Precise Q value determinations for forbidden and low energy β-decays using Penning trap mass spectrometry

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Abstract

Nuclear β-decay provides a laboratory for investigating weak decays occurring inside the nuclear medium. This provides information on the resulting subtle nuclear and atomic effects, and on the underlying interaction and the properties of the particles that are involved, particularly of the neutrino. The Q value of the decay corresponds to the energy equivalent of the mass difference between parent and daughter atoms, and can be precisely and accurately measured using Penning trap mass spectrometry. In this paper we discuss Penning trap Q value measurements for forbidden β-decays of long-lived primordial nuclides, and for a subset of β-unstable nuclides that could potentially undergo a very low energy decay to an excited state in the daughter nucleus. We discuss applications of these measurements to tests of systematics in detectors that perform precise β-spectrum measurements, as inputs for theoretical shape factor, electron branching ratio and half-life calculations, and to identify nuclides that could serve as new candidates in direct neutrino mass determination experiments.

Keywords: Penning trap, atomic mass, forbidden β-decay, low energy β-decay

1 Introduction

Nuclear β-decay studies have a history that is tied to our evolving understanding of subatomic physics, and in the development of the standard model. For example, early β-spectrum measurements led Pauli to postulate the existence of the neutrino, see e.g. Ref. [1], and Fermi to develop the first theory of nuclear β-decay [2–4]; and the proposal of parity violation in β-decay by Lee and Yang [5], and the subsequent experimental confirmation by Wu [6] paved the way for the development of the electroweak theory [7–9].

Experimental β-decay experiments continue to be relevant to fundamental nuclear and particle physics: studies of superallowed $0^+ \rightarrow 0^+$ β-decays [10] and superallowed $T = 1/2$ mirror decays [11, 12], enable a precise determination of the V_{ud} element of the Cabibbo-Kobayashi-Maskawa (CKM) matrix, and a test of its unitarity; precise β-ν correlation measurements provide searches for beyond the standard model scalar and tensor currents, see e.g. Ref. [13, 14]; and precise β-spectrum shape measurements along with theoretical calculations could be used to determine the effective value of the axial vector coupling constant, g_A, via the so-called *spectral shape method* (SSM) [15–18]. In the neutrino sector, precise β-spectroscopy measurements with tritium [19–21] and ^{187}Re [22], and electron capture (EC) spectroscopy measurements with ^{163}Ho [23–25], set upper limits on the mass of the electron anti-neutrino/neutrino; experiments such as BeEST [26, 27] and HUNTER [28] aim to perform precise measurements of the nuclear recoil energy after EC of ^{7}Be and ^{131}Cs, respectively, to search for the signature of sterile neutrinos; and a number of experimental efforts world-wide are developing tonne-scale experiments to search for neutrinoless double β-decay ($0\nu\beta\beta$), see e.g. [29, 30]. Furthermore, rare decays can create background events in sensitive detectors used for $0\nu\beta\beta$ and dark matter searches, and in solar and geo-neutrino experiments, so must be well understood [31–34].

1.1 β-decay definitions

Perhaps the two most fundamental properties of a nuclear β-decay transition are the lifetime of the parent nuclide and the Q value of the decay. Both properties depend on the initial and final nuclear and (usually to a lesser extent) atomic states, and are experimental observables—the Q value essentially corresponds to the endpoint of the energy spectrum in β^- decay, and is related to the discrete energy taken away by the neutrino or recoiling nucleus in EC decay. The lifetime can also be determined theoretically via calculations of the phase space factor, which depends on the Q value, and the relevant nuclear matrix elements, see e.g. Ref. [35]. A precise and independent determination of the Q value can then serve two main purposes: (1) to use in the phase space factor calculations to facilitate a comparison of calculated and measured lifetimes and provide an important test of the theory, and (2) to compare with the experimentally determined decay energy to provide a test of systematics in the detector.

For β^-, EC, and β^+ transitions between parent, X, and daughter, Y, nuclides[1],

$$ {}^{A}_{Z}\mathrm{X}_N \rightarrow {}^{A}_{Z+1}\mathrm{Y}^{+}_{N-1} + e^- + \bar{\nu}_e, \tag{1} $$

$$ {}^{A}_{Z}\mathrm{X}_N + e^- \rightarrow {}^{A}_{Z-1}\mathrm{Y}_{N+1} + \nu_e, \tag{2} $$

$$ {}^{A}_{Z}\mathrm{X}_N \rightarrow {}^{A}_{Z-1}\mathrm{Y}^{-}_{N+1} + e^+ + \nu_e, \tag{3} $$

the decay energy—the *Q-value*—is the energy released as kinetic energy of the final state particles, and can be defined as the energy equivalent of the mass difference between initial and final states [35]:

$$ Q_{\beta^-} = (M_\mathrm{X} - M_\mathrm{Y})c^2, \tag{4} $$

$$ Q_{EC} = (M_\mathrm{X} - M_\mathrm{Y})c^2, \tag{5} $$

$$ Q_{\beta^+} = Q_{EC} - 2m_e c^2. \tag{6} $$

These definitions give the ground-state to ground-state (gs-gs) Q values, Q_{GS}, and can hence be determined directly from the atomic masses of parent and daughter nuclides.[2]

1.2 Q value measurements with a Penning trap

The most precise and accurate method for determining atomic masses for most nuclides is via Penning trap mass spectrometry (PTMS) [36–38]. The fundamental quantity of interest in a PTMS measurement is the cyclotron frequency of an ion of mass-to-charge ratio m/q in a magnetic field of strength, B:

$$ f_c = \frac{qB}{2\pi m}. \tag{7} $$

The magnetic field is typically produced by a superconducting solenoidal magnet, and provides radial confinement. A quadratic electrostatic potential axially confines the ions to a small volume where the magnetic field is uniform, and so that the measurement can be performed. The electrostatic potential is produced by a set of three main electrodes—a ring and two end-caps, along with correction electrodes to compensate for deviations from a purely quadratic field. This combination of magnetic and electric fields results in three normal modes of motion for the ion in the Penning trap: a harmonic axial motion

[1] It is understood that the parent and daughter nuclides, X and Y, in Eqn. (1) are not the same as those in Eqns. (2) and (3) (although it is possible for the two to have a common parent or a common daughter nuclide)

