

Precision Mass Measurements with LEBIT at MSU [★]

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Abstract

The Low-Energy Beam and Ion Trap facility, LEBIT, has been designed to facilitate a variety of experiments at low energies with rare isotopes produced by fast-beam fragmentation. Gas stopping of the fast-fragment beams and modern ion manipulation techniques are used. The first experiments to be performed are high-precision mass measurements possible with a 9.4 T Penning trap mass spectrometer. LEBIT has been recently commissioned and first experiments on stable and unstable nuclides have been performed. Here we present the results of mass measurements on stable krypton isotopes, measured with a precision of better than $\delta m/m = 5 \times 10^{-8}$ that reveal significant deviations from the literature values for ^{83}Kr and ^{84}Kr . The paper also provides an overview of LEBIT and the first Penning trap mass measurements performed on unstable isotopes from fast-beam fragmentation.

Key words:

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1 Introduction

The Coupled Cyclotron Facility at the National Superconducting Cyclotron Lab, NSCL, delivers a large range of rare isotopes with high intensities, pro-

[★] Dedicated to Jürgen Kluge on the occasion of his 65th birthday

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duced by projectile fragmentation or fission with separation in flight at relativistic energies. Compared to other techniques, this approach has the particular advantages of being able to produce isotopes of all elements lighter than the projectile, to be chemistry independent and to not suffer from decay-losses inside production targets. The conversion of fast-projectile fragments into low-energy beams of high quality is important because it has the ability to connect rare isotope beam production via projectile fragmentation to high-precision experimental techniques like laser spectroscopy and ion or atom trapping. With a number of refractory-element beams not available at ISOL facilities, and advantages in the production of the most exotic nuclides, a wealth of new experiments with stopped, and also post-accelerated, ion beams could become possible. This is reflected by the gas-stopping scenario being an integral part of the concept for all new radioactive beam facilities presently under planning [1–3] or construction. At present, several groups around the world are working on projects to stop radioactive ion beams in gas catchers [4–6].

The Low Energy Beam and Ion Trap facility, LEBIT, is the first system designed to convert such fast-fragment beams into low-energy beams with excellent beam properties by using gas stopping, advanced ion guiding and cooling/bunching techniques. Penning trap mass measurements are the first experiments carried out with LEBIT, but in the future other experiments like in-trap decay studies or laser spectroscopy will be possible with low-energy beams at the NSCL as well.

2 The LEBIT Facility

Fig. 1 shows a schematic view of the LEBIT facility. The main components are a gas stopping station, an ion beam cooler and buncher, and a Penning trap system for high-precision mass measurements. The system has been designed to be expandable. Stations for decay studies and laser spectroscopy are foreseen, and are examples of future experimental opportunities.

2.1 *The Gas Stopping Station*

The gas stopping station converts the fast-fragment beams delivered from the A1900 [7] fragment separator into low-energy beams. A set of flat glass degraders and wedged aluminum degraders, a beryllium entrance window and high-purity helium gas at a pressure of up to 1 bar are employed to slow down, stop, and thermalize the high-energy beam. A combination of DC electric fields, created by a set of drift rings and focusing electrodes inside the gas cell, and gas flow through an extraction nozzle is used to transport ions out of

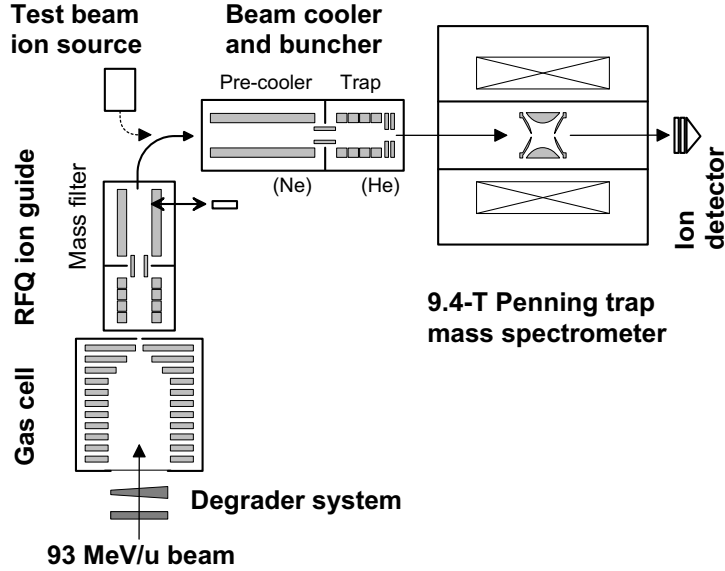


Fig. 1. Layout of the LEBIT facility at the NSCL/MSU.

