ^{q+}, for multiple ionization of He ($q = 1$), Ne ($q = 1$, 2, and 3), Ar ($q = 1$, 2, 3, and 4), Kr ($q = 1$, 2, 3, and 4), and Xe ($q = 1$, 2, 3, 4, and 5) atoms by protons in the 0.75–3.5 MeV impact energy range have been measured. The present results are in good agreement with the previously published data in the case of the single- and double-ionization cross sections, but some non-systematic discrepancies appear for higher charge states. A detailed comparison with electron impact cross sections in these gases is performed, and it shows that the cross sections for single ionization by high-velocity protons are substantially smaller than those corresponding to equi-velocity electrons for heavier targets.

1. Introduction

Ionization by swift protons and electrons is such a basic and important process in several branches of physics that conceptual developments as well as the modelling of different physical environments must have both theory and experiment reasonably well established in, at least, some of its essential aspects. In particular, it is essential that:

(i) the absolute experimental data fulfil the need for giving consistent and reliable cross sections;
(ii) calculations made in the Born high-velocity regime give a good description of experiments and can be used as benchmarks for theoretical estimates;
(iii) differences between equi-velocity fast electrons and protons are negligible, as predicted by first-order theories.

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Nevertheless, an overview of the vast literature on this subject shows that several aspects concerning the above points remain unsatisfactory, even for noble gases, which play the role of basic systems in atomic collision studies. The lack of a proper understanding of the experimental data at high velocities becomes increasingly pronounced with increasing atomic number—not only for multiple ionization, as could be expected, but even for the single-ionization channel. This scenario demanded and motivated several recent investigations on this subject. Indeed, from the electron impact ionization side, we can cite, for example, the extensive absolute measurements of multiple ionization by Rejoub *et al* [1], the first Born approximation (BA) calculations by Bartlett and Stelbovics [2]—both of these for the noble gases from He to Xe—and the distorted-wave calculations for Kr by Loch *et al* [3]. In the proton case, the measurements made by Melo *et al* [4] at 2.0 MeV for the noble gases from He to Xe, the measurements by Cavalcanti *et al* [5] on Ne, and the calculations of Kirchner *et al* for Ne [6] and Ar [7] are the most recent examples.

Melo *et al* [4] reported measurements on multiple ionization of noble gases by 2.0 MeV protons and compared with the case for equi-velocity electrons. They found a surprising result, namely, that the cross sections for *single* ionization by protons become increasingly smaller than the corresponding electron cross sections as the target atomic number increases. The *multiple*-ionization cross sections for both projectiles, on the other hand, seem to agree within the experimental uncertainties for all gases. This result points in the same direction as the earlier findings by Haugen *et al* [8], who reported increasing differences in the ratio between double and single ionization by protons and electrons as the atomic number of the target increases. The absolute measurements of Melo *et al* [4] show that these differences originate from the single-ionization cross sections, a result far from being intuitive. On the other hand, with regard to the multiple ionization, a recent paper concerning protons on Ne shows the importance of both Auger and Coster–Kronig time-delayed post-collisional mechanisms in the double- and triple-ionization cross sections at high velocities [5].

The above studies indicate that the role of some mechanisms which can contribute to the ionization of multi-electron systems by the simplest projectiles, e.g., protons and electrons, needs to be better identified and quantified, both theoretically and experimentally. To this end, comparison between absolute proton and electron multiple-ionization cross sections of rare gases is a basic step towards understanding the mechanisms behind the collision dynamics of multi-electron systems. However, such comparisons are essentially non-existent. The reason for that is the great rarity of absolute measurements on swift protons which have been reported so far. Indeed, to the authors' knowledge, the only systematic absolute measurements on noble gases with swift protons are those of DuBois *et al* [9] and Melo *et al* [4].

In this paper, we report on absolute measurements of multiple-ionization cross sections of protons on He, Ar, Kr, and Xe, made using coincidence techniques, over the energy range from 0.75 to 3.5 MeV. In section 2 the experiment is described. In section 3 the results are presented and compared with some available calculations, previous measurements, and with the ionization by equi-velocity electron impact. In section 4, some concluding remarks are made.