[2] These Q value definitions ignore the binding energies of the missing/additional electron in β^-/β^+ decay, which is ~eV and is typically negligible, and they ignore the binding energy of the captured, typically K or L shell electron, in EC decay, which is ~keV and must be accounted for in some cases.

at frequency, f_z, and two harmonic radial motions (trap-cyclotron and magnetron) with frequencies f_+ and f_-. The frequency of the trap-cyclotron mode, f_+, is reduced compared to the free-space cyclotron frequency, f_c, of Eqn. (7). However, it is possible to recover the free-space cyclotron frequency by combining the measurable normal mode frequencies of an ion in the trap. The two relationships relevant to PTMS experiments are [39, 40]

$$f_c^2 = f_+^2 + f_z^2 + f_-^2, \tag{8}$$

and

$$f_c = f_+ + f_-. \tag{9}$$

Over the last four or five decades, a number of different techniques have been developed to either (i) measure the normal mode trap frequencies directly, so that f_c can be computed from Eqn. (8) or (9), or (ii) probe the frequency combination $f_+ + f_-$ indirectly so that f_c can be deduced from Eqn. (9). Two main classes of measurement techniques exist: non-destructive image charge (IC) detection inside the trap, and destructive ion detection outside of the trap, typically with a microchannel plate (MCP) detector.

For details on IC detection for precise mass measurements we refer the reader to early work by the groups at University of Washington [41, 42], Harvard [43], and MIT [44, 45] (now at FSU [46]), and more recent implementations by the PENTATRAP group in Heidelberg [47], the LIONTRAP group in Mainz [48], and the BASE collaboration at CERN [49]. For details on destructive detection techniques, we refer the reader to descriptions of the traditional time-of-flight ion-cyclotron-resonance (TOF-ICR) technique [50, 51], modifications that include the so-called Ramsey excitation scheme [52–54] and an octupole excitation scheme [55–57], and the more recently developed phase imaging ion cyclotron resonance (PI-ICR) technique [58, 59].

The goal of PTMS is to determine the mass of an ion via a measurement of f_c. However, in order to do this, one must also have precise knowledge of the magnetic field, B. Since it is possible to measure f_c much more precisely than it is to measure B, the experimental procedure is to cancel out the magnetic field in Eqn. (7) by measuring cyclotron frequency ratios of pairs of ions i.e.

$$R = \frac{f_{c0}}{f_{c1}} = \frac{m_1/q_1}{m_0/q_0}. \tag{10}$$

Typically, one of the ions is the ion of interest whose mass is to be determined, and the other is a reference ion with well-known mass[3]. However, Eqn. (10) can also be rearranged to obtain the mass difference between neutral atoms, which is the required quantity for determining the Q value. Thus, if m_0 and

[3]Ideally, this reference mass would be $^{12}C^+$, since the atomic mass unit can be defined precisely in terms of the mass of ^{12}C, but in practice it is better to measure the cyclotron frequency ratio of ions of similar mass to reduce the effect of potential mass-dependent systematic frequency shifts on the ratio.

m_1 are atomic ions of the parent and daughter nuclides, with masses M_P and M_D, respectively, and if we assume singly-charged ions so that $q_0 = q_1 = 1$, then we can obtain:

$$Q_{\beta^-/EC} = (M_{\rm P} - M_{\rm D})c^2 = (M_{\rm P} - m_e)(1 - R)c^2, \tag{11}$$

where m_e is the mass of the electron, and it is understood that M_P and M_D are the relevant parent and daughter masses for either β^- or EC decay. Here we have ignored the ionization energies, since only their difference, typically $\sim$eV, would enter into Eqn. (11), which is usually negligible compared to statistical uncertainty in the measurement.

In this paper, we discuss measurements that have been performed with the MIT-FSU trap at Florida State University [46, 60], LEBIT at the National Superconducting Cyclotron Laboratory/Facility for Rare Isotope Beams (NSCL/FRIB) [61], JYFLTRAP at the University of Jyväskyä [62], SHIPTRAP at GSI [63], the Canadian Penning Trap (CPT) at Argonne National Laboratory (ANL) [64, 65], ISOLTRAP at ISOLDE/CERN [66], TITAN at TRIUMF [67], and SMILETRAP (formerly operated at Stockholm University) [68].

2 Forbidden β-decays of primordial nuclides

On Earth, there exists some 286 primordial nuclides, defined as those nuclides that are stable or have half-lives longer than, or comparable to, the age of the Earth. Of these primordial nuclides, 34 have been observed to decay: thirteen undergo α-decay (^{144}Nd , ^{147}Sm, ^{148}Sm, ^{151}Eu, ^{152}Gd, ^{174}Hf, ^{180}W, ^{186}Os, ^{190}Pt, ^{209}Bi, ^{232}Th, ^{235}U, ^{238}U), eight undergo single β-decay or electron capture (EC) decay (^{40}K, ^{50}V, ^{87}Rb, ^{113}Cd, ^{115}In, ^{138}La, ^{176}Lu, ^{187}Re), see e.g. Ref. [69], and fourteen undergo double β-decay (^{48}Ca, ^{76}Ge, ^{78}Kr, ^{82}Se, ^{96}Zr, ^{100}Mo, ^{116}Cd, ^{124}Xe, ^{128}Te, ^{130}Te, ^{130}Ba, ^{136}Xe, ^{150}Nd, ^{238}U). The double β-decay nuclides include those that have been observed to undergo $2\beta^-$ [70] and 2EC decay [71]. $2\beta^+$ and mixed β^+EC decays are also possible, but have not been observed to date. In fact, there are 35 primordial nuclides for which $2\beta^-$ decay is energetically allowed, and 34 for which 2EC decay is energetically allowed (with a subset of 24 and 6 that could also undergo β^+EC decay, and $2\beta^+$ decay, respectively) [72], but for which single β-decay or EC is energetically forbidden or highly suppressed. The extremely long half-lives of these double β-decays make their experimental confirmation extremely challenging. Precise Q value measurements for double β-decay are discussed by Eliseev in this issue. Here we focus on single β-decay.

