Q-DEPENDENCE OF THE DOUBLE CAPTURE CROSS SECTIONS MEASURED BY ELECTRON SPECTROSCOPY AT 10 qkeV ($q = 4-8$)⁽¹⁾. COMPARISON WITH OTHER EXPERIMENTAL DATA

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Résumé. Les sections efficaces de double capture dans des états autoionisants ont été mesurées par spectroscopie électronique. On montre qu'elles sont indépendantes du cœur de l'ion multichargé. Nos résultats sont comparés avec toutes les autres valeurs publiées à ce jour et obtenues à 10 qkeV.

Abstract. The q dependence of cross sections for double capture into autoionising states has been investigated by electron spectroscopy. It is shown that they are independent of the ionic core. Our results are compared with all other available experimental data obtained at 10 qkeV collision energy.

1 - Introduction

During the last years, it has been observed a growing interest of experimentalists and theoreticians for double capture processes in collisions between multicharged ions and two-electron targets. Electron spectroscopy is certainly the more suitable method to investigate these reactions which generally give autoionising states and it is necessary to have now a critical quantitative knowledge of these processes. A few years ago we have published the first experimental double capture cross sections [1] ; since then, many other collisional systems have been investigated, the reproducibility of our absolute cross sections tested and, also, angular distributions have been measured in order to achieve more meaningful total cross sections (when the angular distribution has not been measured, the estimation of the total cross section relies on 10° data : $O^{7+} + He$, H_2 and $O^{8+} + He$). Therefore one of the aims of this paper is to present and discuss total cross sections for double capture into autoionising states of the projectile (ADC) for various ionic species ranging from $q = 4$ to 8 , with He and H_2 targets. The whole set of values was obtained at a single collision energy equal to 10 qkeV, where q is the projectile charge. Our values will be compared with available data published by other laboratories ; we will also check the consistency with all the other cross sections for single capture (SC), double capture into bound radiative states (BDC) (these states lie below the first ionisation threshold) as well as for charge exchange, the so-called $(q,q-1)$ cross sections which are the sum of SC and ADC processes. This is the first comprehensive survey of total cross sections except for a more limited comparison for H_2 target made by Mack [2]. A compilation of $(q,q-1)$ cross sections in H_2 has been published by Tawara et al [3]. Data obtained with He-like, H-like and bare ions colliding with He and H_2 targets are considered for ion charges $q = 4$ to 8 .

2 - Method and uncertainties for ADC cross sections

Our cross sections are measured with the experimental apparatus already described elsewhere [3]. Absolute values are obtained by normalisation. Ionisation cross sections for H^+ on He system at 100 keV, 160° measured with the same apparatus, are normalised to Rudd's data [4] at 30 eV electron energy ; an averaged cross section equal to $1.5 \cdot 10^{-20} \text{ m}^2/\text{eV}/\text{sr}$ has been adopted as a reference value, known within $\pm 20\%$ [4]. As we measured angular distributions with an effusing beam target, the angular variation of the target volume seen by the analyser must be determined ; this was achieved by comparing the ratio $\sigma(\theta)/\sigma(90^\circ)$ (100 keV, 30 eV) with the one measured by Rudd [4]. An unknown error is also introduced

⁽¹⁾work performed on the ECR source of Lagripa Laboratory at Grenoble.

by the integration over the angles of singly differential cross sections (SDCS) because the angular mesh was not fine enough to ensure high accuracy to this integration (we generally measured electron spectra at five angles between 10° and 160°); fortunately, most of SDCS were found to have small angular anisotropies so that the error is thought to be often unimportant. On the whole, when the various errors are combined in a rms fashion, the uncertainty on the total cross section does not exceed $\pm 35\%$. This normalisation procedure, used at Toulouse, was checked in the ECR source at Grenoble by comparing the $(2p^2\ ^1D + 2s2p\ ^1P)$ cross section in $\text{He}^+ + \text{He}$ collision at 10 keV, 160° with the one previously measured a few years ago [5]; a good reproducibility was always found.

With an hydrogen target, an additional hypothesis concerns the evaluation of the target gas density; it differs from the one initially used [1]. We assume here that the target density is independent of the nature of the gas when the pressure above the capillary array is the same (measured with a Baratron gauge): this results from the so-called transparent mode (molecular flow conditions) [6]. Therefore, the result of the normalisation described above for He is also applied to H_2 .

