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LETTER TO THE EDITOR

Multiple ionization of noble gases by 2.0 MeV proton impact: comparison with equi-velocity electron impact ionization

W S Melo, A C F Santos[1], M M Sant'Anna, G M Sigaud and E C Montenegro

Departamento de Física, Pontifícia Universidade Católica do Rio de Janeiro, Cx. Postal 38071, Rio de Janeiro, RJ, 22452-970, Brazil

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Abstract

Absolute single- and multiple-ionization cross sections of rare gases (He, Ne, Ar, Kr and Xe) have been measured for collisions with 2.0 MeV p^+. A comparison is made with equi-velocity electron impact ionization cross sections as well as with the available proton impact data. For the light rare gases the single-ionization cross sections are essentially the same for both proton and electron impacts, but increasing differences appear for the heavier targets.

The ionization of atoms by electrons and protons has been extensively and intensively studied for almost a century. Since the first quantum mechanical description of the ionizing collisions by bare, light particles, by Bethe in 1930, it has been recognized and almost universally accepted that the ionization cross sections, at high velocities, are the same for electrons and protons. Indeed, within the first Born approximation, the ionization cross section depends on the square of the charge of the incoming particle, varying with the projectile velocity, v, as $Z_1^2 v^{-2} \ln v$ and being essentially the same for equi-velocity electrons, protons and their corresponding antiparticles, if the velocity is much larger than the binding energy of the active electron.

The above assumption is regarded to be correct within the limits of the perturbative regime. However, the boundaries of this region are not specified for cases in which many-electron targets are involved and a large number of open channels are available. On the other hand, from the experimental side, comparisons between electron and proton ionization of multi-electron targets are surprisingly limited due to the small number of independent absolute measurements which have been carried out for both projectiles.

A rather detailed investigation of multiple ionization of noble gases by proton impact, over a broad range of velocities, was presented by DuBois and Manson (1987) and, by electron

[1] Present address: Department of Physics, University of Missouri-Rolla, Rolla, MO 65409, USA.

Table 1. Absolute single- and multiple-ionization cross sections of He, Ne, Ar, Kr and Xe by 2.0 MeV proton impact, in Mb.

Charge state	He	Ne	Ar	Kr	Xe
1	12.8 ± 2.0	29.9 ± 4.6	61.3 ± 9.4	65 ± 10	79 ± 12
2		0.81 ± 0.21	3.33 ± 0.68	6.4 ± 1.3	16.1 ± 2.8
3			0.31 ± 0.12	4.73 ± 0.98	6.9 ± 1.3
4				1.36 ± 0.39	1.39 ± 0.9

impact, by Krishnakumar and Srivastava (1988). From these data it can be observed that, for swift projectiles, there is an increasing discrepancy between the single-ionization cross sections by equi-velocity electrons and protons, as the target atomic number increases. In principle, there are no apparent reasons for such discrepancies. Unfortunately, a further verification of this tendency is prevented because the available proton measurements do not include elements above Kr. Indeed, to the authors knowledge, there are essentially no absolute systematic measurements for single-ionization cross sections, by high-velocity protons, covering the set of noble gases including Xe. In this work we report absolute measurements of the multiple-ionization cross sections of noble gases by 2 MeV ($v = 8.94$ au) protons. These measurements are compared with those obtained with 1 keV ($v = 8.57$ au) electrons.

The experimental apparatus was previously described by Santos *et al* (2001). In brief, a collimated mono-energetic 2.0 MeV H^+ beam is delivered by the 4 MV Van de Graaff accelerator of the Catholic University of Rio de Janeiro. The beam is selected according to its charge, mass and energy by a 90° magnet with energy resolution $E/\Delta E = 200$ and by a switching magnet before entering the beam line. In order to clean the main beam from spurious ones, before entering a windowless gas cell the beams are charge-analysed by a magnet placed just at the entrance of the cell. The emergent beams are again charge-analysed by a second magnet and recorded by a surface barrier detector housed in a detection chamber placed 4 m downstream of the gas cell. In this detection chamber, a x–y position-sensitive microchannel plate detector was used to identify and separate the spurious beams before the proper positioning of the surface barrier detectors. The counting rates of incident projectiles on the surface barrier detectors were kept below 1.5×10^3 s^{-1} in order to prevent pile-up effects. The multiply ionized recoil ions produced by the incident beam, under single-collision conditions, are accelerated by a two-stage electric field, separated in their mass-to-charge ratios by a time-of-flight mass spectrometer, and detected by two microchannel plate detectors in a chevron configuration. They provide a stop signal for two time-to-amplitude converters started by the signal from the surface barrier detectors. The efficiency of the surface barrier detectors was assumed to be unity. The recoil-ion detection efficiency was determined using a C^{3+} beam and looking for the capture channel in the singles and coincidence modes, as described by Santos *et al* (2001).