In addition to the eight nuclides mentioned above for which $\beta^\pm$ or EC decay has been observed, there are four additional primordial nuclides for which β-decay is energetically allowed: ^{48}Ca, ^{96}Zr, ^{123}Te, and ^{180m}Ta [69]. All of the β-decay nuclides are listed in Table 1 along with their half-life, forbiddenness, and Q value. Penning trap measurements of Q values for these nuclides and their relevance are discussed below.

Table 1 Long-lived primordial β-unstable nuclides. The spin and parity (J^{π}) of the parent state and daughter final state are shown in parenthesis and are used to determine the forbiddenness (forbidden unique, FU, or forbidden non-unique, FNU) of the predominant decay. Measured half-life values or lower limits are taken from Ref. [69], except for ^{176}Lu EC. The ground-state to ground-state Q value is taken from the AME2020 [73] unless otherwise noted, and the reference for the Penning trap measurement of the Q value and/or parent/daughter mass, if it has been performed, is listed in the last column.

Decay	Type	Partial half-life (yr)	Q value (keV)	Q Ref
^{40}K(4$^-$) → ^{40}Ca(0$^+$)	3rdFU (β^-)	$1.398(3) \times 10^{9}$	1310.905(60)	[74][a]
^{40}K(4$^-$) → ^{40}Ar(2$^+$)	1stFU (EC)	$1.164(3) \times 10^{10}$	1504.403(56)	[75][a]
^{40}K(4$^-$) → ^{40}Ar(0$^+$)	3rdFU (β^+)	$1.248(3) \times 10^{13}$	482.405(56)	[75][a]
^{48}Ca(0$^+$) → ^{48}Sc(5$^+$)	4thFU (β^-)	$>2.5 \times 10^{20}$	279.2(4.9)	[76][b]
^{50}V(6$^+$) → ^{50}Ti(2$^+$)	4thFU (EC)	$2.67^{+0.16}_{-0.18} \times 10^{17}$	2208.627(58)	[77]
^{50}V(6$^+$) → ^{50}Cr(2$^+$)	4thFU (β^-)	$>1.9 \times 10^{19}$	1038.124(55)	[77]
^{87}Rb(3/2$^-$) → ^{87}Sr(9/2$^+$)	3rdFNU (β^-)	$4.97(3) \times 10^{10}$	282.275(8)	[78, 79]
^{96}Zr(0$^+$) → ^{96}Nb(5$^+$)	4thFU (β^-)	$>3.8 \times 10^{19}$	163.97(10)	[80]
^{113}Cd(1/2$^+$) → ^{113}In(9/2$^+$)	4thFNU (β^-)	$8.04(5) \times 10^{15}$	323.84(27)	[81]
^{115}In(9/2$^+$) → ^{115}Sn(1/2$^+$)	4thFNU (β^-)	$4.41(25) \times 10^{14}$	497.489(10)	[82, 83]
^{123}Te(1/2$^+$) → ^{123}Sb(7/2$^+$)	2ndFU (EC)	$>9.2 \times 10^{16}$	51.913(66)	[84]
^{138}La(5$^+$) → ^{138}Ce(2$^+$)	2ndFU (β^-)	$2.97(3) \times 10^{11}$	1052.46(40)	[85]
^{138}La(5$^+$) → ^{138}Ba(2$^+$)	2ndFU (EC)	$1.55(2) \times 10^{11}$	1748.40(34)	[85]
^{176}Lu(7$^-$) → ^{176}Hf(6$^+$)	1stFNU (β^-)	$3.684(18) \times 10^{10}$	1193.03(55)	[86]
^{176}Lu(7$^-$) → ^{176}Yb(2$^+$)	5thFNU (EC)	$>1 \times 10^{18}$	108.90(73)	[86][c]
^{180m}Ta(9$^-$) → ^{180}W(6$^+$)	3rdFNU (β^-)	$>1.7 \times 10^{17}$	90.9(2.4)	[87][a,d]
^{180m}Ta(9$^-$) → ^{180}Hf(6$^+$)	3rdFNU (EC)	$>2.0 \times 10^{17}$	281.9(2.4)	[d]
^{187}Re(5/2$^+$) → ^{187}Os(1/2$^-$)	1stFU (β^-)	$4.33(7) \times 10^{10}$	2.497(34)	[90][e]

[a]Only the mass of the daughter has been measured via PTMS.
[b]Only the mass of the parent has been measured via PTMS.
[c]^{176}Lu EC partial half-life lower limit estimated from the EC/β^- ratio upper limit from Ref. [86].
[d]The ^{180}W – ^{180}Hf 2EC Q value has been measured via PTMS [87], but the ^{180}Hf mass is predominantly determined from an (n, γ) reaction [88]. Q values are for ^{180m}Ta to ^{180}Hf or ^{180}W ground states with the ^{180m}Ta energy from the PTMS measurement of Ref. [89].
[e]The listed Q value is taken directly from Ref. [90].

^{50}V, ^{113}Cd and ^{115}In—These three nuclides share the property that they are the only known fourth-forbidden β-decays. Decay schemes for these three nuclides are shown in Fig. 1. ^{50}V is unique among the three in that it is unstable against both β^- and EC decay and also that decay to the ground-state of the daughter is even more highly forbidden than the decay to the 2$^+$ excited

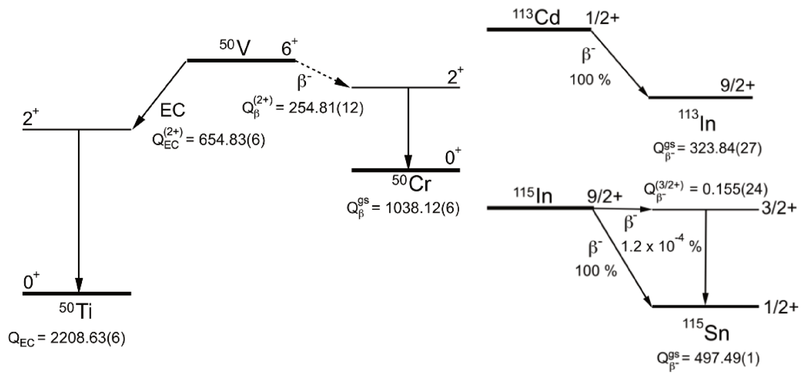

Fig. 1 Decay schemes for ^{50}V (left), ^{113}Cd (top right), and ^{115}In (bottom right). $Q^{gs}_{\beta/EC}$ is the ground-state to ground-state Q value listed in Table 1, and $Q^{J^{\pi}}_{\beta/EC}$ the Q value to the excited daughter state in keV. Percentages give branching ratios, and dotted arrows indicate decays that have not yet been observed.

