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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, $^{73-78}$Kr, and ^{74,76}Rb were performed with the Penning trap mass spectrometer ISOLTRAP. Very accurate Q_{EC} values are needed for the systematic 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 a relative accuracy of $\delta m/m \leq 3 \cdot 10^{-8}$. For most of the measured isotopes presented here this accuracy has been reached. The ^{34}Ar mass has been measured with a relative accuracy of $1.1 \cdot 10^{-8}$. Herewith, the Q_{EC} value of the ^{34}Ar $0^+ \to 0^+$ decay can be determined with an uncertainty of about 0.01%. Furthermore, ^{74}Rb is 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

$$\mathcal{F}t = \frac{K}{2G_V^2(1+\Delta_R^V)} \qquad (1)$$

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 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 nucleaus dependent corrections that are related to the presence of charge-dependent forces in the nucleus. These two 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

$$\mathcal{F}t = ft(1+\delta_R)(1-\delta_C) \qquad (2)$$

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

Until now the parameters of 9 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. That prohibits 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 an uncertainty of 0.1% or better 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 that is better than 0.01%. This requires mass measurements of mother and daughter nuclei with relative uncertainties $\delta m/m \leq 3 \cdot 10^{-8}$.

The very well measured $\mathcal{F}t$ values appear to be constant [1], supporting CVC. However, the extracted V_{ud} together with the much smaller V_{cs} and V_{tb} result in a CKM matrix that is not unitary by 2.2 standard deviations [2]. One important point to be clarified in the characterization of the superallowed β decays is the calculation of the least well know correction, the Coulomb correction δ_C. Today, the 9 well investigated cases have quite low calculated Coulomb corrections in the order of 0.5%. The best cases 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 9 well known emitters. It's study allows the Coulomb corrections in this area to be tested. With ^{74}Rb the Z-dependance of δ_C can be investigated.

In the case of the ^{74}Rb decay, an uncertainty of a few keV would already allow for 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 uses the shell model [3]. The second is based on Hartree-Fock and 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 The 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 that is shown in Fig. 1. It consists of three main parts: (1), a linear gas-filled radiofrequency 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 ultra-high vacuum hyperboloidal Penning trap for the mass measurement. The detailed function and performance of the two Penning traps are described in [6,7]. The recently added linear Paul trap is discussed 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 into the first Penning trap. There the ion cloud is purified and again formed into a cold bunch. 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 $A = 100$. The ion bunch is then delivered to the second trap, which 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 $\nu_c = \frac{1}{2\pi}\frac{q}{m}B$ of an ion with mass m and charge q in a magnetic field of strength B that is determined using a reference mass.

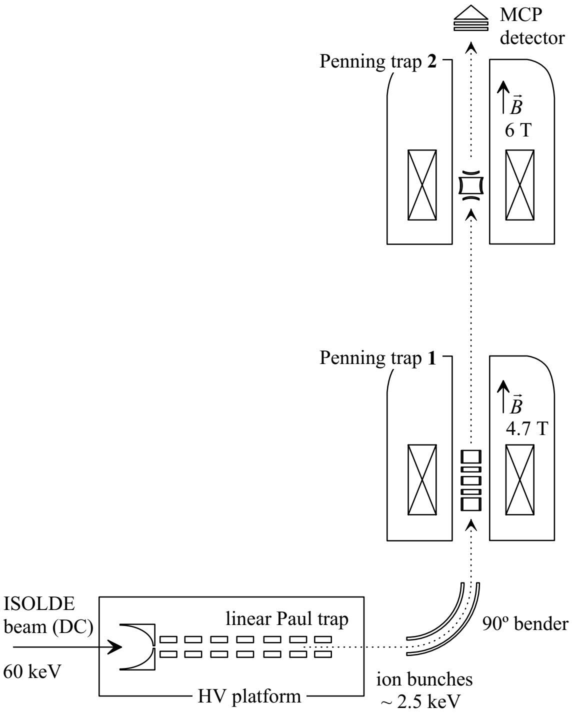

Fig. 1. Experimental setup of the ISOLTRAP Penning trap mass spectrometer. The three main parts are: (1), a linear gas-filled radio-frequency quadrupole (RFQ) trap for retardation of ions, accumulation, cooling and bunched ejection at low energy, (2), a gas-filled cylindrical Penning trap for further cooling and isobaric separation, and (3), an ultra-high-vacuum hyperboloidal Penning trap for the actual mass measurement.

