Z-DEPENDENCE OF THE DOUBLE CAPTURE IN $Aq+(1s2) + He, H2$ SYSTEMS ($A=C,N,O,Ne$) OBSERVED AT 10 qkeV FOR VARIOUS ANGLES. IDENTIFICATION OF $1s23l\ 3l'$ LINES: THEORY AND EXPERIMENT

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Z-DEPENDENCE OF THE DOUBLE CAPTURE IN $Aq^+(1s^2) + He, H_2$ SYSTEMS ($A=C, N, O, Ne$) OBSERVED AT 10 keV FOR VARIOUS ANGLES. IDENTIFICATION OF $1s^2 3\ell 3\ell'$ LINES : THEORY AND EXPERIMENT

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Résumé. La capture de deux électrons observée dans les collisions entre des ions héliumoides ($Z = 6-10$) et une cible He ou H_2 est étudiée à 10 keV en fonction de l'angle d'observation. De nouveaux résultats théoriques pour les états $1s^2 3\ell 3\ell'$ permettent une identification des raies observées.

Abstract. The first accurate identification of $1s^2 3\ell 3\ell'$ states populated by two-electron capture in collisions of He-like ions with helium and hydrogen is made, by comparing new theoretical data with electron spectra. General trends of the double capture are given when Z varies from 6 to 10. The angular dependence of the electron spectra is also shown.

1 - Introduction

The investigation, by electron spectroscopy, of the capture of two electrons by helium-like ions was pursued for several years on the ECR source of Lagrippa Laboratory at Grenoble. Partial results obtained for $N^{5+}(1s^2) + H_2$ [1], $N^{5+}(1s^2)$ and $O^{6+}(1s^2) + He$ [2], and $Ne^{8+}(1s^2) + He, H_2$ [3,4] have already been published together with a preliminary report on the subject of this paper [5]. However this is the first time that an accurate identification of $1s^2 3\ell 3\ell'$ states is given by comparing new theoretical data with electron spectra : for C^{4+} , N^{5+} and O^{6+} ions ; some results have already been published for Ne^{8+} [3,4]. The angular dependence of the electron spectra will also be discussed. Similar studies have been undertaken in other laboratories [6-13].

2 - Electron spectra

In figures 1 to 3 are shown for each system the angular dependence of the n distributions ; these results complete our earlier data on $Ne^{8+} + He, H_2$ [3,4] (see also refs. 6 to 13). Clear anisotropies are observed mainly for hydrogen target with C, N, O ions but also for helium target with Ne^{8+} [4]. As the reaction window peaks on low lying $1s^2 2\ell n'\ell'$ states in $C^{4+} + H_2$ and $N^{5+} + He$ systems [6], small double capture differential cross sections are measured in these cases (fig. 1 and 2a) ; mainly $n' = 3$ terms of the $1s^2 3\ell n'\ell'$ series are seen and double capture processes appear to be rather selective (fig. 1 and 2a). With $O^{6+} + He$, the $n' = 4$ states are excited with a comparable probability as the $n' = 3$ ones (fig. 3a) ; from the known structure of the $n' = 4$ part of the spectrum in $Ne^{8+} + He$ (fig. 5 in [4] and ref. 19), it may be surmised that mainly the $3s4\ell \rightarrow 2p\ell\ell'$ lines are seen in fig. 3a ; in the high energy wing appear the $3p, d4\ell \rightarrow 2p\ell\ell'$ components. The latter states are more strongly excited in $O^{6+} + H_2$ (fig. 3b) and the identification given for $Ne^{8+} + He$ [4] applies here since the two spectra are very similar (compare fig. 3b with fig. 5 in [4]), the $n' = 4$ states being as excited as the $n' = 3$ ones (as for $Ne^{8+} + He$ [4]).

