New Mass Value for ^{7}Li

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A high-accuracy mass measurement of ^{7}Li was performed with the SMILETRAP Penning-trap mass spectrometer via a cyclotron frequency comparison of ^{7}Li^{3+} and H$_2^+$. A new atomic-mass value of ^{7}Li has been determined to be 7.016 003 425 6(45) u with a relative uncertainty of 0.63 ppb. It has uncovered a discrepancy as large as 14σ (1.1 μu) deviation relative to the literature value given in the Atomic-Mass Evaluation AME 2003. The importance of the improved and revised ^{7}Li mass value, for calibration purposes in nuclear-charge radii and atomic-mass measurements of the neutron halos ^{9}Li and ^{11}Li, is discussed.

The mass of an atom and its inherent connection with the atomic and nuclear binding energy is a fundamental property of the atomic nucleus. Accurate mass values are therefore of importance for a variety of applications in nuclear and atomic physics studies ranging from the verification of nuclear models and tests of the standard model to the determination of fundamental constants [1,2]. In nuclear structure studies the nuclear binding energy is the key information and is defined as the missing mass of the bound system $m(N, Z)$ compared to the sum of the masses of the constituent protons Zm_p and neutrons Nm_n:

$$B(N, Z) = [Nm_n + Zm_p - m(N, Z)]c^2. \qquad (1)$$

A most intriguing discovery in the last 20 years related to atomic nuclei is the large nuclear matter distribution of the short-lived nuclide ^{11}Li ($T_{1/2} = 8.94$ ms) [3], which is attributed to a "halo" of neutrons around a compact core of nucleons [4–6]. A halo state can be formed when bound states close to the continuum exist. Since 1985 a large number of high-accuracy experiments have been performed on ^{11}Li in order to observe the halo character also in other nuclear ground state properties, for example, in the nuclear-charge radii [7] and in the quadrupole moment [8] by laser spectroscopy, and in the binding energy, i.e., the neutron-separation energy via direct mass measurements [9]. Common to all of these experiments is the need of a proper reference in order to calibrate the measurement device and to look for systematic uncertainties. Two of the experimental approaches, nuclear-charge radii determination and atomic-mass measurements, are discussed in more detail here. Although ^{11}Li is the best studied halo nucleus, there are only relatively poor and conflicting results regarding its two-neutron-separation energy S_{2n} [9]. This can be resolved with an on-line Penning-trap mass measurement on ^{11}Li, where, for calibration purposes of the magnetic field ^{7}Li is a convenient reference nuclide. It can be obtained simultaneously from the same

ion source as ^{11}Li. All on-line Penning-trap mass spectrometers for short-lived radionuclides use buffer-gas-filled traps or gas cells to decelerate and stop the high-energy incoming ion beam. Thus, other ideally suited reference nuclides like ^{4}He$^+$ or ^{22}Ne^{2+} cannot be used due to tremendous charge exchange losses while stopping a helium or neon beam in a helium (or neon) environment.

A high mass accuracy is also required for a determination of the nuclear-charge radii of the lithium isotopes ^{6,7,9,11}Li via a measurement of the optical isotope shift employing laser spectroscopy [7,10,11]. The isotope shift receives contributions from two sources: the mass shift due to the change of nuclear mass and the field shift due to the change of nuclear-charge radii. Since the mass shift is much larger than the field shift, and in order to extract the difference of charge radii, relating often back to the stable isotopes, one has to know the atomic structure and the nuclear masses with high accuracy [12].

Furthermore, a backbone of very well-known nuclides have been identified by the Atomic-Mass Evaluation (AME) [13], and high-accuracy mass values of suitable stable nuclides are of utmost importance as mass references for on-line mass measurements of radionuclides such as those performed at different radioactive beam facilities worldwide [14].

The literature mass value of ^{7}Li has a relative uncertainty of 11 ppb [13]. It has been derived from two input data, the mass of ^{6}Li measured with an uncertainty of 2.7 ppb in a Penning trap [15] and the Q value of the ^{6}Li$(n, \gamma)^7$Li reaction with 80 eV uncertainty [13]. However, a different Q value has been reported in the literature with 90 eV uncertainty [16], which would result in a greater than 100 ppb different ^{7}Li mass.