3 The experiment

The radioactive isotopes were produced by bombarding a thick target with 1 or 1.4 GeV proton pulses with a length of about 2.4 μs 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. To produce a clean beam of Ar and Kr ions, CaO and ZrO targets, respectively, were coupled via a water-cooled transfer line to a plasma discharge ion source. The ions were then accelerated to 60 keV and mass separated with a mass resolving power $M/\Delta M \approx 4000$.

The realtively long-lived ($T_{1/2} \geq 1$ s) isotopes of Kr and Ar were measured using excitation times of 300, 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 similiar to the one used for ^{33}Ar ($T_{1/2} = 174$ ms) earlier [9]. In this case the excitation time in the measurement trap was choosen to be between 60 and 120 ms, yielding a mass resolving power $M/\Delta M =$ 82000 and 164000, respectively.

The mass reference measurements used for 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 was used for the Rb and Kr measurements. For ^{34}Ar and ^{74}Kr, the cyclotron frequency of the reference ions has been measured about every hour, to minimize the uncertainty caused mainly by unobserved magnetic field changes. In the case of ^{74}Rb reference measurements were taken about every 6 hours.

To reach the best possible accuracy for the mass values of ^{34}Ar and ^{74}Kr 13 and 7 independent frequency measurements have been performed, respectively. Each preceeded and followed by cyclotron frequency determination of the reference ion. Furthermore, to exclude sytematic shifts, the masses of already well known neighboring isotopes have been measured using the same procedure that was used for the ions of interest.

The ^{74}Rb measurement was characterised by the low production yield that limited the number of ions that could be used for the mass measurement. In total 556 ^{74}Rb ions where detected on the multichannel plate detector after the precision trap within about 33 hours measurement time. This compares to the ISOLDE yield of a few times 10^3 ^{74}Rb ions per proton pulse.

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

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

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

To determine the cyclotron frequency ν_c of the ion of interest it is necessary to exclude possible shifts due to contaminating ions. By investigating the cyclotron frequency in dependance of the number of ions stored at the same time in the trap a possible presence of ions with another mass in the precision trap during the frequency measurement can be detected. These contaminating ions would cause a shift of the cyclotron frequency with an increasing number of ions stored simultaneously [10]. 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

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

is the uncertainty of the mean δ_{mean} quadratically added to a remaining systematic uncertainty δ_{sys}. This remaining uncertainty is basically the limit of the reachable uncertainty using the ISOLTRAP spectrometer in the moment. It was determined using more than 300 measurements of carbon clusters of different sizes 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 to low to be significant for the measurements presented here. Using carbon clusters it was measured to be $\delta r/r = 2.2(0.6) \cdot 10^{-10} \Delta m/\mathrm{u}$. The principles of the carbon cluster measurements and first results can be found in [11] while the final numbers that were already used for the presented evaluation will be published in a more comprehensive way shortly.

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 $^{39}\mathrm{K}^+$, respectively. The frequency ratio $\nu_c^{\mathrm{ref}}/\nu_c$ is converted into an atomic mass value for the measured nuclide by

$$m = \frac{\nu_c^{\mathrm{ref}}}{\nu_c} \cdot (m_{\mathrm{ref}} - m_e) + m_e \quad (5)$$

with the electron mass m_e and the atomic mass of the reference nuclide m_{ref}. The resulting mass excesses are also given in Tab. 1 together with literature values.

The masses that were known with high precision before could be reproduced showing the reliability of ISOLTRAP mass measurements and justify, in a way, the renewed uncertainty estimations. The clear discrepancies for ^{73,74,75,76}Kr could be resolved by re-evaluating all prior publications that formed the results in the mass table in [12] and by recalibrating the input into the atomic mass evaluation. However, the deviation of the ^{78}Kr mass persists and requires further investigations. A new set of measurements on Kr isotopes, very recently taken, supports the first measurements presented here and will be published shortly.

