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Electron shakeoff following the $\beta^+$ decay of trapped $^{35}\text{Ar}^+$ ions

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The electron shakeoff of $^{35}\text{Cl}$ atoms resulting from the $\beta^+$ decay of $^{35}\text{Ar}^+$ ions has been investigated using a Paul trap coupled to a recoil-ion spectrometer. The charge-state distribution of the recoiling daughter nuclei is compared to theoretical calculations accounting for shakeoff and Auger processes. The calculations are in excellent agreement with the experimental results and enable to identify the ionization reaction routes leading to the formation of all charge states.

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Precision measurements of the recoil-ion energy spectra in nuclear $\beta$ decay constituted sensitive tools to establish the vector axial vector structure of the weak interaction [1–3]. In particular, these measurements give access to the so-called $\beta - \nu$ angular correlation coefficient, $a_{3\nu}$, which is sensitive to scalar and tensor exotic couplings excluded by the Standard Model of elementary particles [4]. The search for such exotic interactions has motivated new experiments using modern trapping techniques coupled to intense radioactive beams with high production rates [4]. Most of ongoing experiments detect the $\beta$ particles and the recoil-ions in coincidence, providing a precise recoil-ion energy measurement using time of flight (TOF) techniques, and a precise control of systematic effects [5–7]. The LPCTrap setup [8, 9], installed at GANIL, is based on the use of a Paul trap, to confine radioactive ions, coupled to a recoil-ion spectrometer. It has been recently upgraded to perform simultaneous measurements of both the charge-state and the energy of the recoil-ions. Fundamental atomic processes such as electron shakeoff (SO) resulting from the sudden change of the central potential can thus also be addressed through a measurement of the charge-state distribution of the recoiling ions. The setup has already enabled the measurement of electron SO in the decay of $^6\text{He}^+$ [10]. For this ideal textbook case, with only one electron, simple quantum calculations based on the sudden approximation (SA) could be tested with a relative precision better than $4 \times 10^{-4}$. Beyond the prototypical $^6\text{He}^+$ case, heavier systems such as $^{35}\text{Ar}^+$ can reveal the role of more subtle shakeoff dynamics involving several electrons, and of subsequent relaxation processes such as the emission of Auger electrons. These multi-electron processes, of paramount importance in atomic and molecular physics, have mainly been studied as post-collision mechanisms following the absorption of a photon in the X-ray spectral range (see for instance [11–14]). Alternatively, SO and Auger processes can conveniently be decoupled within multi-electron rearrangements induced by sudden nuclear $\beta$-decay. These ionization mechanisms have been explored for a large variety of $\beta^-$ emitters [15]. In contrast, information is scarce for $\beta^+$ decaying parent atoms [6, 16, 17] and even totally missing, up to our knowledge, for multi-electronic singly-charged systems. We thus investigate here the charge-state distribution following $\beta^+$ emission of $^{35}\text{Ar}^+$. In addition to be interesting per se, it is worth noting that SO can further be a source of systematic error for $\beta - \nu$ angular correlation coefficient measurements. This systematic error, that was found to be small in a previous LPCTrap experiment on $^6\text{He}^+$ decay [7], would become problematic for many electron systems. For the WITCH setup [18] installed at ISOLDE-CERN, whose main goal is to measure $a_{3\nu}$ in the decay of $^{35}\text{Ar}^+$ ions confined in a Penning trap, an independent measurement of the charge-state distribution of the recoil ions will ease the analysis and improve the precision [19].