states. The EC decay of ^{50}V has been observed, with recent experiments providing half-life measurements of $2.77^{+0.20}_{-0.19}$ $\times 10^{17}$ yr [91] and $2.67^{+0.16}_{-0.18}$ $\times 10^{17}$ yr [92]. The β^--decay of ^{50}V has not been observed, but lower limits of 8.9 $\times$ 10^{18} yr [91] and 1.9 $\times$ 10^{19} yr [92] have been established. These limits are now becoming comparable to theoretical predictions [93]. The β^- and EC decay Q values for ^{50}V (along with the 2EC Q value for ^{50}Cr) were measured at LEBIT [77], providing precise and accurate data for phase space factor calculations that enter into the theoretical calculations of the partial half-life.

The half-life and spectrum shape for ^{113}Cd β-decay have been particularly well characterized due to the presence of ^{113}Cd in detectors that were developed for $0\nu\beta\beta$ decay searches in Cd and Te nuclides, such as CdTe [94] and CdZnTe [95, 96] semiconductor detectors, $CdWO_4$ low temperature bolometers [97], and $CdWO_4$ scintillator detectors [98, 99]. These studies enable high-precision comparisons of experimental data and theoretical calculations for very rare decays. Moreover, ^{113}Cd was identified as a candidate for applying the spectral shape method (SSM) to extract information on the axial vector coupling constant, g_A [15, 16]. A recent analysis of ^{113}Cd β-spectra taken with CdZnTe semiconductor detectors as part of the COBRA experiment revealed g_A quenching effects [100, 101]. A measurement of the ^{113}Cd Q value was performed at LEBIT [81] and provided a Q value that was a factor of 3.5 more precise than the value obtained from the end-point of the ^{113}Cd β-spectrum from COBRA [96], which agreed with the LEBIT measurement at the 1.7σ level. Hence, the independent Q value measurement provides a test of systematics in these detectors and accurate input data for theoretical calculations.

After two early measurements of the ^{115}In β-spectrum using indium-loaded liquid scintillators in 1961 [102] and 1979 [103], ^{115}In β-decay received little attention. However, in the last couple of decades, theoretical interest has returned [104, 105] and the development of new scintillating bolometer crystals containing indium (developed for $0\nu\beta\beta$-decay searches) have enabled a new measurement of the ^{115}In β-spectrum [106]. Further interest in ^{115}In has been spurred on by the discovery of a weak branching ratio "ultra-low energy" β-decay to the first excited state in ^{115}Sn [107] (see Section 3), and by the possibility of applying the SSM [15, 16] to the ^{115}In β-spectrum. The development of cryogenic In_2O_3 calorimeters is being pursued for this purpose [108]. The ^{115}In β-decay Q value was measured with the FSU-MIT trap [82] and with JYFLTRAP [83]. This Q value has a particular relevance to the ultra-low energy decay branch, but will also provide a test of systematics in these new detectors and an input for theoretical calculations.

^{138}La and ^{176}Lu—Recent developments in detector technology has seen the introduction of high energy resolution lanthanum and lutetium containing scintillator crystals, such as $LaBr_3$, $LaCl_3$, LuAG:Pr, and LSO:Ce. Due to the intrinsic radiation from ^{138}La and ^{176}Lu present in these detectors, they provide the opportunity to perform precise measurements on the β^- and/or EC decay of these nuclides. For both nuclides the β^- and/or EC decays go to an excited state in the daughter that then decays to the ground state. Relevant decay schemes are shown in Fig. 2. The subsequent γ-rays can then be used to gate on different decay modes. Such measurements have been performed by Quarati and collaborators [86, 109, 110] and the β^- and EC Q values were measured at LEBIT [85, 86]. Precise β-spectra were obtained for ^{138}La and ^{176}Lu. In both cases, the end-point energies extracted from the β-spectra were in excellent agreement with the Q value obtained via PTMS. By using the Penning trap Q value in the theoretical fit to the ^{138}La β-spectrum, precise values for the experimental shape factor could be extracted. Furthermore, the new EC Q value measurement for ^{138}La enabled precise calculations of the EC probability ratios for K, L, and M shell capture. The ratios calculated using the new Q value showed improved agreement with the experimental results obtained with the $LaBr_3$ scintillators [110].

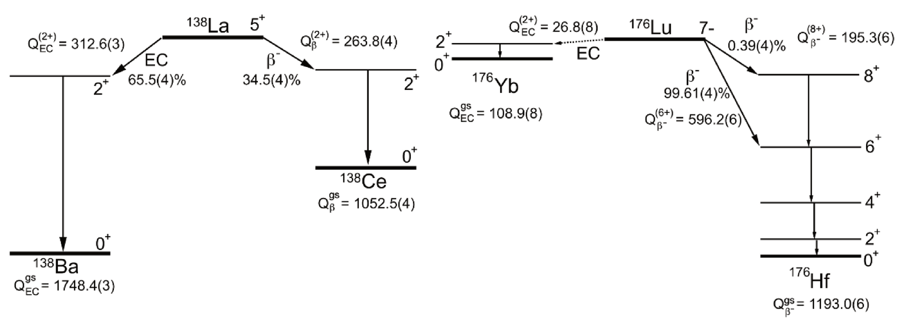

Fig. 2 Decay schemes for ^{138}La (left) and ^{176}Lu (right). $Q^{gs}_{\beta/EC}$ is the ground-state to ground-state Q value listed in Table 1, and $Q^{J^\pi}_{\beta/EC}$ the Q value to the excited daughter state in keV. Percentages give branching ratios, and dotted arrows indicate decays that have not been observed.

