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Improved stability of multi-reflection time-of-flight mass spectrometers through passive and active voltage stabilization

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ABSTRACT

Multi-reflection time-of-flight (MR-ToF) spectrometers are devices in which ions are reflected between two electrostatic mirrors to prolong their flight path. The performance of the apparatus relies on the stability of the voltages that are used to define the mirror potentials. Especially the voltage of the mirror electrodes at the point where the ions reverse the direction of their axial motion needs to be stable in order to minimize fluctuations of their total flight time. Here we present a method to increase the short- and long-term stability of suitable voltage supplies for enhanced performance of the spectrometer.

1. Introduction

Recently, multi-reflection time-of-flight mass spectrometers (MR-ToF MS) gained high attention as these table top devices allowed the extension of the flight path of the ions to reach mass resolving powers $R = m/\Delta m$ on the order of 10^5 in only a few milliseconds [1–5]. This makes the devices suitable for various studies in molecular physics [6–8], cluster physics [9–12] and nuclear physics [13–15].

At the ISOLTRAP experiment [16,17], located at the ISOLDE facility at CERN [18], the MR-ToF device has been used to determine the masses of short-lived as well as stable ion-beam constituents with high precision for many physics topics, such as nuclear structure [13,19–22], nuclear astrophysics [23,24], and to support high precision measurements of decay energies for neutrino physics [25]. In addition to mass spectrometry, the MR-ToF MS also provided purified samples for decay spectroscopy [26,27] and, in combination with the Resonant Ionization Laser Ion Source (RILIS) of ISOLDE [28], assisted measurements of nuclear moments and charge radii with background suppression [29–32]. The fast-identification capabilities also make the MR-ToF device a very attractive tool for target and ion-source optimization and

ion-yield determination [33]. Improvements of the performance and the reliability of the spectrometer will have direct implications to many experimental set-ups exploiting MR-ToF devices. In this paper we demonstrate a method to increase the MR-ToF MS performance with respect to the stability of the mass resolving power by employing passive and active voltage stabilization.

2. Experimental setup and procedure

The measurements were performed with a part of the ISOLTRAP setup which has been described in Refs. [2,1] and is shown in Fig. 1. The RFQ cooler and buncher [34] is used to capture, cool and bunch the continuous ion beam from the offline alkali ion source which delivers singly charged stable rubidium and cesium isotopes. The ions are subsequently captured into and later ejected from the MR-ToF device by use of the in-trap lift technique [1,35]. Once extracted from the MR-ToF apparatus, they are detected by employing a MagneTOF secondary electron multiplier ion detector¹ and a multichannel scaler².

The voltages for the mirror electrodes of the MR-ToF device range from about -4 kV to about 4 kV and are supplied by an EHS 8240x

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¹ DM291, ETP, Ermington, Australia.

² MCS6, Fast ComTec, München, Germany.

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Fig. 1. Schematic overview the ISOLTRAP components relevant for the present studies. Further details can be found in [1,2].

voltage module³. It is specified to have voltage ripples of less than 5 mV for frequencies higher than 10 Hz and a temperature coefficient of 10 ppm/K. The output of the power supply can be set with a resolution of 8 mV.

Since the stability of the total time of flight depends on the stability of the voltages applied to the mirror electrodes (see Fig. 9 in Wolf et al. [36], in this case $0.5 \text{ ppm}_{\text{ToF}}/1 \text{ ppm}_V$), it is of utmost importance to find voltage sources stable enough or, alternatively, to stabilize a given voltage source adequately.

The stability of the power supplies connected to the ion optics of the components in front and behind the MR-ToF device (buncher ejection optics, MR-ToF injection and ejection optics) are influencing the stability of the ion time of flight as well but much less significantly compared to the mirror electrode 5. For this electrode's voltage Fig. 2 shows a strong correlation with the ToF of ^{85}Rb ions after 1000 revolutions in the MR-ToF MS. This suggests that it has the most significant impact on the observed ToF fluctuations, as already suggested in [36]: The voltage of mirror electrode 5 determines the voltage gradient at the point where the ions reverse their axial motion, i.e. the ions turn around and are reflected towards the opposite mirror stack. Thus they are most sensitive to slight changes of this electrode's electrical potential.