2. Experiment

The experimental apparatus as well as the absolute calibration procedure have been described in great detail recently [5, 10, 11] and only a brief summary will be given here. A schematic view of the apparatus is shown in figure 1. Well collimated mono-energetic proton beams with energies ranging from 0.75 to 3.5 MeV delivered by the 4 MV Van de Graaff accelerator of the Catholic University of Rio de Janeiro enter a gas cell after being properly 'cleaned'

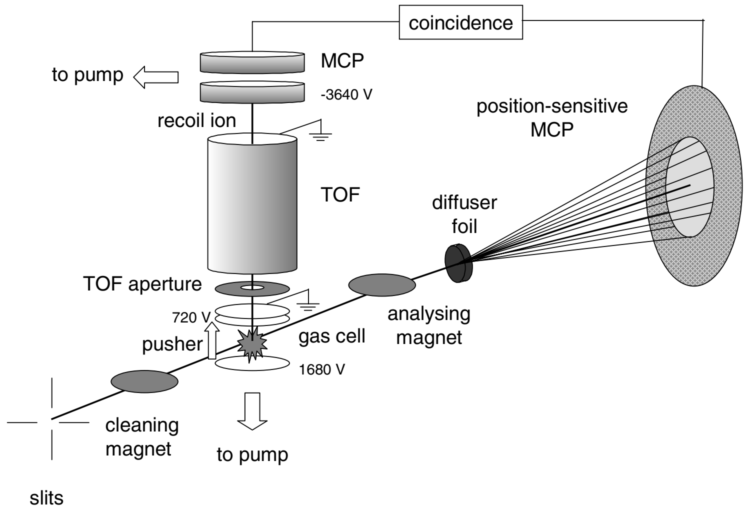

Figure 1. A schematic diagram of the set-up used in this work.

of spurious beams. Recoil ions produced in the gas cell due to the ionization process are pushed towards a microchannel plate detector by a 1680 V potential applied in two stages. After passing through a 9.0 cm time-of-flight tube, the recoil ions are further accelerated by a 3640 V negative potential before impacting on the microchannel plate recoil ion detector (R-MCP). As mentioned previously [10, 11], using these high pushing fields, and taking care to keep the R-MCP at low pressures (approximately 3×10^{-6} Torr), irrespective of the pressure used in the gas cell, there was no observable dependence of the R-MCP efficiency on the recoil ion charge state. The constant low pressure in the R-MCP region is achieved by differential pumping through a 4 mm diameter aperture introduced inside the time-of-flight tube and located at approximately 22 mm from the interaction region [11].

One point affecting the overall recoil ion detection efficiency is worth mentioning. For part of their flight path, the recoil ions travel through a region inside the gas cell. Only after they reach the above-mentioned aperture does the pressure drop abruptly. While they are inside the gas cell, a symmetrical resonant charge transfer can occur, eventually suppressing the detection of those singly charged ions which become neutral. This process, which also occurs with the other charge states, can be identified by the presence of a small broad peak, located on the slower side of the recoil ion peaks in the time spectra. Its influence on the measured yield of each charge state can be estimated by measuring the coincidence spectra for various pressures and looking for the quadratic dependence of the yield on the pressure. We found that the coefficient of the quadratic term presents a clear linear dependence on the recoil ion mass and, for the pressures used in our measurements, from 0.5 to 1.0 mTorr depending on the target gas, the correction associated with this process ranges from 5 to 20% [13].

The beams emerging from the gas cell pass through a 600 Å thick Al diffuser foil before reaching a position-sensitive microchannel plate detector with a delay-line anode (P-MCP) [12]. This foil provides a broad illumination of the P-MCP detector, allowing higher counting rates. The measurements were performed with emergent beam rates between 1.5 and 2.5×10^4 s^{-1}. Standard fast coincidence electronics and time-of-flight spectrometry techniques were used to discriminate among the various recoil ion charge states. The absolute efficiency of the system was determined using the measurements of [4] which were performed in the same manner as described by Santos *et al* [10, 11].