The new and much more accurate 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 nulei masses are know with a relative uncertainty of about $1 \cdot 10^{-8}$. The uncertainty of the $\mathcal{F}t$ value is now dominated by the uncertainty of the half-live measurement. This is going to be tackled soon in an experiment under preparation at Texas A&M [13].

The mass uncertainty of ^{74}Rb is dominated by the number of ions that could be detected during the time allocated to the experiment, i.e. the statistical accuracy of the cyclotron frequency determination at the choosen resolving power. This results in a relative uncertainty of $2.6 \cdot 10^{-7}$ for the mass of ^{74}Rb. Nevertheless, the better knowledge of the ground state masses of ^{74}Rb and ^{74}Kr permits now a much more precise β-decay Q_{EC}-value determination. The Q_{EC} value calculated with the presented ground state masses is $Q_{\mathrm{EC}} = 10\,425 \pm 18$ keV. Though the uncertainty has been improved by a factor of 30 it is still to high for a decisive test on different δ_C calculation 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 it should

Table 1. Frequency ratios relative to ^{85}Rb and ^{39}K for the Rb, Kr and Ar isotopes, respectively, and mass excesses (ME) as determined in this work and literature values from Ref. [12] except for the ^{36}Ar value that comes from Ref. [14].

Nucleus	$T_{1/2}$	freq. ratio $\nu_c^{\rm ref}/\nu_c$	$\rm ME^*_{exp}$ [keV]	$\rm ME_{lit}$ [keV]	$\rm ME^*_{exp}-ME_{lit}$ [keV]
^{74}Rb	65 ms	0.870835567(227)	-51905(18)	-51730(720)	-175
^{76}Rb	36.5 s	0.8942811631(293)	-60480.0(2.4)	-60481.0(8.0)	1.0
^{73}Kr	26 s	0.858999829(114)	-56550.8(9.0)	-56890(140)	339
^{74}Kr	11.5 m	0.8707037618(294)	-62330.3(2.4)	-62170(60)	-160
^{75}Kr	4.5 m	0.882455563(101)	-64323.6(8.0)	-64241(15)	-83
^{76}Kr	14.6 h	0.8941733117(551)	-69010.4(4.4)	-68979(11)	-31
^{77}Kr	1.24 h	0.9059356611(236)	-70169.5(1.9)	-70171.0(9.0)	1.5
^{78}Kr	*stable*	0.9176619688(139)	-74179.2(1.2)	-74160.0(7.0)	-19.2
^{80}Kr	*stable*	0.9411690364(181)	-77891.9(1.5)	-77893.0(4.0)	1.1
^{82}Kr	*stable*	0.9646889211(306)	-80590.8(2.5)	-80588.6(2.6)	-2.2
^{34}Ar	844 ms	0.87209878095(979)	-18377.06(43)	-18378.3(3.0)	1.2
^{36}Ar	*stable*	0.9231027021(115)	-30231.39(49)	-30231.540(27)	0.15
^{38}Ar	*stable*	0.9743097243(102)	-34714.38(46)	-34714.77(49)	0.39
^{41}K	*stable*	1.05128226489(876)	-35558.98(43)	-35558.88(26)	-0.10

*using $m(^{85}\rm Rb) = 84.911\,789\,732(14)\,u$ [15], $m(^{39}\rm K) = 38.963\,706\,86(30)\,u$ [12], and $1\,\rm u = 931.494013\,MeV/c^2$ [16]

be possible to gain a factor of 100 to 150 in sensitivity and thus obtain the desired number of ions within about the same measurement time and reach an uncertainty of only a few keV.

5 Summary

We reported on mass measurements of ^{34}Ar, $^{73-78}$Kr, and ^{74,76}Rb performed with ISOLTRAP. ^{74}Rb is the shortest-lived nuclide ever investigated in a Penning trap ($T_{1/2} = 65\,\rm ms$). The relative accuracy of its mass as obtained in this measurement, $\delta m/m = 2.6 \cdot 10^{-7}$ (i.e. $\delta m = 18\,\rm keV$), 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-live 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 a mass that is accurate enough to distinguish between different aproaches [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 $Q_{\rm EC}$ value for the β decay of ^{34}Ar could be 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 $^{74-78}$Kr a relative uncertainty in the order of $3 \cdot 10^{-8}$ was achieved.

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