

RADIATIVE AND AUGER DECAY CHANNELS IN K-SHELL EXCITED LI-LIKE IONS ($Z = 6-8$)

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Different decay modes for doubly excited Li-like ions ($1s2ln'l'$) produced in slow ($E_{\text{ion}} < 5$ keV/amu) collisions of multiply charged ions with two-electron targets (He, H_2) are discussed. Using the typical differences in the decay of quartet and doublet states we can prove that the spin of the captured electrons is not changed in the collision. It is shown, that the quartet states $1s2s3s$, $1s2s3d$ actually do not autoionise (i.e. the fluorescence yield exceeds 0.95) and there is strong experimental evidence that this should hold as well as for the majority of the other $1s2ln'l'$ -quartet states with $n' > 2$. The radiative quartet decay feeds $1s2p2l'$ quartet states; cascades leading to $1s2p2l'$ doublet states are discussed as well.

1. Introduction

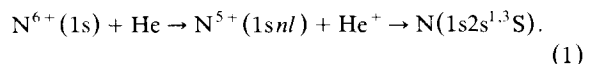
Core excited Li-like ions have attracted interest mostly as test systems for atomic structure calculations, since the relative strength of autoionising and radiative decay depends sensitively on the fine details of the atomic model which is used [1,2]. Our interest here is different: we observe projectile autoionisation spectra from electron capture reactions in slow ($0.1 \text{ a.u.} < v < 0.5 \text{ a.u.}$) collisions with light ($Z \leq 10$) multiply charged ions and we want to know, how strongly these spectra will be affected by the competing radiative decay. We use two-electron targets (He, H_2) and thus there are two capture reactions which produce Li-like ions: either a two-electron capture by a H-like ion or a one-electron capture by a core-excited, metastable He-like ion (configuration $1s2s^{1,3}S$). For the resulting core-excited Li-like ion $1s2ln'l'$ two spin couplings are possible: quartet states (all three spins parallel) and doublet states (two spins antiparallel). Quartet states can not be produced in a two-electron capture from a singlet target as He or H_2 (two spins antiparallel) unless the collision has changed an electron spin. The main decay mode of the ion (in what follows we consider only ions with nuclear charge $Z \leq 10$) will be different for the two spin systems: most of the doublet states will autoionise, except some states for which autoionisation is forbidden in LS-coupling due to conservation of parity and orbital angular momentum, viz. 2P , $^2D^0$, 2F , $^2G^0$ etc. For those states radiative decay will occur, either directly to the singly excited $(1s^2n'l'')^2L$ system by K-X-ray emission, or by L-X-ray emission to other doubly excited states. e.g. $(1s2p2l'')^2L$. (Calculations in intermediate coupling for the $(1s2p^2)^2P$ state by Chen et al. [1] show that the Auger yield brought about by mixing with other autoionising states will not exceed 2% of the total decay).

For all quartet states neither Coulomb autoionisation to the $(1s^2)^2S_0\epsilon l_c$ -continuum nor electric dipole transitions to the singly excited system are allowed. Therefore, one can expect that L-X-ray emission will be a strong decay channel, cascading down to $1s2s2p^4P^0$, which is the lowest quartet state. The corresponding L-X-ray-spectra have been extensively studied (see e.g. B^{2+} [3], C^{3+} [4], N^{4+} [5], O^{5+} [6]). The $^4P^0$ -state itself will decay by slow autoionisation, with different transition rates for its different j -substates [7,8]. Another quartet state known to autoionise is the $1s2p^2^4P_{5/2}$: here, the Auger decay is brought about by configuration interaction with the very fast autoionising $1s2p^2^2D_{5/2}$ -state [1].

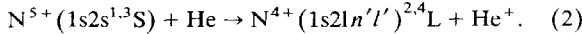
In what follows we will first prove that the capture does not change the spin of the captured electron; this we will use for an investigation of the decay of quartet states and finally we will deal with the non-autoionising doublet states.

