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Mass spectrometry of atomic ions produced by in-trap decay of short-lived nuclides

A Herlert^{1,2,7}, D Beck³, K Blaum^{3,4}, F Carrel², P Delahaye², S George⁵, C Guénaut⁶, F Herfurth³, A Kellerbauer², H-J Kluge³, D Lunney⁶, M Mukherjee³, L Schweikhard¹ and C Yazidjian^{2,3}

¹ Institut für Physik, Ernst-Moritz-Arndt-Universität, 17487 Greifswald, Germany

² CERN, Physics Department, 1211 Geneva 23, Switzerland

³ GSI, Planckstr. 1, 64291 Darmstadt, Germany

⁴ Institut für Physik, Johannes Gutenberg-Universität, 55099 Mainz, Germany

⁵ Institut für Kernphysik, Westfälische Wilhelms-Universität, 48149 Münster, Germany

⁶ CSNSM-IN2P3-CNRS, 91405 Orsay-Campus, France

E-mail: alexander.herlert@cern.ch

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Abstract. The triple-trap mass spectrometer ISOLTRAP at ISOLDE/CERN has demonstrated the feasibility of mass spectrometry of in-trap-decay product ions. This novel technique gives access to radionuclides, which are not produced directly at ISOL-type radioactive ion beam facilities. As a proof of principle, the in-trap decay of $^{37}\text{K}^+$ has been investigated in a Penning trap filled with helium buffer gas. The half-life of the mother nuclide was confirmed and the recoiling $^{37}\text{Ar}^+$ daughter ion was contained within the trap. The ions of either the mother or the daughter nuclide were transferred to a precision Penning trap, where their mass was determined.

⁷ Author to whom any correspondence should be addressed.

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1. Introduction

The determination of nuclear masses is of great importance in many branches of physics [1], ranging from nuclear and astrophysical applications [2] to tests of the weak interaction [3]. In some cases, a relative mass uncertainty of $\delta m/m = 1 \times 10^{-8}$ or even smaller is required. Experiments at radioactive-beam facilities that employ Penning traps for mass spectrometry have proven to be suitable for very accurate mass measurements [4], e.g., as recently demonstrated in the case of ^{22}Mg [5] with the ISOLTRAP mass spectrometer at ISOLDE/CERN [6].

There are currently four experiments that determine the mass of short-lived radionuclides by measurements of the ion cyclotron frequency in a Penning trap: CPT at Argonne National Laboratory [7], JYFLTRAP at IGISOL in Jyväskylä [8], SHIPTRAP at SHIP/GSI [9] and ISOLTRAP as mentioned above [10]. Further on-line Penning-trap experiments for mass determination are under commissioning or construction: TITAN at ISAC/TRIUMF [11], LEBIT at NSCL/MSU [12] and MAFFTRAP at MAFF/FRM-II [13].

The Penning trap permits the confinement of radionuclides in a small volume for very long storage times (limited essentially only by the half-life of the nuclide). This allows one not only to probe the ion motion by applying a radiofrequency (rf) field to measure the cyclotron frequency but also to wait for the decay of the stored radionuclide. The latter will be used in the WITCH experiment at ISOLDE/CERN, where a retardation spectrometer is attached to the Penning trap to measure the energy spectrum of the recoil ions that leave the trap after β decay [14]. In general, detectors for γ rays, electrons or other charged particles can be mounted around the trap to investigate the decay of radionuclides [15]. Recently, the first demonstration of in-trap conversion electron spectroscopy [16, 17] has been performed with REXTRAP at ISOLDE/CERN [18].

We report on a second application of in-trap decay: direct mass measurements of mother and daughter nuclides. Such an achievement has three distinct advantages: (i) a more accurate mass difference measurement can be made since the two species are measured at close time intervals, (ii) concerning the production of radionuclides by the ISOL method, different chemical species could be obtained from the same target/ion-source combination and (iii) elements normally not produced directly by the ISOL method would become accessible.

Two aspects are of importance with respect to the storage of the product nuclide: the kinetic energy of the recoil ion and its charge state after the decay. In the case of β decay, the mass of the daughter is several orders of magnitude larger than the mass of the emitted particle, e.g. an electron or a positron. Thus, the fraction of the decay energy transferred to the recoil

ion is not more than a few 100 eV and, with the appropriate choice of trapping potentials, the capture and storage of the daughter ion is possible. In the case of α decay, the kinetic energy of the recoil ion is orders of magnitude larger than the axial trapping potential. Thus, trapping of the daughter nuclide after α decay strongly depends on the recoil direction relative to the trap axis.

