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Influence of inner-shell electron removal on the multiple ionization of Kr and Xe by protons

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Abstract
The multiple ionization of Kr and Xe by protons has been analysed in detail within the independent particle model. The dependence of the multiple ionization cross sections on the values of the ionization probabilities has been investigated for each target subshell and the most relevant pathways for final quadruple to sextuple ionization are identified. The influence of post-collisional ionization probabilities due to Auger-like effects after the removal of one or more inner-shell target electrons has also been considered. It has been observed that, for large final charge states of the target, deep-core ionization plays an important role, due to the increase in the number of ejected electrons arising from post-collisional effects.

Keywords: multiple ionization, Kr, Xe, inner-shell, post-collisional

(Some figures may appear in colour only in the online journal)

1. Introduction

The description of target multiple ionization in collisions with swift projectiles is a complex task, because there are several mechanisms which may give rise to different pathways, involving both direct-impact and post-collisional mechanisms [1–21]. For low- to intermediate-velocity collisions, this complexity is present even in target double ionization, which is the simplest case possible, where the dependences of the cross sections on the projectile charge states and the influence of competitive collision channels has been shown to be quite strong [22–27].

As the number of electrons removed from a many-electron target increases, the analysis becomes even more difficult, because (1) the number of possible ways to reach a given target final charge state—namely, direct ionization of outer-shell electrons, multiple charge transfer accompanied or not by additional ionization, and inner-shell ionization followed by post-collisional electron emission, like shake-off, Auger, or Coster–Kronig processes—increases geometrically, and (2) other competitive collision channels—such as, electron loss and capture—may play an increasing role in these processes [27]. This is the case, for example, of collisions with highly charged heavy ions, where these latter processes, mainly electron capture from target inner shells, can strongly affect the multiple ionization of the target [28].

Within this scenario, the theoretical analysis of target multiple ionization by protons can be very helpful because not only the electron loss process is not present but, also, for high-velocity protons, electron capture is negligible. Thus, for these collision conditions, the mechanisms leading to multiple ionization can be investigated without the interference of other processes.

For the last 15 years, the importance of post-collisional electron emission after collisional removal of an inner-shell target electron (direct ionization) has been clearly noted by

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several authors [5, 7–11, 14–21]. These processes take place at times much longer than the collisional time and, thus, can be considered as independent of the projectile. Some of these studies have also been spurred by the recent experimental and theoretical advances in photoionization research which have provided a deeper understanding of post-collisional Auger-type electron emission (for a comprehensive review of the most recent photoionization data, the reader is referred to the recent work of Montanari and co-workers [19]).

The effect of post-collisional ionization in the slope of the multiple ionization cross sections at high energy has been found experimentally not only in ion [6–10, 12, 21], but also in electron impact multiple ionization [29–34].

The common approach to most of the above-mentioned theoretical investigations is based on the independent particle model (IPM), where the multiple ionization cross sections are obtained by a combination of the single-electron and post-collisional ionization probabilities using binomial distributions. The main differences between them concern the models which have been employed to calculate the primary single-electron ionization probabilities. The extension of the IPM for multiple ionization of very heavy targets, such as Kr and Xe, and high impact energies (proton energies above 1 MeV) showed discrepancies between theory and experiment for the quadruple and quintuple ionization of Kr and Xe [20].

As the target atomic number increases, on the one hand, the number of electrons potentially active for direct ionization by the projectile also increases. On the other hand, the influence of post-collisional electron emission after the ionization of a deep-shell electron becomes more important as well. It is, then, possible that the contribution of the combined effect of an inner-shell ionization followed by post-collisional electron emission is appreciable, due to the large probability of post-collisional emission, even if the probability of direct ionization of a deep-core target electron is much smaller than that of an outermost one. Thus, it is important to have a way of determining the influence of each possible pathway leading to the emission of a given number of target electrons.

