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Accurate mass measurements of very short-lived nuclei

Prerequisites for high-accuracy investigations of superallowed β-decays

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Abstract. Mass measurements of ^{34}Ar, $^{73\text{-}78}$Kr, and ^{74,76}Rb were performed with the Penning-trap mass spectrometer ISOLTRAP. Very accurate Q_{EC}-values are needed for the investigations of the $\mathcal{F}t$-value of $0^+ \to 0^+$ nuclear β-decays used to test the standard model predictions for weak interactions. The necessary accuracy on the Q_{EC}-value requires the mass of mother and daughter nuclei to be measured with $\delta m/m \leq 3 \cdot 10^{-8}$. For most of the measured nuclides presented here this has been reached. The ^{34}Ar mass has been measured with a relative accuracy of $1.1 \cdot 10^{-8}$. The Q_{EC}-value of the ^{34}Ar $0^+ \to 0^+$ decay can now be determined with an uncertainty of about 0.01%. Furthermore, ^{74}Rb is the shortest-lived nuclide ever investigated in a Penning trap.

PACS. 21.10.Dr Binding energies and masses – 24.80.+y Nuclear tests of fundamental interactions and symmetries

1 Introduction

Nuclear β-decay is a unique, relatively easy-to-access laboratory for investigations of the weak interaction. Of special interest are the superallowed $0^+ \to 0^+$ nuclear β-decays where the axial-vector decay strength is zero. The transition rate $\mathcal{F}t$ for these decays

$$\mathcal{F}t = \frac{K}{2G_{\mathrm{V}}^2(1+\Delta_{\mathrm{R}}^{\mathrm{V}})} \tag{1}$$

should be nucleus independent if the vector current is a conserved quantity (CVC hypothesis). In this case, the vector coupling constant G_{V} can be extracted from eq. (1) and then be used together with the value for the Fermi coupling constant G_{F} extracted from muon decay to determine the V_{ud} element of the Cabibbo-Kobayashi-Maskawa (CKM) matrix. Consequently, nuclear β-decay can serve to test the CVC hypothesis as well as the unitarity of the CKM-matrix. In reality there are small nucleus-dependent corrections that are related to the nuclear structure and to the presence of charge-dependent forces in the nucleus. These corrections, the radiative correction δ_{R} and the Coulomb correction δ_{C}, have to be calculated. The transition rate $\mathcal{F}t$ can then be expressed as

$$\mathcal{F}t = ft(1+\delta_{\mathrm{R}})(1-\delta_{\mathrm{C}}) \tag{2}$$

for a certain nuclear β-decay. The statistical rate function f and the partial half-life t depend on three experimentally accessible parameters: the transition energy Q_{EC} used to calculate f, the nuclear half-life $T_{1/2}$ and the branching ratio R that determine the partial half-life t.

Until now the parameters of nine superallowed β-decays have been measured with sufficiently high accuracy [1]. For many of the other candidates the knowledge of the Q_{EC}-value is limited, prohibiting an accurate determination of the $\mathcal{F}t$-value. Conventional techniques for Q_{EC}-value determinations are often not accurate enough or require too high production yields in the case of very short-lived species. To reach an uncertainty level of 0.1% or better for the ft-value such an uncertainty is required for half-life and branching ratio. But since the statistical rate function f is proportional to Q_{EC}^5, the Q_{EC}-value has to be determined with an uncertainty better

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than 0.01%. This requires direct mass measurements of mother and daughter nuclei with relative uncertainties of $\delta m/m \leq 3 \cdot 10^{-8}$.

The very accurately determined $\mathcal{F}t$-values appear to be constant [1], supporting CVC. However, the extracted V_{ud} together with the much smaller V_{us} and V_{ub} result in a CKM-matrix that is not unitary by 2.3 standard deviations [2]. One important point to be clarified in the characterization of superallowed β-decays is the calculation of the least-known correction, the Coulomb correction δ_{C}. Today, the nine well-investigated cases have quite low calculated Coulomb corrections of the order of 0.5%. The best candidates for testing these calculations are nuclei with Coulomb corrections predicted to be large. Two such cases are ^{34}Ar and ^{74}Rb. ^{34}Ar lies in the same Z region as the nine well-known emitters. Its study allows the Coulomb corrections in this area to be tested. With ^{74}Rb the Z-dependence of δ_{C} can be investigated.