2. Double capture by H-like ions

As an example we take the collision system N^{6+} -He. In a recent measurement [9] the projectile autoionisation spectrum taken at $v \approx 0.4$ a.u. showed strong 4P , $^4P^0$ -peaks, which seem to indicate that the electron spin had been changed in the collision. This is contrary to what is expected based on Wigner's spin rule. In a series of measurements taken at different target gas pressures we could show that the quartet intensity varied quadratically with the target gas pressure (figs. 1a, b). Apparently, two successive single capture events occur: the first one takes place on the passage to the reaction center (after the last selecting magnet) and creates a metastable N^{5+} -beam contamination:



Inside the reaction center a second single capture event takes place:



In such successive capture events ^4L -states may be populated. In our experimental setup it was not possible by reducing the target pressure to completely avoid the contribution from such successive capture events. The reason is that because of their long lifetime, the metastables formed by reaction (1) in the whole region between reaction volume and the last deflecting magnet contribute to the signal due to reaction (2) in the reaction volume. However, comparison of two spectra (s_1, s_2) measured at different gas pressures (p_1, p_2) allows us to separate the double collision contribution (d) due to (2) and the true capture (t); if $q = p_2/p_1$, then

$$s_1 = t_1 + d_1$$

$$s_2 = q t_1 + q^2 d_1, \quad (3)$$

and hence

$$t_1 = (q^2 s_1 - s_2)/(q^2 - q) \quad (4)$$

$$d_1 = (s_2 - q s_1)/(q^2 - q). \quad (5)$$

Fig. 1d shows the extracted double collision signal, in comparison with a spectrum obtained directly by a $\text{N}^{5+}(1s2s^1\text{S})$ -beam (for comparison see as well the electron spectra of the $\text{N}^{5+}\text{-He}$, $\text{N}^{5+}\text{-H}_2$ -systems which have been reported by Bordenave-Montesquieu et al. [10]). Within the accuracy of our measurements both spectra are identical. Thus, the spectra figs. 1a, 1b and the one given in ref. [9] are in fact spectra obtained from a N^{6+} -beam contaminated with a $\text{N}^{5+}(1s2s^1\text{S})$ -fraction. The true spectrum (fig. 1c) extracted according to (5a) shows no longer a $^4\text{P}^0$ peak and the peak at the energy of the $^4\text{P}^e$ state is considerably reduced, the remaining intensity probably being due to the $^2\text{P}^0$ state which accidentally coincides with the $^4\text{P}^e$ -energy. (Note that, according to the calculations in [1], $1s2p^2\ ^4\text{P}_{1/2,3/2}$ decays for more than 90% and $^4\text{P}_{5/2}$ still with about 15% to $1s2s2p\ ^4\text{P}^0$. Therefore the absence of $^4\text{P}^0$ indirectly proves that the peak at 327 eV is due to $[1s(2s2p)^1\text{P}]^2\text{P}^0$.) The same we found for the systems $\text{O}^{7+}\text{-He}$ and $\text{C}^{5+}\text{-He}$. Since any higher quartet state would feed both the $^4\text{P}^0$ and $^4\text{P}^e$ by radiative cascades [5,11], we conclude from the absence of the $^4\text{P}^0$ peak that these spectra are pure doublet spectra; hence, the capture has *not* changed the spin of the captured electrons, i.e. the capture is spin-conserving as predicted by Wigner's spin rule. Evidence, that Wigner's spin rule applies, has recently been found also in translational energy spectroscopy for the system $\text{C}^{4+}\text{-Ne}$ [12] and in electron spectra resulting from double capture by the He-like groundstate ($1s^2$)-ions [13] (therefore we suggest that doubly excited projectile ion triplet states should