In this paper, we present a method of calculating the cross sections for the multiple ionization of many-electron targets in which it is possible to identify the contributions of the different pathways leading to a given target final charge state, including post-collisional time-delayed electron emission. IPM-based calculations for multiple ionization become a heavy task when dealing with many shells. Our goal is to perform an individual analysis of the different contributions to highlight the relevant subshells which contribute to each final state.

The cross sections for the single ionization are calculated using the plane wave Born approximation (PWBA). The method is applied to the quadruple, quintuple and sextuple ionization of Kr and Xe targets by protons. It should be mentioned that, for heavy-ion collisions, first-order perturbative approaches are no longer valid and more sophisticated models should be employed to obtain the single ionization probabilities. In the present calculations we have considered the removal of L, M and N electrons of Kr and M, N and O electrons of Xe. The inclusion, for the first time, of deep-core electrons, namely from the 2s and 2p subshells of Kr and from the 3s, 3p and 3d subshells of Xe, has the aim of solving the discrepancies previously observed in [20].

The paper is organized as follows. In section 2 the calculation method is briefly outlined. In section 3 the results for the quadruple, quintuple and sextuple ionization of Kr and Xe are presented and compared with the available measurements for proton and electron impact. In section 4, some concluding remarks are made.

Table 1. Total number of terms for the q-fold ionization of Kr and Xe according to (1).

Target	$q = 4$	$q = 5$	$q = 6$
Kr	305	683	1358
Xe	467	1151	2510

2. Method of calculation

The general expression for the probability of ionization, $P_n^{(N_1,N_2,\ldots,N_\nu)}$, of n out of N target electrons, distributed in ν subshells with $N_1, N_2, \ldots, N_\nu$ electrons each, $N = \sum_{i=1}^{\nu} N_i$, as a function of the impact parameter of the collision is given by [7, 9, 10]

$$P_n^{(N_1,N_2,\ldots,N_\nu)}(b) = \sum_{\sum_i n_i + n_{\mathrm{post}} = n} \prod_{i=1}^{\nu} \binom{N_i}{n_i} \times [p_i(b)]^{n_i} [1 - p_i(b)]^{N_i - n_i} \mathcal{P}(n_1, n_2, \ldots, n_\nu, n_{\mathrm{post}}). \quad (1)$$

In the above equation, $p_i(b)$ is the probability of removal of at least one electron belonging to subshell i of the target due to the direct interaction with the projectile, n_i is the number of vacancies directly produced in subshell i by the projectile and $\mathcal{P}(n_1, n_2, \ldots, n_\nu, n_{\mathrm{post}})$ is the probability for post-collisional emission of n_{post} electrons after the creation of $n_1, n_2, \ldots, n_\nu$ vacancies in the corresponding target subshells.

As the atomic number of the target increases, so does the number of subshells which can be included in the calculations using (1), which obviously increases geometrically the numbers of terms in this equation. In table 1, the number of terms for the cases covered in the present work, namely, the quadruple, quintuple and sextuple ionizations of Kr and Xe, including the primary direct ionization of subshells 2s through 4p for Kr and subshells 3s through 5p for Xe, are shown. It can be seen that the number of terms ranges from 305, for the simplest 4-fold ionization of Kr, up to more than 2500, for the most complex case, the 6-fold ionization of Xe.

It is thus clear that expressions this size may, in principle, render the use of (1) very ineffective, a situation that gets increasingly worse as the degree of ionization of the target increases. However, as will be shown in the next section, a detailed analysis of the individual terms appearing in the expressions obtained from (1) can provide very useful information about the most relevant pathways which contribute to the total multiple ionization probability for each given final charge state of the target. Thus, with this information one can reduce the calculation of the multiple ionization cross sections to the evaluation of a much more restricted number of terms.

Table 2. Number of terms for the q-fold ionization of Kr and Xe according to (1) considering the vanishing photoionization branching ratios.