The reproducibility of the cross sections was regularly tested by measuring them for the same system at one or several years intervals (apart from the 0^+ and 0^{8+} data which was only measured once). We have also investigated a possible systematic source of error in the determination of the number of ions which interact with the target; indeed, between the collision region and the ion collector, the ions travel a length of about 45 cm in the residual gas at a pressure which does not exceed 10^5 torr. Adopting the procedure described by El Sherbini et al [7], and assuming that the ion beam at the collector is mainly composed of q and $(q-1)$ charges, we found that the correcting factor is negligible even in an unfavourable case ($\text{Ne}^{8+} + \text{H}_2$, SC cross section $= \sigma(q, q-1) = 6 \cdot 10^{15} \text{ cm}^2$, see table 1).

Our total cross sections are given in table 1 (columns 2 and 3). They are compared with other data measured by electron spectroscopy (col. 4 and 5) at the same collision energy 10 qkeV [2,8,9]; the data of Mann [10] obtained at 20 qkeV are also included. All these results have been obtained at a single observation angle (50° in [2,8], 0° in [9,10] and total cross sections for these works assume an angular isotropy. Absolute values measured by Meyer et al [9] are given within $\pm 30\%$; those of Mann [10] can suffer from a largest error since it seems that they result from a normalisation to other data obtained at much lower collision energy. The values of Mack [8] were first normalised to our earlier (and preliminary) data on $\text{N}^{6+} + \text{H}_2$ [1]; however our present value is significantly higher and this procedure is no longer valid. In ref. 2, this author adopts another normalisation to the $1s^2 3s\ ^3P$ part of the $0^{6+} + \text{He}$ electron spectrum measured by Meyer et al [9]; these new values are given in table 1. As the relative values are known within $\pm 25\%$ [8], the error bar on the absolute values can be estimated to be about $\pm 39\%$. In all these works, the quoted uncertainties apply to the SDCS values measured at one angle; the error on the total cross section can be larger than that (angular isotropy hypothesis).

For all the other cross sections given in table 1 (col. 6 to 8), the reader is requested to refer to the original papers for the error bars; only data obtained at 10 qkeV are included in this table.

3 - Present results for total ADC cross sections and comparison with other data

3-1 Helium target

The charge dependence of our results is shown in fig. 1a. Two very clear tendencies are observed:

i) First, ADC cross sections increase quickly with q , and we note a steep increase above $q = 6$. In terms of a reaction window, the strong decrease for low charge He-like ions ($q \leq 6$) comes from a shift of the window for double capture into low-lying non autoionising $1s^2 2n\ ^1S$ bound states [8]. The situation is not so clear for N^{6+} for which a weak ADC value is also observed; this may be due to the fact that the reaction window is centered between the two $1s 2n\ ^1S$ and $1s 3n\ ^1S$ autoionising series, where no state is available [2].

Table 1. Comparison of our total ADC cross sections with other ADC experimental data as well as with BDC, SC and (q,q-1) experimental cross sections (see text). ADC, BDC = double capture into autoionising and bound radiative states respectively ; SC = single capture cross section ; q,q-1 = charge exchange cross section = SC + ADC ; cross sections are given in 10^{-16} cm^2 .

