

Towards high-accuracy mass spectrometry of highly charged short-lived ions at ISOLTRAP

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Dedicated to H.-J. Kluge on the occasion of his 65th birthday.

Abstract

Multiply charged ions of stable xenon isotopes from a plasma ion source have been mass-selected by the on-line mass separator ISOLDE/CERN and delivered to the triple-trap mass spectrometer ISOLTRAP. The doubly charged ions that survived the charge-exchange processes during bunching and ion preparation were transferred to a precision Penning trap for mass determination. Mass values were obtained for the isotopes with mass numbers $A = 126, 129, 130, 136$. They are consistent with previous results except for the case of ^{126}Xe where a significant deviation from the literature value was found. The performance of ISOLTRAP is analyzed with respect to a future application of highly charged ions for mass determination of short-lived radionuclides at ISOLDE.

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1. Introduction

Atomic masses are required in a variety of physics branches. They are of great importance in nuclear physics, where nuclear masses are essential for tests of mass models and theories that describe nuclear structure [1]. A large database of precise values is needed especially for the prediction of atomic masses of short-lived nuclides far away from the valley of stability. Atomic masses are also used as input values for astrophysical models that describe the processes of nucleosynthesis, e.g., in stellar ex-

plosions, where new elements and isotopes are formed by rapid neutron or proton capture.

The relative mass uncertainty required depends on the respective application. For example, $\delta m/m \leq 1 \times 10^{-8}$ is needed for a contribution to the test of the conserved-vector-current hypothesis and the unitarity of the Cabibbo-Kobayashi-Maskawa quark-mixing matrix of the Standard Model [2]. Penning traps can provide mass values at this level of precision [3,4]. For this purpose, the cyclotron frequency [5]

$$\nu_c = \frac{qB}{2\pi m} \quad (1)$$

of an ion with mass m and charge q is measured in a strong and homogeneous magnetic field B . The resolving power $R = \nu_c/\Delta\nu_c$ is related to the observation time T_{obs} of the ion motion

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in the Penning trap via the relation $\Delta\nu_c \propto 1/T_{\text{obs}}$ [6]. Thus, a larger resolving power R can be obtained with either a longer observation time or a higher frequency. For short-lived nuclides the half-life limits the observation time and therefore the frequency remains the only variable parameter.

Although magnetic fields as large as 21 T can be achieved with present superconducting magnets [7], their homogeneity, which is suitable for NMR spectroscopy, might not be sufficient for high-accuracy mass spectrometry. In addition, frequency drifts of the order of a few Hz per hour [7] are too large. Last but not least, the magnets with the highest field amplitudes are also the most expensive. It is much more efficient to increase the cyclotron frequency by increasing the charge state of the ions according to Eq. (1). With an electron-beam ion trap/source (EBIT/EBIS) [8], where ions are ionized in an intense electron beam, up to fully stripped ions for mid-range proton number Z can be provided within tens of milliseconds charge-breeding time.

In addition to the observation time the number of recorded ions N also contributes to the statistical relative mass uncertainty, according to the relation [6]

$$\frac{\delta m}{m} \propto \frac{1}{R\sqrt{N}}. \quad (2)$$

Fig. 1 shows the relative mass uncertainty as a function of the observation time and the number of recorded ions for the two charge states $z = +1$ and $z = +20$ of ^{131}Xe . Note that keeping the observation time fixed, the application of highly charged ions allows the same relative statistical uncertainty for a much smaller number of ions, i.e., a more efficient use of the radioactive ion beam is possible.

The advantage of highly charged ions is already employed at the SMILETRAP experiment [9], where stable nuclides are charge-bred in an electron-beam ion source before they are transferred to a Penning trap for mass determination. Precise mass measurements, for example on $^{24,26}\text{Mg}$ with relative mass uncertainties of 5.8×10^{-10} and 1.3×10^{-9} were achieved, respectively [10].