the gas cell. An RFQ-ion guide system transfers the ions into high vacuum and forms a continuous low-energy ion beam. The ion guides can also be operated as a mass filter. This allows for beam purification by removing contaminant ions from the gas cell created by charge exchange with helium ions generated during the stopping process. Numerous studies have been performed of the stopping and extraction performance of the NSCL gas cell. These tests include range measurements [8,9] in the gas cell employing energy bunching [10] by means of a wedged degrader, and ion stopping and extraction of the rare isotopes [11,4]. As an example, for a mixed $^{38}\text{Ca}/^{37}\text{K}$ beam a stopping efficiency of 50% was observed. Extraction efficiencies up to 8% were measured for beam rates up to 100 pps and found to decrease for increasing beam rates.

2.2 Test Ion Beam Source

As shown in Fig. 1 installed upstream of the buncher is a test ion source station. The station consists of an ion source, an RFQ mass filter for beam selection and acceleration optics. Beams from this station are used extensively in system tuning and optimization, and also to provide reference masses during radioactive mass measurements. At present a plasma ion source is installed to provide ion beams of noble gases. This source also provides beams of alkali metals.

The test ion source is located perpendicular to the main LEBIT beam line. An electrostatic quadrupole deflector is employed to send the beam downstream to the cooler/buncher and Penning trap or upstream towards the gas cell. Sending the beam upstream facilitates systematic studies of the RFQ

system downstream of the gas cell. Similar studies are routinely made of the cooler/buncher and Penning trap systems.

2.3 The LEBIT Ion Cooler and Buncher

The LEBIT ion accumulator and buncher accepts the 5 keV DC beam from the gas cell and converts it into a low-energy, low-emittance pulsed beam. This device is a linear radiofrequency ion trap filled with a buffer gas at low pressure for ion cooling [12,13]. It features two separate vacuum sections, one for beam pre-cooling and one for final cooling, trapping and beam bunching. Before entering the system, the ions are electrostatically decelerated to a few tens of eV. The first section is typically operated with helium or neon at a pressure of a few Pa. Here the ions are transversely cooled and slowed down before they pass through a miniature RFQ ion guide (an efficient differential pumping barrier) into the trap section. The linear trap is operated with helium at a pressure 1-2 orders of magnitude lower than that in the first section to minimize beam heating during the pulsed-beam extraction. The ions are typically stored for 20-30 ms for their final cooling before they are extracted as a micro-second ion pulse. Both the pre-cooler and the trap section have been built as cryogenic devices and can be cooled with LN₂ provided from a stationary cryogenic line. This increases the efficiency of the system by reducing the diameter of the beam in the cooler, thus making the transport between the two sections via the miniature RFQ more efficient. Operating at LN₂ temperatures also decreases the cooling time and significantly reduces the emittance of the resulting pulse which increases the efficiency of injection into the Penning trap. Cooling the system also results in the significant reduction of residual pressures of gases other than the noble gases used for beam cooling. A third feature distinguishing the LEBIT buncher from RFQ ion bunchers used elsewhere is the wedged-electrode design which allows the electric drag potential in the cooling section to be created without the need for segmented rods. More details of the design of the buncher system can be found in [15]. The LEBIT beam cooler and buncher has been extensively tested and its properties were found to be in very good agreement with beam simulations [16] involving realistic ion-atom interactions. In pulsed-mode operation the overall efficiency was found to exceed 50%, while in continuous mode operation values of up to 80% were observed.

