First Use of High Charge States for Mass Measurements of Short-Lived Nuclides in a Penning Trap

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Penning trap mass measurements of short-lived nuclides have been performed for the first time with highly charged ions, using the TITAN facility at TRIUMF. Compared to singly charged ions, this provides an improvement in experimental precision that scales with the charge state q. Neutron-deficient Rb isotopes have been charge bred in an electron beam ion trap to $q = 8 - 12+$ prior to injection into the Penning trap. In combination with the Ramsey excitation scheme, this unique setup creating low energy, highly charged ions at a radioactive beam facility opens the door to unrivaled precision with gains of 1–2 orders of magnitude. The method is particularly suited for short-lived nuclides such as the superallowed β emitter ^{74}Rb ($T_{1/2} = 65$ ms). The determination of its atomic mass and an improved Q_{EC} value are presented.

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Since their introduction into rare isotope research over 20 years ago [1,2], Penning traps have made major contributions to the exploration of the nuclear mass surface as evidenced by the large number of existing and proposed facilities [3] as well as the wealth of experimental results [4]. Advances in experimental techniques now allow measurements for virtually all low energy, rare isotope beams as Penning traps have been able to access nuclides with half-lives below 10 ms [5] as well as superheavies with production yields of less than 1 particle per second [6]. The widespread success of Penning traps is due to their precision following [7]

$$\frac{\delta m}{m} \propto \frac{m}{qBT_{\mathrm{rf}}\sqrt{N_{\mathrm{ion}}}} \tag{1}$$

where $\delta m/m$, is the achievable relative precision in mass m, q is the ion's charge state, and B is the magnetic field strength. The measurement time T_{rf} and the number of ions N_{ion} are limited by a nuclide's half-life and possibly by its production yield at radioactive beam facilities and efficiency of the spectrometer. Measurements are generally performed with singly charged ions (SCI) or in special cases, where coupled to a gas stopper cell, with $q = 2+$. Penning trap mass studies utilizing highly charged ions (HCI) have been successfully pioneered with stable nuclides [8]. Here the requirements of high efficiency and short measurement times are less relevant compared to working with radioactive ions. In the realm of rare isotope science with Penning traps, HCI represent a thus far unexplored opportunity to improve the experimental precision further circumventing constraints imposed by short half-lives and lower yields when probing the limits of nuclear existence.

The superallowed β emitter ^{74}Rb is a prime example where a short half-life of only 65 ms poses a real challenge to experiment. Despite several Penning trap mass measurements [9–11], the total transition energy Q_{EC} still contributes significantly to the uncertainty of its corrected $\mathcal{F}t$ value, only surpassed by theoretical uncertainties of the isospin-symmetry breaking corrections δ_C [12]. These have recently been reduced by experimentally providing the ^{74}Rb rms charge radius as an input for the calculation of δ_C [13]. The Q_{EC} value and δ_c are now close to sharing the same weight to the total uncertainty of the $\mathcal{F}t$ value. Among all superallowed β emitters used to extract V_{ud} of the Cabibbo-Kobayashi-Maskawa matrix [12], ^{74}Rb has the highest atomic number Z. It is hence of special importance in attempts to distinguish between conflicting nuclear models since δ_C approximately scales as Z^2 [14,15]. In this Letter we present the first Penning trap mass measurements of short-lived HCI, performed with TRIUMF's Ion Trap for

Atomic and Nuclear science (TITAN) [16], including a successful mass determination of $^{74}Rb^{8+}$.