At the ISOLDE facility [11] an electron-beam ion source (REXEBIS) [12] is installed for the charge breeding of radionuclides which are further accelerated with the REX-ISOLDE fa-

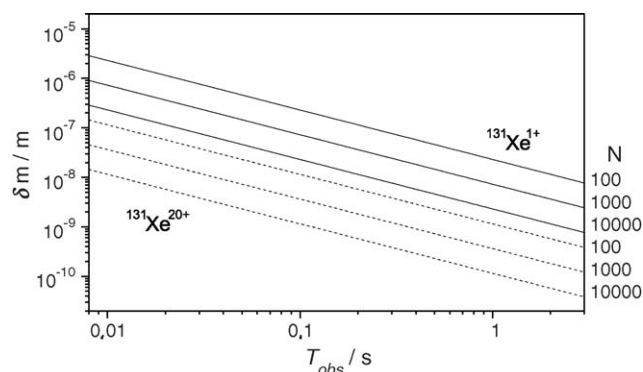


Fig. 1. Relative statistical mass uncertainty $\delta m/m$ according to Eq. (2) for $^{131}\text{Xe}^{1+}$ (solid lines) and $^{131}\text{Xe}^{20+}$ (dashed lines) as a function of the observation time T_{obs} and for different total ion numbers N .

cility for nuclear spectroscopy and solid state physics experiments [13]. Another charge breeding device, an electron cyclotron resonance (ECR) ion source, is currently under investigation [14]. Thus, highly charged radioactive ions might be accessible for the ISOLTRAP mass spectrometer in the near future. As a first test the transfer of multiply charged xenon ions to the ISOLTRAP mass spectrometer has been investigated and mass measurements of doubly charged xenon ions have been carried out.

2. Experimental setup

The mass spectrometer ISOLTRAP employs three ion traps for the preparation and high-accuracy mass measurement of short-lived radionuclides (see Fig. 2). The description of the experimental setup and an overview of performed mass measurements can be found in [15–17]. In the first trap, a linear radiofrequency quadrupole (RFQ) structure, the 60 keV radioactive ion beam from the ISOLDE target/ion-source is stopped, cooled, and bunched in a helium buffer-gas environment [17]. The following preparation Penning trap [18] allows further cooling and selection of ions by removing unwanted isobaric contaminants with a buffer-gas cooling technique [19]. The purified bunch of ions is finally transferred to the precision Penning trap, where the mass determination is performed with a time-of-flight (TOF) cyclotron-resonance detection technique [20].

To this end, the low-frequency magnetron motion of the stored ions is converted by application of a quadrupolar radiofrequency (rf) excitation into the high-frequency cyclotron motion. In the case of resonant excitation, $\nu_{\text{rf}} = \nu_c$, a full conversion occurs and the ions gain radial kinetic energy since the cyclotron frequency is several orders of magnitude higher than the magnetron frequency. Upon ejection of the ions towards a detector, the ions slowly drift through the magnetic field gradient and the energy in the radial motion is converted into an axial acceleration, i.e., resonantly excited ions arrive earlier at the detector than ions excited off-resonance. With a frequency scan a cyclotron resonance is obtained, as shown for $^{131}\text{Xe}^{2+}$ in the inset of Fig. 2. The resonance curve is Fourier limited; the solid line is a fit of the expected line shape to the data points [21]. The frequency ν_c of the unknown nuclide is compared to the frequency $\nu_{c,\text{ref}}$ of a reference ion with well-known mass, usually taken from the stable alkali reference ion source of ISOLTRAP. The frequency ratio finally yields the mass ratio and thus the mass of the nuclide of interest.

In the present investigation the multiply charged xenon ions were produced in a plasma ion source, part of an ISOLDE target. The triply and quadruply charged xenon ions were first selected by setting the separator magnets to the corresponding mass-to-charge ratio and then sent to the ISOLTRAP setup. Micro-channel-plate (MCP) detectors behind the buncher, the preparation Penning trap, and the precision trap were used to monitor the resulting ion ensemble after each section in order to investigate charge-exchange processes in the ion traps. At the RFQ buncher and the preparation Penning trap the helium buffer-gas pressure readings were 5×10^{-6} and 1.2×10^{-6} mbar, respectively.