The experimental setup has been described in detail previously [8–10]. The radioactive $^{35}\text{Ar}$ nuclei were produced at the SPIRAL target-ECR ion source system of GANIL, Caen, France, by a primary beam of $^{36}\text{Ar}$ ions impinging on a graphite target. After mass separation the $^{35}\text{Ar}^+$ ions were guided at 10 keV through the LIRAT low energy beam line. At the entrance of the LPC-Trap apparatus, the $^{35}\text{Ar}^+$ beam intensity was typically $10^6$ pps. The ions were first injected in a Radio Frequency Cooler and Buncher (RFQCB) [20] for the beam preparation. This linear Paul trap, mounted on a high voltage platform to decelerate the ions down to 50 eV, was filled with...
with He buffer gas at a pressure of $1.6 \times 10^{-2}$ mbar to cool down the ions below 1 eV. The $^{35}$Ar$^+$ ions from LIRAT were continuously injected in the RFQCB, cooled, and accumulated into bunches before being extracted with a cycle period of 200 ms. They were then reaccelerated downstream using a pulsed cavity, transported between the two traps with a kinetic energy of about 1 keV, and decelerated down to 100 eV by a second pulsed cavity located at the entrance of the measurement transparent Paul trap (MTPT). For each injection cycle, an average of about $2 \times 10^3$ $^{35}$Ar$^+$ ions were successfully trapped and confined by applying a 0.48 MHz RF voltage of 120 V$_{pp}$ to the two inner rings of the MTPT (Fig. 1). Helium buffer gas at a pressure of $10^{-5}$ mbar was also used in the MTPT chamber to further cool down the trapped ions. The $\beta$ particles and the recoiling ions resulting from the $\beta$ decay of the trapped $^{35}$Ar$^+$ ions were detected in coincidence using detectors located around the trap (Fig.1). The $\beta$ telescope, composed of a thin double sided silicon strip detector followed by a plastic scintillator, provides the position and the energy of the incoming $\beta$ particles. The signal from the plastic scintillator also defines the reference time for a decay event. A recoil ion spectrometer enables to separate the charge-states of the recoiling ions using their time of flight (TOF). Ions emitted towards the recoil ion spectrometer cross a first collimator through a 90% transmission grid (set at ground potential). They are then accelerated by a $-2$ kV potential applied to a free flight tube (Fig. 1) whose entrance and exit are defined by two additional 90% transmission grids. Inside the tube, an electrostatic lens at $-250$ V permits a 100% collection efficiency of the ions by a micro-channel plate position sensitive detector (MCPPSD) [21]. A $-4$ kV voltage applied on the front plate of the MCPPSD ensures a detection efficiency close to maximum for all charge-states of the recoil ions, independently of their initial kinetic energy.

For each detected event, the energy and position of the $\beta$ particle, the time of flight (TOF) and position of the recoil ion, were recorded. The procedure applied for the detector calibrations was identical to that described in Ref. [7]. Only events corresponding to a $\beta$ particle depositing more than 0.4 MeV in the scintillator were kept in the analysis. The TOF distribution measured for the $^{35}$Cl$^+$ recoil ions resulting from $^{35}$Ar$^+$ $\beta$ decay is shown in Fig. 2(a). A constant background in TOF due to uncorrelated signals from the recoil ion and $\beta$ detectors has been subtracted from the data.

The experimental charge-state branching ratios and their associated statistical uncertainty were simply deduced from the integration of counts within the TOF selection windows displayed on Fig. 2(a). Two additional corrections, labeled Tail$_{corr.}$ and MCP$_{corr.}$, in Table I, were then applied. The first takes into account the tails of charge distributions extending beyond their respective integration windows. TOF spectra associated to each charge-state were generated using Monte-Carlo simulations [7, 10] and were adjusted to the experimental data. Several ingredients of the simulations, such as the exact size of the trapped ion cloud, weak decay branches of $^{35}$Ar towards excited states, and possible scattering of the $\beta$ particles on parts of the trapping chamber were neglected or approximated. Therefore, a conservative relative uncertainty of 10 % was applied on these corrections. The second correction concerns the dependence of the MCPPSD detection efficiency on the charge-state of the recoil ions. The loss of detected events due to electronic threshold was precisely estimated by fitting the charge distributions collected from the recoil ion detector with gaussian functions (Fig. 2(b)). The data were then corrected for the relative detection efficiency obtained for each charge-state. For charged recoil ions, the experimental charge-state branching ratios including these corrections are given in Table I. Electron capture probabilities from He buffer gas between the Paul trap and the spectrometer have also been estimated using experimental cross sections measured for Kr$^{q+}$ + He collisions in...
the same velocity regime [22]. Kr and Cl having similar ionization potentials, the charge exchange cross sections for Kr$^{+}+$ ions constitute a good approximation of what one would expect with Cl$^{+}+$ ions. Even for the higher charge states involved here (q=5 and 6), these probabilities are only of the order of a few $10^{-4}$. They were therefore neglected at the present level of precision. For a dominant part of the decay events, there is no electron shakeoff and the $\beta^{-}$ decay of a 35Ar$^{+}$ ion results in the recoil of a neutral 35Cl atom. For Cl recoil atoms, both the collection efficiency and the MCPSD detection efficiency are very low, and depends strongly on their initial energy. The TOF associated to such detected events being larger than 11µs, they do not appear on the spectrum shown in the Fig. 2(a). To estimate the number of 35Cl atoms produced during the experiment, we have used the number of $\beta^{-}$ particles detected in “singles” (without condition on the detection of a recoil). Knowing the overall absolute detection efficiency for ions, the fraction of the “singles” events associated to charged 35Cl recoils could be inferred, the rest being associated to 35Cl atoms. This estimate leads to 72(10)% of neutral 35Cl recoils, with an uncertainty dominated here by the error on the overall ion detection efficiency. This result is in good agreement with the 73.9% ratio obtained from the theoretical calculations that we detail hereinafter.