The charge state of the daughter nuclide depends on the decay process. For a β^+ decay of a singly charged cation, the recoil system is neutral and electron shake-off [19] is necessary for further storage. After a β^- decay, the recoil ion is doubly charged or even multiply charged in case of further electron shake-off. Similarly, electron capture yields singly or multiply charged ions. The probability for the loss of one or more shake-off electrons can be calculated [19, 20]. However, the charge state might be reduced by charge-exchange reactions during the storage.

In the following, the β^+ decay $^{37}\text{K} \rightarrow ^{37}\text{Ar}$ is taken as an example for in-trap-decay mass spectrometry. The determination of the mass of the mother and daughter nuclides as well as the measurement of the half-life of the mother nuclide are presented and discussed.

2. Experimental setup and procedure

The ISOLTRAP mass spectrometer [10, 21, 22] is shown schematically in figure 1. A 60-keV radioactive ion beam from ISOLDE [23] is decelerated, cooled, and bunched in a linear rf trap [24]. From there, the ions are transferred to a preparation Penning trap [25]. By use of buffer-gas-assisted mass-selective centring [26], the ions of interest are separated from isobaric contaminants. After the transfer to a precision Penning trap [10] installed in a 5.9-T superconducting magnet, the ion cyclotron frequency is determined by a TOF detection technique [27]: After a dipolar rf excitation of the ions to a magnetron orbit of about 0.7 mm radius, the initially pure magnetron motion is converted into cyclotron motion by a quadrupolar rf field [28]. At $\nu_{\text{rf}} = \nu_{\text{c}}$, a full conversion from magnetron to cyclotron motion is obtained, where $\nu_{\text{c}} = qB/(2\pi m)$ is the cyclotron frequency of an ion with charge q and mass m in a magnetic field B . Hence, the orbital magnetic moment μ and the radial kinetic energy $E = \mu B$ are increased. Since the axial acceleration of the ions in the fringe field of the superconducting magnet is proportional to μ , the shortest TOF is observed for $\nu_{\text{rf}} = \nu_{\text{c}}$.

In the present experiment, the ^{37}K nuclides were produced by bombarding a Ti-foil target with 1.4-GeV protons from the CERN Proton Synchrotron Booster (3×10^{13} protons/pulse). The potassium atoms were ionized on a hot tungsten surface, accelerated to 60 keV, passed through the high-resolution mass separator [23], and transported to the ISOLTRAP rf trap. Note that, with such an ion source, the production of an argon ion beam is impossible.

For in-trap-decay mass spectrometry, the $^{37}\text{K}^+$ ions were accumulated and cooled in the buncher for about 10 ms and transferred (46 μs) to the preparation Penning trap, where they were stored for up to a few seconds. With a half-life of $T_{1/2} = 1.226(7)$ s [29], about 2/3 of ^{37}K have decayed to ^{37}Ar after 2 s of storage time. The energy release of $Q = 6147.46(23)$ keV [32] results in a recoil energy of the daughter nuclide [30] of no more than $(Q^2/2 - m_e Q)/(Q - m_e + m) \approx 0.5$ keV, where m_e and m are the masses of the electron and recoil atom, respectively. The potential along the axis of the cylindrical trap is plotted in figure 2(a) with the harmonic potential in the centre and a potential well of about 100 V depth at the outermost segments of the trap. Only ions with recoil momentum along the axis and kinetic energies above the axial trapping potential depth are lost.

