

On the Q -value of the tritium β -decay

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Abstract. – We report here the atomic masses of ${}^3\text{H}$ and ${}^3\text{He}$ determined by using the Penning trap mass spectrometer SMILETRAP. The measurements are based on cyclotron frequency determinations of ${}^3\text{H}^{1+}$ and ${}^3\text{He}^{1+}$ using H_2^+ ions as mass reference. The mass values for ${}^3\text{H}$ and ${}^3\text{He}$ are 3.016 049 278 7(25) u and 3.016 029 321 7(26) u, respectively. From these masses a new Q -value of the tritium β -decay was derived resulting in 18.589 8(12) keV, being the most accurate value at present. The Q -value of the tritium β -decay is related to the possible rest mass of the electron antineutrino.

Introduction. – Q -values of nuclear decays and reactions involve masses of atoms in the initial and final states. In some cases such Q -values are related to fundamental questions in current physics requiring an extremely high mass precision. Such a case is the Q -value of the tritium β -decay.

Neutrino oscillations, which recently have been observed [1–3], require the existence of rest masses for the three neutrinos but give only a lower limit of 0.05 eV to the mass of the electron antineutrino. By studying the shape of the β -spectrum of tritium in the region that refers to energy of the last 50 eV with electrostatic electron spectrometers, it has been possible to set an upper limit of the mass to about 2 eV for $m(\nu_e)$ [4–7]. In the planned KATRIN experiment [8] it will be possible to measure a neutrino mass as low as 0.3 eV. This method of determining $m(\nu_e)$ could use a very accurate Q -value for calibration purpose. There are a number of Q -values available from end point determinations of the tritium β -decay. The uncertainty in the weighted average of these measurements is about 1 eV. These measurements should be checked by an independent and at least as accurate method, *i.e.* a Penning trap measurement.

Already in 1993 a Q -value measurement using mass determinations in a Penning trap was reported [9], which was based on the mass measurement of ${}^3\text{He}$ and ${}^3\text{H}$. However, 10 years later in ref. [10] we showed that the mass values of both ${}^4\text{He}$ and ${}^3\text{He}$ were too low by 7 nu and 14 nu, respectively (roughly a 5σ deviation in both cases). Therefore the Q -value reported in ref. [9] did not seem reliable and should thus be remeasured.

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Based on our measurements, a preliminary Q -value of 18.588(3) keV was reported in refs. [11,12]. Due to the fact that the two ions used, ${}^3\text{H}^{1+}$ and ${}^3\text{He}^{2+}$, are not q/A -doublets, these measurements resulted in a relatively large systematic uncertainty in the Q -value. In this paper, a new Q -value is reported which is derived from mass measurements involving ${}^3\text{He}^{1+}$ and ${}^3\text{H}^{1+}$ ions.

Experimental procedure. – The ion production and mass measurements with SMILETRAP have been described in detail in ref. [11]. Here only a short description is given, sufficient for understanding the mass measurements described in this paper.

The ${}^3\text{H}$ and ${}^3\text{He}$ and ${}^4\text{He}$ ions which are used here, were all produced in the electron beam ion source CRYISIS [13–15] at the Manne Sieghbahn Laboratory. Although CRYISIS was designed for the production of highly charged ions, it has now been possible to find ion source conditions such that singly charged ${}^3\text{He}$ ions and ${}^3\text{H}$ ions were produced from disassociated molecules (${}^3\text{H}_2$). Only a small amount of gas is required even for several days of ion production, an important fact when dealing with radioactive elements.

For the production of ${}^4\text{He}$ ions, high-purity ($> 99.99\%$) helium gas was used and for ${}^3\text{He}$ isotopically enriched ($> 99\%$) gas. The tritium ions were produced from a commercial tritium source of about 10 Ci (4 ml) [16]. The tritium was delivered as ${}^3\text{H}_2$ ($> 98\%$) gas absorbed in a uranium bed of 1.7 g inside a tiny gas bottle. The ${}^3\text{He}$ gas created during the transportation and storage of the bottle was removed by pumping on the bottle. After the removal of the ${}^3\text{He}$ gas, the ${}^3\text{H}_2$ gas is released from the uranium by heating the bottle to about 400°C . A buffer volume of ~ 5 ml was filled with 0.5 atm tritium gas from which it was introduced into CRYISIS via a needle valve. From measurements of the buffer volume pressure it was possible to estimate that only 0.14 ml of the tritium gas was used during the 4 days of the experiment.