Fig. 2 also shows the temperature in the laboratory as a function of time. No correlation is visible. For longer time scales, temperature changes are likely to have an influence on the device (the power supplies and the mechanical setup) but the involved time constants may be large so that a delayed reaction seems to manifest itself only later.

To improve the short- and long-term voltage stability, a passive electric low-pass filter together with an active software-based⁴ proportional-integral (PI) regulation was implemented. The principle is shown in the schematic drawing of Fig. 3.

For the precise measurement of the voltage on mirror electrode 5, a high precision 8.5 digit multimeter⁵ is used in combination with a precision high voltage divider⁶, which reduces the input voltage by a factor 1000 with a total impedance of 50 M Ω . The internal resistor chain has a temperature coefficient of less than 5 ppm/K but to avoid any drifts due to the changes of the environment temperature, the divider box was temperature stabilized to about $\pm 5 \text{ mK}$ by use of a Peltier element which was PI-regulated as well. The multimeter was used in the 20 V range where it can measure with a resolution of 1 μV in 20 ms.

For the low-pass filter, a resistance of $R = 1 \text{ M}\Omega$ ⁷ and a capacitor of $C = 40 \mu\text{F}$ ⁸ were used. That results in a time constant of $\tau = R \cdot C = 40 \text{ s}$ and a cut-off frequency as low as $f_c = 1/(2\pi\tau) \approx 4 \text{ mHz}$. The low-pass filter thus suppresses mid- and high- frequency fluctuations which is important since one revolution of ions with mass $A = 85$ in the MR-ToF

device takes about 22.2 μs . On this time scale we cannot correct the fluctuations with the PI-regulation since we cannot measure the voltage fast enough with the necessary precision. For 1000 revolutions in the device the rubidium ions need about 22.2 ms. Fluctuations on this time scale can be corrected with the PI-regulation.

Employing low-pass filters with large time constants to stabilize the potential of electrodes is in many applications impractical since it makes the optimization or scanning of the potential infeasible. In this context the PI-regulation offers a simple solution in which the desired voltage can be reached much faster, since the regulation will set the output of the power supply to the highest value to reach the set-point as fast as possible.

3. Results and discussion

The improvement of the voltage stability of the MR-ToF MS impacts both the resolving power of the apparatus and the accuracy of mass measurements, especially when the reference ToF values (used for calibration) are not recorded in the same spectrum as the ions of interest, i.e. when the references are not isobaric contaminants of the latter. In this section, both aspects of this development will be discussed.

3.1. Enhancement of resolving power

For the first tests of the voltage stabilization, the difference in adding the low-pass filter to the circuit was determined while utilizing the PI-regulation to suppress slower fluctuations. To this end, the voltage of the in-trap lift was varied to determine the time focal point of the ion distribution at the position of the ion detector. This corresponds to the maximal mass resolving power attainable for a given number of revolutions, in this case 1000 revolutions in MR-ToF MS, see Fig. 3 in Wolf et al. [2]. The result is presented in Fig. 4. The measurements were performed with ^{85}Rb and for each spectrum, 300 experimental cycles, corresponding to about 500 ions, were accumulated.

By employing the low-pass filter and thus attenuating voltage fluctuations, a more than 30% higher mass resolving power could be reached. The mass resolving power was determined for each voltage step by fitting a Gaussian profile to the time-of-flight distribution. The relevant parameters of the fit used to describe the mass resolving power as a function of the in-trap lift voltage (again described by a Gaussian) can be found in Table 1. The slight shift of 2.90(63) V between the optimal in-trap lift voltage is probably due to a slight shift of the high-voltage (HV) power supplies of the HV-platform of the buncher, i.e. the slightly different initial ion energy between the measurement series.