With the Penning-trap mass spectrometer SMILETRAP [17] the mass of ^{7}Li has been measured with a relative uncertainty of 0.63 ppb by comparing the cyclotron frequencies of ^{7}Li^{3+} and H$_2^+$. A large deviation of 14σ from

the literature mass [13] has been observed, having a significant effect of a few tens of kHz on the isotope shift calculations [12,18] and the determination of the nuclear-charge radius [7]. The uncovered deviation in the ^{7}Li mass corresponds to about 50% of the total error in the isotope shift for ^{6,8,9}Li and 20% for ^{11}Li. Since the uncertainties in the charge radius (r_c) of all these nuclides are dominated by the uncertainty in the charge radius of the reference nuclide (^{7}Li), each change of $r_c(^7$Li) is important.

In order to find the reasons for the deviation and to look for systematic effects the mass of ^{4}He^{2+} and ^{6}Li^{3+} have also been measured.

SMILETRAP is a double Penning-trap mass spectrometer located at the Manne Siegbahn Laboratory in Stockholm. Our facility has been described in detail elsewhere [17], thus only a brief description shall be given here, relevant for the measurement of the ^{7}Li mass.

The mass measurement is carried out via the determination of the cyclotron frequency, $\nu_c = qeB/(2\pi m)$, of ions stored in a homogeneous and stable magnetic field of a Penning trap. To have access to a wide variety of highly charged ions, an electron beam ion source (CRYSIS) in combination with an external ion injector is used [19]. To produce ^{7}Li^{3+} ions, first singly charged Li ions were created in the external ion source by evaporating LiBr$_3$ from an oven. The extracted singly charged ions were mass separated and then injected into CRYSIS for charge breeding. The injection time was 1.43 s, the confinement time, i.e., the time the ions are exposed to the electron impact inside the source, was 20 ms and the electron beam energy was 14.5 keV. The extracted ion pulse is transported to the double Penning-trap system by use of conventional ion beam optics. Before entering the cylindrical retardation trap (pretrap), the ions are charge state selected in a 90° double-focusing magnet. The pretrap is used to retard the ions from the transportation energy of typically $3.4q \cdot$ keV to ground potential within 30 ms. Then the ions are accelerated again to -1 keV and are transported to the hyperbolic precision Penning trap, where they are finally retarded to ground potential. An aperture with 1 mm diameter prevents ions with too large initial radii to enter the precision trap. In this last stage, the trapped ions are subject to an evaporation process by lowering the trap voltage from 5 to 0.1 V, leaving only the coldest ions in the trap. On average, not more than 1–2 ions are left in the precision trap after this procedure.

The precision Penning trap is located in the homogeneous magnetic field of a superconducting solenoid ($B = 4.7$ T). It consists of a ring electrode and two end-cap electrodes, all with hyperbolic geometry which create an electrostatic quadrupole field. In these fields the ion's motion can be described by three well-defined eigenmotions [20]: an axial motion with frequency ν_z, the so-called magnetron motion with frequency ν_-, and the modified cyclotron motion with frequency ν_+. The two radial frequencies obey the relation $\nu_c = \nu_- + \nu_+$ [21].

The ion's cyclotron frequency is probed directly by exciting the ion's motion by a quadrupolar radiofrequency signal (rf) and measurement of the time of flight to the micro-channel-plate detector placed on top of the magnet [17,21]. Repeating this for different rf frequencies near the true cyclotron frequency, ν_c, and measuring the time of flight as a function of the rf frequency, yields a characteristic time-of-flight cyclotron resonance curve [21]. In order to obtain the mass from the measured frequency, the magnetic field has to be calibrated. This is done by the measurement of the cyclotron frequency, ν_c^{ref}, of a reference ion with well-known mass, which is performed almost simultaneously.

The mass of the reference ion $m(\text{H}_2{}^+) = 2.015\,101\,497\,03(27)$ u has a relative uncertainty of 0.14 ppb [17]. $\text{H}_2{}^+$ is produced in the preparation trap by bombarding the rest gas with 3.4 keV electrons. The measurements on ^{7}Li^{3+} were performed by using a continuous excitation time T_{rf} of 1 s. A typical time-of-flight cyclotron frequency spectrum is shown in Fig. 1. The expected sidebands of the resonance [21] are suppressed. This is mainly due to the initial spread in the magnetron radius, since the ions are not cooled in the pretrap, and an incomplete conversion from magnetron to modified cyclotron motion during excitation.