Target	$q = 4$	$q = 5$	$q = 6$
Kr	111	297	477
Xe	131	406	945

Table 3. Number of terms for the q-fold ionization of Kr and Xe according to (1) considering the vanishing combinations of photoionization branching ratios.

Target	$q = 4$	$q = 5$	$q = 6$
Kr	52	73	69
Xe	69	51	69

3. Results and discussion

As mentioned above, the impact-parameter-dependent ionization probabilities $p_i(b)$ have been calculated using the PWBA; for details of the calculations, the reader is referred to the recent works of Montanari and co-workers [19, 20]. The probabilities for post-collisional electron emission, $\mathcal{P}(n_1, n_2, \ldots, n_\nu, n_{\text{post}})$, which do not depend on the impact parameter of the collision, have been derived from the available data of photoionization compiled in [19]. Since, in the present calculations, we include the 2s and 2p subshells of Kr (besides the M and N shells) and the 3s, 3p and 3d subshells of Xe (besides the N and O shells), the probabilities for manifold post-collisional electron emission after the removal of one electron from those subshells have been taken from the photoionization data of [35, 36] (Kr) and [37, 38] (Xe). Thus, the probabilities $\mathcal{P}(n_1, n_2, \ldots, n_\nu, n_{\text{post}})$ are written, in the present case, as $\mathcal{P}(n_{2s}, n_{2p}, n_{3s}, n_{3p}, n_{3d}, n_{4s}, n_{4p}, n_{\text{post}})$ for Kr and $\mathcal{P}(n_{3s}, n_{3p}, n_{3d}, n_{4s}, n_{4p}, n_{4d}, n_{5s}, n_{5p}, n_{\text{post}})$ for Xe.

From the photoionization data mentioned above, it can be seen that, in several cases, the probabilities of post-collisional emission are zero. For example, when only electrons belonging to the outermost subshells of the target are ejected due to the direct interaction with the projectile, the probabilities for post-collisional electron emission vanish. When these zero probabilities are considered, the number of terms in (1) decreases significantly, as can be seen in table 2.

The branching ratios for post-collisional electron emission in [19, 35–38] refer exclusively to those situations where only one target electron is ejected due to its direct interaction with the impinging projectile. However, in (1) there appear terms which involve the primary direct ionization of more than one target electrons, with or without further post-collisional electron emission. These probabilities have been estimated following a procedure introduced in [10] and which considers that the target electrons are independent. Some examples are given below, in equations (2) and (3), for the simplest case treated here, the 4-fold ionization of Kr, using the values given by El-Shermi *et al* [35] and Morishita *et al* [36] for the L shell, and the values tabulated in [19] for the M and N shells. Equations (2) and (3) correspond to the case of direct triple ionization and the post-collisional emission of one additional electron.

$$\begin{aligned}\mathcal{P}(1,0,0,0,0,1,1,1) = \tfrac{1}{3}[&\mathcal{P}(1,0,0,0,0,0,0,1)\\ &\times\mathcal{P}(0,0,0,0,0,1,0,0)\times\mathcal{P}(0,0,0,0,0,0,1,0)\\ &+\mathcal{P}(1,0,0,0,0,0,0,0)\times\mathcal{P}(0,0,0,0,0,1,0,1)\\ &\times\mathcal{P}(0,0,0,0,0,0,1,0)+\mathcal{P}(1,0,0,0,0,0,0,0)\\ &\times\mathcal{P}(0,0,0,0,0,1,0,0)\times\mathcal{P}(0,0,0,0,0,0,1,1)] = 0.\end{aligned} \tag{2}$$

$$\begin{aligned}\mathcal{P}(1,0,1,0,0,1,0,1) = \tfrac{1}{3}[&\mathcal{P}(1,0,0,0,0,0,0,1)\\ &\times\mathcal{P}(0,0,1,0,0,0,0,0)\times\mathcal{P}(0,0,0,0,0,1,0,0)\\ &+\mathcal{P}(1,0,0,0,0,0,0,0)\times\mathcal{P}(0,0,1,0,0,0,0,1)\\ &\times\mathcal{P}(0,0,0,0,0,1,0,0)+\mathcal{P}(1,0,0,0,0,0,0,0)\\ &\times\mathcal{P}(0,0,1,0,0,0,0,0)\times\mathcal{P}(0,0,0,0,0,1,0,1)] = 0.\end{aligned} \tag{3}$$

