EPJ manuscript No.
(will be inserted by the editor)

Precision electron-capture energy in ^{202}Pb and its relevance for neutrino-mass determination

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Received: May 26, 2017/ Revised version: May 26, 2017

Abstract. Within the framework of an extensive programme devoted to the search for alternative candidates for the neutrino-mass determination, the atomic mass difference between ^{202}Pb and ^{202}Tl has been measured with the Penning-trap mass spectrometer ISOLTRAP at the ISOLDE facility at CERN. The obtained value $Q_{\mathrm{EC}} = 40.2(4.3)$ keV is three times more precise than the AME2012 value. While it will probably not lead to a replacement of ^{163}Ho in modern experiments on the determination of the electron-neutrino mass, the electron capture in ^{202}Pb would however allow a determination of the electron-neutrino mass on the few-eV level using a cryogenic micro-calorimeter.

PACS. PACS-key Atomic masses – PACS-key Penning-trap mass spectrometry – PACS-key Neutrino mass determination

1 Introduction

Among the large variety of experiments exploring neutrino properties, the determination of the neutrino mass is a very ambitious and challenging venture. It will have profound impact on the extension of the Standard Model of particle physics and on our understanding of the origin of fermion masses beyond the Higgs mechanism.

Over the last decades many attempts have been undertaken to measure the neutrino mass, which is known to be small but not zero [1,2]. The electron-antineutrino mass has been determined with an upper limit of about 2 eV at 95% C.L. in tritium-decay experiments [3], whereas for the neutrino mass a limit of 225 eV at 95% C.L. has been obtained from the inner bremsstrahlung spectra of ^{163}Ho [4,5]. If one does not exclude the violation of the CPT-principle [6], the mass of the neutrino and antineutrino must be measured independently with similar sensitivity.

One of the first attempts of the neutrino mass determination in the electron-capture sector (EC) was made at ISOLDE at the beginning of the 1980s [7]. An upper limit of 1.3 keV was determined for the EC in ^{163}Ho by measuring the M-shell X-rays and Auger electrons [8]. A lower limit of 0.5 keV was obtained for the EC in ^{193}Pt in a subsequent experiment [9]. At that time the technical possibilities were not capable of achieving a lower uncertainty. However, the progress over the last years in both Penning-Trap Mass Spectrometry (PTMS) [10,11] and Cryogenic Micro-Calorimetry (CMC) [12], as well as in theory [13], allows determining the neutrino mass at a considerably increased level of precision, i.e. down to the eV region or even below. To determine the neutrino upper mass limit, the results of PTMS and CMC have to be combined, as the decay energy value determined by PTMS influences the atomic de-excitation spectrum following the electron capture [14].

The best candidate for such measurements is a nuclide with a small EC-decay energy which also features a small neutrino total energy and thus a high sensitivity to the

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neutrino rest mass at the high-energy part of the calorimetric spectrum. In this context, ^{163}Ho is the favourable candidate of known electron-capture transitions [15], due to its extremely small Q-value of only 2.833(34) keV [16]. This is, among other things, a reason for several modern projects to determine the neutrino mass based on this process, e.g. ECHo [17], HOLMES [18], and NuMECS [19]. A recent direct and precise measurement of the decay energy of the EC in ^{163}Ho with SHIPTRAP [16] has solved a long-standing puzzle in differences of this quantity obtained by various indirect methods.

Spectacular agreement between the values obtained by PTMS [16] and CMC [17] paves the way to the neutrino mass determination on a sub-eV level by combination of these two methods [20]. Meanwhile, a somewhat simplified model of electron capture, shown later in Equation 3, which was first proposed in [21] for a determination of the neutrino mass must include additional effects accompanying the electron-capture process, e.g. electron shake-up and shake-off effects [22,23]. These effects can hamper the analysis of the de-excitation spectrum when compared to the simple approach [21] and thus, can substantially increase the uncertainty in the neutrino mass determination from the EC in ^{163}Ho. In this context, a search for other candidates for the neutrino mass determination from electron capture, gains significant importance.

The criteria for choosing such alternative nuclei have been discussed in [15] and one of the candidates, viz., ^{194}Hg, has been explored at ISOLDE (CERN). The decay energy $Q_{\mathrm{EC}} = 29(5)$ keV of the EC in this nuclide was determined with ISOLTRAP, which turned out to be considerably smaller than the value 69(14) keV listed in the atomic-mass evaluation 2012 [24]. This highlights the importance of performing high-accuracy PTMS measurements of all promising EC pairs.