In the case of the ^{74}Rb decay, an uncertainty of a few keV would already allow for the discrimination of two different approaches to calculate the charge-dependent correction δ_{C} if one assumes the vector current as conserved and thus $\mathcal{F}t$ as constant. The first approach makes use of the shell model [3]. The second is based on Hartree-Fock (HF) and random phase approximation (RPA) calculations [4]. The Coulomb corrections calculated under different assumptions range from one to two percent in the shell model calculations, while the HF and RPA calculations predict a much lower δ_{C} of only 0.75%.

2 Experimental setup

ISOLTRAP is a Penning-trap mass spectrometer installed at the online isotope separator ISOLDE/CERN [5]. The mass-separated 60 keV ion beam from ISOLDE is guided to the ISOLTRAP setup shown in fig. 1. It consists of three main parts: 1) a linear gas-filled radio-frequency quadrupole (RFQ) trap for retardation, accumulation, cooling and bunched ejection at low energy, 2) a gas-filled cylindrical Penning trap for isobaric separation, and 3) an ultrahigh-vacuum hyperboloidal Penning trap for the mass measurement. The function and performance of the two Penning traps are described in [6,7] and the recently added linear RFQ trap in [8].

The beam delivered from ISOLDE is accumulated, cooled and bunched in the linear RFQ trap. The main task of this device is to transform the 60 keV continuous ISOLDE beam into ion bunches at low energy (2–3 keV) and low emittance ($\leq 10\,\pi$ mm mrad). These bunches can be efficiently transported to and captured in the first Penning trap. Here, a mass-selective buffer gas cooling technique is employed that allows this trap to be operated as an isobar separator with a resolving power of up to $R \approx 10^5$ for ions with mass number $A \approx 100$. The ions are then delivered to the second trap. This is the high-precision trap used for the mass measurements of the ions. The mass measurement is carried out via a determination of the cyclotron frequency $\nu_{\mathrm{c}} = 1/2\pi \cdot q/m \cdot B$ of an ion with mass m and charge q in a magnetic field of strength B using an azimuthal excitation voltage whose duration determines the mass resolving power. The energy gained from the excitation is detected by the corresponding decrease in time of flight when the ions are pulsed out of the trap to a detector. B is determined using a reference ion with known mass.