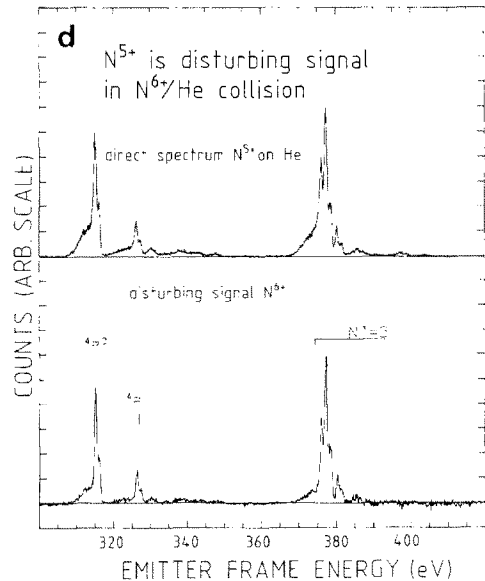
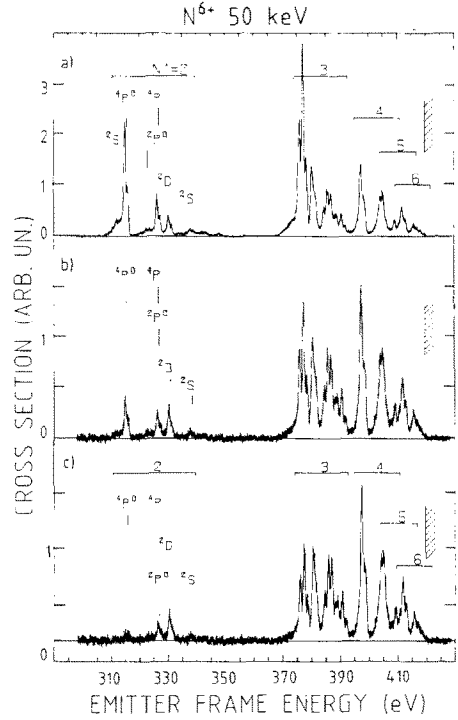


Fig. 1. Autoionising KLV-decay of doubly excited $\text{N}^{4+}(1s2ln'l')$ produced in capture by a N^{6+} -beam (50 keV) from He.

(a) N^{6+} beam with strong N^{5+} -contamination, (b) N^{6+} -beam with weak N^{5+} -contamination, (c) N^{5+} -contamination removed (cf. text), (d) extracted N^{5+} -contamination compared with a spectrum obtained directly from a $\text{N}^{5+}(1s2s^1\text{S})$ -beam. The $1s2s2p\ ^4\text{P}^0$ intensity is not yet corrected from incomplete decay inside the reaction volume.

be discarded from the analysis of the $\text{Ne}^{8-}(1s^2)\text{-He}$ system presented in ref. [14]).

3. Single capture by He-like ions

Formation of quartet states is not forbidden in single capture by He-like ($1s2s\ ^3S$)-ions (see reaction (2)); indeed, all electron spectra in the isoelectronic sequence C^{4-} up to O^{6+} show strong $^4P^o$, 4P population, as can be seen from fig. 2 for H_2 as target. Since these states are situated far outside the "reaction window" as discussed in [15,16], direct population is quite unlikely. Therefore we assume, in accordance with the strong L-X-ray transitions seen in optical emission spectra, that $1s2s2p\ ^4P^o$, $1s2p^2\ ^4P$ are populated exclusively by radiative cascades from higher quartet levels. This has been supposed earlier for the Li-like ions of nitrogen [10], fluorine [17] and neon [18,19]. If there was a significant autoionising probability for these higher quartet levels, then at least some prominent quartet peaks which do not coincide in energy with adjacent doublet peaks should show up as additional peaks in the spectra resulting from the metastable ion beam compared to the pure doublet spectra from the H-like ion beams. In what follows we therefore compare both