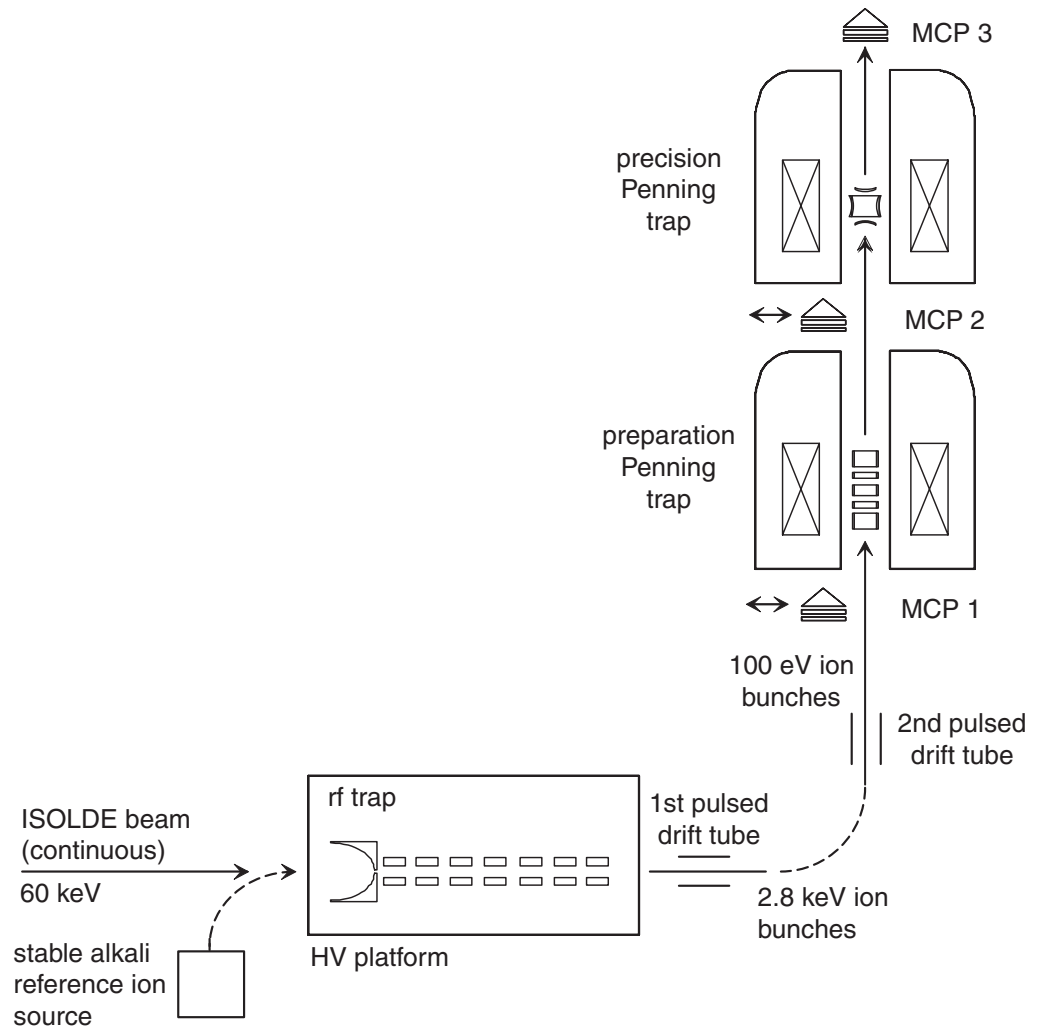


Figure 1. Schematic diagram of the ISOLTRAP setup with the linear rf trap for bunching of the continuous ISOLDE ion beam, the cylindrical preparation Penning trap for cooling and isobaric selection of ions, and the hyperbolic precision Penning trap for cyclotron frequency determination. Micro-channel-plate detectors allow to monitor the transfer (MCP 1 and 2) and to record the time-of-flight (TOF) resonance (MCP 3) for the actual mass measurement.

3. Results and discussion

The decay of $^{37}\text{K}^+$ ions (figure 2(b)) can be monitored by measuring their number in the preparation trap as a function of the storage time (see figure 2(c), full circles). An exponential fit to the data points yields an uncorrected half-life $T'_{1/2} = 1.10(4)$ s. Since the decaying ions are subjected to helium buffer gas at a pressure of about 1×10^{-4} mbar, the collisions lead to an increase of the magnetron radius and thus to a reduced number of detected ions after ejection through the 3-mm exit bore of the trap. This rate of ion loss was determined for the stable nuclide $^{39}\text{K}^+$ from the reference ion source (figure 2(c), open circles). A fit of an exponential decrease of the ion number yields a value $T_{1/2}^{\text{loss}} = 6.5(9)$ s. Taking this ion loss into account, the half-life