^{187}***Re***—^{187}Re is well-known as having the lowest energy ground-state to ground-state β-decay Q value (see decay scheme in Fig. 3, which has made it historically important for direct neutrino mass determination experiments [22]. A number of experiments have investigated ^{187}Re β-decay with two main classes of detectors: proportional counters [111–113] and mircocalorimeters [22, 114–116]. With the exception of Ref. [114], the Q value obtained from the two classes of data showed disagreement, with a difference of 180(39) eV. This discrepancy was resolved with a precise Q value measurement by the SHIPTRAP group [90] that agreed with the microcalorimeter Q value results. The Q value also provides an input for calculations to enable a precise test of β-decay theory, for example environmental effects that can give rise to fine structure in the β-spectrum [117].

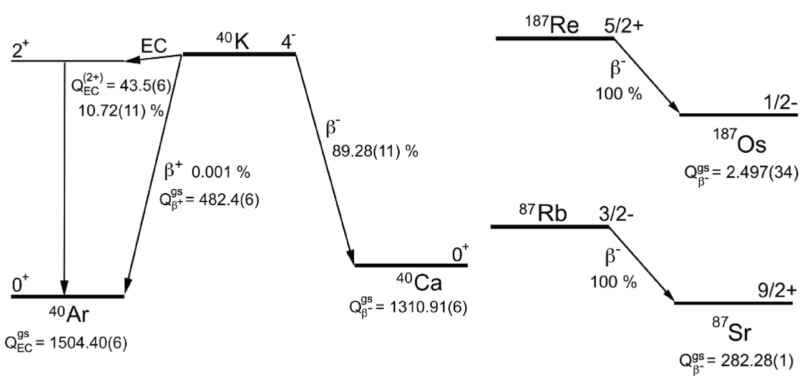

Fig. 3 Decay schemes for ^{40}K (left), ^{187}Re (top right), and ^{87}Rb (bottom right). $Q^{gs}_{\beta/EC}$ is the ground-state to ground-state Q value listed in Table 1, and $Q^{J^\pi}_{\beta/EC}$ the Q value to the excited daughter state in keV. Percentages give branching ratios.

^{40}***K and*** ^{87}***Rb***—^{40}K is the most short-lived of the primordial nuclides, and also has the unique property that it can decay via β^-, β^+, and EC decay. Decay schemes for ^{40}K and ^{87}Rb are given in Fig. 3. Both ^{40}K and ^{87}Rb are important isotopes for geochronological applications, and recent measurements have used liquid scintillation counters to perform spectral shape measurements of their β-spectra [118]. Discrepancies exist in the end-point energies that are extracted from these methods, so precise independent Q value determinations provide an unambiguous reference that can be used to check the accuracy of the β-spectrum measurement and the consequent extraction of the shape factor—experimental and/or theoretical knowledge of β-shapes can have important applications for accurate dose calculations for β-emitters used in medical treatments, and data for calculations in the nuclear power industry and in nuclear waste management [119, 120].

A dedicated direct PTMS measurement of the ^{40}K and ^{87}Rb Q values has not been made. However, the atomic masses of ^{87}Rb and daughter ^{87}Sr have both been measured very precisely with the FSU-MIT Penning trap [78, 79], yielding a Q value with a precision of 8 eV. The atomic masses of the β^- and EC daughters of ^{40}K have been measured precisely with SMILETRAP (^{40}Ca) [74] and the FSU-MIT trap (^{40}Ar) [75]. The mass of ^{40}K, on the other hand, is determined via (n,γ) reactions [121, 122] that link it to the precisely measured masses of ^{39,41}K [78]. Q values obtained from the atomic masses are currently significantly more precise than those obtained from the β-spectrum measurements, but a direct Q value measurement for ^{40}K could be relevant in the future if more precise β-spectrum measurements are developed.

^{123}Te and ^{180m}Ta—Aside from ^{48}Ca and ^{96}Zr (discussed below), that do in fact decay via 2β-decay, ^{123}Te and ^{180m}Ta are the only nuclides listed in Table 1 that have not been observed to undergo β-decay. Both nuclides have some unique characteristics: ^{123}Te is the only primordial nuclide that would decay purely by EC, ^{180m}Ta is the only known nuclide to occur naturally in an isomeric state, and is also the only known isomer that has not been observed to decay [69]. Decay schemes are shown in Fig. 4.

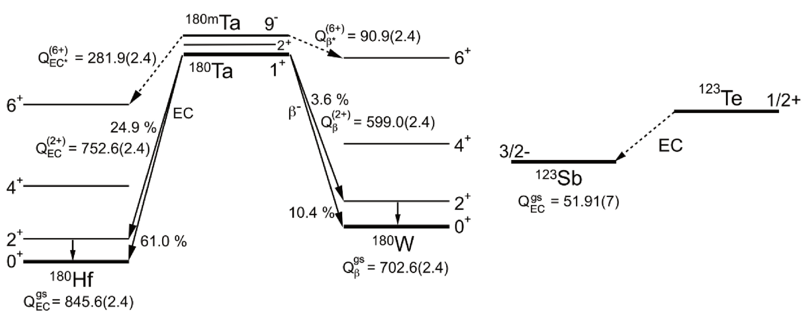

Fig. 4 Decay schemes for ^{180}Ta (left), and ^{123}Te (right). $Q^{gs}_{\beta/EC}$ is Q value listed in Table 1 (ground state to ground state for ^{123}Te and isomer to ground state for ^{180m}Ta), and $Q^{J^\pi}_{\beta/EC}$ the Q value to the excited daughter state in keV. Percentages give branching ratios. Dotted arrows indicate decays that have not yet been observed.

In the case of ^{123}Te, a number of experimental searches for its EC decay have been performed over the years, including claims of an observation that were later shown to be incorrect, see Refs. [123, 124] and references therein. The current best lower limits on the half-life are 9.2×10^{16} yr [124] and 3.2 $\times$ 10^{16} yr[125]. Continued development of detectors for $0\nu\beta\beta$-decay searches with tellurium isotopes are expected to also enable improved searches for ^{123}Te EC. A precise Q value measurement was performed by the SHIPTRAP group in 2016 [84], that eliminated uncertainty from the Q value in evaluating the ^{123}Te EC decay. It also revealed changes in the EC probability for ^{123}Te ions in astrophysical conditions.