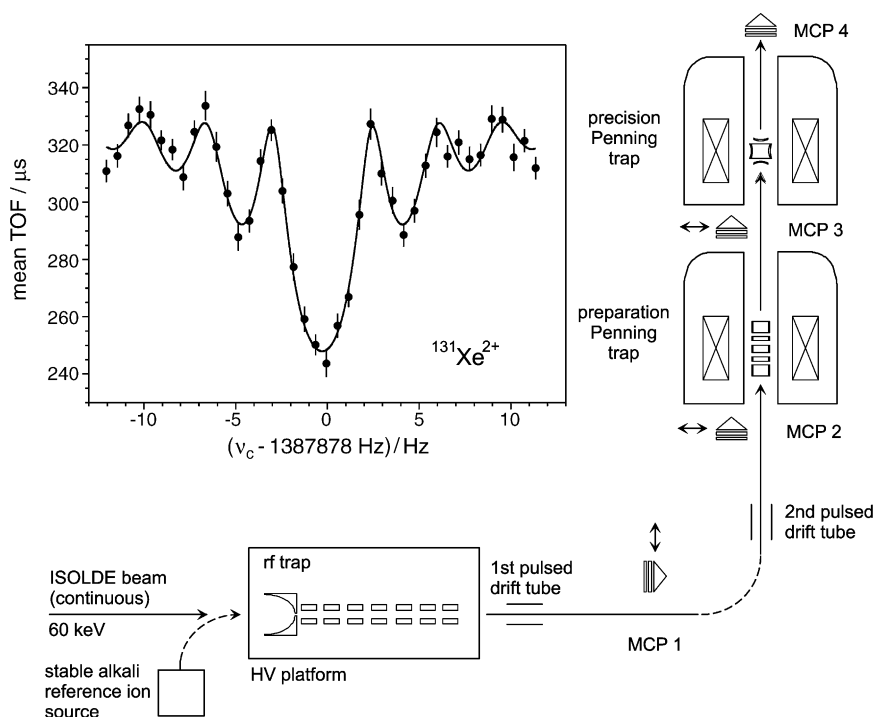


Fig. 2. Sketch of the experimental setup of ISOLTRAP. The inset shows a cyclotron resonance of $^{131}\text{Xe}^{2+}$.

Note that the pressure inside the preparation trap and the RFQ buncher is one to two orders of magnitude higher than measured by the pressure gauges, which are mounted outside the high-field region of the superconducting magnet and the buncher, respectively. The precision Penning trap is separated from the buffer-gas environment by a differential pumping stage and a pressure of the order of 10^{-8} to 10^{-9} mbar is achieved (also measured with a pressure gauge outside the high magnetic field).

3. Charge exchange

3.1. Results

In the RFQ buncher the ions are stored for 8 ms before they are ejected towards the preparation Penning trap. Immediately behind the buncher the ions can be monitored with a MCP detector (MCP 1). As an example, the TOF spectra after injection and storage of $^{131}\text{Xe}^{4+}$ and $^{136}\text{Xe}^{3+}$ are shown in Fig. 3. In both cases the charge states $z = +2$ and $z = +1$ as well as N_2^+ ions are detected, but no $z = +3$ ions are observed (the small signal at $10.1 \mu\text{s}$ is possibly Ar^+). The mass-to-charge ratio of $^{131}\text{Xe}^{4+}$ is 32.74 and thus close to the value of the O_2^+ molecule. Therefore, it cannot be ruled out that O_2^+ is produced by charge exchange which can be falsely identified as the $z = +4$ charge state as indicated in Fig. 3 (top). With the given mass-resolving power of the TOF spectrum the two masses cannot be distinguished. Note, however, that in the case of $^{136}\text{Xe}^{3+}$ this peak does not appear although N_2^+ is observed with a similar yield and therefore O_2^+ is expected, too. Thus, there seems to be a different charge-exchange behavior for $^{131}\text{Xe}^{4+}$ and $^{136}\text{Xe}^{3+}$.