Similar measurements as the one shown in Fig. 2 were performed to determine the fluctuations with and without low-pass filter, with and without activated PI-regulation. Fig. 5 shows a measurement for 1 h using ^{85}Rb ions after 1500 revolutions in the MR-ToF MS with the low-pass filter but without PI-regulation.

One can see that compared to Fig. 2 the short-term fluctuations are reduced but there are still drifts. The correlation to the mirror electrode 5 voltage is still apparent. If the PI-regulation is added, see Fig. 6, again for a 1 h measurement of ^{85}Rb ions after 1500 revolutions in the MR-

³ ISEG, Dresden, Germany.

⁴ LabVIEW, National Instruments, Austin, USA.

⁵ Keithley 2002, Keithley Instruments, Solon, USA.

⁶ KV-30A, Ohm-Labs, Pittsburg, USA.

⁷ Ohmite thick film resistor of type MOX-1 N-131004FE, 2 W, 10 kV, 25 ppm/K.

⁸ General Electric, Gecanol polypropylene-paper dielectric system based series 28F2000 (28F2039), 4.1 kV.

Fig. 2. Experimental values measured as a function of time for 55 min (x-axis) left axis (blue): Time of flight of ^{85}Rb ions after 1000 revolutions in the MR-ToF MS. For each data point, 50 experimental cycles were accumulated which corresponds to about 250 ions per spectrum that were fitted every ≈ 4 s. first right axis (black): Voltage of mirror electrode 5. A measurement was taken every ≈ 100 ms. No low-pass filter was installed and the PI-regulation was not activated. second right axis (red): Temperature of the environment. Here, a data point was taken every 2 s.

Fig. 3. Schematic diagram of the devices used for stabilizing the voltage supply, for further details see text.

Fig. 4. Mass resolving power for different in-trap lift voltages with and without the low-pass filter and an activated PI-regulation. For details see text.

Table 1

Parameters of the Gaussian fits of Fig. 4.

	no low-pass	with low-pass
Centroid	1071.90(35) V	1069.00(28) V
Width	33.41(97) V	24.46(66) V
R _{max}	111.1(4.2) k	172.2(5.7) k

ToF MS with about 150–200 ions from 50 experimental cycles, the long-term drifts are reduced as well.

Now that the main source of the fluctuations is eliminated, the next to leading order fluctuation becomes apparent in the ToF, see Fig. 7. The figure includes the change of the temperature in the laboratory but obviously, there is no correlation with the remaining voltage fluctuations. Most probably, those originate from the voltage source connected to the neighboring mirror electrodes which are supplied by another channel of a EHS 8240x module.

The relative stability for the different scenarios is summarized in Table 2. An improvement of about a factor of 4 has been achieved.

To further characterize the improvement, the Allan deviation [37] for the data presented in Fig. 2, 5 and 6 was determined (Fig. 8). The Allan deviation is a two-sample deviation formed by the average of the squared differences between successive values of a repeatedly measured quantity taken over sampling periods from the measuring interval up to half the maximum measurement time [38,39]. In comparison with the commonly used standard deviation, the Allan deviation is based on measurement to measurement deviation rather than on individual measurement to mean measurement deviation.

The difference compared to using no low-pass and no PI-regulations is clearly visible. The black curve in the left plot of Fig. 8, where the Allan deviation of the voltage measurements are shown, shows a behavior that is compatible with white phase noise ($\sigma(\tau) \propto \tau^{-1}$) for $\tau \geq 1$ s.

This performance improvement of the MR-ToF device can be translated to how long it is able to sustain a certain mass resolving power. This is particularly important when for extended times a purification of the ion distribution shall be applied after the time-of-flight separation, e.g. when delivering a isotopically or isobarically clean beam to a subsequent apparatus for further investigation. To illustrate this, the data presented in Fig. 5 and 6 was used and single time-of-flight spectra were added iteratively. After each addition, the mass resolving power of the resulting ion peak was determined as plotted in Fig. 9. Note that the number of revolutions was chosen to be 1500 which translated to an increased mass resolving power compared to data used in Fig. 4 were the ions traveled for only 1000 revolutions.