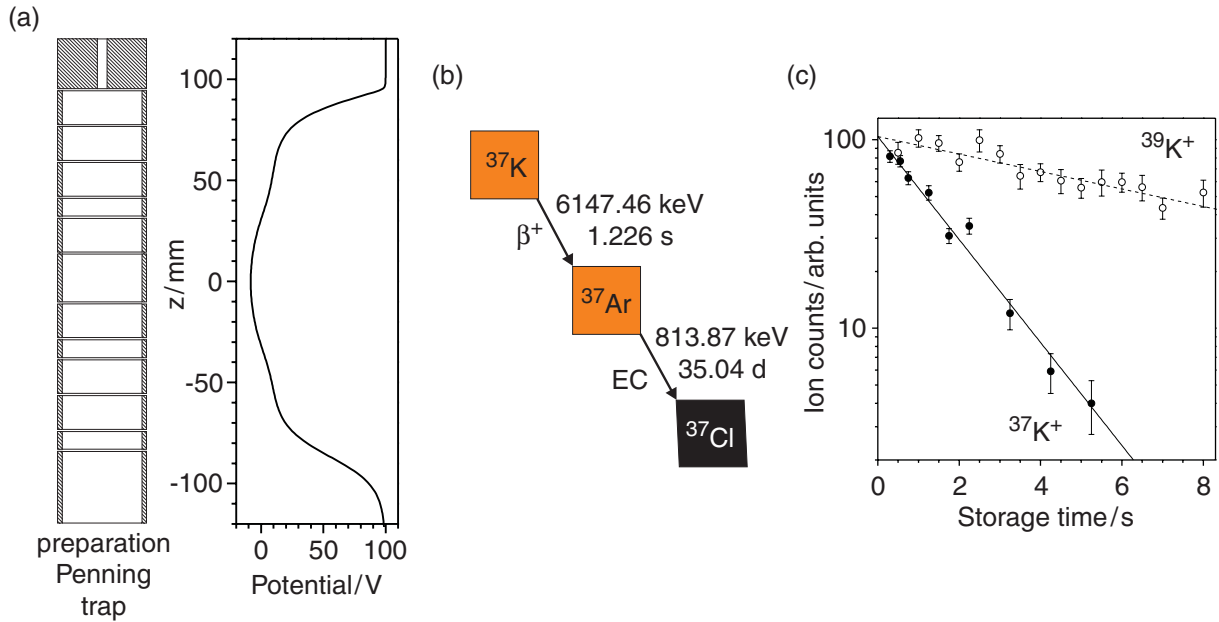


Figure 2. (a) Schematic diagram of the cylindrical preparation Penning trap and the potential along the axis. (b) Decay chain of ^{37}K [29]. (c) Ion counts of $^{37}\text{K}^+$ as a function of the storage time in the preparation Penning trap (full circles). No centring [26] was applied. The ions were detected by ejection from the preparation trap and transferred through the precision trap directly onto the MCP detector. The transfer time has been corrected for. For comparison the ion counts of stable $^{39}\text{K}^+$ as stored under the same conditions have been added (open circles). The data points have been rescaled to the same ion count at $t = 0$. The solid and the dashed lines are exponential fits to the data points.

of $^{37}\text{K}^+$ is corrected to $T_{1/2} = T_{1/2}^{\text{loss}} T'_{1/2} / (T_{1/2}^{\text{loss}} - T'_{1/2}) = 1.32(7)$ s, close to the literature value of 1.226(7) s [29]. Further improvement in the measurement is possible by, e.g., buffer-gas-assisted cooling of the mother nuclides for a reduction of ion loss.

While this measurement demonstrates the possibility of in-trap decay half-life determination, the main focus of the present investigation was set on the capturing and mass spectrometry of the daughter nuclide. For this purpose, a quadrupolar excitation was applied to $^{37}\text{Ar}^+$ to reduce the loss of the product ions [26]. In general, the cooling and centring of the recoil ions is essential for preparation and further transfer to the precision Penning trap for the determination of the cyclotron frequency.

A characteristic property of a cyclotron resonance is the TOF effect

$$\Delta TOF = 1 - \frac{TOF_{v_c}}{TOF_{\text{base}}}, \quad (1)$$

where TOF_{v_c} is the mean TOF in resonance and TOF_{base} the mean TOF ‘baseline’ far from resonance, i.e. without excitation of the cyclotron motion. The TOF effect is a measure for the radial energy gain of the ions: a higher gain leads to an increased acceleration after ejection, a shorter TOF and, consequently, a larger TOF effect.