The doubly charged xenon ions were further transferred to the preparation Penning trap. In normal operation contaminating ions are removed from the ion ensemble by application of a buffer-gas centering technique [19] which lasts typically about 100–200 ms. Afterwards an additional storage time is applied to further cool the ion motion. Due to the helium buffer-gas environment further charge exchange may occur. As an example the TOF spectrum as recorded with MCP 3 after an additional storage time of 25 ms is shown in the bottom of Fig. 4(a). In or-

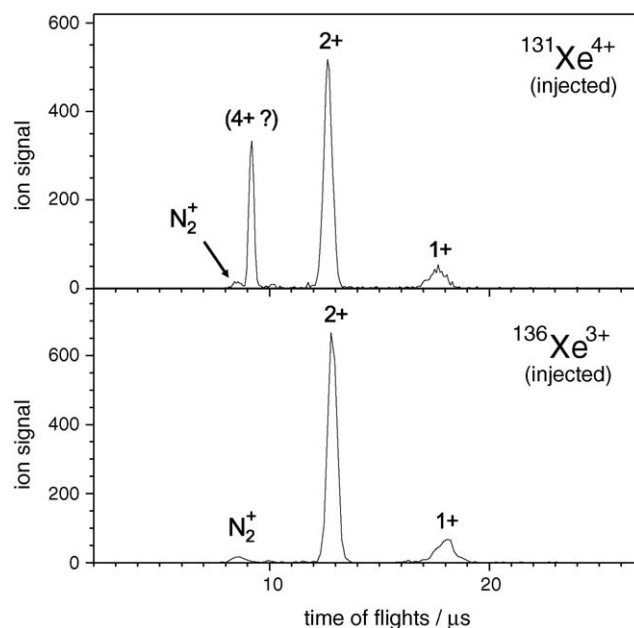


Fig. 3. TOF mass spectrum recorded on MCP 1 (see Fig. 2) after ejection of ions from the RFQ buncher in the case of $^{131}\text{Xe}^{4+}$ (top) and $^{136}\text{Xe}^{3+}$ (bottom).

der to investigate the time dependence of the charge-exchange processes and the loss of doubly charged ions, TOF spectra were recorded after various additional storage times. In Fig. 4(a) the resulting TOF spectra are shown, where for intermediate times the ion-signal intensity has been interpolated. Ion signals marked with an asterisk could not be identified. The graph shows a decrease of the doubly charged xenon ions and an increase of the singly charged ones as well as various residual gas ions created in charge-exchange processes. For clarification the ion yields of $^{131}\text{Xe}^{2+}$ and $^{131}\text{Xe}^{1+}$ have been plotted as a function of the additional storage duration in Fig. 4(b). For times above 500 ms a general ion loss is observed due to the radial drift of all ions caused by collisions with the helium atoms.

For the shortest additional cooling time, 25 ms, the ion ensemble was transferred to the precision trap where the endcap potential was lowered at the corresponding arrival time of the doubly charged xenon ions in order to prevent other ions from entering the second Penning trap. In general, the storage time in the precision trap is defined by the duration of the quadrupolar rf-excitation. In the bottom of Fig. 5 the resulting TOF spectrum (measured with MCP 4) for an excitation time of 50 ms is shown. Although no buffer gas is present in the volume of the precision Penning trap, the residual gas at a pressure of about 10^{-8} to 10^{-9} mbar still leads to charge exchange of the doubly charged ions. As in the case of the preparation trap, TOF spectra have been recorded for various excitation times. The corresponding ion signal intensity is plotted in Fig. 5, where again for intermediate times the signal intensity has been interpolated. A decrease of doubly charged ions is observed; singly charged xenon ions as well as H_2O^+ and N_2^+ are created by charge exchange. For the following mass measurements a quadrupolar rf-excitation period of 300 ms was chosen in order to maximize the amount of doubly charged ions in the precision trap for a sufficiently long observation time.

3.2. Discussion

The space charge produced after charge exchange in the preparation Penning trap may lead to a distortion of the buffer-gas centering technique due to Coulomb interaction of the various ion species in the trap volume [22]. Thus, the mass selection of the ions is not working properly. However, the charge exchange and the resulting loss of the multiply charged ions in the buffer-gas environment is the main challenge (see Fig. 4(b)). Hence, for higher charge states than $z = +2$ new cooling techniques are required that work without collisions with buffer-gas atoms.