In (2), the probability of ejection of one post-collisional electron after the primary ionization of three electrons, one belonging to the 2s and each of the other two from the 4s and 4p subshells (same shell—N), respectively, is estimated as the arithmetic mean of three probabilities, each consisting of the product of the probability of post-collisional emission of one electron after the ionization of one electron from a given subshell (for example, $\mathcal{P}(1,0,0,0,0,0,0,1)$ in the first term on the right-hand side of (2)) and the probabilities of removal of the electrons from the other two subshells with the emission of no post-collisional electrons ($\mathcal{P}(0,0,1,0,0,0,0,0)$ and $\mathcal{P}(0,0,0,0,0,1,0,0)$ in the same term). Since, according to [35], the probabilities of post-collisional emission of less than two electrons after the removal of a 2s electron of Kr as well as those of post-collisional electron emission after the ionization of the outermost shell are zero, all terms in (2) vanish.

A similar evaluation procedure is shown in (3) for the probability of ejection of one post-collisional electron after the primary direct ionization of three electrons, each belonging to different shells, in this case, the 2s, 3s and 4s subshells, which also vanish due to the same reason given above for $\mathcal{P}(1,0,0,0,0,1,1,1)$.

From the examples above, it can be concluded that there is still a considerable number of terms among those in (1) which are zero. Taking this into account, the number of non-vanishing terms is further reduced; those actually evaluated in the present work are listed in table 3.

The results of the present calculations for the quadruple, quintuple and sextuple ionization of Ke and Xe are presented in figures 1–3 as functions of the proton impact energy. In the high-velocity regime, the present calculations using the PWBA for the primary ionization probabilities are expected to coincide for protons and electrons. Thus, in these figures the experimental cross sections refer to protons from DuBois *et al* [40] and Cavalcanti *et al* [8] and electrons from Schram [29], Krishnakumar and Srivastava [30], Syage [31], Rejoub *et al* [32] and Kobayashi *et al* [33] for equal impact velocity.

As has been extensively observed in previous works, the importance of time-delayed post-collisional electron emission is again demonstrated here. As expected, the effects due to post-collisional emission increase as the target final charge state increases.