In its normal mode of operation, continuous or pulsed ion beams from the gas cell or the test ion source are cooled and accumulated in the cooler/buncher and released as short ion bunches for capture in the precision trap of the LEBIT Penning trap mass spectrometer. For diagnostic purposes and buncher optimization time-of-flight distributions are routinely measured with a microchannel plate (MCP) detector downstream of the buncher. Fig. 2 shows the

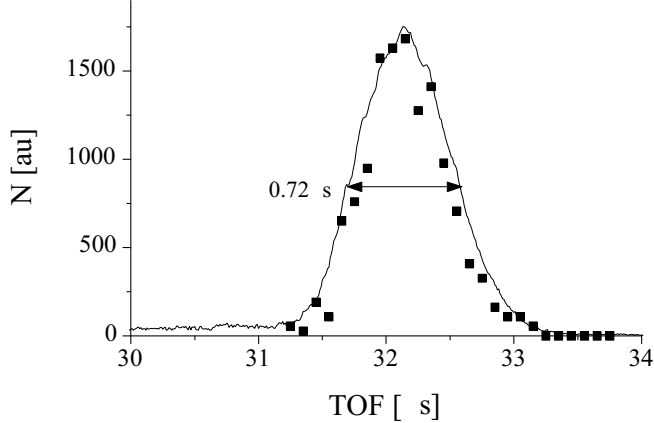


Fig. 2. Time-of-flight distribution of ions accumulated in the buncher and extracted as pulses measured with an MCP detector. The experimental result (solid curve) is compared with that of corresponding simulations (square dots). The latter is corrected for a $0.4 \mu\text{s}$ electronic time delay.

time-of-flight spectrum of a pulse of $^{40}\text{Ar}^+$ ions extracted from the buncher. Again, the result of a beam simulation reproduces the shape of the distribution well except for a shift of about $0.4 \mu\text{s}$, which can be assigned to electronic effects.

2.4 The LEBIT 9.4 T Penning Trap System

Fig. 3 shows the layout of the experimental setup of the LEBIT Penning trap mass spectrometer. The magnetic field is provided by an actively-shielded persistent superconducting 9.4 Tesla magnet system. The magnet system has been upgraded by the addition of external-field compensation coils [14] to reduce the effect of external field changes that may occur in an accelerator environment. The employment of a 9.4 T field, as compared to ~ 6 T which is typical of current systems, has the advantage that a given precision can be achieved in about half the measurement time. The pressure of the helium bath is stabilized by an electric valve operated on a PID loop to eliminate non-linear magnetic field effects, on top of the natural magnetic field decay, due to variations in the helium boil-off rate. A precisely machined vacuum tube, mounted inside of the room-temperature bore of the magnet, serves as an ion optical bench for the trap electrode system. The tube is wrapped with wire allowing either baking or compensation of the natural decay of the main magnetic field during measurements. Two ion-optical packages, one containing the injection optics and Penning trap and the other containing the ejection optics, are inserted into opposite ends of this bore tube. The ion trap and optical elements in its vicinity can be cooled with the help of a cryogenic shield. This aids the creation of an ultra-high vacuum in the center of the bore tube that is pumped by two turbomolecular pumps located on either end

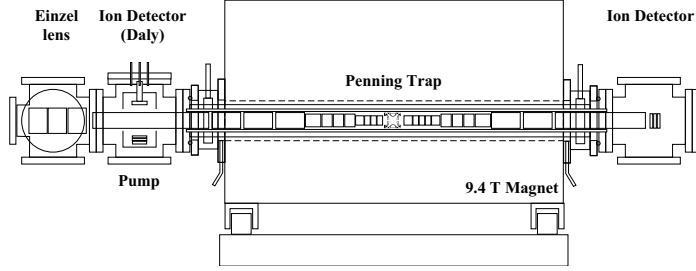


Fig. 3. Schematic layout of the LEBIT Penning trap system. Ions enter from the left.

of the magnet.

Ion bunches that are delivered by the LEBIT cooler/buncher are focused and injected into the magnetic field in a way that minimizes transverse energy pickup before the ions are captured in the Penning trap. The LEBIT Penning trap's electrodes (see Fig. 4) are constructed of high-conductivity copper and plated with gold. The insulators are made of aluminum oxide. The ring electrode has an eight-fold segmentation. This allows not only for the creation of a quadrupole RF field, as required for the excitation of the ion motion at the ion's cyclotron frequency ω_c [17], but also the application of an octupole RF field. The octupole field should allow one to drive the ion motion at $2\omega_c$ and provide a higher resolving power. This new excitation mode is under study at LEBIT and has been verified experimentally. Extensive numerical calculations have been performed to minimize the effect of electric and magnetic field imperfections, and also to determine the optimal voltages to be applied to the electrodes. Deviations from the electric quadrupole field are due to finite electrodes, as well as holes and segments in the electrodes. Two pairs of correction electrodes are used to efficiently compensate for the finite electrodes and the holes. The segmentation of the ring is a negligible effect.