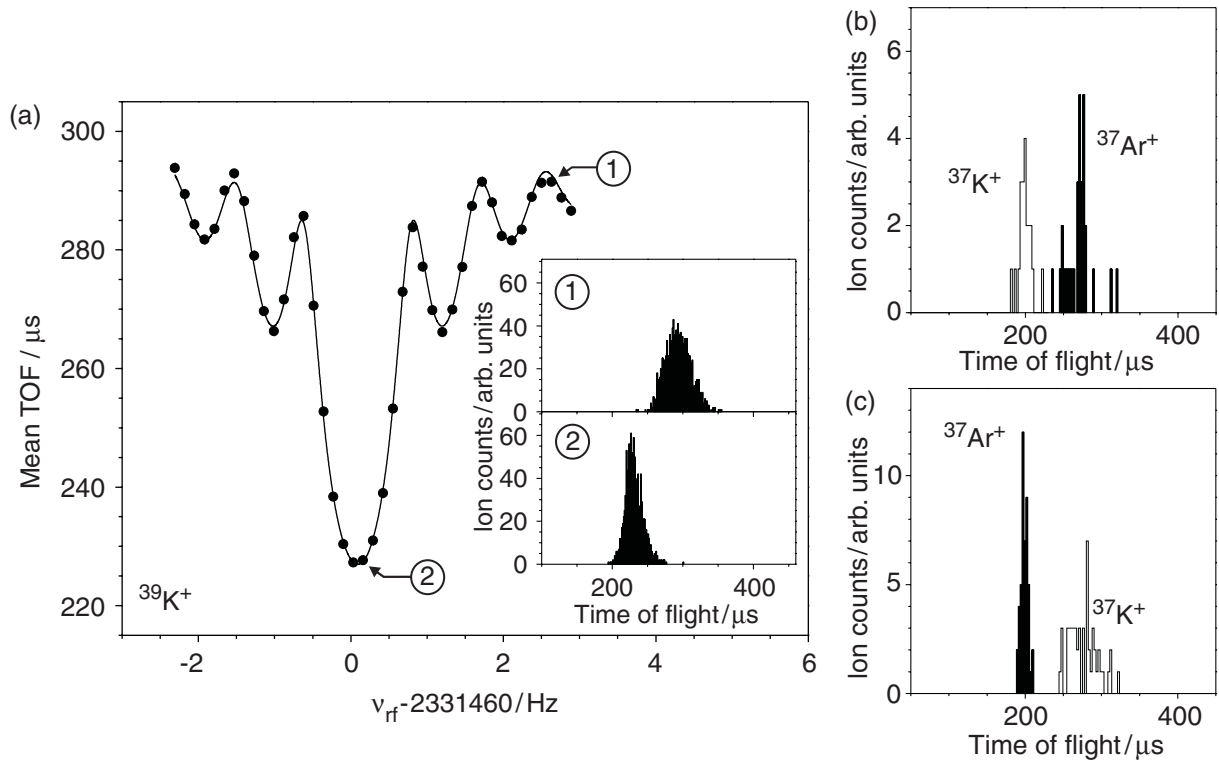


Figure 3. (a) Cyclotron resonance of $^{39}\text{K}^+$. The inset shows two time-of-flight spectra (off and in resonance, labelled with ‘1’ and ‘2’, respectively). (b, c) In-resonance TOF spectra for quadrupolar excitation of $^{37}\text{K}^+$ and $^{37}\text{Ar}^+$ in the precision Penning trap (data are taken from the 3-s cyclotron resonances in figure 4).

In the case of only one species, a cyclotron resonance looks like the example shown in figure 3(a) where $^{39}\text{K}^+$ from the alkali reference ion source was selected and transferred to the precision trap. The corresponding TOF spectra off and in resonance are shown in the inset (marked with ‘1’ and ‘2’, respectively). Resonantly excited ions with $\nu_{\text{rf}} = \nu_c$ arrive after a mean TOF of about $230 \mu\text{s}$. Ions that are not excited arrive at a later time at the detector (mean TOF $\approx 290 \mu\text{s}$) and can be clearly separated from the resonantly excited ions.

In case of in-trap decay, where, in general, both ion species are transferred to the precision trap, only the ions that are in resonance with the applied excitation arrive at a shorter time at the detector. In this case, the TOF signals are split into two components that correspond to the two stored ion species. Figures 3(b) and (c) show TOF spectra at resonance for $^{37}\text{K}^+$ and $^{37}\text{Ar}^+$, respectively.

The TOF effect can be used as a measure of the relative abundance of the two species in the precision Penning trap. In figure 4, the cyclotron resonances of $^{37}\text{K}^+$ and $^{37}\text{Ar}^+$ for storage times of 1, 3 and 5 s are shown. To minimize decay losses in the precision Penning trap, the duration of the quadrupolar excitation was set to 50 ms, which results in a linewidth of $\Delta\nu(\text{FWHM}) \approx 1/T_{\text{rf}} = 20 \text{ Hz}$. The solid lines are fits of the theoretical lineshape to the data points [28]. With the change of ΔTOF as a function of the storage time in the preparation trap, the decay of $^{37}\text{K}^+$ and the buildup of $^{37}\text{Ar}^+$ are demonstrated.