The improvement with PI-regulation on the overall stability translates to a constant overall time-of-flight of the ions in the device. That implies that a high mass resolving power of $2.5 \cdot 10^5$ can be maintained for more than one hour (red curve in Fig. 9). When the PI-regulation

Fig. 5. Experimental values measured as a function of time for 65 min (x-axis) *left axis (blue)*: Time of flight of ^{85}Rb ions after 1500 revolutions in the MR-ToF device. For each fit of the centroid ions from 50 experimental cycles, about 150–200 ions, were accumulated. *right axis (black)*: Simultaneously measured voltage of mirror electrode 5 with low-pass filter but without PI-regulation.

Fig. 6. Experimental values measured as a function of time for 65 min (x-axis) *left axis (blue)*: Time of flight of ^{85}Rb ions after 1500 revolutions in the MR-ToF device. *right axis (black)*: Simultaneously measured voltage of mirror electrode 5 with low-pass filter and PI-regulation.

was off (blue curve in Fig. 9) the mass resolving power fluctuated and dropped significantly (by about 50%) in the last 20 min of this measurement.

3.2. Enhancement of mass-measurement accuracy

Mass measurements with the MR-ToF MS of ISOLTRAP have been reported already in a number of publications [13,24,40,25,22].

The time-of-flight/mass relation of the apparatus is generally of the form:

$$t = a\sqrt{m_i} + b, \quad (1)$$

where m_i is the ion mass and a and b are device parameters, of which a depends also on the number of revolutions at which the measurement is

performed and b incorporates mass-independent offsets, such as electronics or switching delays.

For a mass determination one thus performs three ToF measurements, one for the ion of interest, t_x , and two other for ion species of well-known mass, $t_{1,2}$. From this, the unknown (ionic) mass $m_{i,x}$ is determined as [13]:

$$m_{i,x}^{1/2} = C_{TOF} \Delta_{Ref} + \frac{1}{2} \Sigma_{Ref}, \quad (2)$$

where $\Delta_{Ref} = m_{i,1}^{1/2} - m_{i,2}^{1/2}$, $\Sigma_{Ref} = m_{i,1}^{1/2} + m_{i,2}^{1/2}$ and C_{TOF} is the so-called TOF constant:

$$C_{TOF} = \frac{2t_x - t_1 - t_2}{2(t_1 - t_2)}. \quad (3)$$

Fig. 7. Data of Fig. 6 but with expanded y-axis and in addition the laboratory temperature.

Table 2

Relative stability of the voltage and the ToF for the measurements presented in this manuscript. The values are representing the standard deviation of the measurement series divided by its mean.

	no filter, no PI Fig. 2	filter, no PI Fig. 5	filter and PI Fig. 6
ToF drift	1.5 ppm	1.1 ppm	0.35 ppm
voltage drift	2 ppm	1.8 ppm	0.53 ppm

Drifts in any parameter affecting the ion ToF induce a drift of the value of a between the moments at which t_1 , t_2 and t_x are measured, which leads to mass-measurement errors. At ISOLTRAP, the leading-order effects originate from the drifts of the MR-ToF MS voltages outlined so far and from the periodic effect of the RF field of the cooler-buncher, which produces a variation of the ion ToF with the phase of the RF field at the moment of ejection as displayed in Fig. 10.

To circumvent these problems, in all reported ISOLTRAP measurements at least one of the two references was an isobar of the ion of interest, belonging to the same ToF spectrum. This measurement approach has two advantages: first, the isobaric reference will be measured at the same time as the ion of interest and hence not be affected by a systematic drift of a ; second, it will be very close in mass to the ion of interest and hence carry most of the weight of the two references, minimizing the impact of any possible voltage drift before the measurement of the second reference is performed.