Figures 1–4 show the ionization cross sections corresponding to various final charge states (q) of the recoiling Ne, Ar, Kr and Xe ions, respectively, by 2 MeV protons and 1 keV electrons. Solid circles represent the present measurements, the diamonds proton measurements reported by DuBois *et al* (1984), the open squares absolute electron measurements taken from Schram *et al* (1966) and Schram (1966), the open circles absolute electron measurements taken from Chung and Cho (2001) and the open triangles electron data from Krishnakumar and Srivastava (1988), which are normalized to the results of Rapp and Englander-Golden (1965). In figure 1 the recent calculations by Kirchner *et al* (1999) for protons on Ne, including the KLL Auger contribution are included. The latter results are connected by a line to guide the eye. Table 1 summarizes our present results.

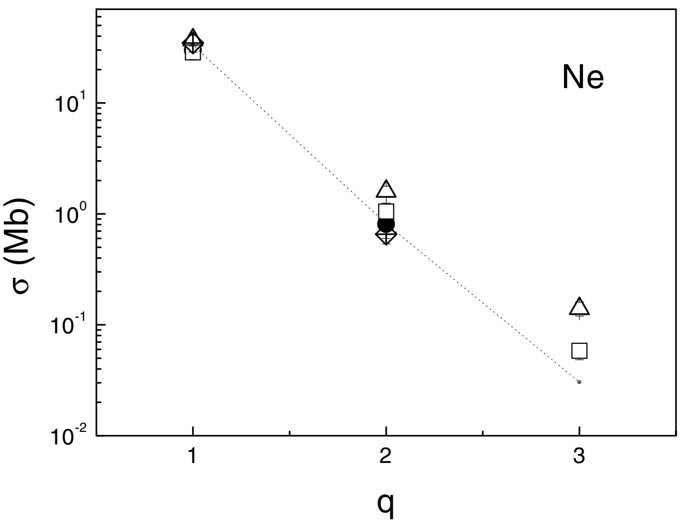

Figure 1. Multiple-ionization cross sections of Ne by 2.0 MeV protons ($v = 8.94$ au) and electrons ($v = 8.57$ au) as a function of the recoil ion charge state. Solid circles, p^+ (this work); diamonds, p^+ (DuBois *et al* 1984); open squares, e^- (Schram *et al* 1966, Schram 1966), open triangles, e^- (Krishnakumar and Srivastava 1988); and open circles, e^- (Chung and Cho 2001). Dotted line, theory (p^+, Kirchner *et al* 1999).

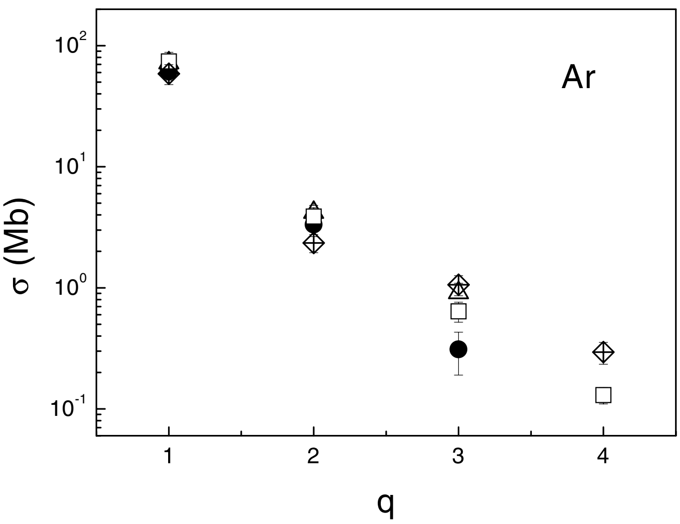

Figure 2. Multiple-ionization cross sections of Ar by 2.0 MeV protons ($v = 8.94$ au) and electrons ($v = 8.57$ au) as a function of the recoil ion charge state. Solid circles, p^+ (this work); diamonds, p^+ (DuBois *et al* 1984); open squares, e^- (Schram *et al* 1966, Schram 1966), open triangles, e^- (Krishnakumar and Srivastava 1988).