^{180m}Ta could potentially undergo either β^- or EC decay to 6^+ states in ^{180}W or ^{180}Hf, respectively. The EC decay is expected to dominate with a calculated half-life of 1.4×10^{20} yr, compared to 5.4×10^{23} yr for β^- decay [126].

A number of experimental searches for the decay of ^{180m}Ta have been performed over the last four decades by looking for γ-rays emitted after β^- or EC decay to the excited daughter states. The most recent measurement using high purity Ge detectors provided the most stringent lower limits of 2.0×10^{17} yr and 1.7×10^{17} yr for the EC and β^- modes, respectively [127]. Penning trap measurements of the mass difference between the daughter nuclides ^{180}W – ^{180}Hf, was performed in the context of determining the ^{180}W 2EC Q value [87], and the mass difference between the ^{180}Ta(1^+) ground state and isomeric state was performed to directly determine the excitation energy of the isomer [89]. However, none of the nuclides in this triplet has been measured directly via PTMS with respect to a secondary reference mass. A direct determination of the ^{180m}Ta β^- and EC Q values could reduce uncertainty in the calculated half lives due to uncertainty in the Q value.

^{48}Ca and ^{96}Zr—These nuclides are unstable against both β-decay and 2β-decay (see the decay schemes in Fig. 5), and while 2β-decay has been observed for both nuclides [128–132], single β-decay has not [133–137]. In both cases, the 0^+ ground state could decay to a 4^+, 5^+, or 6^+ state in the daughter, with the most likely branch being a 4$^{\text{th}}$ forbidden unique decay to the 5^+ state [138]. The high forbiddenness of these decays, along with the low Q values result in calculated β-decay half-lives [80, 138–142] that are indeed longer than the measured 2β-decay half lives [70].

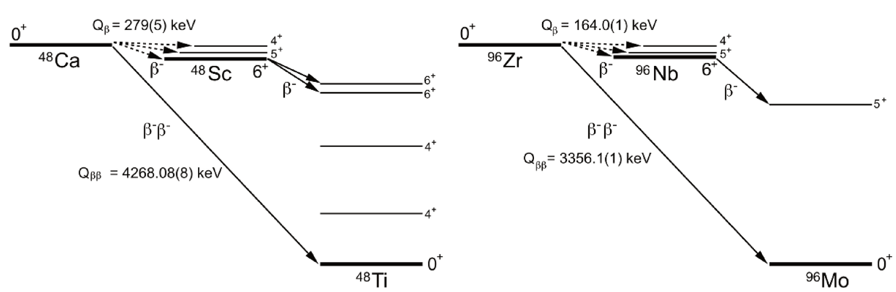

Fig. 5 Decay schemes for ^{48}Ca (left) and ^{96}Zr (right). Q_β^{gs} is the ground-state to ground-state Q value listed in Table 1, and $Q_\beta^{J^\pi}$ the Q value to the excited daughter state in keV. $Q_{\beta\beta}$ is the double β-decay Q value. Dotted arrows indicate decays that have not yet been observed.

The 2β-decay Q values have been measured for ^{48}Ca at LEBIT [143, 144] and TITAN [145], and for ^{96}Zr at LEBIT [146] and JYFLTRAP [80]. The JYFLTRAP group also measured the ^{96}Zr single β-decay Q value [80], which has been important for reducing the uncertainty on the calculated branching ratio and half-life of ^{96}Zr single β-decay. A high precision Penning trap measurement of the mass of ^{48}Ca by the SHIPTRAP group in the context of QED tests in highly-charged ions is available [76]. However, the mass of ^{48}Sc is only known via ^{48}Ca$(p,n)^{48}$Sc reaction data [147, 148] and a measurement of the ^{48}Sc β-spectrum end-point energy [149] to a precision of 5 keV/c^2, making this the least precise Q value for the β-unstable primordial nuclides of Table 1. This Q value limits the uncertainty in the calculated branching ratio of ^{48}Ca single β-decay, which is 7.5(2.8) % [138]. This could impact future plans for experimental searches for ^{48}Ca single β-decay. Hence, a more precise ^{48}Sc mass is called for.

3 Ultra-low Q value β-decays

Beyond the stable and long-lived primordial nuclides, the nuclear landscape is home to a wide range of radionuclides, the large majority of which undergo $\beta^{\pm}$ or electron capture decay. The half-lives of these nuclides span many orders of magnitude, from ms for nuclides close to the drip line, to $\sim$30 million years for nuclides such as ^{92}Nb, which sits along the valley of stability. Similarly the β-decay Q values of these nuclides cover several orders of magnitude from $\sim$1 keV to $\sim$10 MeV.

The nuclides with the lowest ground-state to ground-state decay energies are ^{3}H, ^{187}Re, and ^{163}Ho with Q_{GS} values of 18.59201(7) keV [150], 2.492(34) keV [90], and 2.833(34) keV [151], respectively. These low Q value nuclides are of particular interest for direct neutrino mass determination experiments, since the number of decay events in an energy interval ΔE near the end-point goes as $(\Delta E/Q)^3$ [152] for β-decay, and as $(\Delta E/Q)^2$ for EC decay [153] (but this rate can be enhanced when the binding energy of the captured orbital electron is close to that of the Q value). β-decays often proceed through an excited state in the daughter, with energy E^*, reducing the Q value of the transition to the excited state:

$$Q_{ES} = Q_{GS} - E^* \tag{12}$$

If the β-decay occurs to an excited state with energy E^*, that is similar to the ground-state to ground-state Q value, Q_{GS}, then Q_{ES} can be very small. If $Q_{ES} \lesssim 1$ keV, then it is termed an “ultra-low” Q value decay [154].

3.1 ^{115}In$(9/2^+) \rightarrow {}^{115}Sn(3/2^+)$: the lowest known Q value β-decay

To date, only one ultra-low Q value transition has been observed, that of the ^{115}In$(9/2^+)$ ground-state to the ^{115}Sn$(3/2^+)$ first excited state at 497 keV [107] (see Fig. 1). This decay was discovered serendipitously during developmental stages of the LENS (Low Energy Neutrino Spectrometer) project that aims to detect low-energy solar neutrinos via the inverse electron capture process ^{115}In + $\nu_e \rightarrow {}^{115}$Sn(613 keV) + e^- [155]. Cattadori *et al.* observed a γ-ray at 497 keV that they inferred came from the γ-transition of ^{115}Sn$(3/2^+)$ to the ground state after population via β-decay of ^{115}In, with a deduced branching ratio of $1.2(3) \times 10^{-4}$ %. However, mass data from the Atomic Mass Evaluation available at that time (AME2003), provided a Q value of 2(4) keV [156]. Therefore, it was not known whether the suggested decay branch to the ^{115}Sn($3/2^+$) was energetically allowed.

