

Penning-trap mass measurement of ${}^3\text{He}$ O. Bezrodnova¹, S. Sasidharan¹, W. Quint², S. Sturm¹, and K. Blaum¹¹Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany²GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstraße 1, 64291 Darmstadt, Germany

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We have determined the atomic mass of ${}^3\text{He}$ to 12 parts per trillion relative precision from a measurement of the cyclotron frequency ratio of ${}^3\text{He}^+$ and ${}^{12}\text{C}^{4+}$ in a Penning trap. From the obtained mass of the ${}^3\text{He}^+$ ion, the mass of the neutral atom can be calculated without loss of precision: $m({}^3\text{He}) = 3.016\,029\,322\,011(35)$ u. This result provides independent, high-precision data for the last missing link between the masses in the regime of light ions, thereby resolving discrepancies in the reported masses of light atomic nuclei from various experiments.

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The field of precision mass spectrometry experienced a significant leap forward in the 1980s when single ions, specifically protons, were isolated in a Penning trap [1,2]. Since then the field has advanced remarkably. Today, the leading experiments achieve relative precision as low as a few parts per trillion (ppt) [3–5] for mass values and their ratios [6,7] required in various aspects of fundamental physics. The masses of the lightest nuclei hold particular significance in this context. Precise determinations of the intrinsic properties of the fundamental constituents of the universe, such as hydrogen and helium isotopes, as well as the neutron and electron, are essential for testing the validity of the current understanding of fundamental physics described by the standard model [8].

One example involves the relation between the Rydberg constant R_∞ and the reduced mass of the electron in a hydrogen atom, where the proton-to-electron mass ratio enters [9]. Until recently, the relative atomic mass of the electron, $A_r(e)$, was determined from the ratio of spin-precession and cyclotron frequency measurements on hydrogen-like ${}^{12}\text{C}^{5+}$ in a Penning trap [10,11]. The state-of-the-art experimental techniques employed on lighter hydrogen-like ions, such as ${}^4\text{He}^+$, should enable a reduction in the uncertainty of the electron mass, provided that the ion's mass is known with sufficient precision [12]. Another application for light masses addresses the rest mass of the neutrino. Although presumed to be zero by the standard model, it has been shown to have a finite mass through the solar neutrino oscillation experiments [13,14]. The most direct laboratory method for probing the absolute mass of the antineutrino is through the study of the tritium β -decay spectrum near its endpoint. This is conducted in experiments such as KATRIN [15,16] and Project-8 [17]. A Penning-trap measurement of the Q value of the decay,

given by the mass difference between T and ${}^3\text{He}$, provides an independent cross-check of the systematic effects [8,18,19].

The masses of light nuclei, namely the proton (p), deuteron (d), triton (t), helion (he), and the alpha particle (α), are regarded as fundamental constants. The most precise mass measurements of these nuclei are conducted in Penning traps through the measurement of the ratio of cyclotron frequencies (R_{CF}) of two charged particles. This technique interconnects the masses and enables redundancy checks. Such tests have uncovered inconsistencies among the masses of ${}^3\text{He}$ obtained in various mass spectrometers in the past. This disagreement is recognized in the literature as the “light ion mass puzzle” [3,20]. The discrepancy can be elucidated by examining $\Delta = m_p + m_d - m_{he}$, which represents the proton separation energy in the helion nucleus. Experimentally, this value can be obtained in two distinct ways; see Fig. 1. The masses m_p , m_d , and m_{he} can be directly measured, using ${}^{12}\text{C}$, the standard of atomic mass units (u), as a reference ion. Such measurements were conducted in the past by the group at the University of Washington (UW) [21,22]. Another approach

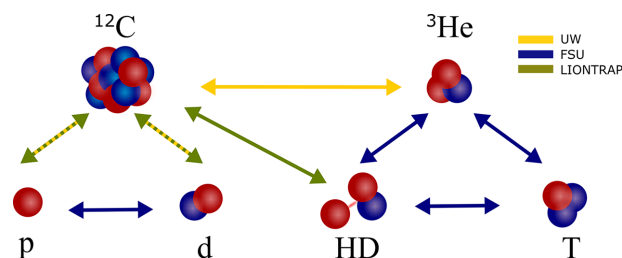


FIG. 1. The puzzle of light-ion masses. The arrows indicate the cyclotron frequency ratios measured by various Penning-trap mass spectrometers prior to this work. Measurements using a carbon ion as reference are performed by the UW [21,22] (yellow) and the LIONTRAP (green) [3,20,23] groups. The blue links are measured by the FSU group [7,18,19,24,37]. The comparison of $\Delta = m_p + m_d - m_{he}$ obtained from direct measurements against carbon and FSU results currently show a 4.8σ discrepancy, with the mass ratio of ${}^3\text{He}$ and ${}^{12}\text{C}$ being the primary candidate for the remaining inconsistency.