Ion species	ADC				BDC	SC	(q,q-1)
	Our results $\pm 35\%$		Other data				
	$3\ell n'\ell'+4\ell n'\ell'$	$2\ell n'\ell'$	$3\ell n'\ell'+4\ell n'\ell'$	$2\ell n'\ell'$			
	<u>Helium target</u>						
C4+(1s2)			0.076 ^c	<0.20 ^a , <0.08 ^b , 0.057 ^c	2 ^d	3.5 ^e	3.2 ^d
C5+(1s)			<0.4 ^b , 0.33 ^e	6.7 ^b , 2.5 ^c			
N5+(1s2)	0.56	0.050	0.88 ^a , 0.8 ^b	0.88 ^a	2.1 ^d	10.7 ^e	11.1 ^d , 10.6 ^f
C6+			$\leq 0.5^b$	$\leq 0.75^b$		12.2 ^t	17 ^l
N6+(1s)	1.1	$\approx 0.9^g$	0.78 ^b	0.75 ^{bs}		7.6 ^h , 15.3 ⁱ	
O6+(1s2)	1.3	0.16	2.2 ^a (=ref) ^b	1.3 ^a	1.7 ^d	12.7 ^e	13 ^d
N7+	9.0				1.5 ^l		24 ^j , 10 ^k
O7+(1s)	10	(1.1)*	6.8 ^a , 3.3 ^b	1.3 ^a , $\leq 0.4^{bs}$			
O8+	23						22 ^l , 17 ^m
Ne8+(1s2)	17	$\leq 0.4^{**}$	8 ^b			16.5 ⁿ , 10 ^u	
	<u>Hydrogen target</u>						
C4+(1s2)	2.0	≈ 0.2	1.5 ^a , 2.4 ^b , 0.13 ^c	1.5 ^a	3 ^s	23.6 ^e	23.2 ^o , 24 ^s , 48 ^j
C5+(1s)			3.4 ^c	0.42 ^c			20.0 ^o , 42 ^j , 23.5 ^q
N5+(1s2)	9.0	0.3	10.0 ^a , 9.2 ^b	3.2 ^a	3P, ^s	16.9 ^e	20.8 ^e , 32.6 ^j , 30P
C6+							22.5 ^o , 23 ^s
N6+(1s)	33		17.7 ^b	< 0.4 ^b			52 ^j , 41.2 ^r
O6+(1s2)	34	0.05	21.2 ^b		2P	30.7 ^e	52 ^j , 65P, 41.6 ^r
							36.8 ^e , 68 ^j , 34 ^o ,
							55P, 38.6 ^q , 37 ^s
N7+	26				90P		48P, 55 ^j , 38.7 ^r
O7+(1s)	26	(5.4)*	12.8 ^b				65 ^j , 38.9 ^r
O8+							73.4 ^j , 49.2 ^r
Ne8+(1s2)	36	seen ^{**}	20.5 ^b			30 ^u	$\approx 60^j$

^g not corrected for radiative cascades.

** superimposed on $4\ell n'\ell' \rightarrow 3\ell \epsilon \ell'$ series ($n' = 4, 5$). With H₂, the $2pn\ell$ contribution is included in the $(3\ell n'\ell' + 4\ell n'\ell')$ data.

* these data, which include a contribution of double collisions $O^{7+}(1s)$ $O^{6+}(1s2s^3S) \rightarrow O^{5+}(1s2\ell n'\ell')$, will not be considered further in this paper.

a : ref. 9 - b : refs. 2,8 - c : ref. 10 - d to n : refs. 15 to 25 respectively o to s : refs. 27 to 31 respectively - t : ref. 36 - u : ref. 37.

ii) Secondly, an equality of the total ADC cross sections for ions with the same charge, that is, with different ionic cores, is observed. This finding applies unambiguously to total DC cross sections for $q \geq 7$ since BDC processes are negligible for these ions. A word must be said on our O^{7+} data ; the value given in fig. 1a only concerns the $1s3\ell n'\ell'$ part of the spectrum (table 1) because double capture into lower $1s2\ell n'\ell'$ series is affected by double collisions with metastable $O^{6+}(1s2s^3S)$ ions. This was discussed in detail by Mack and Niehaus [34] for $N^{6+} + \text{He}$. Contrary to their conclusion we have found in our experiment that a preponderant fraction of metastable ions is independent of the target pressure (and hence the spectrum cannot be corrected) ; they are certainly produced near the ion beam slits [35]. Therefore as the true capture

Fig. 1 : Variation of the ADC cross sections (see text) with the ion charge q .
Fig. 1a : helium target ; Fig. 1b : hydrogen.

into $1s2n'l'$ is much smaller than in $1s3n'l'$ [34] we have neglected this contribution in fig. 1a,b. For $N^{6+} + \text{He}$ we correct our spectrum by comparison with the result obtained by Mack and Niehaus [34] ; for $N^{6+} + \text{H}_2$ the $1s2n'l'$ part is negligible. The equality of O^{6+} and N^{6+} ADC cross sections (fig. 1a) must be considered with care because part of the double capture by O^{6+} occurs into bound states (see section 4). Also for N^{6+} , part of the double capture into $1s2n'l'$ states can be lost by radiative decay of some states on lower singly excited states. Finally, we note that the independence of the total cross sections on the ion core was already observed in many SC processes (see for example [41,42]) and $(q,q-1)$ cross sections [38, and references given therein].