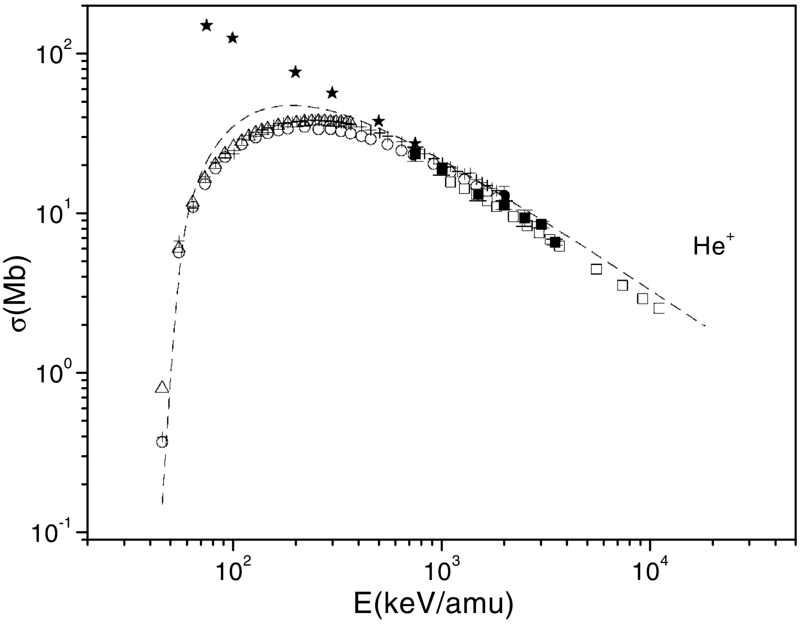

Figure 2. Cross sections for single ionization of He by protons and electrons. Experiments with protons: ■, this work; ●, [4]; ★, [9]. Experiments with electrons: ○, [1]; △, [14]; □, [17]; +, [19]. Theory: - - - -, [2].

3. Results and discussion

Tables 1–4 list the results of our measurements for single and multiple ionization of He, Ar, Kr, and Xe by protons. Figures 2–6 show the present results as well as our previous measurements on Ne [5], for completeness. In these figures we also plot the proton measurements of Melo *et al* [4] and DuBois *et al* [9]. A comparison with single and multiple ionization of these gases by electrons is also presented. The absolute electron data of Rejoub *et al* [1], Wetzel *et al* [14], El-Sherbini *et al* [15], Schram *et al* [16–18], as well as the relative measurements of Krishnakumar and Srivastava [19] normalized to the data from Rapp and Englander-Golden [20], are shown. Although there are several other measurements of multiple ionization of noble gases by electron impact available in the literature, e.g., those of Almeida *et al* [21, 22], we focused our comparison, basically, on those works which reported absolute and independently measured cross sections. Theoretical calculations for electron and proton impact are also included for comparison. These are BA total ionization cross sections for electron impact on all noble gases given by Bartlett and Stelbovics [2], and time-dependent independent-particle model calculations for single ionization of Ne and Ar by proton impact by Kirchner *et al* [6, 7]. The quadruple ionization of Ar, Kr, and Xe and the quintuple ionization of Xe are not shown in order not to overcomplicate the figures.

The following comments can be made after an overall analysis of the figures:

(i) Where the measurements overlap, there is a good agreement—essentially within the experimental uncertainties—between our data and the proton data from [9] for the single-ionization cross sections of all gases. For double ionization, there is also a good general agreement between these two sets of measurements, although a slight tendency of our cross sections to be above those from [9] for Ne and Ar can be observed. Overall, the two sets of data are consistent for single and double ionization. For triple ionization,