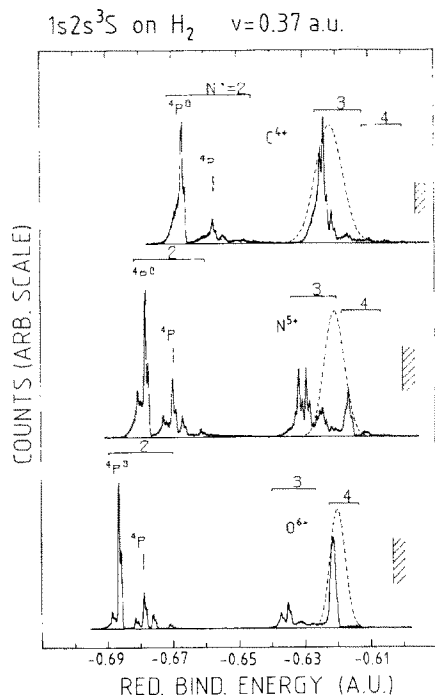


Fig. 2. Autoionising K LX-decay of Li-like ions ($Z = 6-8$) produced by single capture of the He-like core-excited $1s2s^3S$ -ions indicated. The energy scale gives binding energies of the Li-like ions divided by Z^2 , in a.u.. Vertical scales are not comparable. Collision energy about 3.5 keV/amu.

electron spectra; the result of this comparison will be that – within the limits of our energy resolution – no peaks are found in the spectra due to the metastable ion beam, which were not already present in doublet spectra. For some quartet peaks this finding very directly rules out any significant autoionising decay. For other peaks such a direct conclusion is not possible because of overlapping peaks. However, it should be pointed out that, even if the energy resolution is not sufficient to resolve overlapping peaks, it is still possible to rule out a significant contribution from additional states just from the absence of a change of the convoluted spectra. A case where partly unresolved spectra show a drastic change when, according to spin selection rules, additional states may be populated, is the spectrum due to double capture into $\text{N}^{5+}(1s^2)$. For H_2 -targets, only 1L -states may be populated, while for Ar-targets also 3L -states are allowed. Since the 3L -states are allowed to autoionize, the additional population of 3L -states creates additional peaks, which although not resolved clearly lead to drastic differences of the spectra (fig. 3). From the absence of any observable unresolved $1snln'l'$ -spectra when formed by single capture into $1s2s\ ^3S$ -metastable as compared to double capture into $1s$ -ions, we draw the general conclusion that higher quartet states $1snln'l'\ ^4L$ decay predominantly by radiation. A more detailed and quantitative analysis of our spectra regarding a possible contribution from 4L -states ($n > 2$) to the different partly resolved peak groups is outlined below.

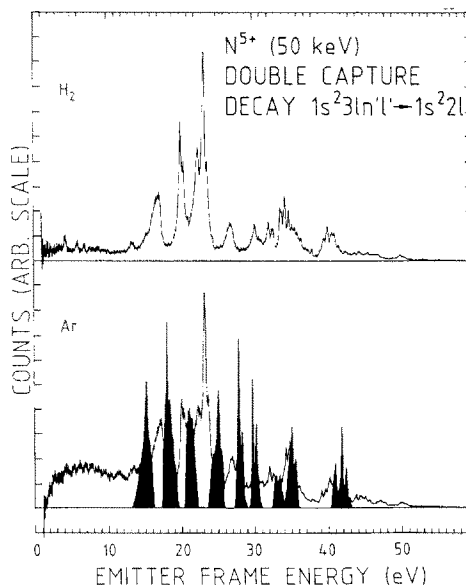


Fig. 3. Autoionising LMX-decay of doubly excited $\text{N}^{3+}(1s^23ln'l')$ produced in collisions of $\text{N}^{5+}(1s^2)$ with Ar and H_2 . Black peaks indicate peaks attributed to triplet states, which do not appear in the $\text{N}^{5+}\text{-H}_2$ spectrum because of spin conservation.