Subsequently to the sudden decay of 35Ar$^{+}$, primary Cl$^{+}$ ions are formed by ionization. In the framework of the independent electron model (IPM,[23]), well suited with the 73.9% ratio obtained from the theoretical calculation describing one electron orbiting in the nl-subshell of Ar$^{+}$ or Cl. Because of the small values of K ($E_{R}^{\max}$ = 452 eV), $e^{-iK\tau}$ can be expanded in eq. (2) in order to highlight the mechanisms underlying $\beta^{-}$-induced SO. Up to second order in K$^{2}$, we obtain [26]

$$p_{i} = 1 - \sum_{n_{i} \leq N} \left\{ |\langle \varphi_{n_{i}l_{i}}^{(Cl)} | e^{-iK\tau} | \varphi_{n_{i}l_{i}}^{(Ar^{+})} \rangle |^{2}ight.$$

$$+ K^{2}|\langle \varphi_{n_{i}l_{i}}^{(Cl)} | e^{-iK\tau} | \varphi_{n_{i}l_{i}}^{(Ar^{+})} \rangle |^{2} -

$$

$$K^{2}Re\langle \varphi_{n_{i}l_{i}}^{(Cl)} | e^{-iK\tau} | \varphi_{n_{i}l_{i}}^{(Ar^{+})} \rangle \langle \varphi_{n_{i}l_{i}}^{(Cl)} | e^{-iK\tau} | \varphi_{n_{i}l_{i}}^{(Ar^{+})} \rangle \right\}.$$  

(3)

It is thus clear that ionization stems from the coherent superposition of two effects: the static Ar$^{+}$/Cl orbital mismatch, through the $|\langle \varphi_{n_{i}l_{i}}^{(Cl)} | e^{-iK\tau} | \varphi_{n_{i}l_{i}}^{(Ar^{+})} \rangle |^{2}$ terms, and the recoil of the Cl daughter nucleus, through the $K^{2}$-dependent terms in (3).

For the calculation of the recoil-induced ionization terms involved in (3), we have used the mean recoil energy obtained in our experiment, $E_{R} = 376$ eV, to define the numerical value of K (0.02074 a.u.). As Ar$^{+}$ and Cl are open-shell valence systems, the wavefunctions $\varphi_{n_{i}l_{i}}^{(Ar^{+},Cl)}$ have been computed by means of Restricted Open-shell Hartree-Fock (ROHF) calculations using the GAMESS-US quantum-chemistry package [27]. These wavefunctions can alternatively be obtained in terms of Unrestricted Hartree-Fock (UHF) computations [28], and we found that the ROHF and UHF $p_{i}$ probabilities differ by less than 1% for all the 1s, ..., 3p levels [29].

Once inner-shell vacancies have been created in Cl through ionization, radiative and Auger transitions involving higher-lying electrons tend to fill these vacancies. The probabilities associated to these transitions have to be properly introduced in our calculations, especially as Auger processes are known to contribute significantly to the production of high charge states [17]. Kastra and Mewe [31] have computed the probabilities $\tilde{p}_{i}^{s,m_{i}}$ corresponding to the ejection of $m_{i}$ electrons through Auger cascades after the electron i, element of the inner nl$_{i}$-subshell, has been removed from Cl$^{+}$. s is the ionization stage of Cl and corresponds to the number of electrons previously pulled out from the outermost subshells. Consistently with the treatment of multiple SO ionization, the description of multiple vacancies, and related Auger cascades, is performed in the IPM framework. The probability for ejection of q$_{S}$ electrons by SO followed by Auger removal of q$_{A}$ electrons thus reads

$$p_{q_{S},q_{A}} = \sum_{m_{i_{1}} = \ldots = m_{i_{N}}} \sum_{i_{1} = 1}^{N} \ldots \sum_{i_{2} > i_{1}}^{N} \ldots \sum_{i_{2} > i_{1}}^{N} \ldots \sum_{i_{N} > i_{N-1}}^{N} \prod_{j \neq i_{1}, \ldots, i_{N}}^{N} (1 - p_{j})$$  

(4)
where \( q_s + q_A = q \), the charge state finally observed.