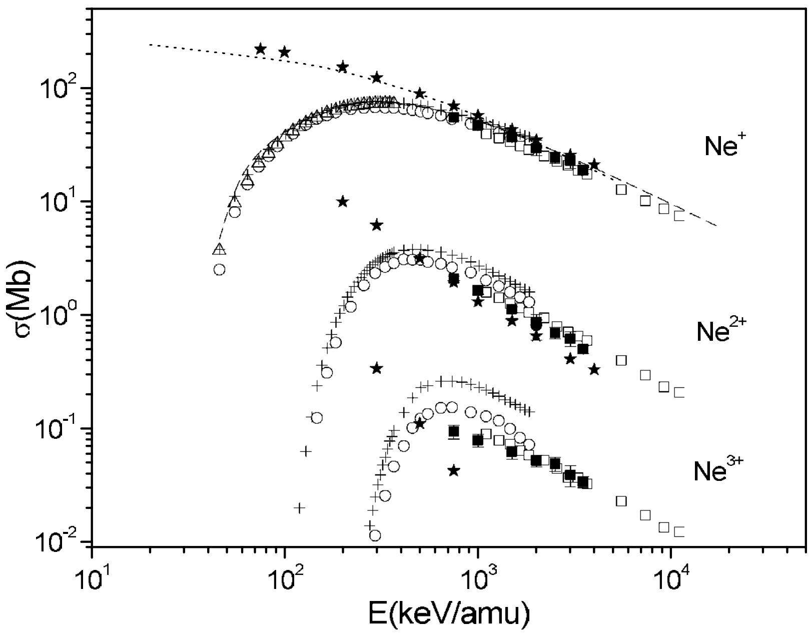

Figure 3. Single, double, and triple ionization of Ne by protons and electrons. Experiments with protons: ■, [5]; ●, [4]; ★, [9]. Experiments with electrons: ○, [1]; △, [14]; □, [17]; +, [19]. Theory: - - - -, [2]; · · · · · ·, [6].

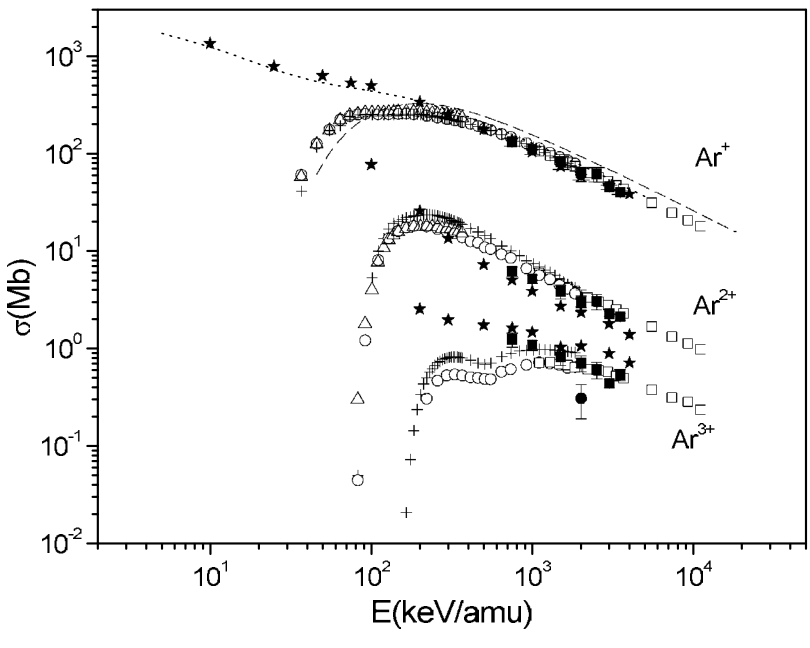

Figure 4. Single, double, and triple ionization of Ar by protons and electrons. Experiments with protons: ■, this work; ●, [4]; ★, [9]. Experiments with electrons: ○, [1]; △, [14]; □, [16, 18]; +, [19]. Theory: - - - -, [2]; · · · · · ·, [7].

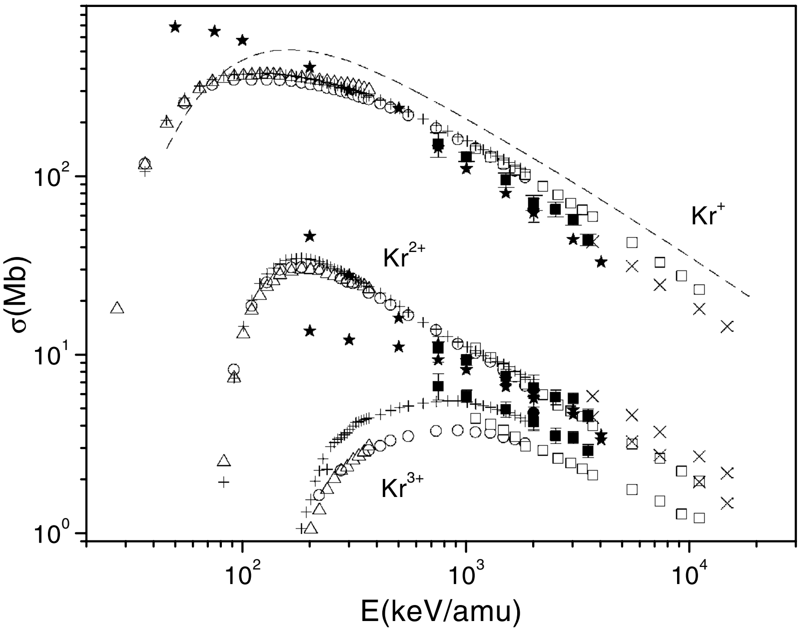

Figure 5. Single, double, and triple ionization of Kr by protons and electrons. Experiments with protons: ■, this work; ●, [4]; ★, [9]. Experiments with electrons: ○, [1]; △, [14]; □, [16, 18]; +, [19]; × [15]. Theory: - - - -, [2].