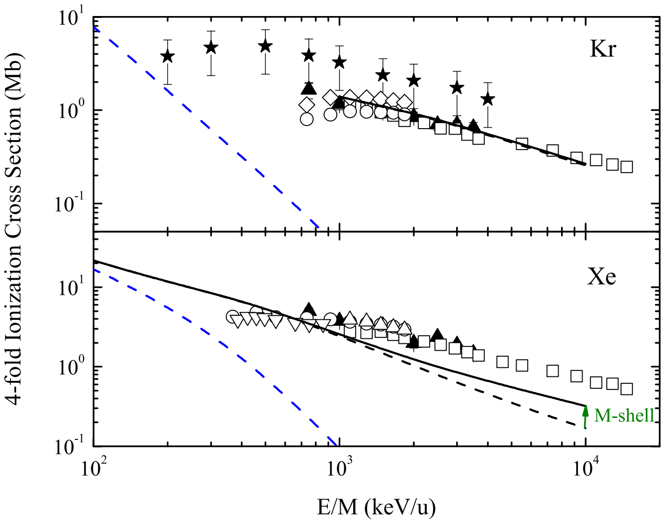

Figure 1. Quadruple ionization cross sections of Kr (upper panel) and Xe (lower panel) by proton impact as a function of half the square of the incident velocity in keV u^{-1}. Theory: solid black line, present calculations using (1), including the direct ionization of the L- and M-shell (for Kr and Xe, respectively); dashed black line, present calculations using (1), excluding the direct ionization of the L- and M-shell (for Kr and Xe, respectively); blue dotted line, calculations from [20] without the inclusion of post-collisional electron emission. Experimental data for protons: solid triangles, [8] (Kr and Xe) and solid stars, [40] (Kr). Experimental data for equi-velocity electrons: open squares, [29] and open circles, [32] (Kr and Xe); open diamonds, [30] (Kr); open down triangles, [31] and open up triangles, [33] (Xe). The green arrow indicates the difference in the calculated cross sections due to the inclusion of Xe M-shell direct ionization.

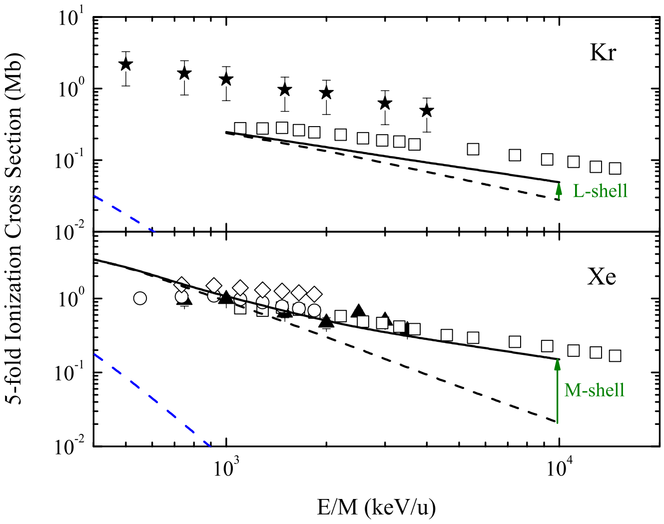

Figure 2. Quintuple ionization cross sections of Kr (upper panel) and Xe (lower panel) by proton impact as a function of half the square of the incident velocity in keV u^{-1}. Theory: the same as in figure 1. Experimental data for protons: solid stars, [40] (Kr) and solid triangles, [8] (Xe). Experimental data for equi-velocity electrons: open squares, [29] (Kr and Xe); open diamonds, [30] (Kr); open circles, [32] and open up triangles, [33] (Xe). The green arrows indicate the differences in the calculated cross sections due to the inclusion of Kr L-shell and Xe M-shell direct ionization.

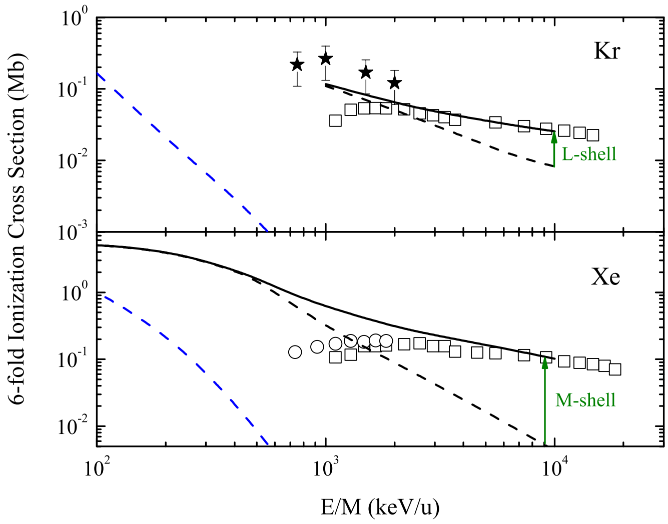

Figure 3. Sextuple ionization cross sections of Kr (upper panel) and Xe (lower panel) by proton impact as a function of half the square of the incident velocity in keV u^{-1}. Theory: the same as in figure 1. Experimental data for protons: solid stars, [40] (Kr). Experimental data for equi-velocity electrons: open squares, [29] (Kr and Xe) and open circles, [32] (Xe). The green arrows indicate the differences in the calculated cross sections due to the inclusion of Kr L-shell and Xe M-shell direct ionization.

