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A new determination of the ^{4}He and ^{3}He masses in a Penning trap

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Abstract. The atomic and nuclear masses of ^{4}He and ^{3}He have been measured using doubly charged ions in a Penning trap connected to an electron beam ion source. Recent technical improvements allow mass determinations with uncertainties of a few parts in 10^{10}. The obtained atomic masses are 4.002 603 256 8(13) u and 3.016 029 323 5(28) u respectively. These values deviate by as much as 5 standard deviations from the accepted values.

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The atomic and nuclear masses of ^{3}He and ^{4}He are fundamental constants appearing in various fields of physics, as for example in nuclear decays and reactions. So for example these masses are used to link heavier atomic masses in the mass tables [1]. The mass of ^{3}He also occurs in the Q-value of the tritium beta decay that might be used in future analysis of the tritium beta spectrum in efforts to determine the mass of the electron antineutrino. The masses of the helium ions have been reported with an uncertainty of a few times 10^{-10} [1]. However, an error in the accepted ^{4}He mass was indicated in our measurement of the proton mass a few years ago [2]. Our Penning trap mass spectrometer SMILETRAP [3] was used with H_2^+ as a carrier for the proton and highly charged ions of ^{4}He, ^{12}C, ^{14}N, ^{20}Ne, ^{28}Si, and ^{40}Ar as mass references. This sequence of ions were selected since they were all reported to have a mass uncertainty close to 10^{-10} [1]. Several of them were fully stripped and thus almost perfect mass doublets to H_2^+. Since the reported uncertainty in the ^{4}He mass was 2.5×10^{-10}, ions of this atom should also have been useful as a mass reference. However, our measurement indicated that the accepted helium mass might be wrong (Fig. 1, Tab. 3). After several improvements of SMILETRAP (better vacuum, stabilized magnetic field and a new, more stable frequency source) it seemed important to repeat the ^{4}He mass measurement adding also a measurement of the ^{3}He mass. In the new measurements we reproduced our previous values of the cyclotron frequency ratios of $^4He^{2+}$ and H_2^+. In this letter we report the revised values for the ^{3}He and ^{4}He masses thus obtained.

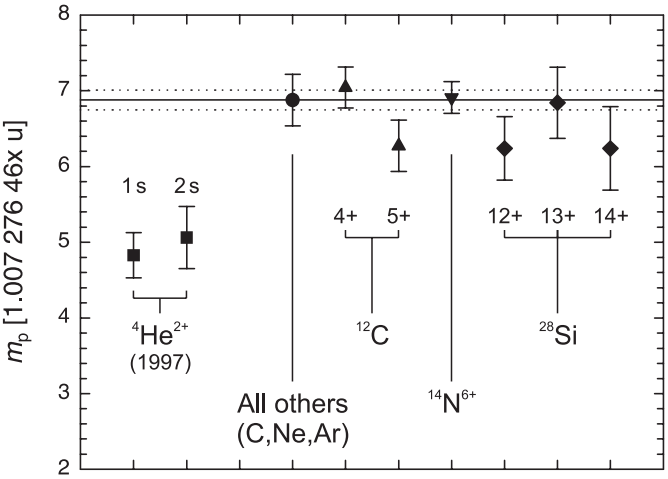

Fig. 1. The proton mass obtained in 1997 [2] using the ions indicated in the figure as mass references. All low statistics data (C^{6+}, $Ne^{9+,10+}$ and, $Ar^{14+,16+}$) are collected into a single point. The solid line shows the present, most accurately determined proton mass (1.007 276 466 88(13) u). The dotted lines corresponds to an uncertainty of 0.13 ppb. Excluding ^{4}He as a reference mass we obtained a proton mass of 1.007 276 466(13)(85) u. The two helium measurements not included in our evaluation of the proton mass indicate that the accepted helium mass may be to low.