Neutron-deficient Rb isotopes were produced at TRIUMF's ISAC facility [17] by bombarding a Nb target with a 98 μA, 500 MeV proton beam from the cyclotron. The surface-ionized Rb beam was accelerated to 20 keV and mass separated prior to injection into TITAN's radio frequency quadrupole cooler and buncher [18] where the ions were accumulated and cooled through collision with a He buffer gas. Extracted ion bunches were transferred with a beam energy of about 2 keV into the electron beam ion trap (EBIT) [19]. Operated with a 10 mA, 2.5 keV electron beam, the EBIT confined the ions radially by the space charge of the electron beam and an axial magnetic field of 3 T. The central, trapping drift tube was biased (at U_{trap}) slightly below the beam energy to remove most of the ions' kinetic energy. To provide confinement in the axial direction, a bias voltage of ≈ 100 V above the central drift tube was applied to the neighboring drift tubes, one of which was lowered during the ions' capture and extraction. Through electron-impact ionization the initially singly charged ions were charge bred to higher charge states. Because of a kinetic energy of qU_{trap} after extraction from the EBIT, different charge states q_i can be identified via time of flight (TOF) as illustrated in Fig. 1. At a fixed electron beam setting, a certain charge state can be maximized in abundance by optimizing the charge breeding time. For instance, the number of Rb ions with $q = 8+$ reached its maximum at a flat plateau of ≈ 20–27 ms of breeding. A Bradbury-Nielson ion gate [20] allowed the selection of one charge state by opening the gate for 300–500 ns during the beam transport from the EBIT to TITAN's measurement Penning trap (MPET). In the MPET, the ion's cyclotron frequency $\nu_c = qB/(2\pi m)$ was determined by the time-of-flight ion-cyclotron resonance (TOF-ICR) technique [21–23]. Typical time-of-flight ion-cyclotron resonances recorded during these measurements are shown in Figs. 2(a) and 2(c). At the TOF minimum the rf frequency ν_{rf} equals ν_c. The width of a resonance, $\Delta\nu_{\mathrm{FWHM}}$, is solely governed by the duration T_{rf} of the rf field [24] but is independent of q, m, or B. Hence, the gain in relative precision $\delta m/m = \delta\nu_c/\nu_c$ when utilizing HCI is due to the larger ν_c for a given $\Delta\nu_{\mathrm{FWHM}}$.

As demonstrated recently, a reduction by a factor of 2–3 in $\delta\nu_c$ can be achieved by the Ramsey method of separated oscillatory fields, in which the rf field is applied during two pulses separated by a waiting period [25,26]. This technique is now used at TITAN, where it can be combined with the advantages offered by HCI. In Figs. 2(b) and 2(d), a Ramsey excitation scheme with two 6 ms rf pulses separated by a 85 ms waiting period has been employed (denoted as 6-85-6 ms throughout this Letter). With a charge state $q = 9+$ in addition to the Ramsey excitation a gain in precision of $\delta\nu_c/\nu_c$ by a factor of about 20 has been achieved compared to the conventional technique. A factor of 36 is possible when optimizing our implementation of the Ramsey technique to the performance reported in [27] and using $q = 12+$, the highest charge state for which a measurement has been performed.

Because of the $\sqrt{N_{\mathrm{ions}}}q$ dependence of the achievable precision [see Eq. (1)], HCI are favorable as long as the loss in efficiency caused by the charge breeding is smaller

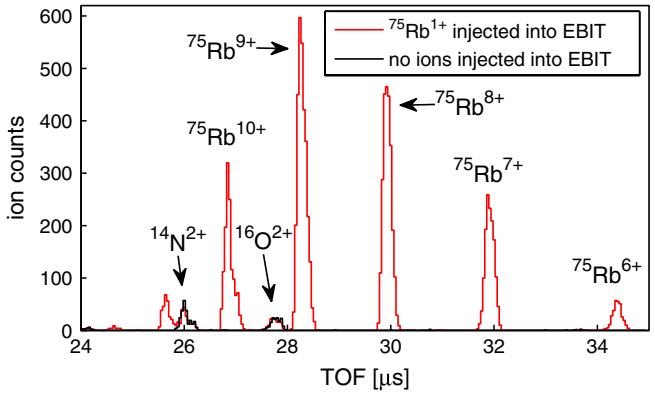

FIG. 1 (color online). TOF spectrum of 500 ion bunches of radioactive ^{75}Rb extracted from the EBIT with an 800 ns extraction pulse after 35 ms of charge breeding with a 10 mA, 2.5 keV electron beam. $^{16}O^{2+}$ and $^{14}N^{2+}$ are due to ionized and further charge-bred residual gas in the EBIT.