However, as shown above, even at high-vacuum conditions with a pressure of the order of 10^{-8} to 10^{-9} mbar in the precision Penning trap charge exchange does occur. The relative abundances of doubly and singly charged xenon ions as well as H_2O^+ and N_2^+ are plotted as a function of the storage time in the precision Penning trap in Fig. 6. The dashed line is an exponential fit to the data points of $^{131}\text{Xe}^{2+}$. Note that ion loss in the precision trap due to collisions with residual gas, which may lead to a drift of the ions out of the trap, is negligible for the time scales and vacuum conditions in the present study. Thus, only

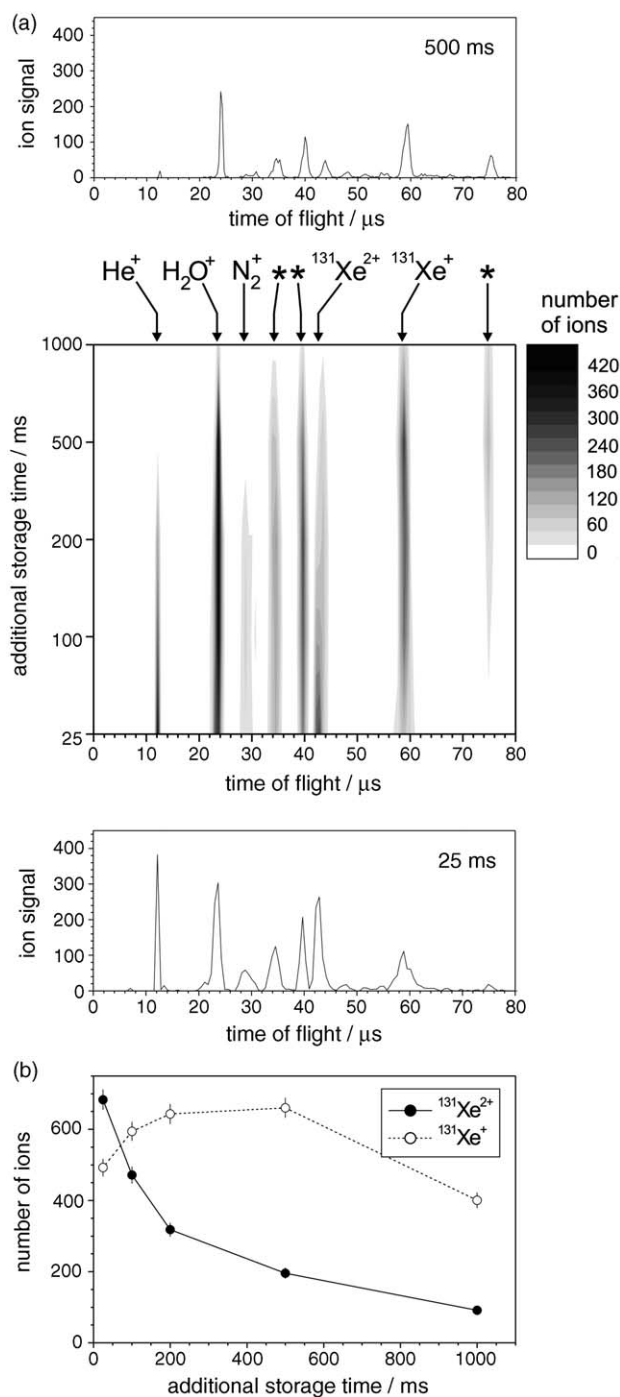


Fig. 4. (a) Middle: Ion signal as a function of the time of flight to the detector MCP 3 (see Fig. 2) and the additional storage time for ion cooling in the preparation Penning trap with indication of expected TOF positions of several ion species. The ion signal between the discrete time steps 25 ms, 100 ms, 200 ms, 500 ms, and 1 s has been interpolated. Bottom and top: TOF spectrum in the case of an additional storage time of 25 and 500 ms, respectively. (b) Number of ions of $^{131}\text{Xe}^{2+}$ (full symbols) and $^{131}\text{Xe}^{1+}$ (open symbols) as a function of the additional ion cooling time in the preparation Penning trap (the data points have been connected by straight lines to guide the eye).