4. Comparison of mixed quartet/doublet spectra with pure doublet spectra

First we want to stress the point, that since the final states of the quartet cascades are not present in the double-capture spectra produced by H-like ions (C^{5+} -He, N^{6+} -He, O^{7+} -He) these spectra are definitely pure doublet spectra and all peaks will be brought about by doublet states. Thus, we have a set of – not always fully resolved – doublet peaks and our analysis does not have to rely on any theoretical calculation of doublet term energies (like e.g. [20,21]). On the other hand the presence of $1s2s2p\ ^4P^0$ in the spectra produced by the 3S metastable ion beam indicates that quartet states $1s2l3l'$ and higher are populated; the experimental results of X-ray spectroscopy [4–6] provide us with a set of exceptionally accurate (uncertainty typically better 20 meV) experimental quartet term energies up to $1s2p4l'$. It is at these energy positions that we will look for the additional quartet peaks. Let us first describe how we determine energy positions in the doublet spectra; then we will compare the groups $1s2s3l'$, $1s2p3l'$ and $1s2s4l'$ separately. Here we will only treat the N V-decay in detail; detailed information concerning C IV and O VI will be deferred to a forthcoming paper.

4.1. Determination of experimental doublet energies

The spectra are shown in fig. 4 and on an enlarged energy scale in fig. 4b. All peaks are kinematically broadened; however, due to the high spectral resolution ($\Delta E/E \leq 5 \times 10^{-3}$) of our electron spectrometer (which is an electrostatic cylindrical mirror analyzer, accepting electrons under an angle of $50^\circ \pm 2^\circ$ with respect to the beam axis) a substructure is observed in the Doppler-broadened peaks: two maxima with a sharp dip in between. The two maxima are due to the finite divergence of the ion beam (maximum divergence angle 10 mrad); they correspond to the cases that the velocity component of the ion in the direction of the emitted electron is at its maximum or minimum, i.e. electron and emitting ion moving in the same or in opposite azimuthal directions, respectively. The dip in between corresponds – as has been verified in computer simulations – to a Doppler-angle of precisely 50° and it is this accurate energy position which is used for evaluation. The asymmetry of the double peak is partly caused by the low-energy tail inherent to the spectrometer function which will enhance the low-energy half of the double structure; a slightly asymmetric detection efficiency for electrons above/below 50° brought about by imperfections of the lens system which guides the electrons from the energy-selecting diaphragm to the detector contributes as well. A fixed instrumental double-peak shape taken from well isolated peaks (e.g.