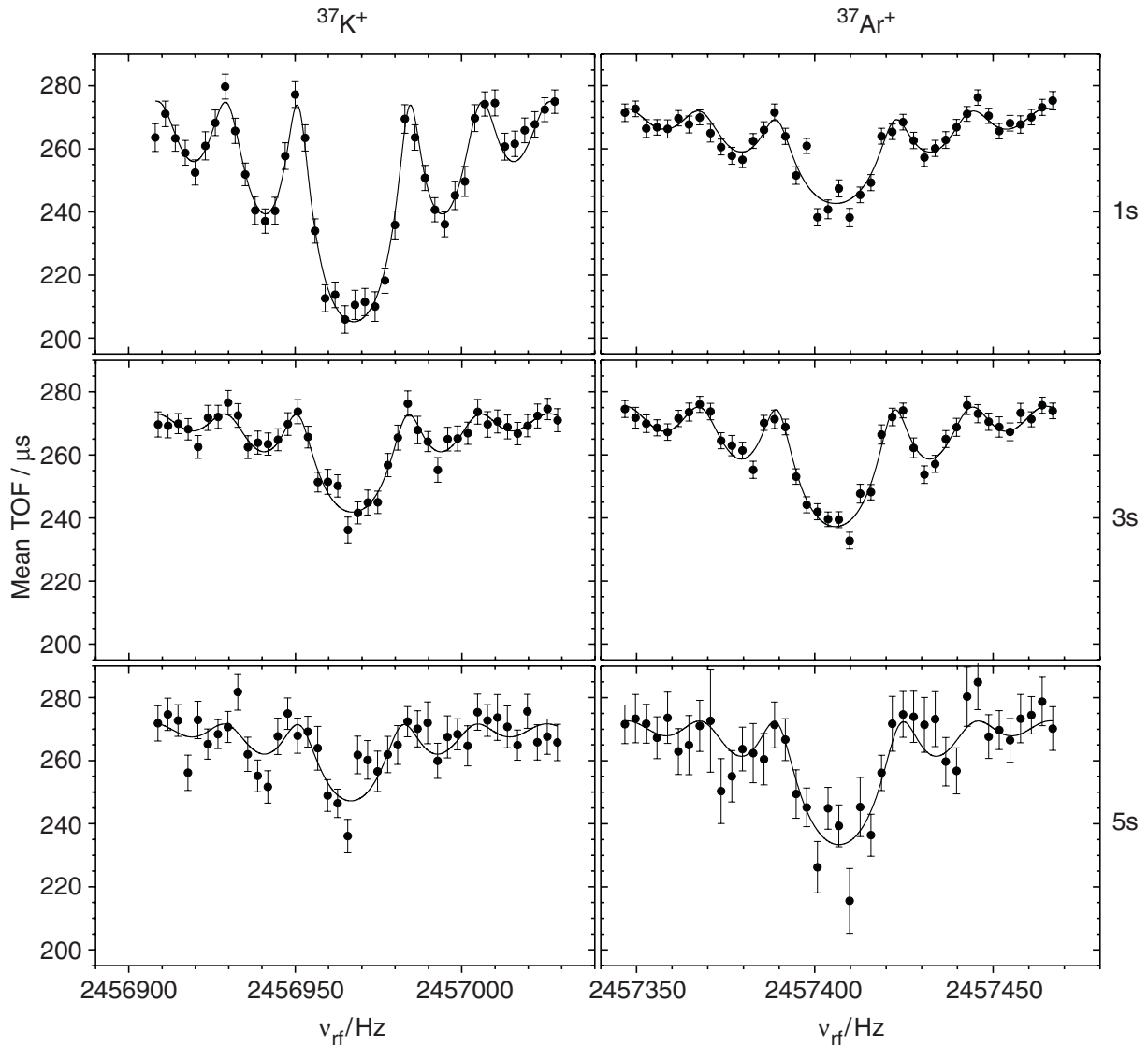


Figure 4. Cyclotron resonances for $^{37}\text{K}^+$ and $^{37}\text{Ar}^+$ after a storage time in the preparation trap of 1, 3, and 5 s. The solid lines are fits of the theoretical line shape to the data points [28].