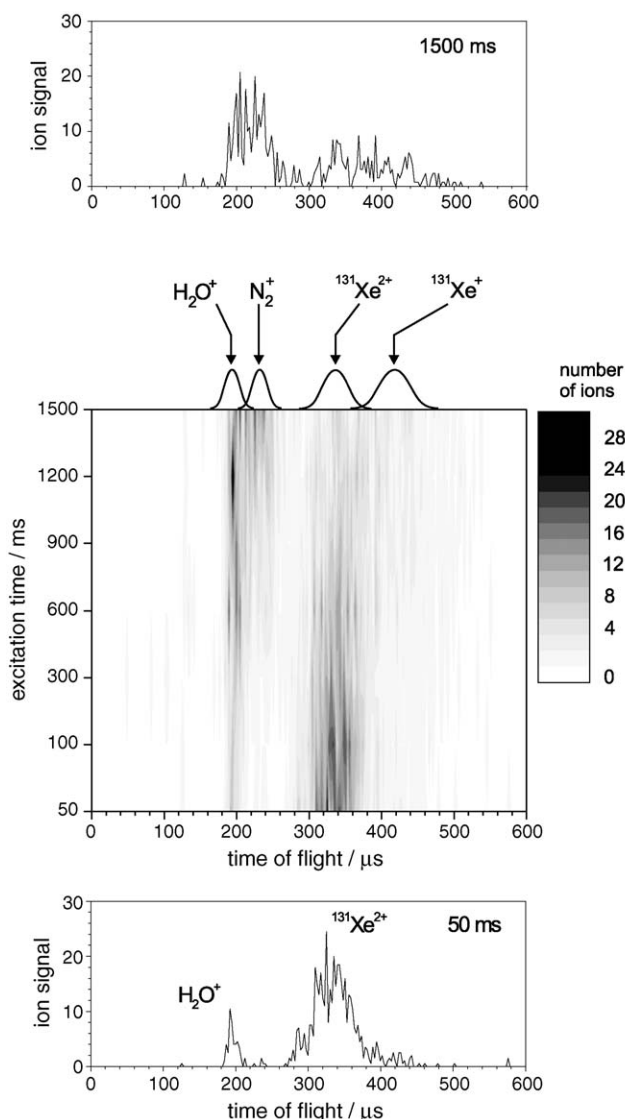


Fig. 5. Middle: Ion signal as a function of the time of flight to the detector MCP 4 (see Fig. 2) and the excitation time in the precision Penning trap with indication of expected TOF positions of several ion species. The ion signal between the discrete time steps 50 ms, 100 ms, 300 ms, 600 ms, 900 ms, 1.2 s, and 1.5 s has been interpolated. Bottom and top: TOF spectrum in the case of a 50 and 1500 ms quadrupolar rf-excitation period, respectively.

charge exchange is taken into account. From the deduced time constant $\tau = 1.46(6)$ s a cross-section for the charge exchange can be estimated with

$$\sigma \approx \frac{k_B T}{p v \tau}, \quad (3)$$

where p is the pressure in the trap volume, $T = 295$ K the temperature, and v is the relative velocity of the doubly charged ions and the helium gas atoms. As an estimate, a cross-section of the order of $\sigma = 10^{-14}$ cm² is obtained for the given experimental parameters, which is a reasonable value as compared to other experimental results, e.g., 5×10^{-15} cm² for collisions of Xe²⁺ with rare gas atoms [23] at velocities of less than 10⁸ cm/s.

Thus, under better vacuum conditions, i.e., a lower pressure p due to an improved vacuum system and without buffer gas,

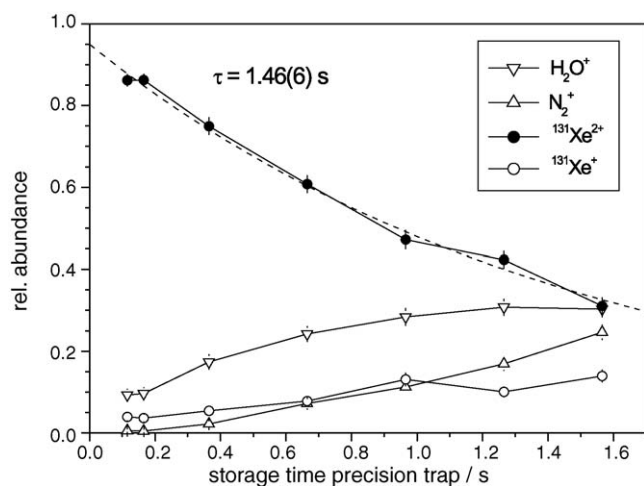


Fig. 6. Relative abundance of ¹³¹Xe²⁺ (full circles), ¹³¹Xe¹⁺ (open circles), N₂⁺ (open up-triangle), and H₂O⁺ (open down-triangle) as a function of the storage time in the precision Penning trap. The dashed line is an exponential fit, $R(t) = a \exp(-t/\tau)$, to the data points of ¹³¹Xe²⁺ with a life-time of $\tau = 1.46(6)$ s.