At low electron energies ($E < 10$ eV) additional lines are observed (10° spectra in fig. 1 to 3) corresponding to the autoionisation of upper states of $1s^2 2p n\ell$ series into $1s^2 2s\ell\ell'$ continuum. These lines have recently been analysed by Meyer et al [13] for $O^{6+} + He$ system, they come from a direct double capture

into $1s^2 2n'l'$ series (this also applies to $C^{4+} + H_2$, $N^{5+} + He$ and H_2 systems). For $O^{6+} + H_2$ (fig. 3b) and Ne^{8+} spectra (fig. 5 and 6 in [4]), their meaning is less clear and radiative cascades from upper terms of $1s^2 3n'l'$ series must be put forward [12]. This is particularly true in Ne^{8+} spectra where $2pnl$ lines are observed to be more strongly excited with H_2 than with He target (compare fig. 5 and 6 at 10° in [4]). Whereas the reaction window shifts in hydrogen towards the upper terms of $1s^2 3n'l'$ series and the $1s^2 4n'l'$ one [4]. We also recall the strong discrepancies noted between the various experimental results for the absolute strength of the $3pnl$ lines [4,6,7,9,11-13, see also the companion paper, ref. 20].

From this discussion it appears that an identification of the $3l3l'$ states will be very interesting to obtain valuable informations on the double capture process itself. This implies that accurate theoretical calculations of the positions and lifetimes of $3l3l'$ states are made.

Figure 1: Electron spectra observed in $C^{4+}(1s^2) + H_2$ collision at 10 keV, for different observation angles θ_{lab} . Labels "A", "a", ... see text, 1 = $3s^2 1S$, 2 = $3s3p 1P$, 3 = $3s3d 1D$, 4 = $3p^2 1S$, 5 = $3p3d 1D$, 6 = $3p^2 1D$, 7 = $3d^2 1G$, 8 = $3p3d 1F$, 9 = $3d^2 1D$, 10 = $3p3d 1P$, 11 = $3d^2 1S$, 12 = $3d^2 3P$.

Figure 2 - As fig. 1 for $N^{5+}(1s^2) + He$ (fig. 2a) and $N^{5+}(1s^2) + H_2$ (fig. 2b).

Figure 3 : As fig. 1 for $O^{6+}(1s^2) + He$ (fig. 3a) and $O^{6+}(1s^2) + H_2$ (fig. 3b).

3 - Theoretical positions and lifetimes for $1s^2 3l 3l'$ states of C^{2+} , N^{3+} and O^{4+} ions

3-1 Calculation of the atomic parameters

The atomic parameters, energy levels, wavelengths λ and radiative transition probabilities A_r , have been calculated using the SUPERSTRUCTURE code developed by Eissner et al. [14]. The program includes in the hamiltonian some of the Breit-Pauli relativistic corrections. The autoionisation probabilities A_a for all the available continua have been obtained using the AUTOLSJ program described in a paper by the TFR group [15]. These two programs use the same wavefunctions and model potentials, and are well suited for the computation of large quantities of atomic data for multiply charged ions. They used scaled Thomas - Fermi - Dirac potentials for which the scaling parameters λ_e are determined by an energy minimization. For C^{2+} , N^{3+} and O^{4+} ions λ_s and λ_p are adjusted through a minimization of the sum of the energies of all the terms belonging to the configurations $1s^2 2s^2$, $1s^2 2s 2p$, $1s^2 2p^2$, $1s 2s^2 2p$, $1s 2s 2p^2$ and $1s 2p^3$ (λ_d was set equal to λ_p). The values of the scaling parameters are given in table 1.

Table 1 - Thomas - Fermi scaling parameters used for the calculations.

	C^{2+}	N^{3+}	O^{4+}
λ_s	1.875	1.875	1.877
$\lambda_p = \lambda_d$	1.569	1.572	1.575

The energy levels, wavelengths and transition probabilities have been calculated using multiconfiguration expansions containing, for the four-electron systems, the configurations $1s^2 n n' l'$ with n and n' equal to 2 and 3 and $l' = 0, 1, 2$. The relative accuracy on energy levels is about 10^{-3} .