The relative abundance of each ion species in the trap is reflected by the corresponding TOF effects

$$\frac{N_i}{N_1 + N_2} = \frac{\Delta TOF(v_{c,i})}{\Delta TOF(v_{c,1}) + \Delta TOF(v_{c,2})}, \quad (2)$$

where $i = 1, 2$ denotes the two different species in the trap, here $^{37}\text{K}^+$ and $^{37}\text{Ar}^+$. From the TOF effects of the cyclotron resonances in figure 4, the relative abundance of $^{37}\text{Ar}^+$ in the precision trap can be calculated and is plotted in figure 5 as a function of the storage time in the preparation Penning trap. In the case that all daughters are charged and remain stored in the trap, a behaviour given by the dashed line is expected. The deviation of the experimental

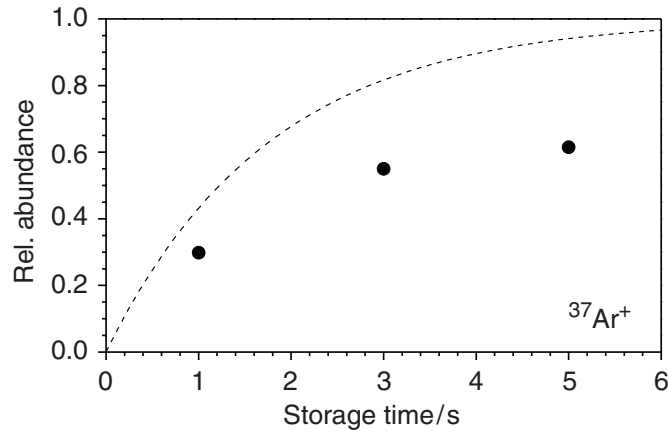


Figure 5. Relative abundance of $^{37}\text{Ar}^+$ as a function of the storage time in the preparation trap as calculated with equation (2) (data are taken from the cyclotron resonances in figure 4). The dashed line shows the increase in the relative abundance following $f(t) = 1 - \exp[-\ln(2)t/T_{1/2}]$ with the half-life $T_{1/2} = 1.226$ s of ^{37}K .

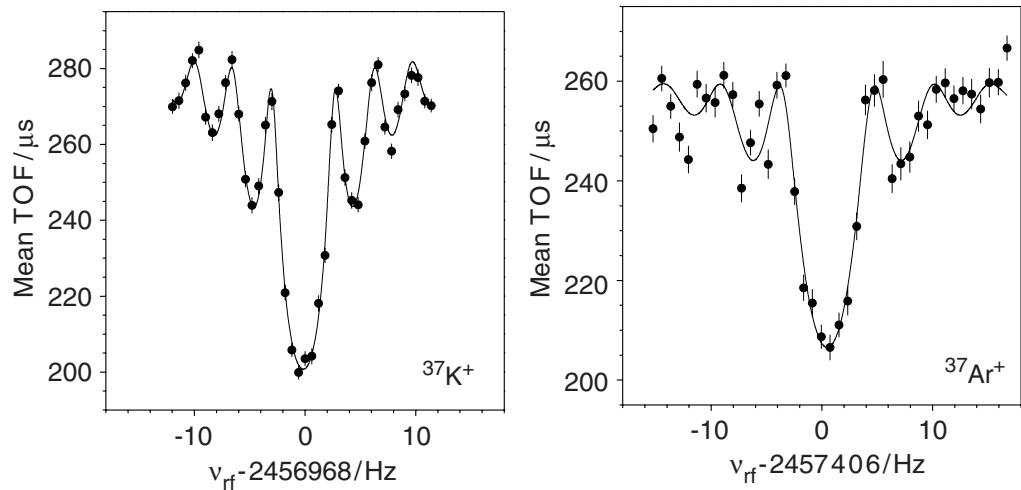


Figure 6. Cyclotron resonances for $^{37}\text{K}^+$ (excitation duration $T_{\text{rf}} = 300$ ms) and $^{37}\text{Ar}^+$ ions ($T_{\text{rf}} = 200$ ms), which were obtained directly from ISOLDE and by in-trap decay, respectively. The solid lines are fits of the theoretical line shape to the data points [28].

result is due to axial loss of $^{37}\text{Ar}^+$ for kinetic recoil energies exceeding the trapping potential, a fraction of daughters produced as neutrals and charge-exchange reactions with the buffer gas. For a quantitative analysis, more data are needed but, in general, the electron shake-off probability and the rate of charge exchange in the buffer gas of the preparation trap can be deduced from the relative abundance of the daughter ions.