the time constant τ for ion loss will increase and consequently a longer duration of the quadrupolar rf-excitation can be chosen. In the present study of stable nuclides, charge exchange in the precision trap, especially during the application of the quadrupolar rf-excitation, limits the achievable precision in the frequency determination: The factor two expected for the higher charge state is not reached due to a reduced excitation duration. Note that for short-lived nuclides the situation is somewhat different since the excitation duration is limited by the half-life.

4. Mass measurements

4.1. Results

Despite charge-exchange losses doubly charged xenon ions were successfully transferred to the precision Penning trap for mass determination. ¹³⁴Xe²⁺ was taken as a reference-mass nuclide. Cyclotron resonances for the nuclides ¹²⁶Xe²⁺, ¹²⁹Xe²⁺, ¹³⁰Xe²⁺, and ¹³⁶Xe²⁺ were recorded. From the frequency ratio relative to ¹³⁴Xe²⁺ the mass was deduced as

$$m = \frac{\nu_{c,\text{ref}}}{\nu_c} (m_{\text{ref}} - 2m_e) + 2m_e, \quad (4)$$

where m_e is the electron mass. Note that with increasing charge state, the binding energy of the electron has to be taken into account. In the case of xenon the first two ionization energies are 12.1 and 21.0 eV, respectively [24]. For the present study they are negligible since their contribution is two orders of magnitude smaller than the obtained overall mass uncertainties (see below).

The analysis procedure is described in detail in Ref. [25]. Especially important is the correction of the influence of contaminant ions that are stored simultaneously with the ions of interest. The main contaminant ions are N₂⁺, H₂O⁺, and Xe⁺, all created by charge exchange in the precision trap. Their presence might shift the observed cyclotron frequency. The center frequency is determined for various numbers of ions per experimental cycle and an extrapolation to only one ion in the

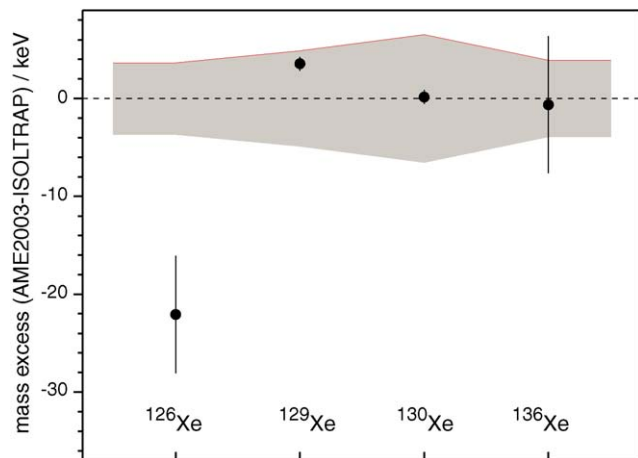


Fig. 7. Deviation $\Delta = \text{ME}_{\text{lit}} - \text{ME}$ of the literature value [26] of the mass excess with respect to the ISOLTRAP results for the investigated xenon nuclides. The shaded area indicates the uncertainty of the ISOLTRAP values. The uncertainties of the literature values are indicated by the error bars (as stated in Ref. [26]).

trap yields the corresponding value of the cyclotron frequency [25].

The results of the mass analysis are summarized in Table 1. The deviations of the mass excesses as compiled in the recent atomic-mass evaluation (AME2003) [26] to the present experimental values are shown in Fig. 7. The shaded area gives the uncertainty of the experimental values of the present study. The ISOLTRAP values are in agreement with the literature values except for the case of $^{126}\text{Xe}^{2+}$, where a deviation of more than 20 keV (about 3σ) is observed.