This situation was resolved in 2009 by two independent Penning trap measurements of the ^{115}In – ^{115}Sn mass difference by groups at Florida State University [82] and Jyväskylä [83] that determined Q_{GS} values of 497.489(10) keV and 497.68(17) keV, respectively. Accounting for the 497.334(22) keV energy of the ^{115}Sn($3/2^+$) state [157], Q_{ES} values of 155(24) eV and 350(170) eV were determined by the two groups for the decay to the excited state. The two results agree at the 1σ level, and confirm that ^{115}In($9/2^+$) $\rightarrow$ ^{115}Sn($3/2^+$) β-decay is the lowest known Q value nuclear decay. The Jyväskylä group also confirmed the original observation of Cattadori *et al.* and reduced the uncertainty on the partial half-life and branching ratio of this decay branch via ultra-low background γ-ray spectroscopy at the HADES underground laboratory [83, 158]. From this work, the partial half-life was determined to be 4.5(5) $\times$ 10^{20} yr, and the branching ratio 1.02(13) $\times$ 10^{-4} %.

Theoretical studies of the ^{115}In ultra-low Q value decay by Suhonen and collaborators determined a partial half-life that was $1 - 2$ orders of magnitude smaller than the measured value [83, 154, 159]. A particular challenge is the calculation of the phase space factor for this second forbidden unique decay with ultra-low Q value. Effects that could significantly influence the phase space factor, and hence calculated half-life, include electron screening corrections, mismatch between initial and final atomic states, exchange effects, and final state interactions. Some of these atomic effects had been studied for low energy allowed β-decay, but have not been extended into the ultra-low energy regime and not for forbidden decays [160–162].

Experimental interest in this decay has continued in recent years with new measurements of the ^{115}Sn($3/2^+$) first excited state energy performed in 2016 and 2018 with the results $E^* = 497.316(7)$ keV [163] and 497.342(3) keV [164], respectively. The former experiment populated the ^{115}Sn($3/2^+$) state via the ^{114}Sn$(n,\gamma)^{115}$Sn reaction, while the latter populated the state of interest via β-decay of ^{115}Sb obtained via the ^{115}Sn$(p,n)^{115}$Sb reaction. While both results agree with the previously accepted value within uncertainty, they differ from each other by 3.4σ. Further work is needed in order to resolve this discrepancy.

The potential use of the ^{115}In ultra-low β-decay for neutrino mass determination was noted in Ref. [107] and discussed in further detail in Ref. [165]. An experimental procedure similar to that used in the ^{3}H or ^{187}Re β-decay experiments could be employed to extract a value for the neutrino mass from a fit to the shape of the β-spectrum near the end-point. Background from the ^{115}In – ^{115}Sn ground state decay could be reduced by gating on the 497 keV γ-ray released after β-decay to the ^{115}Sn ($3/2^+$) state. However, this scheme would require a precise theoretical calculation of the second forbidden β-spectrum shape, which itself is challenging and, as mentioned above, the theoretical calculation is further complicated by the ultra-low Q value.

The discovery of this first, and currently only known ultra-low Q value β-decay has raised interesting possibilities for the introduction of new decay candidates for use in neutrino mass determination experiments. Furthermore, it has revealed that subtle effects in nuclear β-decay theory could play a more significant role in the ultra-low energy regime. Hence, there is a need for the identification and study of other ultra-low Q value decays that could be better suited for neutrino physics experiments, and could provide a testing ground for investigating atomic effects in ultra-low energy decays.

3.2 Identification of potential ultra-low Q value β-decays

In order to observe additional ultra-low Q value β-decay transitions, dedicated low background experiments will be required. Therefore, candidate transitions must first be identified. From Eqn. (12), such a candidate should have a nuclear level with energy E^* within 1 keV of the Q value. However, in reality E^*, and more often Q_{GS}, are not known to the required precision to make this identification. Instead, researchers have identified nuclides for which an ultra-low Q value transition could potentially occur, but for which more precise Q value and/or energy level data are required.

Not long after the confirmation of the ultra-low Q value transition in ^{115}In, Suhonen and collaborators identified potential ultra-low Q value transitions in ^{135}Cs [166], ^{115}Cd [167], and 12 other nuclides [159, 168]. Some of these candidates, and additional ones, were also identified in a study by Kopp and Merle in 2010 investigating the possibility of using ultra-low Q value transitions for neutrino mass measurements [169]. Later, Gamage *et al.* performed a study of atomic mass and nuclear energy level data on $\beta^{\pm}$ and EC decay nuclides across the nuclear chart to provide a comprehensive list of potential ultra-low Q value decays [170]. This evaluation was recently updated [171] to include data from the most recent atomic mass evaluation (AME2020) [73]. In both cases around 80 nuclides were identified with $Q_{ES} \lesssim$10 keV and uncertainties on Q_{ES} of $\sim$1 − 10 keV, making them potential candidates to have a decay with $Q_{ES} \leq$1 keV.

Since the identification of potential ultra-low Q β-decay candidates in Refs. [159, 166–170], a number of Penning trap groups have performed measurements to provide precise and accurate Q values for some of these systems. In total, fifteen candidates have been investigated by the JYFLTRAP, LEBIT, CPT, and ISOLTRAP groups. Five nuclides were identified as potential candidates for experimental searches for an ultra-low Q value decay and for use in neutrino mass determination experiments. These nuclides, their measured Q_{GS} values and resulting Q_{ES} values are listed in Table 2.