The time-of-flight resonance curve of both the ion of interest and reference ion is measured with 21 equidistant frequency steps around the center of the resonance frequency. One scan, involving 21 frequency steps, takes about 40 s which is repeated twice. After two complete scans the settings are switched between the two ion species: the reference ion $\text{H}_2{}^+$ and the ion of interest ^{7}Li^{3+}. Switching between ion species takes only about 1 s, thus the total cycle time is shorter than 3 min. In this way effects from magnetic field drifts due to temperature or pressure fluctuations between two measurements can be reduced.
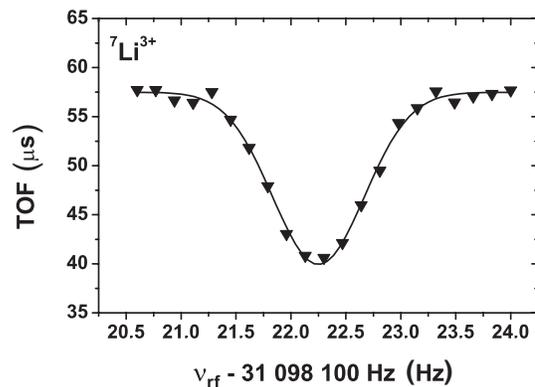

FIG. 1. The time-of-flight cyclotron frequency resonance of ^{7}Li^{3+} from 100 scans, representing only <10% of the overall data. The central part of the resonance is approximated with a Gaussian (solid line) in the data evaluation resulting in a FWHM of less than 1 Hz. The absence of well-pronounced sidebands is explained in the text.

TABLE I. The measured cyclotron frequency ratio R to the H_2^+ reference ion, and the determined atomic mass m_{exp}, which is compared to the value m_{lit} taken from AME2003 [13]. The error in R is only the statistical error, while m_{exp} includes the systematic uncertainties as well.

	^{7}Li	^{6}Li	^{4}He
R	0.861 847 167 21(31)	1.005 292 631 83(80)	1.007 171 503 45(53)
m_{exp}	7.016 003 425 6(45) u	6.015 122 890(40) u	4.002 603 253 3(26) u
m_{lit}	7.016 004 550 (80) u	6.015 122 795(16) u	4.002 603 254 15(16) u

The mass of the ^{7}Li^{3+} is obtained from the observed cyclotron frequency ratio of the two ion species:

$$R = \frac{\nu_1}{\nu_2} = \frac{q_1 m_2}{q_2 m_1}, \qquad (2)$$

where the subscript 1 denotes the Li ion and subscript 2 the H_2 ion.

Since the two frequency measurements are performed in similar ways, certain systematic uncertainties in the frequency ratio cancel to a large extent. This is, in particular, the case for ions which have the same q/A value [17]. The ^{7}Li^{3+} ion is close to this requirement having $q/A = 0.43$ compared to 0.5 for H_2^+. To obtain the atomic mass $m(^7$Li$)$, one has to correct for the missing q electrons, each with mass m_e, and their total binding energy E_B according to

$$m(^7\text{Li}) = m(^7\text{Li}^{3+}) + q m_e - E_B, \qquad (3)$$

where $m(^7$Li$^{3+})$ is the experimentally determined ion mass obtained using Eq. (2). The electron mass is 5.485 799 094 5(24) $\times 10^{-4}$ u with a relative standard uncertainty of 4.4×10^{-10} [22]. The error introduced by the electron mass can be neglected. The total electron binding energy is calculated by summing up the relevant experimental ionization energies tabulated in [23].

The results of the three data taking periods are summarized in Table I. The table gives the resulting frequency ratio R of the ion of interest relative to the reference ion H_2^+, as well as the atomic mass of the studied nuclide, and compares them with the values given in the literature [13]. The uncertainty of the ^{7}Li mass includes the relative statistical uncertainty (0.4 ppb) and the relative overall systematic uncertainty (0.52 ppb). The dominant part of the latter comes from relativistic effects, ion-number dependency, and the q/A asymmetry. The different contributions to the systematic uncertainty are listed in Table II, and were

TABLE II. Systematic uncertainty budget for ^{7}Li in units of ppb.

Reference mass	0.18	Binding energy (E_B)	0.1
Relativistic effect	0.2	ion-number dependence	0.25
q/A asymmetry	0.33	contaminating ions	0.1
Total systematic	0.52		

estimated using the procedures described in Ref. [17]. At the time of the ^{6}Li run, we were affected by an uncontrollable internal helium leak in CRYSIS which had not been present while running ^{7}Li. Since the ^{6}Li^{3+} and ^{4}He^{2+} are q/A doublets, unwanted ^{4}He^{2+} ions were present in the beam, and in the trap, mixed together with ^{6}Li ions, leading to the large systematic uncertainty in the mass of ^{6}Li.