The computed charge-state branching ratios, which consist of the relative populations of Cl\(^{q+}\) species among the total ion yield, are compared to their experimental counterparts in Table I. Eq. (4) provides a very good agreement with the measurements.

The calculations can then be used to discriminate between the roles of SO and Auger transitions in the production of Cl\(^{q+}\) ions. Neglecting Auger decays, i.e., computing the charge-state distribution according to Eq. (1), severely distorts the Cl\(^{q+}\) populations: the \( q = +1 \) population is overestimated by \( \sim 13\% \) while multiple SO is inefficient to explain the abundance of \( q \geq 3 \) states. Such an importance of Auger processes has to be contrasted with previous studies [17] in the near future. Besides the intrinsic interest of such investigations to nuclear physics, this will allow obtaining a more complete and \( Z \)-dependent picture of the underlying ionization mechanisms.

The authors thank the LPC staff for their strong involvement in the LPCTrap project and the GANIL staff for the preparation of a high quality ion beam. The authors also acknowledge the computational facilities provided by the Mésocentre de Calcul Intensif Aquitain at University of Bordeaux [32]. D.R. acknowledges support from the Spanish ministry of Economy and competitiveness. The authors thank the LPC staff for their strong involvement in the LPCTrap project and the GANIL staff for the preparation of a high quality ion beam. The authors also acknowledge the computational facilities provided by the Mésocentre de Calcul Intensif Aquitain at University of Bordeaux [32]. D.R. acknowledges support from the Spanish ministry of Economy and competitiveness. The authors thank the LPC staff for their strong involvement in the LPCTrap project and the GANIL staff for the preparation of a high quality ion beam. The authors also acknowledge the computational facilities provided by the Mésocentre de Calcul Intensif Aquitain at University of Bordeaux [32]. D.R. acknowledges support from the Spanish ministry of Economy and competitiveness.

### Table I: Experimental ion charge-state relative branching ratios (%) and included corrections (see text) compared to calculations with and without recoil and Auger ionizations.

<table>
<thead>
<tr>
<th>Charge</th>
<th>MCP(_{corr.})</th>
<th>Tail(_{corr.})</th>
<th>Exp. results</th>
<th>With recoil With Auger</th>
<th>Without recoil With Auger</th>
<th>With recoil Without Auger</th>
<th>Without recoil Without Auger</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.37</td>
<td>-0.17</td>
<td>74.75 ± 1.07</td>
<td>74.37</td>
<td>74.44</td>
<td>87.07</td>
<td>87.37</td>
</tr>
<tr>
<td>2</td>
<td>-0.24</td>
<td>-0.09</td>
<td>17.24 ± 0.44</td>
<td>16.98</td>
<td>16.91</td>
<td>11.92</td>
<td>11.66</td>
</tr>
<tr>
<td>3</td>
<td>-0.09</td>
<td>0.03</td>
<td>5.71 ± 0.27</td>
<td>6.03</td>
<td>6.04</td>
<td>0.95</td>
<td>0.91</td>
</tr>
<tr>
<td>4</td>
<td>-0.03</td>
<td>0.13</td>
<td>1.58 ± 0.21</td>
<td>1.79</td>
<td>1.79</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>&gt;4</td>
<td>-0.01</td>
<td>0.10</td>
<td>0.71 ± 0.18</td>
<td>0.82</td>
<td>0.82</td>
<td>&lt;0.002</td>
<td>&lt;0.002</td>
</tr>
</tbody>
</table>

For Cl\(^{2+}\), twofold SO from the outer \( n = 3 \) shell, eventually followed by radiative stabilization when 3s holes are involved, represents \( \sim 50\% \) of the population and single Auger transitions filling the 2p\(^-1\) SO-hole make the rest. The relevance of SO ionization with multiplicity greater than 2 rapidly falls down (see Table I). As a result, the creation of Cl\(^{3+}\) ions with \( q \geq 3 \) mostly involves Auger decays subsequent to one- and two-fold SO ionization. Moreover, multiple Auger emission, involving intermediate core-hole states and emission of several electrons during a single-hole decay, becomes increasingly important for high \( q \); it participates to almost 100\% of Cl\(^{5+}\) ion creation.