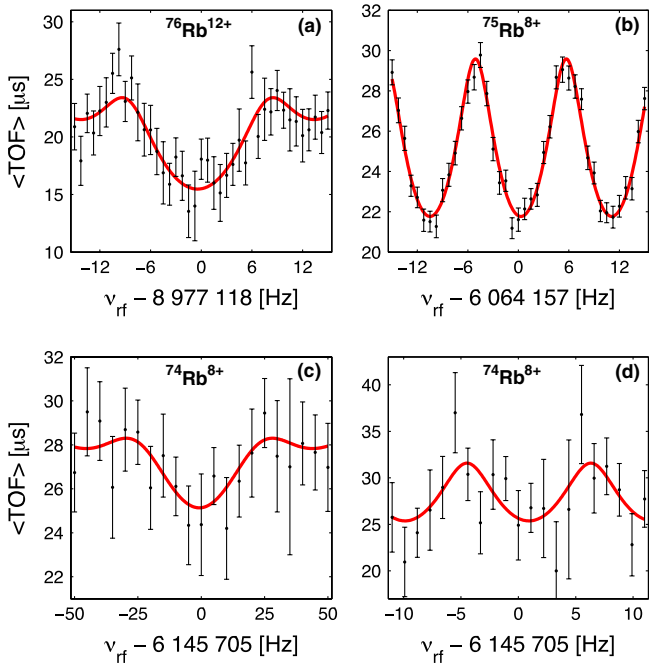

FIG. 2 (color online). Time-of-flight ion-cyclotron resonances for Rb isotopes in charge state $q = 8+$ and $q = 12+$. During (a) and (c) the rf field was continuously applied for $T_{\mathrm{rf}} = 97$ and 30 ms, respectively, and a Ramsey excitation scheme with 6-85-6 ms was utilized in (b) and (d). The (red) lines represent the fit to the theoretical line shapes [22,25].

than q^2. However, these measurements must be performed with only a few ions (1–5) at a time. If ^{75,76}Rb had been done with SCI, beam attenuation would have been required. Hence, a lower efficiency for $^{75,76}Rb^{+8}$ could be compensated by reducing this beam attenuation. The total number of ions is affected by the breeding in three ways: (1) nonunity efficiency for the chosen charge state due to the charge-state distribution (see Fig. 1), (2) increased beam emittance causing a reduced transport and trapping efficiency at MPET, and (3) loss of ions due the radioactive decay in the EBIT. For the ^{74}Rb measurement, ions were kept for 23 ms in the EBIT ($\sim 0.35T_{1/2}$) and the breeding was done in parallel to a measurement in MPET.

Since HCI are more likely to exchange charge with the residual gas in MPET, the use of HCI demands severe vacuum requirements compared to SCI. When an ion recombines with one or more electrons during the measurement period, its cyclotron frequency changes and the ion is subsequently unaffected by the rf-excitation scheme. These partially recombined ions add to the detected background reducing the sensitivity and possibly induce frequency shifts, e.g., through ion-ion interactions. In preparation for these measurements, we baked the trap, the vacuum vessel, and the extraction beam line. Despite an improved vacuum of $\approx 6 \times 10^{-11}$ mbar in the MPET vacuum section, charge exchange occurred, and in the TOF spectrum we observed an increasing abundance of H_2^+ with longer storage time. Even though the current vacuum allowed the recording of a TOF resonance of $^{76}Rb^{+8}$ with $T_{rf} = 1$ s, ions were typically trapped for 97 ms in MPET as a compromise between increased precision and recombination for longer T_{rf}. A summary of the trapping and rf schemes are listed in Table I together with the measured frequency ratio $R = \nu_c^{ref}/\nu_c^{meas}$ to the reference ion, $^{85}Rb^{9+}$. $^{85}Rb^{+}$ was delivered at the same beam energy from TRIUMF's off-line ion source [28]. A fraction of the mass $A = 74$ beam from ISAC was ^{74}Ga, and we have determined its mass as well. In order to purify the beam and to push the contaminant out of the trap, a dipole rf field at the reduced cyclotron frequency ν_+ [3] of $^{74}Ga^{8+}$ was applied for 20 ms during one set of ^{74}Rb measurements.