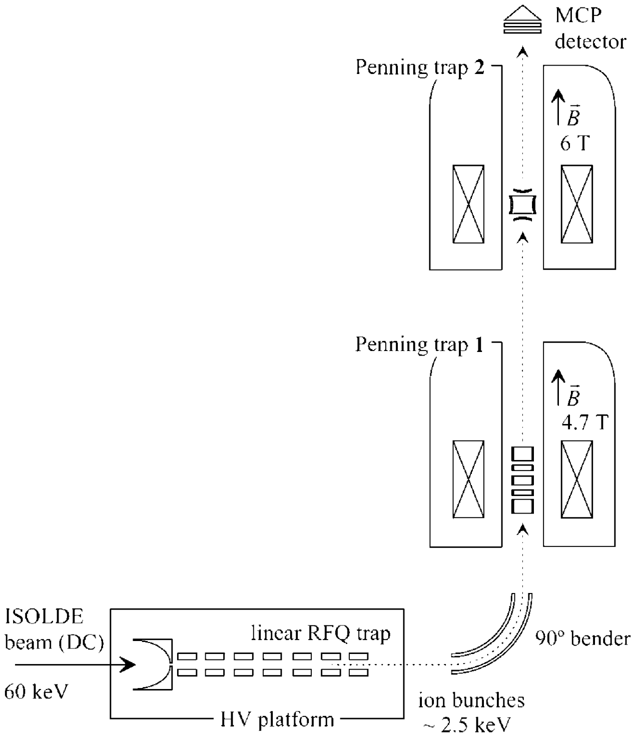

Fig. 1. Experimental setup of the ISOLTRAP Penning-trap mass spectrometer. The three main parts are 1) a linear gas-filled radio-frequency quadrupole (RFQ) trap for retardation of ions, accumulation, cooling and bunched ejection at low energy, 2) a gas-filled cylindrical Penning trap for further cooling and isobaric separation, and 3) an ultrahigh-vacuum hyperboloidal Penning trap for the actual mass measurement. For this, the cyclotron frequency is determined by a measurement of the time of flight of the ions ejected out of the Penning trap to a micro-channel plate (MCP) detector [6].

3 Experiment

The radioactive isotopes were produced by bombarding a thick target with 1 or 1.4 GeV proton pulses containing up to $3 \cdot 10^{13}$ protons. For the production of ^{74}Rb a Nb foil target was used in conjunction with a hot W surface ion source. In the case of Ar and Kr, the respective targets were CaO and ZrO. Though the plasma ion source is required for these elements, some selectivity was obtained by cooling the transfer line from target to source. The ions have been mass separated with a resolving power $M/\Delta M \approx 4000$ and were delivered to the ISOLTRAP setup.

The relatively long-lived ($T_{1/2} \geq 1$ s) isotopes of Kr and Ar were measured using excitation times of 300,

Table 1. Frequency ratios and mass excesses (ME) as determined in this work. In the case of Rb and Kr isotopes, ^{85}Rb was used for calibration of the magnetic field and ^{39}K for the Ar isotopes. Literature values are from ref. [13] except for ^{36}Ar [16].

Nucleus	$T_{1/2}$	Frequency ratio $\nu_c^{\mathrm{ref}}/\nu_c$	$\mathrm{ME}_{\mathrm{exp}}^{(a)}$ (keV)	$\mathrm{ME}_{\mathrm{lit}}$ (keV)	$\mathrm{ME}_{\mathrm{exp}}^{(a)} - \mathrm{ME}_{\mathrm{lit}}$ (keV)
^{74}Rb	65 ms	0.870835571(226)	−51905(18)	−51730(720)	−175
^{76}Rb	36.5 s	0.8942811649(232)	−60479.8(1.8)	−60481.0(8.0)	1.2
^{73}Kr	26 s	0.858999829(114)	−56550.8(9.0)	−56890(140)	339
^{74}Kr	11.5 min	0.8707037618(303)	−62330.3(2.4)	−62170(60)	−160
^{75}Kr	4.5 min	0.882455563(102)	−64323.6(8.0)	−64241(15)	− 83
^{76}Kr	14.6 h	0.8941733117(557)	−69010.4(4.4)	−68979(11)	−31
^{77}Kr	1.24 h	0.9059356611(248)	−70169.5(2.0)	−70171.0(9.0)	1.5
^{78}Kr	*stable*	0.9176619688(159)	−74179.2(1.3)	−74160.0(7.0)	−19.2
^{80}Kr	*stable*	0.9411690366(199)	−77891.9(1.6)	−77893.0(4.0)	1.1
^{82}Kr	*stable*	0.9646889212(305)	−80590.8(2.5)	−80588.6(2.6)	−2.2
^{34}Ar	844 ms	0.87209878056(922)	−18377.10(41)	−18378.3(3.0)	1.2
^{36}Ar	*stable*	0.9231027017(105)	−30231.44(46)	−30231.540(27)	0.11
^{38}Ar	*stable*	0.97430972477(952)	−34714.40(44)	−34714.77(49)	0.37
^{41}K	*stable*	1.05128226487(825)	−35559.02(45)	−35558.88(26)	−0.14

(a) Using $m(^{85}\mathrm{Rb}) = 84.911\,789\,738(12)$ u [17,13], $m(^{39}\mathrm{K}) = 38.963\,706\,82(30)$ u [13], and 1 u = 931.494009(7) MeV/c^2 [18].