The most important result of the present calculations is that, for the highest degrees of target ionization, the contribution from the removal of deep-core electrons becomes increasingly relevant. On the one hand, at 1.0 MeV u^{-1}, the primary ionization of Kr L-shell electrons contributes with 0.3%, 3% and 6% to the 4-, 5- and 6-fold ionization, respectively, while for Xe, the contributions of the initial removal of M-shell electrons are 5% for the quadruple, 14% for the quintuple, reaching almost a factor of 2 for the sextuple ionization. On the other hand, at 10.0 MeV u^{-1}, the increase in the 4-, 5- and 6-fold ionization cross sections are of 4%, 75% and a factor of 3 for Kr, and 50%, and factors of 7 and 20 for Xe, respectively. The reason for these large differences in the increases in the cross sections are obviously due to the fact that, while the probabilities for the direct electron removal decrease significantly with the projectile energy, those for post-collisional electron emission are velocity-independent, so that their relative influences become increasingly larger as the projectile energy increases. These numbers show that, even when the probability of direct removal of a deep-core electron is one or even two orders of magnitude smaller than those of outer-shell electrons, the contribution to the final target charge state can be quite substantial, due to the high probability of multiple electron post-collisional emission. The differences between the calculations at around 10.0 MeV u^{-1} without and with the inclusion of deep-core ionization are shown by green arrows in figures 1–3.

In general, the agreement between the present results and the experimental data is very good in the high-velocity region, even for the new results concerning the sextuple ionization cross sections. There are still some discrepancies, namely in the 4-fold ionization of Xe and the 5-fold ionization of Kr, where the present calculations including L-shell (Kr) and M-shell (Xe) ionization, although presenting better results than

Table 4. Most relevant pathways for the q-fold ionization of Kr and Xe (figures in parentheses denote the number of post-collisional emitted electrons). In the bottom line the total contribution of the pathways are shown.

	$q = 4$		$q = 5$		$q = 6$	
	Kr	Xe	Kr	Xe	Kr	Xe
	2p (3)	3d (3)	$2p4p^2$ (2)	3d (4)	2s (5)	3p (5)
	3s (3)	4s (3)	3s (4)	3d5p (3)	2p (5)	3d (5)
	3p (3)	4p (3)	3s3p (3)	4s (4)	3s3p (4)	3d4d (4)
	3p4s (2)	4p5s (2)	3s3d (3)	4s5s (3)	3s3d (4)	3d5p (4)
	$3d^2$ (2)	$4d^2$ (2)	3s4s (3)	4s5p (3)	$3s3d^2$ (3)	4s4d (4)
	3d4s (2)	4d5s (2)	3p3d (3)	4p4d (3)	3s4p (4)	4s4d5p (3)
	3d4p (2)	4d5p (2)	$3p4s^2$ (2)	4p5p (3)	$3p^2$ (4)	$4p^2$ (4)
	3d4s4p (1)	$4d^25p$ (1)	$3p4p^2$ (2)	$4p5p^2$ (2)	3p3d (4)	4p4d (4)
	$3d4p^2$ (1)	$4d5p^2$ (1)	$3p4p^3$ (1)	$4d^2$ (3)	3p3d4s (3)	4p4d5s (3)
			3d4s4p (2)	4d5s (3)	3p3d4p (3)	4p4d5p (3)
			$3d4s^24p$ (1)	$4d^25s$ (2)	$3d^2$ (4)	$4d^2$ (4)
			$3d4s4p^2$ (1)	$4d^25p$ (2)	$3d^24p$ (3)	$4d^3$ (3)
			$3d4p^3$ (1)	$4d5p^2$ (2)		$4d^25s$ (3)
			$3d4p^4$ (0)			$4d^25p$ (3)
						$4d5p^2$ (3)
Total	99%	98%	98%	95%	92%	91%