involves measuring the ratio of ${}^3\text{He}$ to HD^+ and utilizing the binding energy of the molecular ion HD^+ and the mass of the electron, both of which are known with sufficient precision [11]. Such measurements were carried out by the group at Florida State University (FSU) [18]. The comparison of the results from these two groups showed a deviation at the 4.2σ (combined standard deviation) level: $\Delta_{\text{UW}} - \Delta_{\text{FSU}} = 860(210)$ pu. The identified discrepancy raised questions about the reliability of these values and motivated independent measurements. The LIONTRAP experiment was established to remeasure the masses of light ions relative to ${}^{12}\text{C}$. We have measured the masses m_p [20,23], m_d , and HD^+ [3] at this facility, showing consistency with the direct measurement of the mass difference between the molecular ion H_2^+ and d at the FSU facility [7,24]. Additionally, LIONTRAP's measurements showed reasonable agreement with laser spectroscopy experiments that determine transition frequencies in the rovibrational spectrum of the molecular ion HD^+ , providing the ratios of the involved masses [25–28]. As of now, due to the reduced uncertainty in the absolute mass values from the latest FSU [19] and LIONTRAP results [3,20], the deviation in the value of Δ is 4.8σ : $\Delta_{12\text{C}} - \Delta_{\text{FSU}} = 263(61)$ pu. The primary candidate for the remaining deviation is the mass ratio between ${}^3\text{He}$ and ${}^{12}\text{C}$.

While the mass of the α -particle does not directly contribute to the described puzzle, due to the persistent discrepancies between the LIONTRAP and UW groups, the mass of the ${}^4\text{He}$ nucleus was also measured at LIONTRAP [29]. This measurement revealed a 6.6σ deviation from the literature value [11], given by the measurement of the UW group [2]. This provided strong motivation to measure the mass ratio of ${}^3\text{He}$ to ${}^{12}\text{C}$ and thus to perform an independent check.

Our measurement principle relies on comparing the cyclotron frequencies of two ions placed in the same, mostly homogeneous magnetic field B , generated by a superconducting magnet. In the plane perpendicular to the “axial” magnetic field lines, a particle with charge q and mass m is forced into motion with the cyclotron frequency [30]:

$$\omega_c = \frac{qB}{m}. \quad (1)$$

Static voltages applied to cylindrical electrodes superimpose an electrostatic quadrupole potential, which provides confinement of the particle. The circular motion is modified by the applied electrostatic potential, resulting in three distinct eigenmotions. Two of these occur in the radial plane, perpendicular to the magnetic field lines: the modified cyclotron motion, characterized by the frequency ω_+ and the slow $\vec{E} \times \vec{B}$ drift with the magnetron frequency ω_- . The third axial eigenmotion is a harmonic oscillation along the magnetic field lines, related to the strength of the electrostatic potential U : $\omega_z \propto \sqrt{\frac{qU}{m}}$. The invariance theorem [31] enables the determination of the true cyclotron frequency when all three eigenfrequencies are measured even in the presence of certain unavoidable field imperfections, thus establishing a relation to the mass of the particle:

$$\omega_c = \sqrt{\omega_+^2 + \omega_z^2 + \omega_-^2}. \quad (2)$$

To eliminate the magnetic field B from Eq. (1), Penning-trap mass spectrometry relies on measuring the ratio of cyclotron frequencies for two different species. In LIONTRAP, the carbon ion serves as a reference due to its standardized mass in atomic mass units. To determine the mass of the particle of interest, such as ${}^3\text{He}^+$, with ${}^{12}\text{C}^{4+}$ as the reference, the following relation is employed:

$$m({}^3\text{He}^+) = m({}^{12}\text{C}^{4+}) \times \frac{\omega_c({}^{12}\text{C}^{4+})}{\omega_c({}^3\text{He}^+)} \times \frac{1}{4}. \quad (3)$$