600 and 900 ms. For the very short-lived ^{74}Rb ($T_{1/2}$ = 65 ms) a short measurement cycle had to be used for the ISOLTRAP measurement procedure similar to the one used earlier for ^{33}Ar ($T_{1/2}$ = 174 ms) [9]. In this case the excitation time in the measurement trap was chosen to be between 60 and 120 ms, yielding a mass resolving power of $M/\Delta M = 82000$ up to 164000.

The reference measurements for the calibration of the magnetic field were performed with an excitation time of 900 ms, thus increasing the resolving power to about 10^6. ^{39}K was used as reference for the Ar measurements and ^{85}Rb for the Rb and Kr measurements.

To achieve high accuracy for ^{34}Ar and ^{74}Kr, several independent frequency measurements were performed (7 and 13, respectively). Each was preceded and followed by a reference measurement. In the latter, the cyclotron frequency of the reference ions was measured about every hour, in order to minimize the uncertainty caused by unobserved magnetic-field fluctuations. Furthermore, to exclude systematic shifts, the masses of already well-known neighboring isotopes were measured using the same procedure that was used for the ions of interest.

The ^{74}Rb measurement suffered from a low production yield that limited the number of available ions. In total 556 ^{74}Rb ions were detected during 33 hours of measurement time. In this case, low statistics prevented obtaining accuracy as high as for the two previous cases and reference measurements were taken every six hours.

4 Results

The result of a measurement is the frequency ratio $r_i = \nu_c^{\mathrm{ref}}/\nu_c$ of the cyclotron frequencies of reference ion and ion of interest.

The reference frequency is measured before and after the measurement of the radioactive nuclide. To obtain the reference frequency at the time the radioactive nuclide is measured a linear interpolation is used. The uncertainty of the resulting reference frequency

$$(\delta\nu_{\mathrm{ref}})^2 = (\delta_{\mathrm{int}})^2 + (\delta_B)^2 \tag{3}$$

is given by the interpolation uncertainty δ_{int}, determined by the statistical uncertainty of the two frequency measurements, quadratically added to the uncertainty that is due to unobserved magnetic-field fluctuations δ_B. The uncertainty δ_B depends on the time interval between two reference measurements. For two hours $\delta_B/\nu_{\mathrm{ref}}$ is about $7 \cdot 10^{-9}$ [10,11].

To determine the cyclotron frequency ν_c of the ion of interest it is necessary to exclude possible shifts due to contaminant ions. By investigating the cyclotron frequency in dependence of the number of ions stored simultaneously in the trap a possible presence of ions with another mass in the precision trap can be detected. These contaminant ions would cause a shift of the cyclotron frequency with an increasing number of ions stored simultaneously [12]. Therefore, the cyclotron frequency of the ion of interest is the result of a linear extrapolation to the case of only a single stored ion.

This procedure is repeated for all measurements, *i.e.* for all triplets $[\nu_c^{\mathrm{ref1}}, \nu_c, \nu_c^{\mathrm{ref2}}]$ that yield a ratio r_i. The final ratio r is the weighted mean of all frequency ratios r_i. The final uncertainty

$$(\delta r)^2 = (\delta_{\mathrm{mean}})^2 + (\delta_{\mathrm{sys}})^2 \tag{4}$$

is the uncertainty of the mean δ_{mean} quadratically added to a remaining systematic uncertainty δ_{sys}. This remaining uncertainty is given by the accuracy limit of the ISOLTRAP spectrometer. It was investigated using more than 300 measurements of carbon clusters of different sizes [10,11] and determined to be $\delta_{\mathrm{sys}}/r = 8 \cdot 10^{-9}$ excluding effects due to the mass difference Δm between reference ion and measured ion. This mass difference is causing an uncertainty too low to be significant for the measurements presented here. Using carbon clusters it was measured to be $\delta r/r = 2.0(7)\cdot 10^{-10}\Delta m/\mathrm{u}$ [10,11].