The ions are ejected from CRYISIS in a bunch of $\sim 10^8$ ions with $100\ \mu\text{s}$ pulse length at an energy of 3.5 keV. This ion pulse is transported over a distance of about 15 meters to the SMILETRAP area, using electrostatic quadrupole lenses and deflectors. After charge selection in a 90° magnet the ions are decelerated in an electrostatic lens system before they enter the pre-trap and are captured. The pre-trap is an open-ended cylindrical Penning trap inside a warm solenoid of 0.25 T. Due to phase space properties and the fact that the ion pulse is much longer than the dimension of the pre-trap, only a small fraction of the beam is captured, typically 1000 ions. After the ions are captured the potential of the trap is lowered from 3.5 keV to 0 V in about 30 ms. The ions are then transported through a series of drift tubes at -1 keV to the hyperbolic precision Penning trap inside a 4.7 T superconducting magnet. The ions are again decelerated before entering the precision trap. An entrance aperture with a diameter of 1 mm prevents ions with too large radial energies to enter the trap. The ions are then subject to an evaporation process by changing the trap potential from 5 to 0.1 V, leaving only the coldest ions in the trap. After this procedure on average 1–2 ions are left in the trap.

In a Penning trap a homogeneous magnetic field confines the ions in a plane perpendicular to \mathbf{B} , and an electric quadrupole field confines the ions axially. The mass of an ion is derived from cyclotron frequency determinations. The cyclotron frequency of an ion with mass m and charge qe moving perpendicular to a magnetic field \mathbf{B} is given by the well-known equation, $\nu_c = qeB/(2\pi m)$. In the combined magnetic and electric field the ions move in three independent modes, each one with its own frequency: an axial motion ν_z , independent of the magnetic field, and two radial motions, the so-called magnetron and reduced cyclotron motions, denoted ν_- and ν_+ , respectively. In SMILETRAP these frequencies are for an ion with $q/A = 1/2$ about 240 kHz, 810 Hz and 36 MHz, respectively. It can be shown [17] that the sum of these frequencies is equal to the cyclotron frequency, $\nu_c = \nu_- + \nu_+$. The cyclotron frequency is measured by the time-of-flight technique developed by Gräff *et al.* [18]. The segmented ring electrode of

the trap is used for an azimuthal quadrupole excitation near the true cyclotron frequency ν_c . After the excitation the ion is ejected from the trap and the flight time to a detector located 500 mm above the trap is measured. If the ion is in resonance it gains radial energy which is converted into axial kinetic energy in the fringe field of the magnet [19]. Therefore, ions in resonance have a shorter time of flight. A frequency scan using a 1 second non-interrupted excitation time results in a resonance with a FWHM of about 1 Hz. A typical time-of-flight resonance spectrum is given in fig. 1. The expected sidebands of the resonance [19] are suppressed. This is mainly due to the initial spread in the magnetron radius, since the ions are not cooled in the pre-trap, and due to an incomplete conversion from magnetron to reduced cyclotron motion during excitation.

The most precise measurement of the magnetic field in the trap is done by measuring the cyclotron frequency of an ion with a sufficiently well-known mass. In this work we have used H_2^+ ions since its mass is known to 1.4×10^{-10} [11]. The H_2^+ ions are produced by rest gas electron-impact ionization in the pre-trap. In order to eliminate a time dependence in the magnetic field, the cyclotron frequencies of the reference ion and the ions of interest is measured alternatively in 3 min. Typically, the time of flight is measured at 21 equidistant frequencies around the resonance center twice for the first ion specie, before the other ion specie is measured after which the procedure is repeated. For each ion specie this takes 1.5 min and it takes only a few seconds to reprogram all settings that differ for the two ion species.