Point i) is partly verified by data of other authors [2,9]. Above $q = 6$, Mack's results do not increase as steeply as ours and $q = 7,8$ cross sections are a factor of two smaller. N^{5+} cross section is found to be an order of magnitude smaller than the C^{5+} one (fig. 1a, table 1) ; the small N^{5+} ADC value is related to strong BDC processes for such He-like ions. The C^{6+} cross section is near that of N^{6+} and O^{6+} . The results of Meyer et al [9] concern He-like ions with an additional value for O^{7+} . For this ion, it is known that the reaction window peaks between $1s3l3l'$ and $1s3l4l'$ [8] ; the $1s2n'l'$ series is outside this window except for the upper states. It is the autoionisation of these states lying above the $1s2s^3S$ limit which is observed by Meyer et al [9] (it is not observed in [8]) to have a stronger probability of excitation than expected from the reaction window calculated by Mack [8] (table 1) ; also the excitation of lower $1s2n'l'$ ($n' \leq 5$) states is found to be weak in [8] (it is not mentioned in [9]). We note that the $q = 7$ cross section of Meyer et al is in good agreement with ours even if our value is only a lower bound since it does not include $1s2n'l'$ series (table 1) ; it is a factor of two higher than that measured by Mack [2] as for O^{6+} ions. For O^{6+} this is because the cross section normalisation in [2] (see section 2) only applies to the $1s^23n'l'$ part of the spectrum ; Meyer et al. also found a strong double capture into $1s^22pn'l'$ states which equals the $1s^23n'l'$ one (table 1), an observation also made by Mann and Schulte [13] but verified neither in our work [14] nor by Mack [2,8]. This strong discrepancy (table 1)

is due to the difficulty of measuring a true intensity for low energy electrons ($E_e \leq 20$ eV) [14]. The $q = 4$ data of Mann [10] agrees well with the result of Meyer et al. [9] even if it is obtained at a higher collision energy (20 keV); the C^{5+} value is larger than the He-like cross sections as already noted for Mack's value (see section 4) but these two data disagree within a factor of two.

On the whole, total ADC cross sections for the helium target only agree within a factor of two but it is necessary to remember that for He-like systems, partial total cross sections can suffer much larger discrepancies (capture into $1s^2 2p n l$; table 1).

3-2 Hydrogen target

Our results (fig. 1b) show a rapid increase of ADC cross sections up to $q = 6$; the following interesting features must be noted:

i) Contrary to the helium case, an almost constant value is noted in the q -range 6 to 8. This means to our opinion that the sharp increase noted in helium above $q = 6$ (fig. 1a) is not due to an experimental artefact related to a measurement error with high charge ions. A small oscillatory behaviour is seen with a minimum value for $q = 7$. Even if this structure lies within the error bar, it is repeatedly observed for various ion species: N^{7+} and O^{7+} cross sections are about 30 % lower than those measured for O^{6+} , N^{6+} and 38 % lower than the Ne^{8+} value.

This oscillation is confirmed by the data of Mack [2] where the O^{7+} cross section is 40 % lower than $q = 6$ and 8 ones. As in our work, the equality of O^{6+} and N^{6+} cross sections is well observed. The absolute values for $q = 6$ to 8 are smaller than ours by a factor slightly smaller than two. We note the very good agreement for $q = 4$ and 5 values between the two sets of data. Such a minimum value for $q = 7$ cross sections was also observed in $(q, q-1)$ cross sections measured for bare ions on H_2 target by Meyer et al [38]. It was interpreted in terms of a simple Landau-Zener model [39] (see also [40]) identifying $(q, q-1)$ cross sections with SC ones; the large ADC value measured for $N^{7+} + H_2$ together with the same kind of oscillation noted in the ADC cross section means that the interpretation must be revised. The results of Meyer et al. for $q = 4$ and 5 [9] are near our results although systematically above as in helium and for the same reasons. The cross section obtained by Mann [10] for C^{5+} seems too small by a factor of two; also, the value for C^{4+} is an order of magnitude too small.