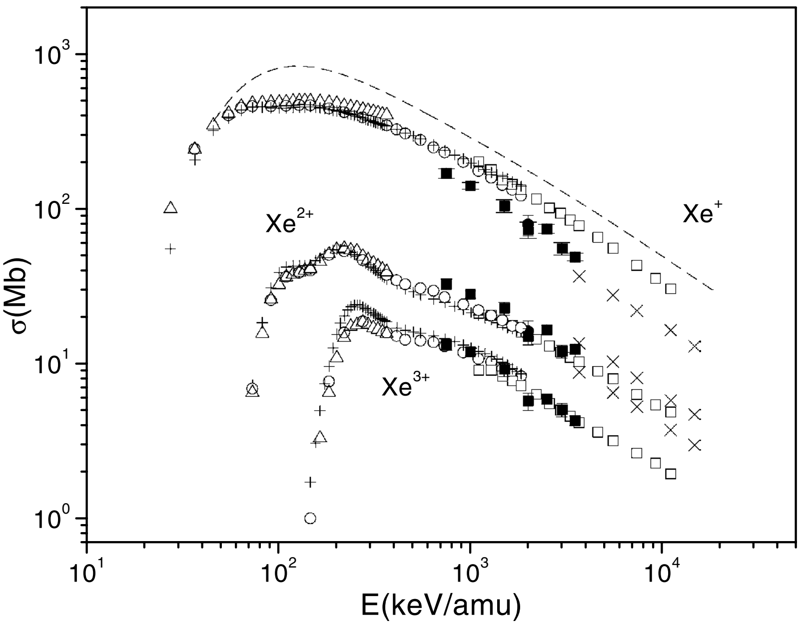

Figure 6. Single, double, and triple ionization of Xe by protons and electrons. Experiments with protons: ■, this work; ●, [4]. Experiments with electrons: ○, [1]; △, [14]; □, [16, 18]; +, [19]; × [15]. Theory: - - - -, [2].

Table 1. Absolute cross sections for single ionization of He by protons (Mb).

E (MeV)	He^+
0.75	23.5 ± 2.4
1.00	18.9 ± 1.5
1.50	13.1 ± 1.1
2.00	11.3 ± 0.7
2.50	9.4 ± 1.1
3.00	8.5 ± 0.4
3.50	6.6 ± 0.3

Table 2. Absolute cross sections for multiple ionization of Ar by protons (Mb).

E (MeV)	Ar^+	Ar^{2+}	Ar^{3+}	Ar^{4+}
0.75	132 ± 13	6.22 ± 0.69	1.24 ± 0.20	0.20 ± 0.07
1.00	110 ± 11	5.16 ± 0.53	1.09 ± 0.13	0.25 ± 0.04
1.50	81.0 ± 12	3.86 ± 0.62	0.82 ± 0.15	
2.00	63.3 ± 9.4	3.00 ± 0.48	0.71 ± 0.13	0.10 ± 0.03
2.50	62.0 ± 10	3.06 ± 0.53	0.61 ± 0.12	0.15 ± 0.04
3.00	45.3 ± 2.4	2.28 ± 0.14	0.44 ± 0.04	
3.50	40.2 ± 1.8	2.12 ± 0.16	0.54 ± 0.07	0.07 ± 0.02

Table 3. Absolute cross sections for multiple ionization of Kr by protons (Mb).