The mass of the ion is determined from the cyclotron frequency ratio between the ion of interest and that of the reference ion. To deduce the mass of the neutral atom one has to correct for the mass ($q_{ion} \cdot m_e$) of the missing electrons and their atomic binding energies E :

$$m_{atom} = \frac{\nu_{ref} q_{ion}}{\nu_{ion} q_{ref}} m_{ref} + q_{ion} \cdot m_e - E. \quad (1)$$

The uncertainty in the electron mass is known so accurately that it contributes to an uncertainty in the mass which is much less than 1×10^{-10} [20]. For light ions such as ${}^3\text{He}^{1+}$ and

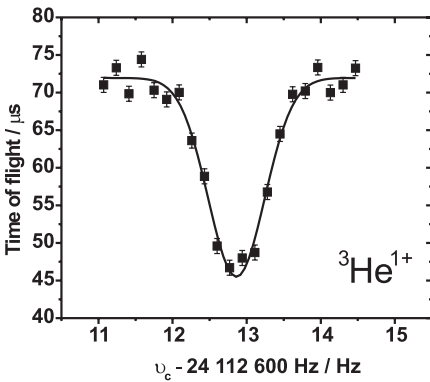


Fig. 1

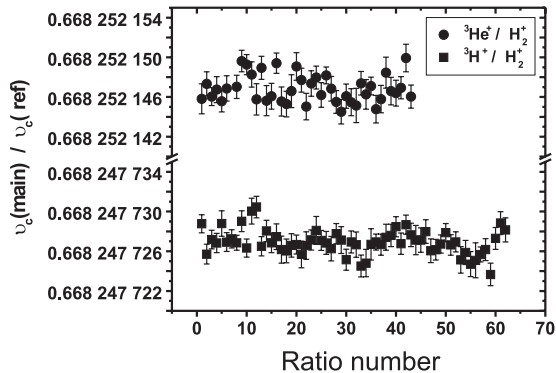


Fig. 2

Fig. 1 – The time-of-flight cyclotron resonance frequency for ${}^3\text{He}^{1+}$ ions generated from 50 scans, representing about 2% of the total data. The applied excitation time is 1 s. The FWHM is < 1 Hz and the center frequency can be obtained with ± 0.03 Hz. A similar spectrum is recorded for the reference ion H_2^+ .

Fig. 2 – Measured cyclotron frequency ratios $\nu_c({}^3\text{He}^{1+})/\nu_c(\text{H}_2^+)$ and $\nu_c({}^3\text{H}^{1+})/\nu_c(\text{H}_2^+)$; the error bars represent only the statistical uncertainty.

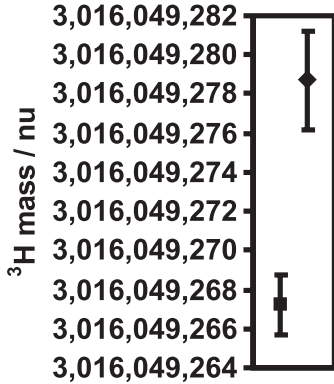


Fig. 3

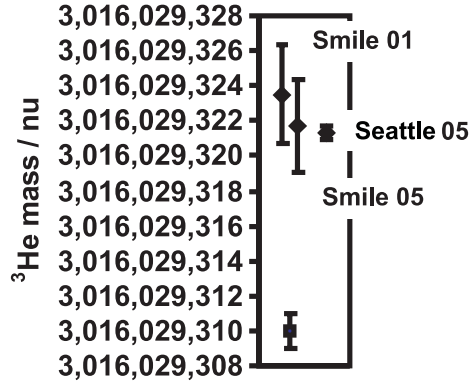


Fig. 4

Fig. 3 – The mass of ${}^3\text{H}$. The lower value (square) is from refs. [9, 21]; the higher value (diamond) is from the work presented in this paper.

Fig. 4 – The mass of ${}^3\text{He}$. The lower value (square) is from refs. [9, 21]; the higher values are from left to right, SMILETRAP using ${}^3\text{He}^{2+}$ [10], this paper using ${}^3\text{He}^{1+}$ ions and a preliminary value from the Seattle group [22].

${}^3\text{H}^{1+}$ the binding energies are known from precise experiments, and contribute therefore to an uncertainty less than 1×10^{-10} .