The SMILETRAP facility is a hyperboloidal Penning trap mass spectrometer with a 4.7 T magnet. The trap is connected to an electron beam ion source, able of producing highly charged ions. Helium gas (pure ^{3}He or ^{4}He) is directly injected into the ion source which then

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Table 1. Observed frequency ratios with statistical uncertainties.

Ion	Frequency ratio	Relative unc. (ppb)
$^3He^{2+}/H_2^+$	1.336 747 448 58(26)	0.19
$^4He^{2+}/H_2^+$	1.007 171 502 79(18)	0.18

delivers pulses of $\sim 10^8$ ions at a 5 Hz repetition rate. Selected pulses of these ions are transported to SMILETRAP where a 90° magnet selects the desired charge state. The ions are first retarded and then trapped in a cylindrical Penning trap. A few thousand of them are then sent to the hyperboloidal trap, where as an average one ion is trapped. An ion with mass m and charge qe that is moving perpendicular to a magnetic field B has a cyclotron frequency ν given by:

$$\nu = \frac{1}{2\pi}\frac{qeB}{m} \cdot \tag{1}$$

After exciting the ion cyclotron motion with an azimuthal quadrupole radio frequency field the ions are ejected out from the trap. The ion flight time from the trap to a detector is measured. In the gradient of the magnetic field the radial kinetic energy of the ions is converted into an axial kinetic energy [3,5]. Therefore, ions in resonance with the applied excitation have a shorter flight-time than ions out of resonance. By scanning the frequency and measuring the average ion flight-time for each frequency it is possible to detect a resonance. In this experiment we are using an excitation time of 1 second which results in a frequency line width of ∼1 Hz, since it is a Fourier-limited process. The corresponding resolving power is 3.6×10^7 but the center can be determined to ∼1% of the FWHM and thus it should be possible to reach a statistical uncertainty of a few parts in 10^{10}.

In order to eliminate a possible B-field dependence we alternatively measured (in a time shorter than 1.5 min) the cyclotron frequencies of the helium and the reference ion, H_2^+. This ion is produced by rest gas electron-impact ionization in the first trap.

The mass of the ion is then deduced from the observed frequency ratios (Tab. 1):

$$R = \frac{\nu_1}{\nu_2} = \frac{q_1 m_2}{q_2 m_1} \tag{2}$$

where the helium ion and the reference ion are denoted with subscript 1 and 2, respectively. It should be noted that the relevant observable is a frequency ratio and thus several systematic errors cancel when the measurements are performed under similar conditions.

The mass of H_2^+ can be calculated very accurately from the proton mass [4], the hydrogen ionization energy, the molecular binding energy [6] and the average molecular vibrational energy [7]. The value 2.015 101 497 03(36) u has been used here.

To deduce the atomic masses (M) of ^{4}He and ^{3}He one has to correct for the mass $q_1 m_e$ of the missing electrons and their binding energies E_B:

$$M = \frac{1}{R}\frac{q_1}{q_2} m_2 + q_1 m_e - \frac{E_B}{c^2} \cdot \tag{3}$$

Table 2. Uncertainty budget in ppb.

Uncertainties	$^3He^{2+}$	$^4He^{2+}$
Reference mass	0.18	0.18
Relativistic mass	0.45	0.10
Ion number dependence	0.10	0.10
q/A asymmetry	0.77	0.00
Contaminant ions	< 0.10	< 0.10
Magnetic field drift	< 0.06	< 0.06
Total systematic unc.	< 0.92	< 0.26
Statistical unc.	0.19	0.18
Total unc.	0.94	0.32

The nuclear and atomic masses obtained from the frequency ratios presented in Table 1 are given in Table 3 and the estimated systematic uncertainties in Table 2. The size and limits of possible systematic errors have been investigated before [2] using the ions mentioned above to measure the proton mass. The analysis of these measurements reveals the presence of four main possible systematic errors discussed in detail in reference [8].