E (MeV)	Kr^+	Kr^{2+}	Kr^{3+}	Kr^{4+}
0.75	151 ± 24	10.9 ± 1.8	6.70 ± 1.1	1.65 ± 0.33
1.00	128.5 ± 7.6	9.41 ± 0.67	5.86 ± 0.46	1.16 ± 0.15
1.50	95.2 ± 8.8	7.61 ± 0.72	4.96 ± 0.48	0.97 ± 0.11
2.00	71.0 ± 6.8	6.55 ± 0.65	4.24 ± 0.43	0.85 ± 0.10
2.50	65.4 ± 6.0	5.78 ± 0.56	3.53 ± 0.35	0.70 ± 0.09
3.00	57.1 ± 4.0	5.74 ± 0.42	3.44 ± 0.26	0.70 ± 0.07
3.50	44.1 ± 3.1	4.60 ± 0.38	2.90 ± 0.25	0.65 ± 0.09

Table 4. Absolute cross sections for multiple ionization of Xe by protons (Mb).

E (MeV)	Xe^+	Xe^{2+}	Xe^{3+}	Xe^{4+}	Xe^{5+}
0.75	170 ± 13	32.7 ± 2.5	13.3 ± 1.1	5.00 ± 0.49	0.95 ± 0.16
1.00	140.9 ± 6.7	28.0 ± 1.4	12.0 ± 0.69	3.75 ± 0.29	0.98 ± 0.12
1.50	104.7 ± 9.8	22.8 ± 2.2	9.17 ± 0.90	3.27 ± 0.34	0.64 ± 0.09
2.00	73.4 ± 8.9	15.0 ± 1.8	5.74 ± 0.72	1.98 ± 0.26	0.47 ± 0.08
2.50	74.2 ± 5.0	16.5 ± 1.1	5.91 ± 0.42	2.36 ± 0.18	0.64 ± 0.06
3.00	55.2 ± 5.1	12.1 ± 1.1	5.06 ± 0.49	1.87 ± 0.19	0.50 ± 0.06
3.50	48.9 ± 2.9	12.4 ± 0.8	4.30 ± 0.31	1.47 ± 0.14	0.37 ± 0.06

some clear disagreement can be seen in the energy dependence for the Ne case and in the absolute values in the Kr case. For Ar, however, the two measurements agree well.

(ii) There is a very good and consistent agreement between the measured cross sections for protons (this work and [9]) and those for equi-velocity electrons (see e.g. [1, 17–19]) for single ionization of He, Ne, and Ar, for velocities corresponding to proton energies above 1 MeV. The same is not true for the Kr and Xe cases, which will be further discussed below.

(iii) An excellent agreement between our measurement and the equi-velocity electron measurements of Shram *et al* [16–18] can be observed for *double- and triple-ionization cross sections* of all heavy gases, with the exception of the triple ionization of Kr, where our data lie slightly above theirs. The same can be said for the electron data of Rejoub *et al* [1] for the Ar, Kr, and Xe targets. The data from [1] are slightly above our measurements only for the double and triple ionization corresponding to the Ne case. This same general behaviour also appears in the data from [19]. As a general conclusion, it can be said that the measured data for swift equi-velocity protons and electrons consistently agree for *double and triple ionization* with no clear *systematic* discrepancies observed.

(iv) A *systematic* disagreement among the *single-ionization cross sections* for swift equi-velocity protons and electrons for Kr and Xe targets clearly emerges from the data. This tendency has been reported previously by Melo *et al* [4] for 2.0 MeV protons showing a behaviour which deviates from the general pattern described in the points above. It is worth noting that, contrary to the majority of electron data, the electron data from El-Sherbini *et al* [15] agree with the equi-velocity proton single- and double-ionization cross sections for both Kr and Xe targets[4].

(v) The calculations of Kirchner *et al* for Ne [6] and Ar [7] for cross sections for single ionization by protons show a good agreement with the data. The agreement seems to be excellent at high velocities. The electron impact BA calculations by Bartlett and Stelbovics [2] for the ionization cross sections, although giving a good description for the He and Ne targets, show an increasing tendency to give too large cross sections as the target atomic number increases. This tendency is also seen in the electron impact configuration-average distorted-wave results reported by Loch *et al* [3] (not shown) for Kr, as well as in the continuum-distorted-wave (CDW) proton cross sections obtained by Gulyás *et al* [23] (not shown) for proton impact on Ne and Ar.

The consistency of our measurements can be further verified through the double-to-single-ionization ratios. Figure 7 displays our measured ratios, as a function of the proton energy, for Ne, Ar, Kr, and Xe, together with those reported by Haugen *et al* [8]. A very good agreement is observed for all gases as well as a sharp increase in the ratio for the heavier gases. As shown in [8], this increase is significantly less pronounced in the electron case. What the present work shows, through the measurement of absolute values of the cross sections, is that the origin of the differences in behaviour of the double-to-single-ionization ratios lies in the single-ionization cross section, a result previously indicated by Melo *et al* [4].