In the present work, we report on the determination of the Q_{EC} value of the EC in ^{202}Pb, already proposed as a good candidate for the neutrino mass evaluation in [15]. Its AME2012 value is $Q_{\mathrm{EC}} = 46(14)$ keV [24]. The relevant values were achieved by combining the results of a previous mass measurement of ^{202}Pb in 2008 at ISOLTRAP, described in [25] and the precision mass measurement of ^{202}Tl which will be explained in the following.

2 Experimental procedure

The experiment was accomplished by use of ISOLDE's high-precision Penning-trap mass spectrometer ISOLTRAP at CERN. All required steps for the mass measurements at ISOLTRAP, including the radioactive ion-beam production at the ISOLDE facility and data analysis routine are described in the references [7,26,27,28]. Here, only a brief summary of the experimental procedure is given.

The isotopes of interest were produced at ISOLDE by a proton beam with intensity of up to 2 μA and an energy of 1.4 GeV impinging on a, 50 g/cm^2, uranium-carbide ($\mathrm{UC_X}$) target. The reaction products were released from the target at a temperature of about 2000°C by thermal diffusion and effusion. Subsequently, the atoms were ionized by different techniques, including plasma, surface and laser ionization. The ^{202}Pb-ions were produced in 2008 by a hot plasma ion source [29], while the ^{202}Tl ions were produced in 2015 by RILIS [30], ISOLDE's laser ion source. After the ionization process, the ions were accelerated and mass separated by the general purpose magnetic separator.

The mass-selected ions were delivered as a 30-keV continuous beam to the ISOLTRAP setup, composed of four ion traps. The ions were accumulated in a linear radio-frequency-quadrupole (RFQ) [31], cooled by collisions with helium buffer gas, and bunched. The bunches were released from the RFQ and injected into the multi-reflection time-of-flight mass separator (MR-ToF MS) [28,32], where the bunches of different isobars were mass separated, with a resolving power of up to $3{\cdot}10^5$ [33]. In this way the contaminants ^{202}Fr, ^{202}Bi were successfully separated with the MR-ToF MS. They were 100 times less abundant than the ion of interest ^{202}Tl. The separation was achieved using 1000 revolutions in the MR-ToF MS, which corresponds to a trapping time of about 36 ms. The isobarically purified ion bunch was then transferred to the first, cylindrical Penning trap [34] for cooling, re-centering and additional isobaric cleaning by mass-selective resonant buffer-gas cooling [35]. The cleaned bunch of singly charged ions of interest was afterwards injected into the hyperbolic precision Penning trap.

Here, a precise measurement of the cyclotron frequency $\nu_c = qB/(2\pi m)$ of ions with mass m and charge q, stored in a strong and homogeneous magnetic field B, was performed with the time-of-flight ion cyclotron-resonance detection technique [36] using single-pulse- and Ramsey-type excitation schemes [37,38] with various excitation times. The magnetic field strength was calibrated by measuring the cyclotron frequency ν_{ref} of a reference ion with well-known mass, here ^{133}Cs. The mass of the ion of interest was deduced from the cyclotron-frequency ratio:

$$r_{ref,x} = \frac{\nu_{c,ref}}{\nu_{c,x}} = \frac{m_{ion,x}}{m_{ion,ref}}. \quad (1)$$

The frequency ratio can be converted to the atomic mass m_{atom} of the nuclide of interest according to:

$$m_{\mathrm{atom}} = r_{ref,x} \cdot (m_{\mathrm{ref}} - m_e) + m_e, \quad (2)$$

where m_{ref} is the mass of the reference nuclide and m_e the electron mass. In Eq. 2 the atomic binding energies are not included, as their contribution is negligible compared to the precision of the measurements presented here.

In order to determine the Q-value of the EC in ^{202}Pb, two independent campaigns on the absolute mass measurements of the mother ^{202}Pb and daughter nuclide ^{202}Tl have been performed at ISOLTRAP. Between the two independent measurement campaigns, a simultaneous measurement of ^{202}Pb and ^{202}Tl was attempted, which would have allowed a direct Q-value determination with very high precision. Both species were in this case produced