As part of the cyclotron frequency detection scheme used in LEBIT and most other Penning trap mass spectrometers for the study of rare isotopes it is necessary to have the trapped ions perform an initial magnetron motion. This is normally achieved by driving the ion motion of the captured ions with an azimuthal RF field at the ion's magnetron frequency. In LEBIT this time-consuming step is avoided with a new technique based on a so-called Lorentz steerer. This steerer is a quartered cylindrical tube at the end of the retardation optics preceding the Penning trap that can create an electric dipole field perpendicular to the magnetic field. Because it is located in the strong magnetic field region the ions experience a force in the $\mathbf{E} \times \mathbf{B}$ direction. This results in an off-axis capture and eliminates the necessity of magnetron excitation. The trapped ion ensemble is purified, if necessary, and a time-of-flight resonance detection technique [18–20] is used to determine the ions' cyclotron frequency ω_c . Ion purification uses RF azimuthal dipole fields applied inside the trap that drive undesired ions mass-selectively to large orbits by exciting

their modified cyclotron motion at frequency ω_+ . The removal of unwanted ions avoids frequency shifts due to ion-ion interactions of unequal species [21]. After this cleaning process the ions are exposed to an azimuthal quadrupole RF field with a frequency $\omega_{RF} \approx \omega_c = \omega_+ + \omega_-$. In resonance, i.e., $\omega_{RF} = \omega_c$, there is a periodic conversion between the initial magnetron and the modified cyclotron motion. The excitation time, T_{RF} , and RF amplitude, A_{RF} , are chosen such that the initial magnetron motion is converted into cyclotron motion. This process is accompanied by a significant change of the ions' radial energy, which can be used to detect the cyclotron resonance when the ions are ejected from the trap and allowed to drift through the inhomogeneous section of the magnetic field. The radial energy is converted into axial energy during this drift which is measured via the ions' time-of-flight to a detector system. A maximum time-of-flight change is observed at $\omega_{RF} = \omega_c$ which is then used to determine the mass of the ions via $\omega_c = q/m \times B$, where q and m are charge and mass of the ion and B is the strength of the magnetic field. Currently, a micro channel plate detector, in a Daly configuration [22], located downstream of the trap is used for the time-of-flight measurement. In the near future it is planned to eject the ions upstream and to use a Daly detector with switchable voltage supply, mounted perpendicular to the beam axis, as indicated in Fig. 3. This would free the downstream exit of the ion trap allowing, for example, detectors for in-trap decay studies to be permanently installed.



Fig. 4. The LEBIT high-precision Penning trap with the endcap electrode removed.

The total efficiency of the cooler/buncher and Penning trap system is estimated to be in the range of 30 – 60%. These values have been obtained by operating the ion source at a beam current of a few picoamps. An ion-source beam gate was used to produce short pulses of a few microsecond length, containing a few tens of ions. These pulses were sent into the cooler/buncher, extracted, captured in the Penning trap, extracted again and the number of ions detected was determined with the MCP at the end of the system. The uncertainty in the efficiency is largely determined by the uncertainty in the MCP efficiency. Furthermore, the efficiency of capturing ion pulses in the Penning trap was measured relative to continuous beam transmission through the

Penning trap. These measurements were performed with the cooler/buncher in continuous extraction mode. A continuous beam current of a few pA was measured in a Faraday cup just upstream of the Penning trap. The current was again measured at the end of the system with another Faraday cup with the continuous beam being transmitted through the Penning trap. There was no measurable loss. The ion-source beam gate was again used to produce short ion pulses. These pulses were captured in the Penning trap and then measured with the MCP at the end of the system. Taking the MCP efficiency ($\sim 40 - 50\%$) into account it was determined that there was no measurable difference in efficiency between the transmission and capture scenarios.

After careful tuning of the potentials of the Penning trap and optimization of parameters for beam transport, ion capture and ejection, excellent line shapes and high resolving powers have been obtained with the LEBIT Penning trap mass spectrometer. A sample cyclotron resonance curve for $^{82}\text{Kr}^+$ ions is shown in Fig. 5. For an excitation time of $T_{RF}=200$ ms a line width of $\Delta\nu = 5$ Hz is obtained, resulting in a resolving power of $R = \nu/\Delta\nu \approx 450,000$. Perfect agreement is observed between the data points and the fit with the theoretical line shape [20]. The highest resolving power observed thus far is about 3 million for a 1-s excitation of $^{40}\text{Ar}^+$ ions.