To avoid potential frequency shifts induced by ion-ion interactions, isobaric contamination, or charge exchange, we only considered ion bunches with 5 or less detected ions per detection cycle and performed a count-class analysis [29] where statistics allowed. For all $A = 74$ measurements, the low count rate made it rare that two or more ions were stored at the same time. To be conservative, we have nevertheless added the difference in ν_c between 1 and 1–5 detected ions per ion bunch to the systematic uncertainty. Ions of different charge states were produced due to charge exchange in MPET as mentioned above and extracted onto the MPET detection multichannel plate. They could not be resolved in TOF because the ions' initial energy spread when captured in the MPET was large enough to smear out differences in TOF between adjacent charge states. Thus, the cut on the TOF range was varied in the analysis, and when shifts in R were observed, they were added in quadrature as systematic uncertainties. By far the largest of these shifts was found in ^{74}Ga, where it accounts for 60 ppb. Other systematic effects due to improper electric field compensation, misalignment between magnetic and trap axes, or harmonic distortions of the electrode structure as well as relativistic effects were minimized by choosing a reference ion, $^{85}Rb^{9+}$, with similar m/q.

TABLE I. Mean frequency ratios $\bar{R}$ of $^{76,75,74}Rb^{q+}$ and $^{74}Ga^{8+}$ with respect to $^{85}Rb^{9+}$. Where applicable, the ratio and error are the result of a count-class analysis. Uncertainties are displayed as (stat) and {stat + syst}.

Species	Excitation [ms]	$\bar{R} = \nu_c^{ref}/\nu_c^{meas}$	No. of measurements
$^{76}Rb^{8+}$	97 (conventional)	1.006 067 401(15)	5
	6-85-6 (Ramsey)	1.006 067 422(12)	4
	Previous combined	1.006 067 414{22}	
$^{76}Rb^{12+}$	97 (conventional)	0.670 692 259(23)	1
$^{75}Rb^{8+}$	6-85-6 (Ramsey)	0.992 864 003(10){25}[a]	5
$^{74}Rb^{8+}$	30 (conventional)	0.979 689 909(318)[b]	4
	6-85-6 (Ramsey)	0.979 689 552(79){98}[bc]	3
	20(dip) and 6-65-6(Ram)	0.979 689 66(10){13}[bc]	2
	Previous combined	0.979 689 609{86}	
$^{74}Ga^{8+}$	97 (conventional)	0.979 460 129(29){71}[b]	4

[a] ν_c^{meas} was determined for $^{75}Rb^{8+}$ by a 40-7-40 ms Ramsey scheme, in which the sidebands are less pronounced [25].
[b] Too few ions to perform a count-class analysis.
[c] Too few data to study TOF-range dependence. As an upper limit the TOF range dependence for $^{74}Ga^{8+}$ was added in quadrature.

Remaining errors were investigated by measuring $^{85}Rb^{10,8+}$ and $^{87}Rb^{9+}$ versus $^{85}Rb^{9+}$ in various experimental settings. Within 1 standard deviation, all of these measured R agree with the literature values constraining these systematic uncertainties to less than 42 ppb for $^{74}Rb^{8+}$ and 20 ppb for the other on-line measurements with $q = 8+$. The different upper limits are due to a turbo pump failure that necessitated the reconditioning of the electron beam in the EBIT and subsequent retuning of the injection into MPET. According to [30,31], the image charges do not alter the measurement of ν_c. Details about all mentioned systematic uncertainties as well as correlations in R due to shared reference frequency measurements will be discussed in a forthcoming publication.