It can be seen from these figures that there is indeed a good general agreement between protons ($v = 8.94$ au) and electrons ($v = 8.57$ au), with approximately the same velocity, for all gases and for all charge states except for $q = 1$. In fact, for $q > 1$, apart from an increase of the dispersion in the measured cross sections as q increases and the cross section steeply decreases, there are no observed *systematic* differences between the proton and the

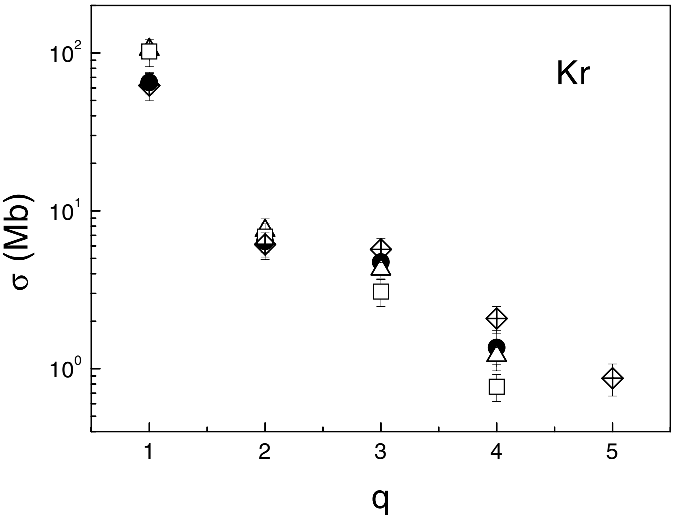

Figure 3. The same as figure 2 for Kr targets.

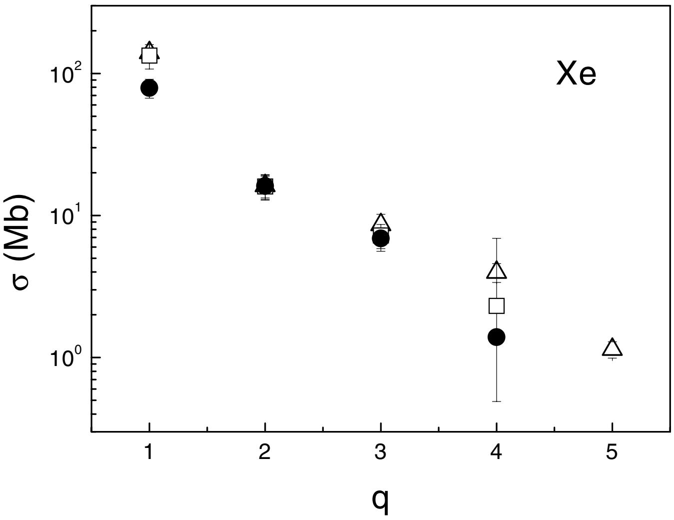

Figure 4. Multiple-ionization cross sections of Xe by 2.0 MeV protons ($v = 8.94$ au) and electrons ($v = 8.57$ au) as a function of the recoil ion charge state. Solid circles, p^+ (this work); open squares, e^- (Schram *et al* 1966, Schram 1966), open triangles, e^- (Krishnakumar and Srivastava 1988).

electron data. For $q = 1$, on the other hand, it is quite clear that the electron cross sections are significantly larger than the proton cross sections as the target becomes heavier. This unexpected behaviour is better seen in figure 5, where the *single* ionization cross sections ($q = 1$) are plotted as a function of the target atomic number. Although there is a good agreement between protons and electrons for the light targets, as expected from first-order theories, for Kr and Xe the electron cross sections are about 70% larger than the corresponding proton cross sections.

Experimental effort has recently been applied to elucidate the origin of the reported discrepancies occurring in multiple ionization of gases by electrons when measurements by