In order to study the achievable precision and accuracy a large number of test measurements have been performed with stable ions, in particular $^{23}\text{Na}^+$, $^{39}\text{K}^+$, $^{40}\text{Ar}^+$, $^{40}\text{Ar}^{2+}$, and $^A\text{Kr}^+$. From these tests we conclude that LEBIT has the potential to achieve a very high accuracy. For example, the measured frequency ratio of stable $^{40}\text{Ar}^{2+}$ and $^{23}\text{Na}^+$, both known with sub-ppb precision, is in full agreement with the expected ratio obtained from literature mass values [23], showing an insignificant deviation of only $3(5) \times 10^{-9}$. This example corresponds to a close-doublet situation. The results obtained for a $^A\text{Kr}^+$ - $^{39}\text{K}^+$ mass comparison, discussed below, shows excellent agreement even for large mass differences, indicating that mass-dependent systematic errors are well under control.

3 Mass Measurements of Stable Krypton Isotopes

Krypton beams from the test ion source have been used extensively to characterize LEBIT's performance in the regime of medium heavy masses. Mass measurements utilizing ^{86}Kr as a reference mass, measured to high precision by SMILETRAP [24], were performed on $^{78,80,82,83,84}\text{Kr}$, revealing somewhat surprising results.

Previous preliminary LEBIT measurements had indicated that the AME03 mass values of $^{83,84}\text{Kr}$ were too large. To explore this discrepancy in greater

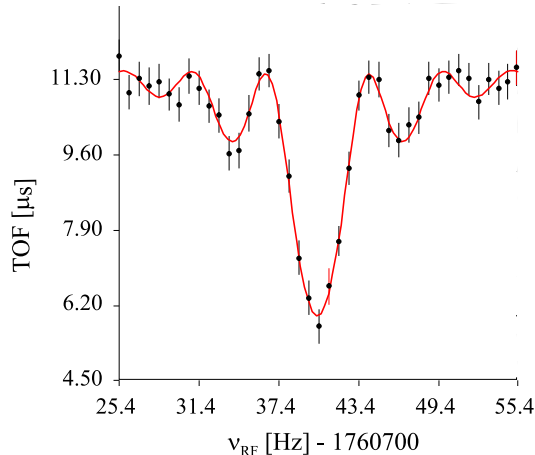


Fig. 5. Cyclotron resonance curve of $^{82}\text{Kr}^+$ ions with a fit of the theoretical line shape. An excitation time of 200 ms was used in this measurement.

detail 4-5 cyclotron frequency measurements were made of $^{78,80,82}\text{Kr}^+$ and 11 cyclotron frequency measurements were made of $^{83,84}\text{Kr}^+$, the Kr isotopes in question. Each of these individual measurements was bracketed by a cyclotron frequency measurement of the reference ion, $^{86}\text{Kr}^+$. The time separating each reference measurement was approximately half an hour. During the measurement of any specific Kr isotope, possible contamination from the other isotopes was cleaned away via dipole excitation as discussed above. Table 1 provides the frequency ratios obtained in the run with their statistical uncertainties in parentheses, and an additional uncertainty of 1×10^{-8} , added in quadrature, in curled brackets. This additional uncertainty accounts for any additional systematic uncertainties that we have not been able to rule out at that level.

Using the known mass value for ^{86}Kr and the frequency ratios obtained here, mass values for $^{78,80,82,83,84}\text{Kr}$ were obtained. They are listed in Table 2 together with the AME03 value and the difference between these values. This difference is shown as a function of mass number in Fig. 6.

Excellent agreement is observed for the three isotopes $^{78,80,82}\text{Kr}$ within measurement uncertainties. The mass values for these isotopes are known with very high precision and are dominated by data from other Penning traps. A significant but equally large deviation is observed for ^{83}Kr and ^{84}Kr which have been determined by many different and partially inconsistent data (discussed below). The observed averaged relative deviation of the LEBIT results from the AME03 values is less than 6×10^{-9} , if ^{83}Kr and ^{84}Kr are excluded. As a part of this measurement series, mass comparison of $^{86}\text{Kr}^+$ and $^{39}\text{K}^+$ yields a mass-dependent systematic effect of less than $10^{-9}/u$. Assuming that the AME03 mass values for ^{83}Kr and ^{84}Kr are suspect, this provides great confidence in the data obtained with LEBIT.