Results. – In fig. 2 the measured cyclotron frequency ratios data are shown for ${}^3\text{H}^{1+}$ and ${}^3\text{He}^{1+}$ ions. In table I the mass values and uncertainties obtained for ${}^3\text{H}$ and ${}^3\text{He}$ are listed. They are compared to the values from ref. [9] in fig. 3 and fig. 4. Using the masses from table I we derive a new Q -value of 18.5898(12) keV. It should be emphasized that since the Q -value is a mass difference between ions of almost the same mass, which were measured in the same way using H_2^+ reference ions in both cases, certain systematic uncertainties in the Q -value cancel to a large extent.

It was shown in ref. [11] that the mass measurements using SMILETRAP are effected by four main systematic errors. The first one is a mass shift that depends on the kinetic energy of the ion, thus being a relativistic effect. The second one is a change of the observed frequency that depends on the number of simultaneously stored ions (charges) in the trap. A third effect is a shift depending on the q/A ratio of the observed ion. Finally, it has been observed that the presence of a *large* amount of contaminant ions in the trap can produce a shift of the resonance.

TABLE I – Frequency ratios, atomic masses and uncertainties for the measured ions. Only the statistical uncertainties are given for the frequency ratios.

Ion pair	Frequency ratio Atomic mass/u	Statistical uncertainty/nu	Systematic uncertainty/nu	Year
$\text{H}_2^+ - {}^3\text{H}^{1+}$	0.668 247 726 86(15) 3.016 049 278 7(25)	0.7	2.4	2000
$\text{H}_2^+ - {}^3\text{He}^{1+}$	0.668 252 146 82(16) 3.016 029 321 7(25)	0.7	2.5	2005
$\text{H}_2^+ - {}^3\text{He}^{2+}$	1.336 747 448 58(26) 3.016 029 323 5(28)	0.6	2.7	2000

TABLE II – *The measured mass of ${}^4\text{He}$ from 2000 and 2005 and the literature value from Van Dyck et al./AME2003 [23, 24].*

Ion pair	Atomic masses	Year
$\text{H}_2^+ - {}^4\text{He}^{2+}$	4.002 603 256 8(13)	2000
$\text{H}_2^+ - {}^4\text{He}^{2+}$	4.002 603 253 6(26)	2005
Accepted	4.002 603 254 15(6)	2003

Relativistic mass increase: The measured ion mass is not the rest mass since the ion moves with an increased velocity during excitation. The ion may even have a radial energy before being excited, due to a poor injection into the magnetic field. The correction due to this relativistic effect can be done either by using a method in which the energy is measured directly by a decelerating potential or by using previously measured time of flight *vs.* energy. The total correction for both ion species and reference is usually in the region 0.1–0.5 ppb. In the final mass of ${}^3\text{H}$ and ${}^3\text{He}$ this effect causes a shift of 0.10 ppb and 0.15 ppb, respectively, with an uncertainty of 0.1 ppb.

Ion number dependence: The cyclotron frequency decreases for an increased number of trapped ions (charges). For singly charged ions, as in the case of ${}^3\text{H}$, ${}^3\text{He}$ and the H_2 reference ion, the effect is small, 0.127(14) ppb/ion [11]. All data is evaluated using events with 1–3 ions to ensure identical conditions. Since the shift is similar for the measured ion specie and the reference ion, the net effect in the atomic mass is close to zero. The estimated uncertainty due to this effect is 0.1 ppb.

q/A asymmetry: In the analysis of all SMILETRAP measurements, it cannot be excluded that the result depends on the q/A ratio difference between the ion of interest and the reference ion. In the tritium Q -value determination, since it is a mass difference involving masses with identical mass-to-charge ratios, this effect cancels. However, in the determination of the individual atomic masses this effect gives the largest contribution to the total uncertainty, the q/A difference $1/2$ – $1/3$ gives an uncertainty of 0.77 ppb.