The JYFLTRAP group found that ^{131}I and ^{135}Cs do have $Q_{ES} \lesssim 1$ keV for allowed and 1$^{\text{st}}$ forbidden unique β^- decays to excited states in their respective daughters. They also found that ^{111}In and ^{159}Dy have allowed EC decays to daughter states with Q_{ES} values of $\approx$1–4 keV. However, these Q_{ES} values are further reduced when the binding energy of the captured electron is included, see Refs. [153, 174] for further discussion. Measurements at both JYFLTRAP and LEBIT indicate that ^{75}Se could also have an energetically allowed 1$^{\text{st}}$ forbidden unique EC decay to the 865.4(5) keV ($3/2^-$ or $5/2^-$)

Table 2 Potential ultra-low Q value decay candidates that were identified based on mass data from the atomic mass evaluation and have since had their Q_{GS} values precisely measured via PTMS as listed under Ref.

Decay	Type	Q_{GS}	E^*	Q_{ES}	Ref.
^{75}Se($5/2^+$) → ^{75}As{$3/2^-$;$5/2^-$}	1^{st}FNU (EC)	866.041(81)	865.40(50)	0.64(51)	[172]
		866.50(44)	865.40(50)	1.10(67)	[173]
^{111}In($9/2^+$) → ^{111}Cd($7/2^+$)	Allowed (EC)	857.63(17)	853.94(7)	3.69(19)	[174]
^{111}In($9/2^+$) → ^{111}Cd($3/2^+$)	2^{nd}FU (EC)	857.63(17)	855.6(10)	2.0(1.0)	[174]
^{131}I($7/2^+$) → ^{131}Xe($9/2^+$)	Allowed (β^-)	972.25(19)	971.22(13)	1.03(23)	[175]
^{135}Cs($7/2^+$) → ^{135}Ba($11/2^-$)	1^{st}FU (β^-)	268.66(30)	268.218(20)	0.44(31)	[176]
^{159}Dy($3/2^-$) → ^{159}Tb($5/2^-$)	Allowed (EC)	364.73(19)	363.545(1)	1.18(19)	[153]

state in ^{75}As. Again, the binding energy of the captured electron will affect the Q_{ES} value, but a more precise determination of the energy of the 865.4 keV level is also required to fully evaluate this potential decay. In addition to the five nuclides listed in Table 2 for which decay with $Q_{ES} \approx 1$ keV is possible, PTMS measurements have also ruled out nine additional candidates: ^{72}As [177], ^{75}Ge [172, 173], ^{76}As, ^{155}Tb [178], ^{89}Sr, ^{139}Ba [179], and ^{112,113}Ag,^{115}Cd [180].

Of the many potential ultra-low Q value decay candidates identified in Ref. [171] for which more precise Q value measurements are needed, a short list of some of the most promising ones to investigate are listed in Table 3. These have been selected because (i) they have a relatively long total half-life, which would make an experiment with this nuclide more feasible; (ii) they have Q_{ES} values obtained from AME data that are close to zero with uncertainties of a few keV or less, increasing the likelihood that they do have a $Q_{ES} \lesssim$ 1 keV, and (iii) they are allowed or first forbidden transitions, resulting in a longer partial half-life for the ultra-low transition, and a β-spectrum that can be readily described theoretically.

4 Summary and Outlook

Penning trap mass spectrometry provides the means to determine the Q value for a β-decay transition by measuring the cyclotron frequency ratio between ions of the parent and daughter nuclides. This direct Q value determination, based on the energy equivalence of the mass difference between initial and final states, can be compared to Q value measurements made via the kinetic energy of the decay products. Such a comparison provides a strong test of systematics in the detectors that are employed. Furthermore, these precise Q values can be used as reliable inputs for theoretical calculations. This enables

Table 3 List of some of the most promising allowed and first forbidden potential ultra-low Q value $\beta^{\pm}$ and EC decay candidates to investigate via PTMS.

nuclide	Decay	Forbiddenness	Half-life	Q_{ES} (keV)
^{136}Cs	β^-	Allowed	13 dy	3.7(19)
^{188}W	β^-	Allowed	70 dy	-4.6(32)
^{155}Eu	β^-	1st Forbidden	5 yr	0.3(16)
^{156}Eu	β^-	1st Forbidden	15 dy	1.0(37)
^{56}Co	EC	Allowed	78 dy	4.76(55)
^{97}Tc	EC	Allowed	4.2 Myr	-0.1(42)
^{175}Hf	EC	Allowed	70 dy	1.0(26)
^{81}Kr	EC	1st Forbidden	229 kyr	3.2(15)
^{146}Pm	EC	1st Forbidden	6 yr	-0.3(45)
^{157}Tb	EC	1st Forbidden	71 yr	-2.3(14)
^{173}Lu	EC	1st Forbidden	1.5 yr	1.0(18)
^{183}Re	EC	1st Forbidden	70 dy	2.5(81)
^{195}Au	EC	1st Forbidden	186 dy	1.9(12)
^{148}Eu	β^+	Allowed	55 dy	-15(10)
^{105}Ag	β^+	1st Forbidden	41 dy	5.7(47)
^{144}Pm	β^+	1st Forbidden	1 yr	-4.8(32)
^{146}Pm	β^+	1st Forbidden	6 yr	-4.3(45)

a comparison of theoretical and experimentally determined half-lives, which can shed light on contributions from subtle effects such as atomic interference effects. It also enables a precise comparison of calculated and measured β-spectra, which could help determine the quenching of the axial vector coupling constant, g_A, and provide information on backgrounds in sensitive detectors used for $0\nu\beta\beta$ and dark matter searches.

PTMS has provided precise Q values for the β-decays of nearly all primordial nuclides, the main exception being ^{48}Ca. A more precise Q value here would help determine the possibility of detecting ^{48}Ca single β-decay in future experiments. An interesting new area both on the theoretical and experimental front is that of ultra-low Q value β-decays. Comparison of experimental and theoretical half-lives for these systems can provide a strong test of the theory, and the experimental observation of such decays could lead to new candidates for neutrino mass studies. Several Penning groups have begun evaluating potential candidates by performing precise Q value measurements to determine if they do have Q_{ES} values that are $\lesssim 1$ keV, and several have been identified so far. There are still many other potential candidates to investigate, which will require dedicated precision measurements with Penning traps, and in some cases precise energy level measurements. A comprehensive list of potential candidates would then enable the selection of the best system to use for an ultra-low Q value decay search.

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