The energies, autoionisation and radiative decay probabilities are given in tables 2 to 4 for C^{2+} , N^{3+} and O^{4+} respectively.

Table 2 - Theoretical energies and decay probabilities for $C^{2+}(1s^2 3\ell 3\ell')$ states. E is given with respect to the ground state $1s^2 2s^2$. Aa(2 ℓ) = autoionisation probability towards $2\ell\ell'$ continuum; $\Sigma Aa = Aa(2s) + Aa(2p)$. ΣAr = total radiative decay probability. Notation used : $1.34+14$ means $1.34 \cdot 10^{14}$.

$C^{2+}(1s^2 3\ell 3\ell')$	E (Ryd)	Aa(2s) (s^{-1})	Aa(2p) (s^{-1})	ΣAa (s^{-1})	ΣAr (s^{-1})
$3s^2 \ 1S$	4.690	$1.34+14$	# 0	$1.34+14$	$8.76+9$
$3s3p^3P$	4.797	$5.69+13$	$8.20+13$	$1.39+14$	$9.18+9$
$3s3p^1P$	4.926	$1.62+14$	$3.59+14$	$5.21+14$	$9.85+9$
$3s3d^1D$	4.928	$8.68+13$	$2.88+14$	$3.75+14$	$1.30+10$
$3s3d^3D$	4.976	$1.50+13$	$1.01+13$	$2.51+13$	$1.51+10$
$3p^2 \ 3P$	5.007	0	$2.32+14$	$2.32+14$	$9.75+9$
$3p3d^3F$	5.078	$2.45+12$	$2.48+12$	$4.93+12$	$1.52+10$
$3p3d^1D$	5.089	0	$1.14+13$	$1.14+13$	$1.55+10$
$3p^2 \ 1S$	5.095	$1.38+14$	$7.01+14$	$8.39+14$	$1.33+10$
$3p^2 \ 1D$	5.132	$3.51+13$	$1.09+14$	$1.44+14$	$1.64+10$
$3p3d^3D$	5.174	0	$6.96+13$	$6.96+13$	$1.56+10$
$3p3d^3P$	5.183	$5.16+12$	$3.32+13$	$3.84+13$	$1.58+10$
$3d^2 \ 3F$	5.237	0	$6.02+13$	$6.02+13$	$2.22+10$
$3d^2 \ 1G$	5.309	$6.48+13$	$2.04+14$	$4.21+14$	$1.72+10$
$3p3d^1F$	5.321	$2.94+13$	$2.13+14$	$2.42+14$	$1.39+10$
$3d^2 \ 3P$	5.325	0	$1.23+12$	$1.23+12$	$2.56+10$
$3d^2 \ 1D$	5.405	$6.11+12$	$1.27+14$	$1.33+14$	$2.01+10$
$3p3d^1P$	5.432	$1.78+13$	$9.49+13$	$1.13+14$	$1.54+10$
$3d^2 \ 1S$	5.611	$1.49+13$	$3.78+13$	$5.28+13$	$2.31+10$

Table 3 - As table 2 for $N^{3+}(1s^2 3\ell 3\ell')$ states.