To sum up, our joined experimental/theoretical endeavour has provided a quite complete picture of ion formation resulting from the \( \beta^+ \) decay of \(^{35}\)Ar\(^+\). We plan to apply the same techniques to \(^{19}\)Ne\(^+\) and also revisit previous studies [17] in the near future. Besides the intrinsic interest of such investigations to nuclear physics, this will allow obtaining a more complete and \( Z \)-dependent picture of the underlying ionization mechanisms.

The authors thank the LPC staff for their strong involvement in the LPCTrap project and the GANIL staff for the preparation of a high quality ion beam. The authors also acknowledge the computational facilities provided by the Mésocentre de Calcul Intensif Aquitain at University of Bordeaux [32]. D.R. acknowledges support from the Spanish ministry of Economy and competitiveness under the project FPA2010-14803 and the action AIC10-D-000562.

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TABLE II: Main ionization routes leading to Cl$^{n^+}$ formation (in %). $nl^{-1}$ refers to primary SO hole creation in the $nl$-subshell of Cl while $m \times e_A$ means emission of $m$ Auger electrons. The asterisk indicates multiple Auger emission.

<table>
<thead>
<tr>
<th>Cl$^+$</th>
<th>Cl$^{2+}$</th>
<th>Cl$^{3+}$</th>
<th>Cl$^{4+}$</th>
<th>Cl$^{5+}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$3p^{-1}$: 79.83</td>
<td>2$p^{-1} + e_A$: 49.71</td>
<td>2$s^{-1} + 2e_A$: 52.21</td>
<td>2$s^{-1}3p^{-1} + 2e_A$: 36.95</td>
<td>1$s^{-1} + 4e_A$: 35.56</td>
</tr>
<tr>
<td>$3s^{-1}$: 20.16</td>
<td>3$p^{-2} + 2$</td>
<td>2$p^{-1}3p^{-1} + e_A$: 29.36</td>
<td>1$s^{-1} + 3e_A$: 31.02</td>
<td>1$s^{-1}3p^{-1} + 3e_A$: 19.69</td>
</tr>
<tr>
<td>$3s^{-1}3p^{-1}$: 18.51</td>
<td>2$s^{-1}3s^{-1} + e_A$: 7.41</td>
<td>2$s^{-1}3s^{-1} + 2e_A$: 9.31</td>
<td>2$s^{-1}2p^{-1} + 3e_A$: 15.64</td>
<td></td>
</tr>
<tr>
<td>$3s^{-2}$: 1.17</td>
<td>3$s^{-1}3p^{-2}$: 4.37</td>
<td>2$p^{-1}3p^{-2} + e_A$: 8.28</td>
<td>2$s^{-1}3p^{-2} + 2e_A$: 9.26</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3$p^{-3}$: 3.46</td>
<td>2$p^{-2} + 2e_A$: 5.86</td>
<td>2$s^{-1}3s^{-1}3p^{-1} + 2e_A^*$: 5.84</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>1$s^{-1} + 2e_A^*$: 1.56</td>
<td>2$p^{-1}3s^{-1}3p^{-1} + 1e_A$: 5.23</td>
<td>1$s^{-1}3s^{-1} + 3e_A$: 4.90</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2$p^{-2}3p^{-1} + 3e_A^*$: 3.66</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2$p^{-1}3s^{-1}3p^{-2} + e_A$: 1.31</td>
</tr>
</tbody>
</table>

[29] It is quite common to employ the Roothan-Hartree-Fock (RHF) orbitals of Clementi and Roetti [30] to compute the ionization probabilities $p_i$. Even though we employ a different (and larger) underlying basis to construct the Ar$^+$ and Cl orbitals, we have verified that the $p_i$ values obtained with the orbitals of Clementi and Roetti differ by less than 1% from our ROHF and UHF values.