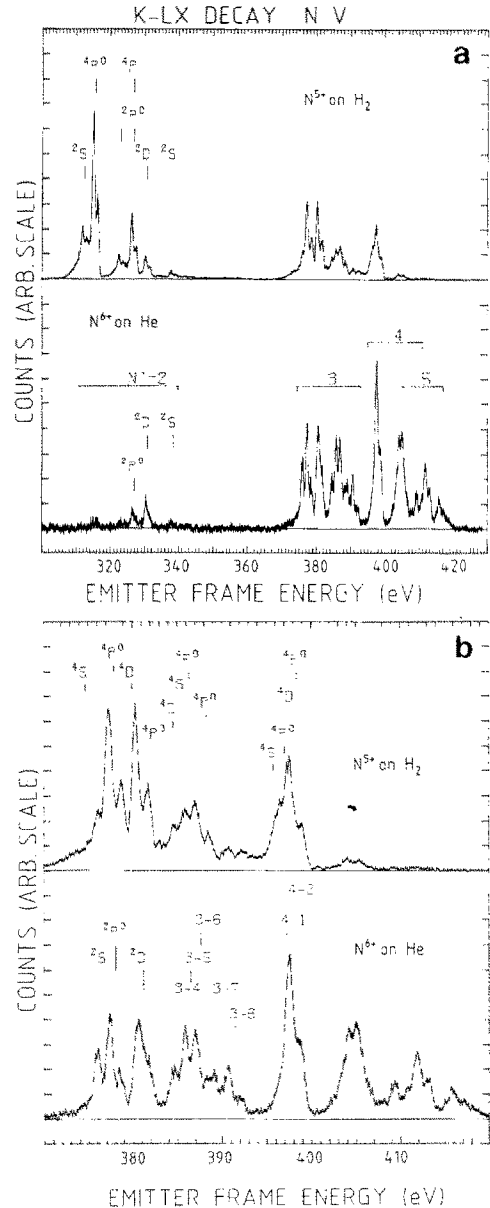


Fig. 4. (a) Comparison of the K-LX-spectra of core excited N^{4+} obtained from N^{6+} on He and $N^{5+}(1s2s^3S)$ on H_2 . Collision energy 50 keV. (b) The same as (a) energy range 370–420 eV.

$1s2p^2\ ^2S$) is used to fit peak intensities and energy positions where peaks start to overlap. In this way we obtain the energy positions of the $1s2l2l'$ and $1s2s3l'$ terms. For the $1s2p3l'$ group (made up by $^1S3l'$, $^3P3l'$ and $^1P3l'$) we can recognize six maxima of obviously overlapping peaks; a fit using five different peaks (labeled 3-4 to 3-8) was sufficient to accurately reproduce this group – which has a very similar structure for C IV, N V and O VI. Therefore we assume, that this part of the spectrum may be described by a set of five dominant peaks; each of them may contain several

states and we cannot exclude masking of less pronounced peaks. In a similar way we use two peaks (labeled 4-1, 4-2) to describe the $1s2s^3S4l'$ -part of the spectrum. Relative energy calibration is achieved using theoretical energy differences between theoretical $1s2s2p^4P^0$ -energies; for each ion $1s2s2p^4P^0$ is used as a common reference point for the quartet and doublet term energies (note that the doublet spectrum is obtained from the weighted difference of spectra which do contain the $4P^0$ -peak). Details of the calibration procedure are given in a separate paper [22]; there we compared our $1s2l2l'$, $1s2s3l'$ doublet term energies with other measurements and energies derived from X-ray spectroscopy, and from this comparison we can infer that our energy scale is consistent with the energy scale of the quartet term energies; hence, we can compare directly. The uncertainty of the doublet peaks is estimated from the variation of the peak energy seen in several different measurement and amounts typically to 100 meV.

4.2. The group $1s2s3l'$

The spectra obtained by the metastable ion ($N^{5+}-He, N^{5+}-H_2$) show precisely the same peak energies for the three peaks $1s2s^3S3s, 1s3s^3S3p, 1s2s^3S3d$ as for the pure doublet spectrum. No peaks appear at the – known – positions of the $1s2s3s^4S$ and the $1s2s3d^4D$, which are about 2 eV below the corresponding doublet peaks. Our fit does not allow to fit more than 5% of the corresponding doublet intensity to the quartet line position, even if we shift the quartet lines by the uncertainty of the doublet lines. Assuming an initially equal population of doublet and quartet states we conclude that the fluorescence yield of $4S(1), 4D(1)$ exceeds 0.95. For C IV and O VI too, the mixed quartet/doublet spectrum shows only doublet lines for these peaks and about the same estimate for the fluorescence yield is obtained. What concerns the configuration $1s2s3p$, the energy difference between quartet and doublet peak is only about 200 meV for all three ions, which makes a quantitative estimate of the fluorescence yield by a comparison of relative peak intensities impossible. However, we can state that the $1s2s^3S3p$ -peak seen in the mixed spectrum stays for all ions safely within the experimental uncertainty of the doublet peak (± 100 meV) on the position of the doublet peak. This indicates at least that the quartet peak does not auto-ionise strongly, because we expect not only an intensity comparable to the doublet peak but strong cascade contributions from the $1s2s4s, 1s2s4d$ quartet states as well [5]; thus, the peak position should vary according to whether $n' = 4$ states are populated ($N^{5+}-H_2$) or not ($N^{5+}-He$), which is not the case. Note that our analysis indicates that the tentative peak assignment for the mixed spectrum given in [10] (which uses quartet peaks) should be modified.