When the buffer-gas-assisted mass-selective centring is applied only for either $^{37}\text{K}^+$ or $^{37}\text{Ar}^+$, i.e. only one species is transferred to the precision Penning trap, cyclotron resonances as shown in figure 6 are obtained. From four resonances of $^{37}\text{Ar}^+$ measured after in-trap decay

Table 1. Candidates for mass spectrometry of in-trap-decay product ions with ISOLTRAP at ISOLDE: β^- decay of manganese to iron (data are taken from [29, 32, 33]).

Nuclide	J^π	Half-life	Yield (at/ μ C)	Mass excess (keV)	Decay energy (keV)
^{61}Mn	$5/2^{-\text{a}}$	670(40) ms	$\sim 10^6$	-51560(230)	7370(230)
^{61}Fe	$3/2^-, 5/2^-$	5.98(6) min		-58921(20)	3977(20)
$^{61\text{m}}\text{Fe}$	$9/2^{+\text{b}}$	250(10) ns		-58060(20)	
^{62}Mn	$3^{+\text{a,c}}$	671(5) ms	$\sim 10^6$	-48040(220)	10860(220)
$^{62\text{m}}\text{Mn}$	$1^{+\text{a,c}}$	92(13) ms		-48040(230) ^b	
^{62}Fe	0^+	68(2) s		-58901(14)	2531(25)
^{63}Mn	$5/2^{-\text{b}}$	275(4) ms	$\sim 10^5$	-46350(260)	9190(310)
^{63}Fe	$5/2^{-\text{a}}$	6.1(6) s		-55550(170)	6290(170)

^a Value is uncertain.

^b Estimated value.

^c Assignment to ground state or isomeric state is uncertain.

(with excitation times from 50 ms up to 200 ms in the precision Penning trap), a mass excess of $D = -30948.7(2.4)$ keV was deduced by use of the evaluation procedure as described in [31]. Two different reference masses were taken: $^{39}\text{K}^+$ from the stable alkali reference-ion source and $^{37}\text{K}^+$ from the continuous ISOLDE ion beam. The mass excess of $^{37}\text{Ar}^+$ agrees with the AME2003 value $D(^{37}\text{Ar}) = -30947.66(21)$ keV [32]. Note that whether the singly charged daughter ion is produced by electron shake-off or charge exchange does not contribute any systematic effect to the mass measurement due to the buffer-gas centring in the preparation trap.

4. Summary and outlook

For the first time, the recoiling daughter of a trapped, beta-decaying nuclide has been held in a Penning trap and its mass has also been measured. Ions of daughter nuclides after in-trap decay offer a new approach to mass spectrometry of short-lived nuclides which are otherwise not available: they either require a different type of ion source or do not diffuse out of the thick ISOL target. As a proof of principle, the decay of $^{37}\text{K}^+$ was used to produce $^{37}\text{Ar}^+$ in the preparation Penning trap of ISOLTRAP. The measured half-life of $^{37}\text{K}^+$ agrees with the literature value. The daughter nuclides were captured and stored with an appropriate trapping potential. They were further transferred to the precision Penning trap of ISOLTRAP; a measurement of the cyclotron frequency and thus a determination of the mass of $^{37}\text{Ar}^+$ was performed which is in agreement with the literature value.

As an example of possible further mass measurements after in-trap decay at ISOLTRAP, table 1 lists some iron isotopes that are not available at ISOL facilities such as ISOLDE. Although the ISOL technique allows to obtain a large variety of radioactive species, some nuclides, especially of refractory elements, will not be available due to excessive release times from the target matrix. In this particular example, the yields for the manganese mother isotopes are sufficient to obtain enough ions for the mass determination of the daughter nuclides [33].

Note that the lifetime can change significantly when the environment of the decaying nuclide is varied. An extreme example is the bound-state β decay of fully ionized $^{187}\text{Re}^{75+}$ observed at the ESR at GSI/Darmstadt which has a half-life several orders of magnitude shorter than the neutral nuclide [34]. But even a small change in the chemical surroundings may alter the half-life as observed for ^7Be encapsulated in C_{60} [35]. Thus, the determination of the half-life of free ions could add another facet to these observations. In addition, it should be possible to study the breakup patterns of chemical bonds in connection with the in-trap decay of a nucleus within a molecular ion and thus the nature of the structure and the stability of these molecules.

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