The axial motion functions as an observational window into all modes of the ion within the trap. It can be directly detected by connecting an LC -resonance circuit to one of the off-central electrodes of the trap within the cryogenic environment of the experiment. When an ion oscillates in the axial direction, it induces an image current on the neighboring electrodes. When the LC circuit resonates with the frequency of the particle, they exchange energy and the ion is consequently cooled to thermal equilibrium with the circuit. Then, the thermal noise of the resonance circuit is effectively shorted, resulting in a characteristic “dip signal.” A dedicated line-shape fit enables the direct acquisition of the axial frequency. The radial modes can be determined by dressing the axial motion with any of the radial motions using a radio frequency sideband drive [30,32]. To measure the modified cyclotron frequency, which is the fastest of all eigenfrequencies and thus requires the highest precision, a phase-sensitive “pulse and amplify” (PnA) technique is employed [10].

The charge states of the particles are selected such that the charge-to-mass ratio is nearly identical for both ions. This way, we can use the same voltage configuration for both particles, placing them in the same location within the trap and allowing the use of a single axial detector for both ions. The remaining difference in frequencies is compensated by fine tuning the resonator's frequency using voltage-controlled capacitors. To compensate for the low charge of the ${}^3\text{He}^+$ ion, a new axial detection system was developed, which includes a toroidal superconducting coil and a cryogenic amplifier [33]. This upgrade resulted in an increase in the Q value of the detection system to about 6000, which is more than twice the value compared to the system used in the previous measurement campaigns [3,29].

The LIONTRAP setup comprises a Penning-trap stack composed of five interconnected traps [20]. Three traps are specifically dedicated to precision measurements, namely two storage traps and the precision trap. Ideally, cyclotron frequency measurements for both ions should be performed simultaneously at the same place to cancel out the effects of the spatial and temporal variations of the magnetic field. While this is not exactly possible due to the Coulomb repulsion of the ions, two ions of similar mass can be crystallized on a small common magnetron orbit [6,7,34]. However, due to the large mass mismatch of the ${}^3\text{He}^+$ and ${}^{12}\text{C}^{4+}$, we have employed a fast shuttling technique. While the cyclotron frequency of one ion is being measured in the precision trap, the other ion is stored in one of the storage traps. The ions are then alternately shuttled between the precision and storage traps to minimize the time between modified cyclotron frequency measurements for both ions [29].

The trap tower is enclosed within a sealed chamber and positioned inside a superconducting magnet. Along with cryopumping, the sealing enables a near-perfect vacuum with pressures better than 10^{-17} mbar, allowing for ion storage times of several months. However, this setup necessitates the *in situ* production of ions within the trap tower. This is particularly challenging for ^3He , which remains gaseous down to 3.3 K. Therefore, it must be confined within a closed volume and released into the trap tower on demand for ion production. For this purpose, a specialized two-chamber source utilizing charcoal as an adsorption agent was designed [35]. It was initially used to produce ^4He ions and later repurposed for ^3He . Additionally, a fused silica sphere filled with 50 mbar of ^3He gas was incorporated into the production section of the trap tower. A temperature-dependent permeability of the glass allows helium atoms to be released only when the sphere is heated significantly above 4.2 K by applying a voltage to resistors attached to it [36]. The free atoms are then ionized by an electron beam emitted from a field emission point. Both the chamber and glass sphere sources were successfully used to produce $^3\text{He}^+$ ions. Carbon ions were produced by ablating atoms from a solid-state target with the electron beam.

The measurement sequence comprises recurring cyclotron frequency ratio measurements:

$$R_{\text{CF}} = \frac{\omega_c(^{12}\text{C}^{4+})}{\omega_c(^3\text{He}^+)}. \quad (4)$$

One R_{CF} denotes a measurement cycle. These cycles are grouped into runs, which are temporally limited by the requirements of experiment maintenance, specifically the refilling of cryogenic liquids into the reservoirs of the magnet and experimental apparatus. In various runs, the excited amplitudes r_+ of the modified cyclotron motion for both ions, which depend on the excitation strength, are varied from 14.6 to 83.4 μm . The variation of the amplitudes allows for the extrapolation of energy-dependent frequency shifts during the phase sensitive PnA measurement to the zero energy by fitting a plane to a number of excitation strength combinations [20]. Two ion pairs in opposite spatial order in the trap tower were used to ensure the absence of transport-related systematic effects and frequency shifts arising from potentially unnoticed contaminant ions. The surface fit, illustrated in Fig. 2, yields a value for the R_{CF} with the statistical uncertainty given in parentheses:

$$R_{\text{CF}}^{\text{stat}} = 1.005\,344\,079\,966\,8(101). \quad (5)$$

This extrapolated value still requires correction for various systematic effects, summarized in Table I. A significant shift arises from the finite temperature of the ion, resulting in a relativistic mass increase. This shift, which scales with q/m^2 , is particularly pronounced for the light ^3He ion. Prior to each cycle the ions' axial motion is cooled through thermalization with the tank circuit. This temperature, equal to that of the surrounding cryogenic environment, is further reduced through negative electronic feedback. This way, the axial temperature of a ^3He ion amounts to $T_z = 1.3(3)$ K. The temperature of the modified cyclotron motion, which mainly contributes to the relativistic shift, is decreased by coupling it to the axial mode with a radio-frequency excitation drive [32].

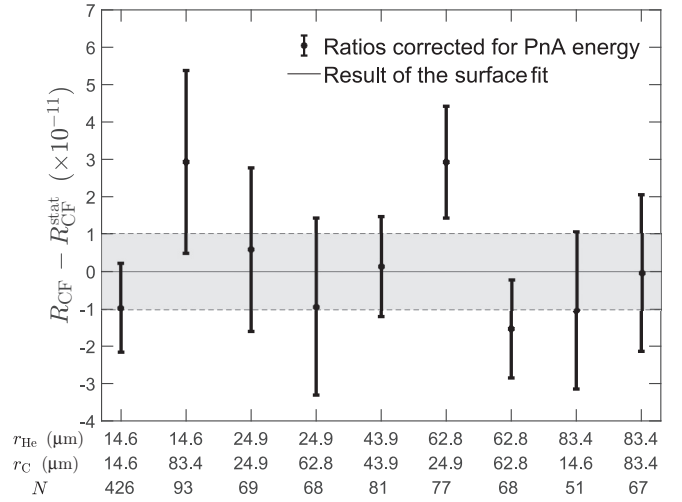


FIG. 2. The averaged R_{CF} residuals, grouped by equal excitation strength. A three-parameter fit allows the correction of R_{CF} to zero-excitation amplitude. On the x axis, the corresponding excited cyclotron radii of helium (r_{He}) and carbon (r_{C}) and the number of cyclotron frequency ratios (N) in each point are given. The error bars denote the standard error of the mean. The grey band denotes the 1σ uncertainty for the fitted frequency ratio.

The thermal oscillation amplitude contributes to the frequency shift due to the residual quadratic component of the magnetic field. The superconducting shim coil [3] minimizes the corresponding uncertainty of R_{CF} down to 0.9×10^{-12} .

Another significant effect is the back action of charges induced by the ion on the electrodes. This phenomenon, known as the image charge effect, was studied in [38], where a numerical simulation of the effect for the particular trap geometry in the LIONTRAP setup was experimentally tested to a relative precision of 5%, which is set as an uncertainty of the frequency shift.

The seven-electrode design of the precision trap enables an extremely harmonic electrostatic potential, effectively canceling out the leading-order anharmonicities [20]. Consequently, the associated uncertainties are in the order of 10^{-13} on R_{CF} .

The dominant systematic uncertainty in the recent LIONTRAP measurement campaigns was the image current effect, also known as the “dip line shape” [3,29]. This effect stems

TABLE I. Relative shifts and uncertainties in R_{CF} (in ppt) due to systematic effects. $R_{\text{CF}}^{\text{corr}}$ denotes the ratio corrected for systematic shifts.

Effect	Relative shift $\left(\frac{R_{\text{CF}}^{\text{corr}} - R_{\text{CF}}^{\text{stat}}}{R_{\text{CF}}^{\text{stat}}}\right)$ (in ppt)	Relative uncertainty (in ppt)
Image charge	73.88	3.69
Relativistic effect	-1.15	0.27
Magnetic inhomogeneity	0.18	0.92
Electrostatic anharmonicity	0	0.16
Axial frequency	0	4.02
Total	72.91	5.54