With the measured $\bar{R}$, the atomic masses of the respective nuclides are calculated in Table II taking into account the total electron binding energies (0.5, 0.7, and 1.6 keV for $Rb^{8,9,12+}$ [35], respectively, as well as 0.6 keV for Ga^{8+} [36]). We also performed a complete atomic mass evaluation based on the procedures in [32], but adding the electron binding energies to the linear equations which were neglected in previous evaluations. Our results are in agreement with ISOLTRAP's measurements [11,37] (see Fig. 3). Because of the use of HCI and the Ramsey excitation, they are comparable in precision despite our significantly shorter measurement time (< 22 h for ^{74}Rb). Combined with the ISOLTRAP mass value, the total transition energy Q_{EC} of the superallowed decay in ^{74}Rb is 10 416.8(3.9) keV, an improvement by $\approx 12\%$. This results in a statistical rate function of $f = 47\,283(94)$ [34] and translates together with the recent improvements in δ_C due to the laser spectroscopy work [13] to a corrected $\mathcal{F}t$ value of 3077(11) s, when considering δ_C based on shell model calculations with Woods-Saxon radial wave functions [12]. To reduce its uncertainty further, a new mass measurement of ^{74}Rb and its daughter, ^{74}Kr, is planned with charge states up to $q \approx 30+$, reachable by a more intense electron beam ($I_e = 400$ mA). However, to take full advantage of HCI, it will be necessary to gain better control over systematic effects. Work is under way to further improve the MPET vacuum to avoid charge exchange and its associated uncertainties. It was demonstrated that m/q dependent shifts at TITAN for SCI are at the level of a few ppb [23,38] and an experimental confirmation of this accuracy for HCI is expected. In light of the purely statistical uncertainties of ^{76,75}Rb in Fig. 3, such a new measurement could provide knowledge of the Q_{EC} value of ^{74}Rb within 0.5 keV. Uncertainties of half-life, branching ratio, or theoretical corrections would then surpass the Q_{EC} value's contribution to the $\mathcal{F}t$ value's error, stimulating new branching ratio measurements. At this level of experimental precision f's uncertainty in ^{74}Rb would be dominated by theory and not by the Q_{EC} value [34,39]. Perhaps most importantly, more stringent comparisons of conflicting theoretical models of δ_c similar to [15] could challenge perceived consistencies between a set of δ_C calculations, experimental results, and the conserved vector current hypothesis since ^{74}Rb, with its largest δ_C among all superallowed β emitters, would carry particular weight were it not limited by the current precision in its Q_{EC} value.

In summary, Penning trap mass measurements of highly charged, short-lived nuclides have been performed for the first time, opening a new class of on-line mass measurements with potentially up to 2 orders of magnitude improved precision versus conventional SCI-TOF-ICR spectroscopy when combined with the Ramsey excitation. This is essential for fundamental symmetries studies, such as presented here for the superallowed β emitter ^{74}Rb. For nuclear structure and nuclear astrophysics, where the experimental precision is already sufficient, this novel technique will reduce the measurement time and thus allow one to map the nuclear mass landscape more quickly. In addition, the same precision can be achieved for lower production yields and/or shorter half-lives. At this time, TITAN is

TABLE II. Mass excess for measured nuclides. $^{76}Rb^{12+}$ was a feasibility test of higher charge states. Because of its larger systematic uncertainty, only those with $q = 8+$ were considered.

Nuclide	This work [keV]	Mass evaluation [keV]
^{76}Rb	−60 481.0(1.6)	−60 479.1(0.9)
^{75}Rb	−57 218.7(1.7)	−57 218.7(1.2)
^{74}Rb	−51 916.5(6.0)	−51 916.0(3.0)[a]
^{74}Ga	−68 049.7(5.0)	−68 049.6(3.0)[b]

[a]Reference [32] included a mass value determined from the $\mathcal{F}t$ value of other superallowed β emitters and ^{74}Rb's half-life and branching ratio [33]. This evaluation does the same with an updated Q-value estimate of 10 413.8(7.0) keV [34] based on [12]. For the discussion of superallowed decays, we of course do not consider this estimate.
[b]Yield measurements determined an isomer to ground state ratio of about 1:190.

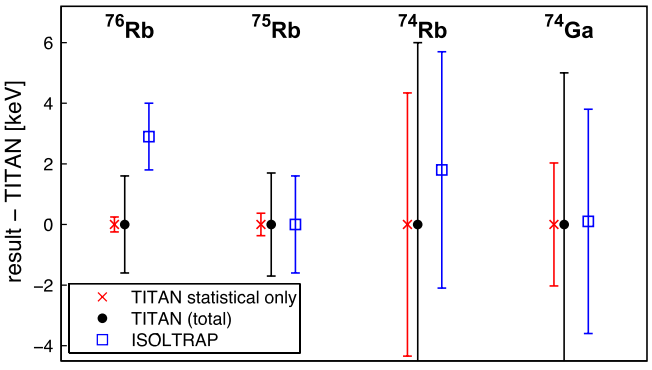

FIG. 3 (color online). Atomic masses of ^{76,75,74}Rb and ^{74}Ga in comparison to their respective ISOLTRAP measurements [11,37]. Statistical uncertainties are based on fits of ion bunches with 1–5 detected ions but without a count-class analysis.

unique in providing the possibility for high charge states in Penning trap mass spectroscopy of radionuclides.

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