The theoretical description of the ionization of heavy targets is still a challenge. Our experiment relates to the measurement of exclusive cross sections [24, 25], and the apparent failure of the BA and CDW to describe the single-ionization cross sections might be related to the need to include this constraint in the calculations, since multiple ionization becomes substantial for heavy targets [26]. Indeed, a general scaling law for single ionization of H, He, Ne, Ar, and Kr by alpha particles can be successfully obtained through the independent-particle model, even if a rather crude analysis is used to calculate the ionization probabilities [25]. Furthermore, as shown in our previous work on Ne [5], multiple ionization at high velocities is strongly influenced by delayed post-collisional decay of inner electrons through Coster–Kronig and Auger transitions. There are no reasons for this effect not being present also in heavier targets. A more intriguing aspect of the results reported here is the observed charge and/or mass dependence of the *single*-ionization cross sections of heavy atoms. We should mention that the measurements reported by Santos *et al* [10] on ionization by He^+ show a very good agreement with the present proton measurements, at the same projectile velocity.

[4] See a comment about the El-Sherbini *et al* [15] results at the end of [19].

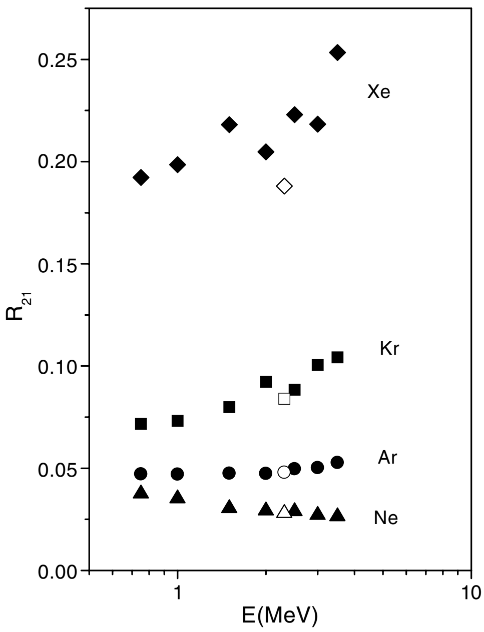

Figure 7. Cross section ratios of double to single ionization of Ne (triangles), Ar (circles), Kr (squares), and Xe (diamonds) by protons. Closed symbols, this work; open symbols, [8].

This agreement indicates that small mass differences are not important and distant collisions play a major role in the single-ionization cross sections since only these collisions are able to keep a dressed projectile, such as He^+, intact during its collision with a heavy atom [27, 28]. For distant collisions, He^+ behaves like a unit-charge bare particle and we should expect the same contribution as that from equi-velocity protons to the single-ionization cross section.

4. Summary and conclusions

Independent, absolute cross sections for single and multiple ionization of He, Ar, Kr, and Xe by proton impact in the 0.75–3.5 MeV energy range have been measured. The analysis of the present measurements together with previous, independent, absolute measurements using high-velocity proton and electron projectiles shows that cross sections for *single* ionization by electrons become increasingly larger than those for equi-velocity protons as the target atomic number increases. This tendency is not followed by double- or triple-ionization cross sections, where an overall agreement between the results for the two kinds of projectile is observed, without any clear systematic differences between them.

The general understanding that electrons, protons, and the corresponding antiparticles have identical single-ionization cross sections at high impact velocities [29] cannot be justified with the present experimentally available, independent, absolute data in the case of heavy targets. Although our findings point towards the current understanding that the cross section shapes for all these projectiles tend to merge at high velocities [29], the same cannot be said as regards the absolute values of single-ionization cross sections.

The identification of the mechanisms which lie behind the observed differences between the single ionizations of heavy targets by proton and electron impacts demands differential experiments of higher orders. For example, fine structures in the low-energy electron continuum [30], signatures of initial or final state correlations [31], and projectile energy loss [32] have been investigated, in detail, for ionizing collisions of Au^{53+} with Ne and Ar, through kinematically complete experiments. These are important signatures and mechanisms which can be suggested to help with the understanding of the observed differences reported in this paper.

Acknowledgments

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