An analysis of the data used in the Atomic Mass Evaluation [23] shows that

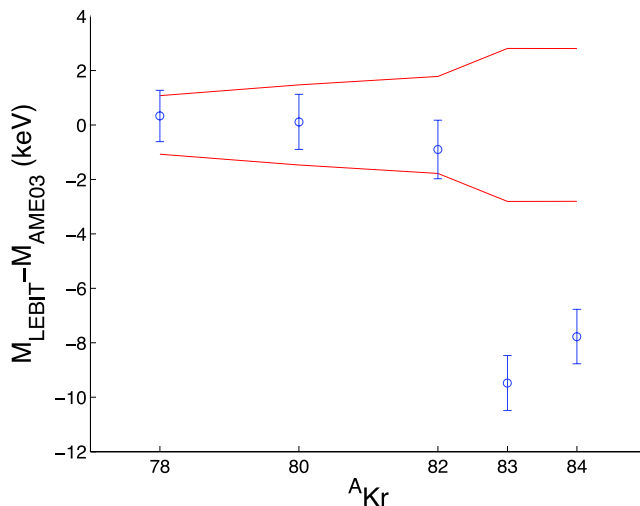


Fig. 6. The difference between mass values measured with LEBIT and AME03 [23] for the stable krypton isotopes. The error band corresponds to the uncertainty of the literature values, the error bars to the uncertainty of the mass values determined with LEBIT.

mass values for ^{83}Kr and ^{84}Kr are determined via a network of Q-values from decays and reactions combined with data from doublet mass measurements. The mass values for ^{83}Kr and ^{84}Kr are linked strongly by an (n,γ) reaction [25] which has an uncertainty of 0.3 keV. The Q-value agrees within 1.5σ with the value calculated from the LEBIT data. The absolute mass of ^{84}Kr is determined primarily by the Q-value of the β -decay $^{84}\text{Rb}(\beta^+)^{84}\text{Kr}$ [26,27], and the result of a doublet mass spectrometer measurement, $\text{C}_6\text{H}_{12}\text{-}^{84}\text{Kr}$ [28]. The mass of ^{83}Kr is primarily determined by the link to ^{84}Kr , but also by a doublet mass measurement $\text{C}_6\text{H}_{11}\text{-}^{83}\text{Kr}$ [28]. Two recent measurements of ^{84}Kr are not included in the AME03. One was made at Florida State University, FSU [29], and another [30] at ISOLTRAP [31].

Table 1

Cyclotron frequency ratios $R = \nu(^{86}\text{Kr}^+)/\nu(A\text{Kr}^+)$ obtained in this work. Column 1 contains the atomic number of the krypton isotope. Column two contains the weighted average of the measured ratios with the statistical uncertainties in parentheses and an additional 1×10^{-8} uncertainty, added in quadrature, in curled brackets.

A	\bar{R}
78	0.906992902(7){12}
80	0.930226664(8){13}
82	0.953473109(9){13}
83	0.965120677(8){13}
84	0.976730172(8){12}

Table 2

Mass excess values ME for krypton isotopes with mass number A as obtained from the measured frequency ratios and compared to their AME03 values [23].

A	ME_{LEBIT} keV	ME_{AME03} keV	ΔME keV
78	-74179.4(0.9)	-74179.7(1.1)	0.3(1.4)
80	-77892.4(1.0)	-77892.5(1.5)	0.1(1.8)
82	-80590.4(1.1)	-80589.5(1.8)	-0.9(2.1)
83	-79991.2(1.0)	-79981.7(2.8)	-9.5(3.0)
84	-82438.8(1.0)	-82431.0(2.8)	-7.8(3.0)

Fig. 7 displays the deviation of mass excesses of $^{83,84}\text{Kr}$, as determined by individual measurements, from AME03 values. The three measurements made using Penning traps, FSU, ISOLTRAP and LEBIT, of ^{84}Kr agree very well. The doublet measurements of both krypton isotopes in question also agree with the Penning trap measurements, but may have a slight systematic bias towards heavier masses. The mass excesses calculated from the results of the β decay measurements are obtained by using the AME03 values of the parent nuclei. The fact that they do not agree well with the Penning trap measurements could indicate that there is one or more incorrect input data used in the AME03 in the region of ^{84}Rb and ^{83}Br .