4.3. The group $1s2p3l'$

Comparison of this group shows again, that the mixed spectrum can be described by the doublet energies taken from the N^{6+} -spectrum with only slight adjustments in relative intensity – the peaks labelled 3-7, 3-8 which contain mainly states with $1P$ -core decrease in intensity. This holds for C IV and O VI as well. The question than arises, whether by a fortuitous coincidence (which had to occur for all systems) quartet peaks fall close to doublet peaks. We have to deal with five quartet peaks for which autoionisation is not forbidden: $1s2p3s^4P^0, 1s2p3p^4D, 4S$ and $1s2p3d^4F^0, 4P^0$. For all three ions, $1s2p3s$ is lower than the first doublet peak (3-4). For C IV, the other quartet states are situated within 0.4 eV around the doublet peaks, for NV and OVI only the $1s2p3p^4S$ is close to 3-4 (about 0.2 eV) and the $1s2p3d^4F^0$ close to 3-5 (again about 0.2 eV). However, the small uncertainty of the quartet peaks, the limited uncertainty of our doublet peaks (≤ 150 meV) and the fact that there is no dramatic change of the relative peak intensities inside the range of the quartet peaks makes it in our view highly improbable that strong quartet peaks would replace doublet peaks. Again, our finding would be consistent with the hypothesis that all five quartet peaks had a fluorescence yield approaching unity.

4.4. The group $1s2s4l'$

Here as well the doublet peaks are sufficient to reproduce the mixed spectrum simply by adjusting relative intensity. Besides that, none of the known quartet peaks coincides with one of our doublet peaks, and the peak labeled 4-1 is at higher energy than $1s2p4p^4P^0$, which means that at least we can definitely rule out $1s2s4s^4S$. Hence, we again suppose, that the peaks of the mixed spectrum should be attributed to doublet states only.

4.5. Higher levels

As the autoionisation probability for higher levels decreases roughly as $1/n^3$ whereas the dipole transition probability does not, a fluorescence yield of nearly 100% for the $1s2l3l'$ -quartet terms would suggest a similar high fluorescence yield for all higher quartet states.

4.6. Conclusion

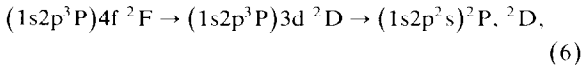
In conclusion, we may say, that the mixed spectra contain only peaks which are also present in the pure doublet spectrum.

A comparison of the $N^{6+}-He$ doublet spectrum with a spectrum of the $N^{6+}-Ar$ -system yielded the same

results: although quartet states may be populated in case of the latter system, the peak structure of the electron spectra of the two systems is identical for groups $1s2ln'l'$ with $n' > 2$, proving that 4L -states ($n' > 2$) have a fluorescence yield close to one. Our findings are consistent with a very recent theoretical calculation [21] of lifetimes for autoionisation and radiative decay (the latter are found to be in excellent agreement with directly measured lifetime [10]): for $Z = 6-8$, the fluorescence yield of any quartet state $1s2l3l'$ exceeds 0.9, except for ${}^4F^0$, where it amounts to roughly 0.6. (We think that $1s2p3d {}^4F^0$ is the state Sellin et al. [23,24] have seen in their delayed electron spectra of beam-foil excited O^{5+} ; lifetime, fluorescence yield and electron energy would fit this assignment better than the original $1s2p3p {}^4P$ assignment. We would like to point out that the quartet intensity detected in their experiment amounts only to a fraction of some percent of the direct autoionising decay; hence, their measurement would still be consistent with a fluorescence yield for all quartet states of nearly one).