from a UC_x target, and ^{202}Pb was laser ionized. The measurement was, however, hampered by the very low surface-ionization yield of ^{202}Tl and by the contamination of the ^{202}Pb ground state by its long-lived isomeric state ($T_{1/2}$ = 3.54 h [39]). The masses of both nuclei were determined by measuring the cyclotron-frequency ratio between the ion of interest and a corresponding singly charged reference ion. In the case of ^{202}Pb the reference mass was $^{133}Cs^+$, whereas the reference ions in the case of ^{202}Tl were $^{181}Ta^{16}O^+$ and $^{203}Tl^+$. The latter choice of reference avoids a significant mass-dependent frequency shift of the frequency ratio. In total 10 ToF-ICR measurements of $^{202}Tl^+$ were performed, the first six being sandwiched between ToF-ICR measurements of $^{203}Tl^+$, the last four between measurements of $^{181}Ta^{16}O^+$. Excitation times up to T_{ex} = 3 s were applied, either by a single-pulse- or a Ramsey-type excitation scheme. The choice of using two reference ions for the measurement series ($^{203}Tl^+$ and $^{181}Ta^{16}O^+$) was made in order to avoid any bias of the determined Q-value, due to a potential error of the literature value of the reference mass. Nevertheless, all 10 ToF-ICR measurements of $^{202}Tl^+$ were also sandwiched between ToF-ICR spectra of $^{133}Cs^+$, in order to monitor whether there were any significant changes in the measurement conditions; which would appear as sudden jumps in the frequency ratio with respect to $^{133}Cs^+$. The measured cyclotron-frequency ratios of $^{202}Tl^+$ with respect to $^{133}Cs^+$ are plotted in Fig. 1.

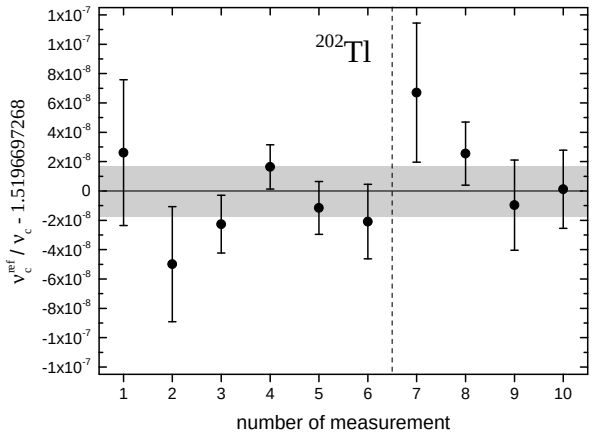

Fig. 1. Measured cyclotron-frequency ratios of $^{202}Tl^+$ with respect to $^{133}Cs^+$. The error bars of the individual measurements are the statistical uncertainties. The dashed line in the figure separates the data points for which ^{203}Tl and $^{181}Ta^{16}O$ were used as reference. The Grey shaded band represents the standard deviation of the average cyclotron-frequency ratio. For details see text.

Table 1 lists the final cyclotron-frequency ratios ν_{ref}/ν_c. The table separately shows both statistical and systematic uncertainties. In the case of the frequency ratios, the systematic uncertainty originates from two sources. The first is a mass-dependent error, which is due to the misalignment between the electric and magnetic symmetry axes of the Penning trap. The second error has no clear mass dependence and is due to the fact that ions of different masses probe different volumes of the Penning trap and hence their frequency ratios are affected by the trap inhomogeneities. Both relative errors were determined in [27] and were applied for the measurement of ^{202}Pb. The mass-dependent error was determined to have the value 1.6×10^{-10}/u, while the mass-independent one was determined to be 8×10^{-9}. Before the ^{202}Tl measurement, however, the superconducting magnet of ISOLTRAP underwent a maintenance operation and the trap tower was realigned. The mass-dependent error was preliminarily re-estimated by use of alkali ions to be 3.5×10^{-10}/u and this value is used for ^{202}Tl. As the general layout of the apparatus has not changed, the mass-independent error is expected to have the same value as determined in [27]. The statistical uncertainty of the mass-excess values also includes the contribution of the uncertainty of the reference mass. The evaluated ^{202}Tl mass-excess value, used within the article, was obtained using the AME algorithm, which accounts also for the consistency of the two corresponding reference masses ^{203}Tl and $^{181}Ta^{16}O$, in the entire AME16 network [39]. We note that the two mass-excess values exhibit a discrepancy larger than one standard deviation. The cause of this discrepancy could well be due to errors in the reference-mass values. This is supported by Fig. 1, where one notices that the frequency ratios of ^{202}Tl with respect to ^{133}Cs do not have any systematic shift between the set corresponding to ^{203}Tl and $^{181}Ta^{16}O$. The Q-value of the EC in ^{202}Pb is given by the mass difference between ^{202}Pb and ^{202}Tl and amounts to Q_{EC} = 40.2(4.3) keV.

3 Evaluation

The following formula addresses how the total Q_ν-value can be determined, where E_{B_x} is the binding energy of an electron from the xth atomic shell of the daughter nuclide:

$$Q_\nu = Q_{EC} - E_{B_x} \tag{3}$$

The binding energy of the K-electron is higher than Q_{EC}, E_{B_K} = 85.530 keV [40] for ^{202}Tl, hence the K-capture is energetically forbidden and thus electron capture only from higher atomic shells for ^{202}Pb is allowed.