from the interaction of an ion with the LC circuit. If the detection circuit is not perfectly tuned to the ion's axial frequency, the latter is "pushed," resulting in a systematic shift. While this effect is nominally corrected for by the line-shape model employed to fit the dip spectra, the model needs the exact parameters of the resonator as input parameters. The uncertainty of these parameters introduces an error in the corrected axial frequency, which is subsequently transferred to the cyclotron frequency via Eq. (2). The resonator parameters are determined regularly in independent measurements. We have doubled the number of such determinations compared to previous campaigns to reduce the impact of small random variations. Additionally, we implemented a new analysis approach, which takes advantage of the similarity of the charge-to-mass ratios of both ions, and so eliminated the back action of the LC circuit as a dominant systematic effect. Initially, R_{CF} is determined in the classical way by obtaining ω_z of both particles through fitting the dip spectra. The obtained ratio is used to calculate a new set of $\tilde{R}_{CF}$, where the axial frequency of one arbitrarily chosen particle is still obtained from the dip measurement (ω_z^{ion1}), while for the other particle it is calculated from the initial cyclotron frequency ratio and ω_z^{ion1} via the equation

$$\omega_z^{\text{ion2}} = \omega_z^{\text{ion1}} \sqrt{\frac{\omega_c^{\text{ion2}}}{\omega_c^{\text{ion1}}}}. \quad (6)$$

This relation is valid under the assumption that both ions are placed in the exact same electrostatic potential, and allows for the suppression of the effect of image current shift on R_{CF} by a factor of $(R_{CF} - 1)^{-1} > 100$. R_{CF} and $\tilde{R}_{CF}$ agree within their uncertainties. To support the approach, we ensured that there were no significant systematic potential differences present during the measurement. While the nominal voltages are identical for both ions, we investigate possible drifts connected to the transport process. We evaluate modified cyclotron frequency data by excluding a number of phase measurements performed in a close time proximity to the transport of ions, when the trap voltages are being changed [29]. From comparison of the resulting cyclotron frequency ratios, a conservative relative uncertainty is estimated to be 4×10^{-12} .

Since the magnetron frequency is the smallest of the eigenfrequencies, its uncertainty has the least impact on the cyclotron frequency. For the final R_{CF} , magnetron frequencies are calculated from axial and modified cyclotron frequencies: $\omega_- = \frac{\omega_z^2}{2\omega_+}$. Although this relationship holds true only for the ideal trap, not accounting for imperfections such as ellipticity and tilt of the experiment, the measured and calculated values deviate by less than 150 mHz, contributing only 10^{-14} to R_{CF} .

Given the systematic shifts and uncertainties considered, the final corrected cyclotron frequency ratio is

$$R_{CF}^{\text{fin}} = 1.005\,344\,080\,040\,1(101)_{\text{stat}}(56)_{\text{sys}}(115)_{\text{tot}}. \quad (7)$$

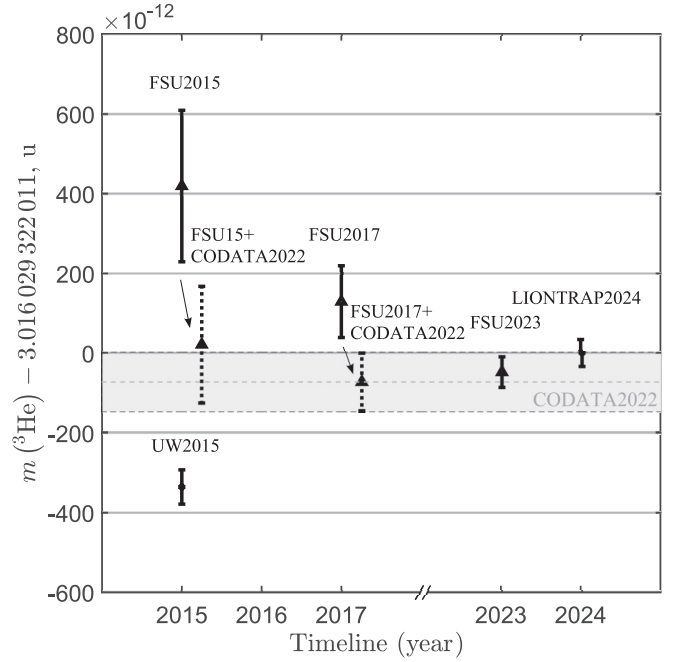


FIG. 3. High-precision values of the atomic mass of ${}^3\text{He}$. The initial discrepancy between FSU2015 [18] and UW2015 [22] is 3.9σ . The result FSU2017 is obtained using the $m({}^3\text{He})/m(\text{HD})$ ratio from [37], the $m(\text{T})/m({}^3\text{He})$ ratio from [18], the deuteron mass from [22], and the proton mass from [23]. The FSU2017 and the FSU2015 results are also recalculated using current literature values for m_p and m_d from CODATA2022 [39]. The corrected FSU2015 and FSU2017 values, as well as the latest measurement FSU2023 [19], are consistent with the result of this work LIONTRAP2024. The results LIONTRAP2024 and UW2015 show a discrepancy of 6.1σ .