5. Radiative decay of doublet states

As can be seen from fig. 3, doublet states $1s2l2l'$ are strongly populated in case of the $N^{5+}-H_2$ -system. Since these states lie outside the reaction window [15,16] we assume that they are fed by cascades as well. *Radiative* cascades have been observed in optical emission spectra of Li-like ions with lower Z . For Be^+ and B^{2+} a radiative cascade



has been found ([3]). There are no calculations available for the L-X-ray branching of the radiative doublet decay in C IV, N V and O VI. A calculation for Ne VII [25] shows that L-X-ray branching could be appreciable for the $1s2p^3P3d$ configurations and the $1s2p3p {}^2P$. Therefore, we attribute the weak KLL-decay seen in the spectrum of N^{6+} on He to radiative decay of these states. For the more pronounced KLL doublet peaks seen in the spectra due to the metastable ions, an *autoionizing* cascade from triply excited Be-like states may provide another feeding cascade, which would especially explain the presence of $1s2s^2 {}^2S$, viz.



The strength of this autoionizing cascade could be determined if one measured the emitted low energy electron in coincidence with the target ion.

6. Some consequences

Let us summarize the consequences from our experimental findings in four points:

- (1) Due to the contamination of the N^{6+} -beam with $N^{5+}(1s2s)^{1,3}S$ the K LX-cross sections given in ref. [26] are too high; we derive a K LX-cross section (using the calibration of ref. [26]) $\sigma = (0.97 \pm 0.25) \times 10^{-16} \text{ cm}^2$. The intensity of the extracted double collision signal enables us to estimate a cross section for single capture by $N^{5+}(1s2s)^{1,3}S$ from He. We found a cross section which is roughly equal to the one for single capture by $N^{5+}(1s^2)$ from He [27]. Using this value, we can estimate the $1s2s^3S$ -content of the N^{5+} -beam supplied by our ECR-source: We obtain a value of approximately 5%, which agrees fairly well with estimates from other work [28]. Because of the rather long transit time of ions from the ECR-source to our reaction volume – it is considerably longer than the $He(2^1S)$ -lifetime – the main metastable component will be 2^3S .
- (2) Assignments of the $1s2ln'l'$ -($n' > 2$)-peaks in electron spectra of Li-like ions to quartet peaks are probably wrong: at least peaks $1s2s3s$ and $1s2s3d$ should be attributed to doublet states only
- (3) The population of states originally produced in the collision will in general be drastically changed by radiative decay of quartet and doublet states; therefore, n' -distributions, angular momentum substate distributions or the angular distribution of the emitted electrons [29] contain – in general – only indirect information about the capture process.
- (4) How much of the intensity of the originally populated quartet or doublet state will show up in an electron spectrum, depends on the branching into L-X-ray and K-X-ray emission. Cross sections derived from electron spectra alone, therefore, constitute only lower bounds for the capture cross section to doubly excited states.

7. Conclusion

In conclusion we have demonstrated that the spin of the captured electron is not changed in slow collisions of multiply charged H-like ions. Taken the experimental evidence that this holds as well for He-like ions (groundstate ions and metastables) we suggest to see this as a general feature for all collision systems involving light multiply charged ions. By an appropriate choice of the target, it is therefore possible to obtain electron spectra which consist only of doublet lines. We have used these “pure” doublet spectra to show the absence of the quartet lines $1s2s3s$ and $1s2s3d$ in cases where quartet population is not forbidden by spin conservation. For all $1s2ln'l'$ -states up to $1s2s4l'$ we find strong

evidence that the fluorescence yield should be close to unity. This result is consistent with a recent theoretical calculation of lifetimes. We suggest that the $1s2s2p^4P^0$, $1s2p^2^4P$ are populated by radiative cascades from these higher quartet levels. For the observed $1s2l2l'$ doublet intensity outside the reaction window we assume indirect population from higher states as well viz. by radiative cascades or by an autoionizing decay of a triply excited Be-like ion.

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