The first one is a frequency shift that is given by the kinetic energy of the ion motion, thus being a relativistic effect. This energy is measured either by applying a retardation potential on a grid in front of the detector or calculated from the flight time.

The second one is a change of the observed frequency that depends on the number of ions simultaneously stored in the trap. For this reason only events with one or two ions are used to deduce the resonance as a compromise between statistics and the smallest possible correction.

The third effect is a frequency shift depending on the q/A ratio difference between the observed ion species. This could come from a misalignment of the axis of the trap and the magnetic field axis. Therefore, when possible, ion species with similar q/A should be used, for example $^4He^{2+}/H_2^+$, for which there is no correction of this kind.

Finally it has been observed that the presence of a large number of contaminant ions in the trap can produce a considerable shift of the resonance. An upper limit of this effect is set by applying a strong dipole excitation at the resonance frequency shifting all ^{4}He ions to shorter flight times. In this way the ^{4}He ions are entirely resolved from any impurity ions. It was concluded that the impurities could introduce a frequency shift of less than 0.1 ppb [3].

The total systematic uncertainties (Tab. 2) are 0.92 ppb for $^3He^{2+}$ and 0.26 ppb for $^4He^{2+}$. The relatively large uncertainty in the $^3He^{2+}$ mass originates from comparisons between two ion species with different q/A-ratios: one with $q/A = 0.5$ (H_2^+) and the other one with $q/A \approx 0.67$ ($^3He^{2+}$). As seen in Table 2 the other systematic uncertainties are relatively small.

Table 3. Obtained masses of $^3He^{2+}$, ^{3}He, $^4He^{2+}$ and ^{4}He compared to the accepted masses [1,10] and our values from 1997.

	Mass (u)	Relative unc. (ppb)
	$^3He^{2+}$	
Accepted	3.014 932 234 69(86)	0.28
SMILETRAP 2000	3.014 932 248 5(28)	0.94
	^{3}He	
Accepted	3.016 029 309 70(86)	0.28
SMILETRAP 2000	3.016 029 323 5(28)	0.94
	$^4He^{2+}$	
Accepted	4.001 506 174 7(10)	0.25
SMILETRAP 2000	4.001 506 181 8(13)	0.32
SMILETRAP 1997 1 s data	4.001 506 183 6(13)[a]	0.33[a]
SMILETRAP 1997 2 s data	4.001 506 182 7(18)[a]	0.45[a]
	^{4}He	
Accepted	4.002 603 249 7(10)	0.25
S. Brunner *et al.* [11]	4.002 603 254 5(62)	1.56
SMILETRAP 2000	4.002 603 256 8(13)	0.32

[a]Only statistical uncertainties.

Each measurement corresponds to about 24 hours of data acquisition. As can be seen in Table 1 the statistical uncertainty in the frequency ratios are < 0.20 ppb. From these observed ratios and the known reference mass it is possible to use equation (2) to extract the mass of the ions (Tab. 3). The atomic masses are calculated using equation (3). The uncertainties in the electron mass and the electron binding energy are $\ll 0.1$ ppb [9].

In conclusion, the obtained atomic mass of ^{4}He and ^{3}He becomes 4.002 603 256 8(13) u and 3.016 029 323 5(28) u, respectively, using the most recent determination of the proton mass [4]. The value of the mass of ^{4}He is consistent with the value obtained in 1997 for 1 s and 2 s excitation times. However, our values for the ^{4}He and ^{3}He masses deviate by as much as 5 standard deviations from the presently accepted values of these masses [1,10].

We have been informed by van Dyck Jr [12] that the large discrepancy between the accepted mass values and our data might be due to a daily variation in the magnetic field unknown at the time of their measurement.

Since our value of the ^{3}He mass deviates considerably from the accepted value we are also preparing a measurement of the tritium mass. In both the $^3He^+$ and $^3H^+$ measurements there is a relatively large q/A uncertainty which, however, will cancel in the Q-value determination of the ^{3}H β-decay if these ions are measured under similar conditions.

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