Using Eq. (3), we now derive the mass of ${}^3\text{He}^+$:

$$m({}^3\text{He}^+) = 3.015\,480\,768\,496(30)_{\text{stat}}(17)_{\text{sys}}(35)_{\text{tot}} \text{ u}. \quad (8)$$

The masses of the nucleus and the neutral atom can be calculated using the electron mass [39] and electron binding energies [40] without loss of precision:

$$m({}^3\text{He}^{2+}) = 3.014\,932\,247\,004(30)_{\text{stat}}(17)_{\text{sys}}(35)_{\text{tot}} \text{ u}, \quad (9)$$

$$m({}^3\text{He}) = 3.016\,029\,322\,011(30)_{\text{stat}}(17)_{\text{sys}}(35)_{\text{tot}} \text{ u}. \quad (10)$$

The comparison of the mass of the neutral atom ${}^3\text{He}$, as measured by different experiments, is depicted in Fig. 3.

While the LIONTRAP result agrees well with the FSU results [18,19,37], updated using the latest proton and deuteron masses [39], it reveals a 6.1σ deviation from the UW result [21,22]. The literature CODATA2022 value [39], primarily given by the corrected FSU2017 measurement [37], is in agreement with the result of this work.

The inconsistency in the measurements of light ion masses is visually represented in Fig. 4 by tracking the change in the value of Δ as measured by different experiments. Analysis of various measurements suggests that the results from LIONTRAP and FSU are mutually consistent and deviate from the results obtained by the UW group. This interpretation is further supported by the measurement of the mass of ^4He , which, although not directly part of the puzzle, aligns with the trend observed [29].

This work concludes a series of measurements on light ion species conducted at the Penning-trap mass spectrometer LIONTRAP. Further mass measurements of light ions are planned at the PENTATRAP experiment [41].

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The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

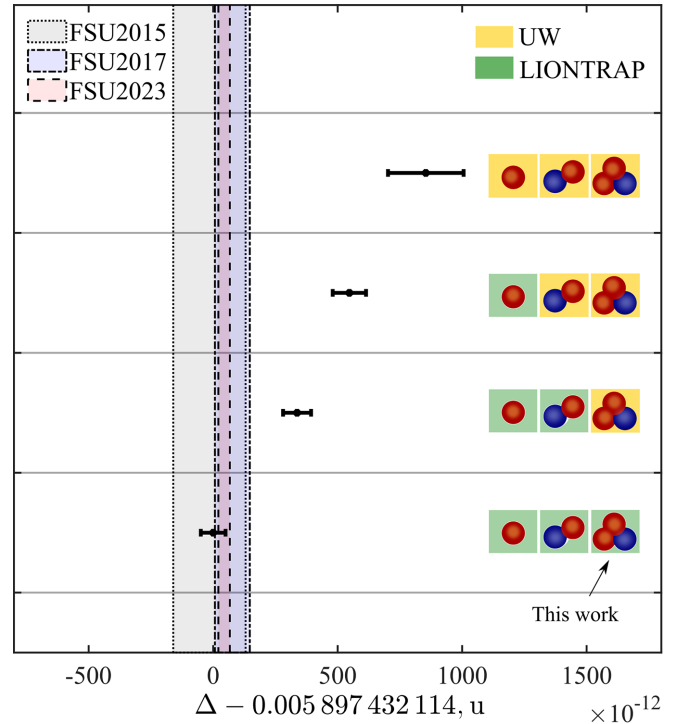


FIG. 4. The value of $\Delta = m_p + m_d - m_{\text{he}}$, obtained by different groups. Points with error bars represent the measurements with ^{12}C as a reference ion. The diagram on the right side indicates which group measured the mass of the respective ion (yellow: UW [21,22]; green: LIONTRAP [3,20]). Grey, blue and red bands are results of the FSU group from 2015, 2017, and 2023, respectively [18,19,37]. While combinations that include UW results are inconsistent, the value determined solely from previous LIONTRAP results [3,20], as well as from this work, agree with the FSU group's results within 1σ .

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