Table 1 summarizes the frequency ratios r and their combined uncertainties for the measured Rb, Kr and Ar ions using the reference ions $^{85}\mathrm{Rb}^+$ and $^{39}\mathrm{K}^+$. The frequency ratio $\nu_\mathrm{c}^\mathrm{ref}/\nu_\mathrm{c}$ is converted into an atomic mass value for the measured nuclide by

$$m = \frac{\nu_\mathrm{c}^\mathrm{ref}}{\nu_\mathrm{c}}\cdot(m_\mathrm{ref} - m_\mathrm{e}) + m_\mathrm{e} \qquad (5)$$

with the electron mass m_e and the atomic mass of the reference nuclide m_ref. The resulting mass excesses are also given in table 1 together with literature values.

The well-known masses are reproduced showing the reliability of ISOLTRAP mass measurements and justifying the revised uncertainty estimations. The clear discrepancies for ^{73,74,75,76}Kr could be resolved by re-evaluating all prior publications used in the mass table in [13] and by recalibrating the input to the atomic mass evaluation. However, the deviation of the ^{78}Kr mass persists and requires further investigations. A more recent set of ISOLTRAP Kr measurements also confirms this deviation.

The new mass value for ^{34}Ar is now accurate enough to permit the calculation of a Q_EC-value with an uncertainty of about 0.01%. Both mother and daughter nuclear masses are now known with a relative uncertainty of about 10^{-8}. The uncertainty of the $\mathcal{F}t$-value is now dominated by the uncertainty of the half-life measurement, soon to be remeasured at Texas A&M [14].

The mass uncertainty of ^{74}Rb is dominated by the number of detected ions, *i.e.* the statistical uncertainty of the cyclotron frequency determination at the chosen resolving power. This results in a relative uncertainty of $2.6\cdot 10^{-7}$ (*i.e.* $\delta m = 18$ keV). The mass of ^{74}Rb was also determined by the MISTRAL experiment, ME $= -51950(100)$ keV [15]. Though not as precise, it is in complete agreement with ours, providing an important independent verification necessary for high-accuracy measurements. Despite the statistical limitation, improved knowledge of the ground-state masses of ^{74}Rb and ^{74}Kr now permits a much more precise β-decay Q_EC-value determination. The Q_EC-value calculated with the presented ground-state masses is $Q_\mathrm{EC} = 10\,425(18)$ keV. Though the uncertainty has been improved by a factor of 30 it is still too large for a decisive test on different δ_C calculations or on CVC itself. For this the uncertainty should be decreased by another factor of 10 to 20. By the implementation of a more efficient transfer and a new detector system (secondary electron multiplier) it should be possible to gain about two orders of magnitude in sensitivity and thus to reach an uncertainty of only a few keV.

5 Summary

We reported on mass measurements of ^{34}Ar, $^{73\text{-}78}$Kr, and ^{74,76}Rb performed with ISOLTRAP. ^{74}Rb is the shortest-lived nuclide ever investigated in a Penning trap ($T_{1/2} = 65$ ms). The relative accuracy of its mass as obtained in this measurement, $\delta m/m = 2.6\cdot 10^{-7}$ (*i.e.* $\delta m = 18$ keV), is governed mainly by statistics.

The conclusion to be drawn from the first mass measurements at this uncertainty level is that the method is well adapted to the low yield and short half-life of ^{74}Rb. The ongoing improvements of the sensitivity of ISOLTRAP should make it possible to reach an uncertainty of a few keV. That will yield masses accurate enough to distinguish between different approaches [3,4] for the calculation of the Coulomb correction δ_C.

The mass of ^{34}Ar was determined with a relative uncertainty of only $1.1\cdot 10^{-8}$. This is the highest accuracy ever obtained in mass measurements of short-lived nuclides. With this result the Q_EC-value for the β-decay of ^{34}Ar is determined with an uncertainty well below 1 keV, sufficient to be used in a decisive test of Coulomb-correction calculations. For the longer-lived Kr isotopes $^{74\text{-}78}$Kr a relative uncertainty in the order of $3\cdot 10^{-8}$ was achieved.

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