4.2. Discussion

For the only significant deviation to the literature value, that was found in the case of ^{126}Xe , a comparison to other experimental results is shown in Fig. 8. The value in the AME2003 is determined entirely from the Q -value of the β decay $^{126}\text{I}(\beta^-)^{126}\text{Xe}$ measured by Koerts et al. [27]. The recent result from Litvinov et al. performed at the ESR storage ring experiment at GSI Darmstadt [28] is in disagreement with the present ISOLTRAP result by about 2σ . However, the mass excess determined via the β decay $^{126}\text{Cs}(\beta^+)^{126}\text{Xe}$ by Osa et al. [29] is in agreement with the ISOLTRAP value. Thus, further independent mass measurements on the stable xenon isotopes should be performed to resolve the current discrepancy. With regard to the mass uncer-

Table 1
Frequency ratios of the investigated doubly charged xenon isotopes relative to the reference nuclide $^{134}\text{Xe}^{2+}$ and the corresponding mass excess

A	$\nu_c/\nu_{c,\text{ref}}$	ME (keV)	ME_{lit} (keV)	Δ (keV)
126	0.940247647 (28)	-89146.1 (3.6)	-89169.0 (6.0)	-22.9 (7.0)
129	0.962655277 (38)	-88701.0 (4.9)	-88697.4 (0.7)	3.6 (4.9)
130	0.970113830 (52)	-89881.9 (6.5)	-89881.7 (0.7)	0.2 (6.5)
136	1.014949671 (30)	-86424.3 (3.9)	-86425.0 (7.0)	-0.7(8.0)

For comparison the literature value of the mass excess [26] is given and the difference $\Delta = \text{ME}_{\text{lit}} - \text{ME}$.

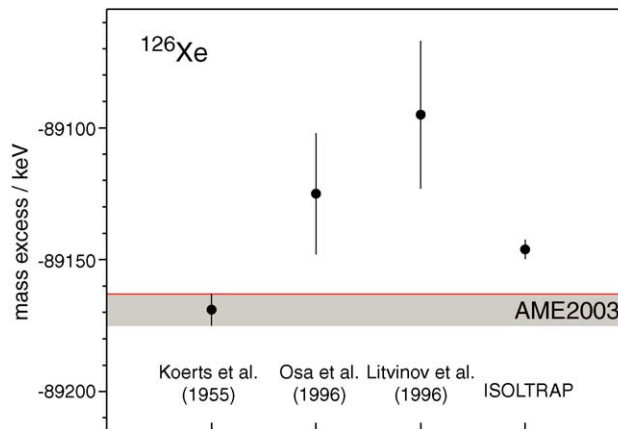


Fig. 8. Experimental values for the mass excess of ^{126}Xe [27–29] as compared to the literature value given in the latest atomic-mass evaluation [26] (the shaded area indicates the uncertainty of the AME2003 value).

tainties, the masses for ^{126}Xe and ^{136}Xe were improved (see Table 1).

5. Summary and outlook

Stable multiply-charged xenon ions from a plasma ion source have been transferred to the triple-trap mass spectrometer ISOLTRAP after mass selection with the on-line separator ISOLDE at CERN. Although charge-exchange processes were observed during ion preparation, a mass determination on doubly charged xenon ions was possible. With the exception of ^{126}Xe , the measured masses are in agreement with the literature values. Two mass values were improved.

With the present investigation the feasibility of mass measurements on doubly charged ions at ISOLTRAP has been demonstrated. For short-lived nuclides, where the measurement duration is limited by the half-life, the application of highly charged ions would allow to achieve the required relative mass uncertainty with a smaller number of ions. A faster measurement and thus a more efficient use of beam-time is possible. However, excellent vacuum conditions are required to prevent ion loss due to charge exchange with residual-gas atoms. In addition, the buffer-gas cooling technique [17,19] presently applied at ISOLTRAP needs to be replaced by other cooling schemes, e.g., evaporative cooling as performed at SMILETRAP [9] or electron cooling as shown for protons stored in a nested Penning trap [30].

A new ion source is currently being commissioned which allows the production of stable multiply charged ions by ionization with an electron beam and thus further tests will be possible. In addition, the external production of highly charged ions at ISOLDE with existing charge-breeding devices, e.g., the REXEBIS, and subsequent ion transfer to ISOLTRAP is under consideration.

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