those without these shells, still lie below the experimental data. In principle, these discrepancies could be attributed either to the values of the branching ratios used here or to the exclusion of the direct ionization of electrons from even deeper shells. However, the different experimental (see [35] for Kr and [38, 39] for Xe) and theoretical works [37] agree in that the removal of electrons from deeper shells contribute to higher degrees of target multiple ionization.

The present calculations using (1) allow the evaluation of the individual contribution of each of the non-vanishing terms to the multiple ionization cross sections. We have observed that, in all the cases considered here, no more than 15 terms contribute with more than 90% of the total, indicating that there is no need to evaluate all the terms listed in table 3. In table 4, the most relevant pathways which lead to the target final charge states are shown for the 4-, 5- and 6-fold ionization of Kr and Xe by protons at the high velocity regime. It should be noted that, except in one case—the 5-fold ionization of Kr, where the term with no post-collisional emission contributes with less than 3% of the total cross section −, all of the most relevant pathways involve the removal of at least one inner-shell electron followed by the emission of post-collisional electrons. This is the reason why, for energies above 700 keV u^{-1}, the most relevant pathways which contribute to the multiple ionization cross sections are not dependent on the projectile energy.

In the first row of table 4, the deepest subshells which contribute to each q-fold ionization can be observed. For example, in the cases where the present results underestimate the experimental data, these subshells are the 3d for the 4-fold ionization of Xe and the 2p for the 5-fold ionization of Kr. Although the present calculations include the primary ionization probabilities of the whole L shell of Kr and M shell of Xe, the contributions from the even deeper subshells (namely, 2s for Kr and 3s and 3p for Xe) proved to be negligible in these cases, being, however, quite relevant for the 6-fold ionization of both targets.

An important feature which can be observed from this table is that there is a strong similarity between the most relevant pathways of both targets for a given ionization degree, with the obvious correspondence between shells with $n = 2$, 3 and 4 of Kr and those with $n = 3$, 4 and 5 of Xe. The most striking example is the case of the quadruple ionization, where the pathways for Kr and Xe are almost identical; similar correspondences also occur for the other cases. One can thus conclude that, in order to have estimates of multiple ionization cross sections of atoms which cannot be easily obtained either experimentally or theoretically, it is not necessary to perform a full calculation which includes all the possible ionization pathways, but only those few that are presented in table 4.

4. Summary and conclusions

We have calculated the cross sections for the multiple ionization of many-electron targets identifying and weighting the importance of the contributions of the different pathways leading to a given target final charge state, including post-collisional time-delayed electron emission. In the present work, the primary ionization probabilities have been calculated using the PWBA.

The method has been successfully applied to the quadruple, quintuple and sextuple ionization of Kr and Xe targets by protons and electrons, taking into account the removal of deep-core electrons, namely from the 2s and 2p subshells (besides the M and N shells) of Kr and from the 3s, 3p and 3d subshells (besides the N and O shells) of Xe, providing in general a very good agreement with experiment and thus solving some of the discrepancies previously observed in the calculations of Montanari and co-workers [20] which did not include these contributions.

It was observed that, in all cases, no more than 15 pathways are enough to account for more than 90% of the total multiple ionization cross sections, almost all of them including the removal of at least one inner-shell electron with the subsequent post-collisional emission of electrons. These pathways for Kr and Xe are very similar, which indicates that, not only one does not need to make a full calculation involving all the possible pathways, but, also, the most relevant pathways do not seem to change much from one target to another, a fact that can be useful if one wants to estimate multiple ionization cross sections in situations where either experimental or theoretical absolute cross sections are hard to obtain.

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