A comparison of our result for the ^{7}Li atomic mass with previous data is shown in Fig. 2. The AME83 [24] value of ^{7}Li is based on a reaction energy and has an uncertainty of 0.8 μu. Similarly, the AME93 [25] value, which is derived from a ^{7}Li$(p, n)^7$Be reaction Q value, has an uncertainty of 0.5 μu. The most recent AME2003 value has a much reduced uncertainty of only 0.08 μu. In this case, the mass of ^{7}Li has been derived using as input data the mass of ^{6}Li and the ^{6}Li$(n, \gamma)^7$Li reaction Q value [13]. The ^{7}Li mass value from AME2003 deviates significantly (>1 μu) from our result, which means that at least one of the two input data used to derive the ^{7}Li mass must be wrong.

Different Q values for the latter reaction exist in the literature [16,26–28]; see Fig. 3. Note that the Q value from 1985 [16] deviates by about 1 keV from the value in AME2003 [13] which is claimed to be based upon recalibrated data from Ref. [16] and the recent data from Ref. [28]. Furthermore, the work in Ref. [28] is not pub-
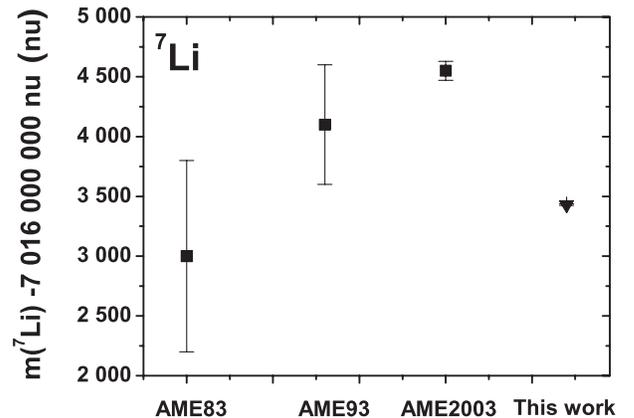

FIG. 2. Comparison of the atomic mass value of ^{7}Li from the measurement reported here with previous results published in the Atomic-Mass Evaluations AME83 [24], AME93 [25], and AME2003 [13]. The recent AME2003 value deviates by 160 ppb from our measurement.
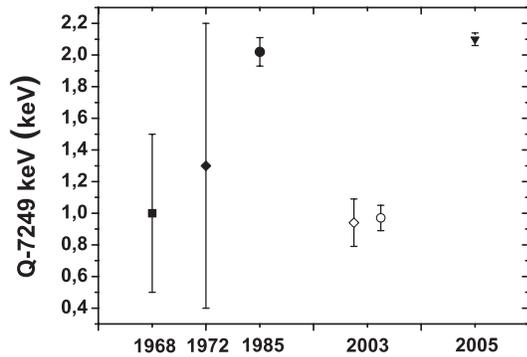
FIG. 3. The Q value of the ^{6}Li$(n, \gamma)^7$Li reaction in chronological order. The values are 1968 from Ref. [26], 1972 from Ref. [27], 1985 from Ref. [16]. The first point at 2003 is from Ref. [28] and the second from Ref. [13]. The 2005 data are the values derived from our direct mass measurements using ^{6}Li and ^{7}Li.

lished. The mass of ^{6}Li is known to 2.7 ppb uncertainty [15]. However, to shed light upon this large deviation we have measured the mass of the ^{6}Li and found an agreement within 2.4σ compared to the literature value (Table I). Using our mass values for ^{6}Li and ^{7}Li reported here, a Q value of 7251.10(4) keV is derived. For the 2003 ^{7}Li mass calculation a Q value of 7249.97(8) keV [13] has been used which deviates by more than 1 keV from our result, and can explain the large discrepancy observed. Note that the Q value derived from our mass measurement is in agreement with the Q value from Ref. [16] of 7251.02(9) keV.

The excellent agreement of our simultaneously measured ^{4}He mass with the literature value gives further confidence in the ^{7}Li mass value reported here, where both measurements are at exactly the same level of precision.

Summarizing, the result of a high-accuracy atomic-mass measurement of ^{7}Li with the Penning-trap mass spectrometer SMILETRAP has been reported. The mass of ^{7}Li was measured directly with unprecedented precision and the result has been compared to previous published mass values. Our value deviates by 1.1 μu compared to the AME2003 value which seems to be due to a wrong ^{6}Li$(n, \gamma)^7$Li reaction Q value used in the latest atomic-mass evaluation [13]. The new mass value for ^{7}Li is an important input parameter for transition isotope shift and nuclear-charge radii measurements of the Li isotopes [7,10,11]. It can also be used as reference mass for calibration purposes in high-accuracy Penning-trap mass spectrometry of short-lived nuclides [1]. Furthermore, in the evaluation of the masses of ^{7}Be and ^{8}Li, the mass of ^{7}Li is used as input parameter [13].

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