4 - Total double capture cross sections $DC = ADC + BDC$

It has already been mentioned that low charge He-like cross sections ($q \leq 6$) must be corrected for double capture into $1s^2 2p n l$ bound states (BDC) by adding BDC cross sections (table 1, col. 6) to corresponding ADC values.

4-1 Helium target

When this correction is made, we note for $q = 4$ a clustering of different data around the same value which is due to the preponderant weight of BDC cross section over the ADC ones for this ion charge. For $q = 5$, the apparent discrepancy between N^{5+} and C^{5+} values observed in fig. 1a is removed when considering the C^{5+} value of Mann [10]; the C^{5+} cross section of Mack [2] seems too high by a factor of two. For $q = 6$, the order of N^{6+} and O^{6+} cross sections is reversed using our data but the final DC values are again very near each other; on the other hand, the equality of $q = 6$ values is not so well verified using Mack's results; also the C^{6+} value appears too small. For $q \geq 7$ no correction has to be applied to ADC values which can be considered equal to DC values: $ADC \approx DC$. We don't consider the BDC value of Afrosimov et al. [23] for N^{7+} because it seems surprisingly high since electron spectroscopy shows that about 70 % of the double capture occurs into $3l3l'$ non radiative states [1,26].

In summary, even when ADC are corrected for BDC processes, the equality of DC cross sections seems to be always verified whatever the ion core is; the only serious discrepancies come from some of the values of Mack [2] which are too high (C^{5+}) or too low (C^{6+}, O^{7+}) with respect to the other data.

4-2 Hydrogen target

Nothing has to be changed in the previous discussion of fig. 1b since ADC cross sections are much larger than the BDC ones (capture into $1s^2 2n'\ell'$) except for C^{4+} (table 1). As in helium, the N^{7+} BDC value [28] seems very large (table 1); however it is recognised that there exists a possibility of radiative decay of upper $3n'\ell'$ states ($n' > 8$) if these states are excited (they don't appear in the electron spectrum [1,32]). Adding such a large BDC value to our ADC cross section makes the above mentioned oscillation less clear.

5 - Comparison of $(q,q-1)$ cross sections with the sum (SC + ADC)

It is well known that the $(q,q-1)$ cross sections measured in charge exchange experiments by only detecting the variation of the ion charge gives the sum (SC + ADC). A comparison of all the experimental data is made in table 1. In figures 2a,b, these values are only represented by vertical bars which join the limit values; the ADC cross sections are recalled by thin lines disregarding the C^{5+} values which cannot be tested since no SC cross section is available for this system.

5-1 Helium target (fig. 2a)

For $q \leq 6$ the agreement between (SC + ADC) and $(q,q-1)$ cross sections is remarkable; for N^{6+} we have taken the largest SC value [20]. It is worth noting that the agreement for $q = 6$ concerns various ionic species. No SC value exists for $q = 7$; we note that there exists a large discrepancy between the two $(q,q-1)$ cross sections for N^{7+} ; the plot of all the data show that an average value of about $1.7 \cdot 10^{-15} \text{ cm}^2$ seems the more correct one. For $q = 8$, taking an average SC value give a reasonable agreement with $(q,q-1)$ cross sections well within the error bar which is about $\pm 46\%$ for the summed (SC + ADC) cross section. The ratio ADC/SC for $q = 8$ lies between 1.3 to 0.6 due to the large scattering of ADC data (1.3 with our data); nevertheless it indicates the large part taken by the ADC processes at 10 qkeV for $q \geq 7$ ionic species.

From this comparison we conclude that the overall agreement between all the data is reasonable even if the sum (SC + ADC) is systematically higher than the $(q,q-1)$ cross section; this discrepancy is however within the error bars.

Figure 2. Comparison of (ADC + SC) and $(q,q-1)$ cross sections at 10 qkeV. The same notations as in fig. 1 are used.

5-2 Hydrogen target (fig. 2b)

The same agreement as for helium is observed and the same remarks apply here. We note that the large vertical bars for $(q, q-1)$ data are often due to the values measured by Panov et al. [21] which seem too high, particularly for C^{4+} and C^{5+} values (table 1). Also, as for helium, the ratio ADC/SC is near unity for $q \geq 6$ multicharged ions at 10 qkeV (table 1).

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