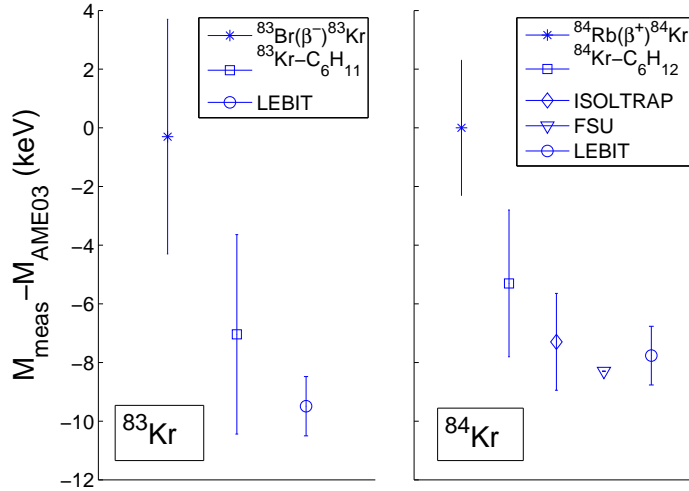


Fig. 7. Deviation of individual mass measurements of $^{83,84}\text{Kr}$ from AME03. Values obtained from β decays use AME03 values for the masses of the parent nuclei.

4 First Mass Measurements of Unstable Isotopes and Perspectives

LEBIT has recently seen its first very successful measurements on short-lived rare isotopes. This was preceded by a substantial one-year effort to reduce the intensity of contaminant molecular ion beams extracted from the gas cell. Initially these beams were found to have intensities several orders of magnitude greater than the incoming rate of rare isotopes. These molecular ion beams are produced by charge exchange of the helium ions generated in the stopping process with residual gas molecules present in the gas cell. Intensive physical cleaning, the assembly of gas cell components in a clean room environment, the employment of additional mass filtering techniques and the use of Collision Induced Dissociation (CID) was decisive to reduce the stable beam currents to an acceptably low level.

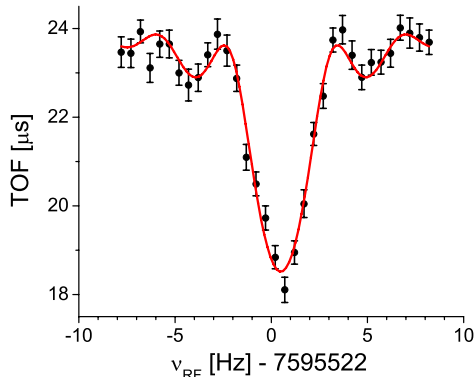


Fig. 8. $^{38}\text{Ca}^{2+}$ cyclotron resonance taken with 300 ms RF excitation. The observed linewidth corresponds to a resolving power of $R \approx 2,000,000$

The first radioactive nuclide measured by LEBIT was the super-allowed β -emitter ^{38}Ca [32]. Fig. 8 shows a cyclotron resonance curve of this isotope, which has a half-life of 440 ms. Doubly-charged $^{38}\text{Ca}^{2+}$ ions were used and compared to H_3O^+ ions, allowing for a true doublet measurement. With the LEBIT measurement the mass of this nuclide is now known with an uncertainty of less than 10^{-8} which makes it a new candidate for testing the Conserved Vector Current hypothesis [33]. During the same run the mass of ^{37}Ca was also measured and its mass uncertainty considerably improved. ^{37}Ca has a half life of only 102 ms and is a candidate for testing the Isobaric Mass Multiplet Equation, IMME [34].

In further runs $^{66,67}\text{As}$ and ^{65}Ge were also measured. ^{66}As is one of the two shortest-lived nuclides ever studied with a Penning trap, with a half-life of 96 ms. This is second only to the ^{74}Rb [35], half life of 65 ms, measurement by ISOLTRAP. Yet the ^{66}As measurement was performed with only a few detected ions per hour, demonstrating LEBIT's high degree of sensitivity.

These initial mass measurements of unstable isotopes have proven that a va-

riety of thermalized radioactive beams from fast-beam fragmentation can be produced with the excellent beam properties and high purity necessary for high-precision mass measurements and other experiments with similar requirements. LEBIT's experimental program will continue with more measurements relevant to fundamental interactions, nuclear structure and astrophysics.

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