$N^{3+}(1s^2 3\ell 3\ell')$	E (Ryd)	Aa(2s) (s^{-1})	Aa(2p) (s^{-1})	ΣAa (s^{-1})	ΣAr (s^{-1})
$3s^2 \ 1S$	7.322	$1.34+14$	# 0	$1.34+14$	$1.97+10$
$3s3p^3P$	7.460	$5.99+13$	$8.11+13$	$1.41+14$	$2.20+10$
$3s3p^1P$	7.625	$1.63+14$	$3.71+14$	$5.34+14$	$2.41+10$
$3s3d^1D$	7.627	$9.60+13$	$2.59+14$	$3.55+14$	$3.34+10$
$3s3d^3D$	7.688	$1.71+13$	$1.05+13$	$2.76+13$	$3.91+10$
$3p^2 \ 3P$	7.729	0	$2.43+14$	$2.43+14$	$2.53+10$
$3p3d^3F$	7.819	$2.02+12$	$1.51+12$	$3.53+12$	$4.08+10$
$3p3d^1D$	7.831	0	$1.24+13$	$1.24+13$	$4.24+10$
$3p^2 \ 1S$	7.847	$1.41+14$	$7.53+14$	$8.94+14$	$3.42+10$
$3p^2 \ 1D$	7.888	$5.77+13$	$1.48+14$	$2.06+14$	$4.25+10$
$3p3d^3D$	7.937	0	$9.13+13$	$9.13+13$	$4.14+10$
$3p3d^3P$	7.952	$5.82+12$	$4.17+13$	$4.75+13$	$4.21+10$
$3d^2 \ 3F$	8.018	0	$8.72+13$	$8.72+13$	$5.99+10$
$3d^2 \ 1G$	8.114	$9.72+13$	$5.00+14$	$5.98+14$	$4.85+10$
$3p3d^1F$	8.124	$1.93+13$	$3.09+14$	$3.28+14$	$3.73+10$
$3d^2 \ 3P$	8.133	0	$2.54+11$	$2.54+11$	$6.71+10$
$3d^2 \ 1D$	8.230	$9.46+12$	$1.54+14$	$1.64+14$	$5.26+10$
$3p3d^1P$	8.264	$1.54+13$	$1.26+14$	$1.41+14$	$3.98+10$
$3d^2 \ 1S$	8.497	$6.61+12$	$1.77+13$	$2.43+13$	$6.05+10$

Table 4 - As table 2 for $0^{4+}(1s^2 3s 3p')$ states.

$0^{4+}(1s^2 3s 3p')$	E (Ryd)	Aa(2s) (s ⁻¹)	Aa(2p) (s ⁻¹)	Σ Aa (s ⁻¹)	Σ Ar (s ⁻¹)
3s ² 1s	10.512	1.36+14	# 0	1.36+14	3.75+10
3s3p ³ p	10.682	6.14+13	8.12+13	1.43+14	4.49+10
3s3p ¹ p	10.884	1.66+14	3.79+14	5.45+14	5.02+10
3s3d ¹ D	10.885	1.02+14	2.37+14	3.39+14	7.13+10
3s3d ³ D	10.958	1.82+13	1.12+13	2.94+13	8.37+10
3p ² 3p	11.010	0	2.49+14	2.49+14	5.45+10
3p3d ³ F	11.122	1.51+12	9.47+11	2.46+12	8.98+10
3p3d ¹ D	11.133	0	1.28+13	1.28+13	9.29+10
3p ² 1s	11.160	1.41+14	7.90+14	9.31+14	7.34+10
3p ² 1p	11.204	7.75+13	1.80+14	2.58+14	9.08+10
3p3d ³ D	11.260	0	1.07+14	1.07+14	9.07+10
3p3d ³ P	11.281	6.05+12	4.78+13	5.38+13	9.21+10
3d ² 3F	11.362	0	1.06+14	1.06+14	1.32+11
3d ² 1G	11.482	1.23+14	6.11+14	7.34+14	1.11+11
3p3d ¹ F	11.485	2.80+13	3.67+14	3.95+14	8.21+10
3d ² 3p	11.505	0	7.86+11	7.86+11	1.46+11
3d ² 1p	11.612	1.17+13	1.73+14	1.85+14	1.14+11
3p3d ¹ P	11.652	1.33+13	1.48+14	1.61+14	8.57+11
3d ² 1s	11.940	2.18+12	7.37+12	9.55+12	1.30+11