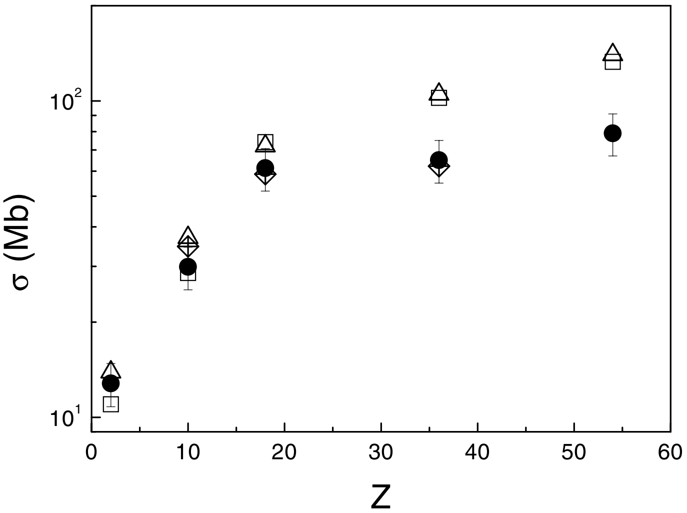

Figure 5. Single-ionization cross sections of Ne, Ar, Kr and Xe by 2.0 MeV protons ($v = 8.94$ au) and electrons ($v = 8.57$ au) as a function of the target atomic number. Solid circles, p^+ (this work); diamonds, p^+ (DuBois *et al* 1984); open squares, e^- (Schram *et al* 1966, Schram 1966), open triangles, e^- (Krishnakumar and Srivastava 1988).

different groups are compared (Almeida *et al* 1997, Almeida 1999, Chung and Cho 2001). As these discrepancies generally increase with the charge state of the recoil ions, most of these experiments normalize the gross cross sections to previously reported measurements and are focused on the relative distribution for the various charge states. Only a few of them present absolutely independent measured values for the *single-ionization* cross sections. In the proton case, our present absolute measurements are in excellent agreement with those of DuBois *et al* (1984) for the single-ionization channel but some discrepancies also appear when the recoil ion charge state increases.

If the present partial cross sections, σ_q, are used to obtain the gross ionization cross section, $\sigma_g = \Sigma q\sigma_q$, the resulting cross sections are smaller than those reported by the compilation of Rudd *et al* (1985). The same tendency occurs with the measurements of DuBois *et al* (1984) as well as when the partial electron ionization cross sections of Chung and Cho (2001) are compared with those from Krishnakumar and Srivastava (1988). Apparently, the absolute gross cross sections obtained from partial cross sections measurements using time-of-flight spectrometers have the tendency of being smaller than those obtained using different arrangements, such as the transverse-field method, for example. Following this tendency, the single-ionization cross section of Xe by protons reported by Manson and DuBois (1987), which was normalized to the gross cross section given by Rudd *et al* (1985), is 33% larger than our present result.

Our results show that the proton ratio between double- and single-ionization cross sections increases relative to that of electrons as the target becomes heavier, a result which was observed some time ago by Haugen *et al* (1982). However, our findings show that the origin of this behaviour is in the single-ionization cross section, which seems to contradict the idea that distant collisions play a minor role in the determination of that ratio. Indeed, it is not clear which element of the collision dynamics could privilege electrons compared to protons in the single ionization but not in double ionization. Preliminary CTMC calculations performed by Olson and Fiol (2001) do not show any difference between the two projectiles, either for single or double ionization, in the Ar case. Many of the conceptual reasonings raised to explain differences in the double-to-single ionization ratios of electrons and protons (see Haugen *et al* (1982), for example), such as the change of projectile trajectory due to different target fields, particle mass or momentum transfer to the target, changes in the binding energies of the active electrons, inner shell contributions, or differences coming from post-collision interaction are not yet presently available in a reliable quantitative form for heavy targets. It is worth mentioning that for proton and antiproton impact on light targets, such as He, the opposite occurs and the enhancement of this ratio is attributed to the larger cross section for double ionization by antiprotons, as discussed by Reading and Ford (1987) and Olson (1987).

In summary, cross sections for single and multiple ionization of noble gases by 2.0 MeV proton impact have been measured. The absolute values of the present set of data are not normalized to any previous measurements, thus providing an independent reference for studies of multielectronic processes in the intermediate-to-high velocity regime.

Differences between results for equi-velocity electrons and protons are observed for increasing target atomic number in the single-ionization cross sections. In contrast, the double- and triple-ionization cross sections of rare gases by both electron and proton impact do not show any significant systematic differences.

Single-ionization cross sections by electron impact have been used to normalize positron cross sections at high velocities (see, for example, Bluhme *et al* (1999)) based on the assumption that both cross sections should merge at high velocities, according to the first-order Born approximation. As the same reasoning can be applied for protons, it would be important to clarify the origin of the differences between proton and electron impact ionization measurements in heavy targets in order to confirm normalization procedures based on results given by the independent electron model associated with first-order theories.

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