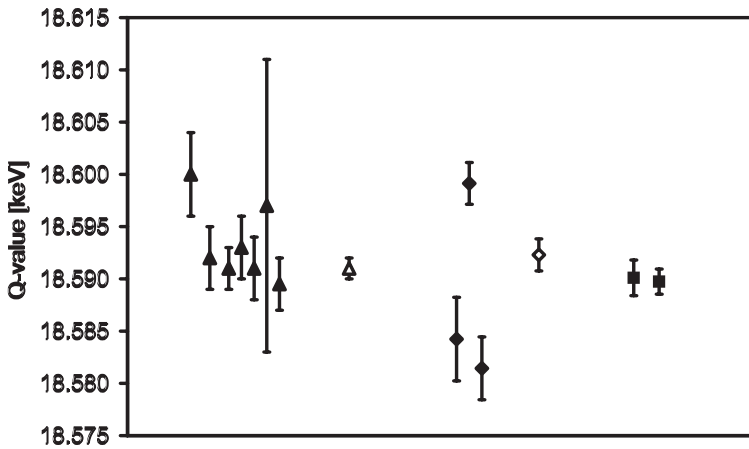


Fig. 5 – Tritium Q -values from β -spectrometers (triangles) [25–31], FTICR measurements (diamonds) [32–34], and Penning traps (squares). The first Penning trap value is the one reported by the Seattle group, 18.590 1(17) keV [9], and the last value is our Q -value of 18.589 8(12) keV, reported in this paper. The open symbol is the weighted average of the corresponding data.

Contaminant ions: Finally, contaminant ions can either be injected from the ion source or created by charge exchange processes in the trap system. As an example, 30% ${}^4\text{He}^{2+}$ in a ${}^{12}\text{C}^{6+}$ resonance shifted the center by 7 ppb. The impurity is checked by driving the reduced cyclotron frequency at resonance and then studying the flight time spectra. The excited ions will all be collected in a narrow peak at a short flight time, entirely resolved from the impurity ions that are out of resonance. A relative impurity concentration is obtained from which a limit of the impurity ions can be estimated. In the case of ${}^3\text{H}$ and ${}^3\text{He}$ the effect of impurities is estimated to be < 0.1 ppb.

Other effects: Remaining sources for systematic uncertainty are from the uncertainty in the reference ion mass (0.18 ppb only in the mass of ${}^3\text{H}$ and ${}^3\text{He}$), electron mass, electron binding energies, and magnetic field drift. In the here presented mass measurements these are all ≤ 0.1 ppb. The effect from the natural decay of the magnetic field can be neglected, $10^{-11}/\text{h}$.

The statistical and systematic uncertainties are given in table I. To look for possible further systematic errors the mass of ${}^4\text{He}$ was measured at different times also in connection with this measurement. The values are listed in table II together with the literature mass value [23,24]. As seen from the table, the ${}^4\text{He}$ mass values measured at different times are in good agreement.

Discussion and conclusion. – In fig. 5, Q -values from different sources are compared. The average of these values is the Q -value, 18.591(1) keV, presented in AME2003 [24]. The weighted average of the β -spectrometer [25–31], and FTICR (Fourier Transform Ion Cyclotron Resonance) measurements [32–34] are 18.591 0(9) keV and 18.590 0(8) keV, respectively. They are in agreement with the value reported here, 18.589 8(12) keV. Our Q -value is also in numerical agreement with the previous Penning trap value of 18.590 1(17) keV [9], although this agreement is accidental and it is due to the fact that the day-to-night effect in early Seattle group measurements introduced similar shifts for the ${}^3\text{H}$ and ${}^3\text{He}$ masses, see fig. 3 and fig. 4. The mass measurement of singly charged ${}^3\text{He}$ ions presented in this article agrees with our previous value obtained by using $2+$ ions, see fig. 4, and more importantly, it has been confirmed by the Seattle group [22] in an ongoing measurement. By using ${}^3\text{He}^{1+}$ ions instead of $2+$ we could improve our preliminary Q -value [11,12] by a factor of 2. In order to be comparable to the expected sensitivity of the KATRIN experiment, further precision improvements are necessary and envisaged.

It should be possible to further improve the Q -value by using SMILETRAP, by doubling the excitation time to 2 s and by implementing the Ramsey excitation technique [35,36]. A precision gain about a factor of 3–5 can be expected due to the narrower line width and multiple resonance fringes obtained by using this technique. This is being tested by comparing the cyclotron frequencies of H_2^+ using Gaussian and Ramsey fringes at different excitation times.

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