3-2 Comparison with experiment

Comparison of experimental line positions with theoretical data implies that the angular dependent post-collision interaction (PCI) between the electron and the doubly ionised target is taken into account (see [16] and references given therein, together with [17]). This was recently applied to $N^{7+} + He$ [16] and $Ne^{8+} + He$ [4]. In this work we used the method described in [16] without doing a fit; a detailed analysis of the line shapes is in progress. The positions given in figures 1 to 3 are the final positions of the theoretical functions. In the case of H_2 target, a special detailed theoretical investigation of the PCI effect has shown that the dissociating H_2^{2+} molecules ion can be considered to have an effective charge (+2) as for the helium ion [18]. The experimental spectra have been calibrated against theoretical level positions: the 3p3d¹D state is chosen for $C^{4+} + H_2$ (fig. 1) and N^{5+} , $O^{6+} + He$ (fig. 2a and 3a); the 3p² 1D state for $N^{5+} + H_2$ (fig. 2b) and, finally, the 3d² 3P state for $O^{6+} + H_2$ (fig. 3b). The theoretical positions used as references are indicated by an arrow in these figures in the 10° part. For other angles, the same was done, recalculating when necessary the angular dependent post-collision shift for the reference lines.

4 - Experimental results

From the comparison with theoretical positions, most of the observed lines are clearly identified: capital letters "A" to "E" are used for decay into 2pε continuum (except for the 3s² 1S state decaying into 2sε continuum which contributes to line "C" as appeared in the following); corresponding small letters are used for decay into 2sε continuum.

4-1 Helium target

i) Lines "A" and "a". For C , N and O ions the first line corresponds to an excitation of 3s3p¹P and 3s3d¹D states. As we have confidence in the theoretical relative positions and decay probabilities, it is seen in the 10° spectrum of $N^{5+} + He$ (fig. 2a, see also $O^{6+} + He$ in fig. 3a) that the maximum of the structure "A" appears at a too high energy and that the bump in the low energy wing is too broad to be explained by the well-known PCI shapes. It is also observed that the shape and position of this structure evolves with the angle of emission (compare 45° spectrum with 10° spectrum in fig. 2a), with Z (no problem arises in $Ne^{8+} + He$ [4,19]), with the nature of the target (compare 10° spectra

in fig. 2a and 2b), as well as the continuum of autoionisation (in fig. 2a compare the shapes of lines "A" and "a"). This new phenomenon is tentatively ascribed to an interference between the transition amplitudes of $1P$ and $1D$ states; if the interpretation is valid, this is the first time that such interferences are observed in electron spectra obtained with multicharged ions. It is certainly related to the fact that for low Z ions, these two states are found to be almost degenerate with a separation smaller than 0.03 eV, much less than their decay widths which are equal to about 0.3 eV (tables 2 to 4); in figures 1 to 3, it is the PCI shift which separates somewhat their positions.

ii) Lines "B" and "b". Line B is always very clearly seen in all the spectra. It is mainly explained by the $3p3d^1D$ state; it can be verified that the $3p^2\ ^1D$ state weakly contributes because its decay into $2s\epsilon\ell$ continuum is observed to be very weak (fig. 2a and 3a; remember that decay of $3p3d^1D$ into this continuum is forbidden due to parity conservation selection rule). The contribution of the $3p^2\ ^1S$ state is never seen as a line but, if excited, it must give a more diffuse contribution between lines "A" and "B" because of the large PCI broadening (see decay probabilities in tables 2 to 4).

iii) Line "C". At 10° for N^{5+} and at all angles for O^{6+} , this weak line is due to the decay of the $3s^2\ ^1S$ state alone into $2s\epsilon\ell$ continuum (tables 3,4). The $3d^2\ ^1G$ state does not contribute directly to this line intensity; as with Ne^{8+} [4], it gives a more diffuse contribution under this line C. From N^{5+} to Ne^{8+} it is observed a strong enhancement of the excitation of the $3s^2\ ^1S$ with Z .