To be a preferable candidate for the neutrino mass determination, the total energy of the emitted neutrino Q_ν should be as small as possible. In the case of ^{202}Pb the lowest Q_ν is for the $L1$-capture and is equal to 24.6(4.3) keV [40]. This number is far from the value for ^{163}Ho, which is only 0.79(3) keV [16]. Thus, the EC in ^{202}Pb is expected to have much less sensitivity to the neutrino rest mass than the EC in ^{163}Ho even taking into consideration the additional effects, like the shake-off of a second electron, elaborated on in [22,23].

Table 1. Cyclotron-frequency ratios of $^{202}Pb^+$, with respect to $^{133}Cs^+$ (ME(^{133}Cs) = -88070.931 (0.008) keV/c^2 [24]) and $^{202}Tl^+$, with respect to $^{181}Ta^{16}O^+$ (ME($^{181}Ta^{16}O$) = -53178.6 (1.8) keV/c^2 [24]), as well as $^{203}Tl^+$ (ME(^{203}Tl) = -25760.8 (1.3) keV/c^2 [24]). The $^{202}Pb^+$ result is taken from [25]. The mass excess deduced in this work is also shown (ME$_{exp}$), compared to the literature value (ME$_{lit}$) [24]. In the sixth column are listed the differences between the experimental value and the AME value, whereas in the last column the Birge ratios ($B.r.$) of the individual measurements are presented. The statistical uncertainties are given between parentheses, while the systematic uncertainties are given between square brackets.

Ion	Reference	Freq. Ratio ν_{ref}/ν_c	ME$_{exp}$ / keV	ME$_{lit}$ / keV	Δ / keV	$B.r.$
$^{202}Pb^+$	$^{133}Cs^+$	1.5196700577(284)[164]	-25941.4(3.5)[2]	-25940(4)	1.1(5.7)	
$^{202}Tl^+$	$^{181}Ta^{16}O^+$	1.0255363752(99)[84]	-25984.5(2.6)[1.5]*	-25986(14)	1.5(14)	0.36
$^{202}Tl^+$	$^{203}Tl^+$	0.9950720535(63)[80]	-25978.9(1.8)[1.5]*	-25986(14)	7.1(14)	0.43

*The used mass excess within this article is -25980.2(1.6) keV [39].

Moreover, there are several additional issues in neutrino-mass measurements by means of PTMS/CMC. First, the Q_{EC}-value of the EC in ^{202}Pb is relatively large for current CMC experiments [17,18,19]. Thus, either another micro-calorimetric experiment must be built, which is suitable for this energy regime, or the current CMC projects have to be adapted for this higher absorption energy value. Second, the daughter nuclide ^{202}Tl is not stable and decays with a half-life of $T_{1/2}$ = 12.3 days, which produces significant background in the calorimetric spectrum.

4 Conclusion and outlook

In this work we reported a new result from the measurement campaign searching for alternative candidates to determine the neutrino mass. The mass difference of ^{202}Pb-^{202}Tl had been evaluated in [24] as Q_{EC} = 46(14) keV with a relatively large uncertainty. The Penning-trap mass spectrometer ISOLTRAP at ISOLDE/CERN was used to improve this value. The obtained Q_{EC} = 40.2(4.3) keV value has a threefold higher precision. The direct measurements of the Q_{EC}-value has shown that ^{202}Pb is applicable for the neutrino mass determination on the level of a few eV, however the Q_{EC}-value has to be determined even more precisely with the new generation of PTMS (e.g. [41,42]). Nevertheless, ^{163}Ho remains the best candidate for micro-calorimetry experiments from the point of view of its low Q_{EC}-E$_{B_x}$ value. The two reference masses ^{203}Tl and $^{181}Ta^{16}O$ should be measured in a separate beam time to exclude any mass deviation that might exist. For this purpose, a carbon cluster reference (e.g. C_{18}) could be used to minimize the contribution of the systematic errors.

5 Acknowledgements

The authors thank the Max-Planck Society for financial support and the German BMBF for the grant WTZ:01DJ-14002, 05P12HGCI1, 05P12HGFNE, 05P15ODCIA, the Wolfgang Gentner Program, the French IN2P3, the UK Science and Technology Facilities Council, the grant of the Russian Minobrnauki, the RILIS team, the ISOLDE Collaboration, as well as G. Audi and W. Huang (AME) for discussions about the involved masses.

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