iv) Line "D". In N^{5+} spectra, this line peaks between the $3p3d^1F$ and $3d^2\ ^3P$ positions. With O^{6+} , it is clearly related to the triplet excitation. Compared with a similar finding in Ne^{8+} [4], it can be concluded that this triplet is really excited and its relative intensity strongly increases with Z as it must be since its excitation is thought to come from a spin-orbit coupling in the ion in the exit channel [4], if we disregard spin dependent couplings in the quasimolecule.

v) Line "E". A small line appears in N^{5+} spectrum at 90° which cannot be explained by the $3d^2\ ^1G$ or $3s^2\ ^1S$ state. The only possibility is to identify it as a triplet $3d^2\ ^3F$ contribution (table 3).

4-2 Hydrogen target

i) Lines "A" and "a". The relative weight of $3s3p^1P$ and $3s3d^1D$ states increases quickly with Z . It also appears that the perturbation of this line decreases when Z increases. With C^{4+} , the broad structure at 10° looks like the one observed in $N^{5+} + He$ (fig. 12a); again at 50° , as for $N^{5+} + He$ (fig. 2a, 45°), the shape is more symmetrical (fig. 4 in Mack and Niehaus [7]). The disagreement for the positions between theory and experiment decreases when Z increases, and a good agreement is achieved for Ne^{8+} [4].

ii) Lines "B" and "b". It has been assumed for C^{4+} spectra that line "B" is mainly due to $3p3d^1D$ state. For N^{5+} and O^{6+} , the larger width of this line, with respect to the corresponding helium lines, together with the observation of the "b" component (decay into $2s\epsilon\ell$ continuum of $3p^2\ ^1D$ state) imply that the $3p^2\ ^1D$ state contributes to "B". This is the reason why we change the reference line for N^{5+} and $O^{6+} + H_2$.

iii) Line "C". With C^{4+} , this small structure does not correspond to any singlet state (a more clear observation with lower statistical noise is reported by Mack and Niehaus [7]). A weak excitation of $3p3d^3P$ and $3d^2\ ^3F$ states can explain it. With N^{5+} , line "C" is clearly due to the $3s^2\ ^1S$ state (fig. 2b). The $3d^2\ ^1G$ contribution seems rather weak at 10° ; at 90° , the separation between lines "C,D" and "B" deteriorates due to the increase of the broad $1G$ line intensity; at 160° , the $1G$ line becomes predominant over all the other singlet lines and certainly explains the very particular shape observed for the $1s^23\ell3\ell'$ spectrum. This identification also holds for O^{6+} ; again at 160° , the $1s^23\ell3\ell'$ spectrum presents a strong variation of shape, the lines "A", "B" and "C" being less resolved, but it seems that the $1G$ line is not as anisotropic as in N^{5+} spectrum. The angular variation of the $1s^23\ell3\ell'$ part of the $O^{6+} + H_2$ spectrum is similar to the one already observed in $Ne^{8+} + He$ (fig. 5 in [4]).

iv) Line "D". For C^{4+} and N^{5+} , this line includes a contribution of $3d^2\ 3p$ and $3p3d^1F$ states. The $3p$ state alone contributes to this line in $O^{6+} + H_2$ as in $Ne^{8+} + He$ [4].

v) Line "E". Finally, we must mention a new structure labelled "E" in the 160° spectrum of O^{6+} ; it can be identified as a $3s^2\ 1S \rightarrow 2p\epsilon\lambda$ contribution even if the theoretical result indicates a very unfavourable branching ratio (table 4). A weak contribution of this transition is also seen in $N^{5+} + H_2$ (fig. 2b, 10° and 90° , without label "E").

5 - Conclusion

This qualitative analysis of the electron spectra allows to know the general trends of the double capture processes into $1s^23\epsilon3\epsilon'$ configuration. More quantitative results will be obtained with the analysis of the line shapes which is now in progress in our laboratory.

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