Nevertheless, this approach also has a series of shortcomings. First of all, the reliance on isobaric contaminants as references goes against the goal of target-ion-source and mass-spectrometer developments which is to provide pure beams of radioactive isotopes. Second of all, isobaric contaminants can't often be unambiguously identified, case in which the approach is to determine their mass first using Penning-trap mass spectrometry, before passing to the MR-ToF mass measurement, which wastes precious on-line measurement time. Finally, even when

Fig. 8. left figure: Allan deviation of the measured voltage data from Fig. 6, 5 and 2. right figure: Allan deviation of the measured time-of-flight data presented in the same figures as mentioned before.

Fig. 9. Mass resolving power as a function of time. Statistics for the ^{85}Rb ion peak was accumulated with time and the mass resolving power was determined after every 4s. For the spectrum with PI-regulation a total of 131161 ions and for one without PI-regulation a total of 151935 ions were accumulated in 50,000 experimental cycles. The time of flight and voltage values for this data are the ones presented in Fig. 5 and 6.

Fig. 10. Time-of-flight of ^{85}Rb with 1000 revolutions inside the MR-ToF MS with the phase of the RF field of the cooler-buncher at the moment of ion ejection scanned over three RF periods. The blue line is a sinusoidal fit to the data. For this measurement, the RF generator was running until the ions had left the cooler-buncher.

present in the spectrum and identified, the isobaric contaminants are most of the time much more abundant than the ion of interest and require limiting artificially the ion rate through of the MR-ToF MS in order to avoid saturation effects of the detector and the data acquisition or Coulomb effects.

All these aspects make the use of well-known, off-line references highly desirable on the long term. This, however, makes the voltage stability of the MR-ToF MS a critical aspect which directly impacts its accuracy. Consequently, a study of the accuracy of MR-ToF MS measurements when using only off-line references was performed, in connection to the reported improvement of voltage stability.

Such a accuracy measurement is performed in a cycle of three subsequent ToF recordings, one for each reference species and, in between, one for the ion of interest. In this study, the mass of ^{87}Rb was measured with ^{85}Rb and ^{133}Cs as references and then compared to the literature value. All masses were taken from the AME2016 [41].

In order to minimize the impact of the RF field of the cooler-buncher on the ToF of the ions, the buncher was operated in phase-locked mode. This means that the RF generator is triggered, with fixed initial phase, and is stopped close to the moment of ion ejection. This ensures that between different ion bunches of the same mass, the ToF shift due to the

Fig. 11. Schematic of the timing pattern of the ion preparation in the ISOLT-RAP buncher and amplitude of the RF field during the ion extraction. During the decrease of the RF amplitude, the ion cloud expands.

RF phase has the same magnitude and the same direction, which solves the issue of ToF stability. However for ions of very different mass, because of different transport times in the buncher extraction section, the shift can have different magnitude and even sign. This latter aspect poses a serious issue for mass measurements.

In order to solve this latter problem one exploits the fact that the stopping of the RF field is not instantaneous, due to the energy stored in the RF system, electrodes and resonance circuits. The electric field amplitude decreases exponentially with a time constant of $\tau = 8.7 \mu\text{s}$. In order to minimize the effect of the RF field on the ion ToF, the RF field is thus switched off before the extraction of the ions. The exact time difference between the end of the RF gate and ion extraction is optimized to be as large as possible, without sacrificing ion count rate [42]. The timing pattern is schematically displayed in Fig. 11.

The weaker electric RF field at the moment of ion extraction leads to a strong reduction of the ToF dependence on the RF phase as can be seen in Fig. 12. Finally, in order to average out any remaining effect, during each ToF measurement the buncher holding time is scanned in steps of 25ns over an integer number of RF periods (in our case, the frequency was 1.405 MHz). This effectively translates to a scan of the last RF phase seen by the ions at extraction, as the initial phase is fixed. This alone would not have been a sufficient remedy without optimizing the RF stopping time, because the RF field produces an out-of-phase oscillation of both ion ToF and count rate, which when averaged over many phases leads to a ToF bias with respect to the correct value (as shown in [42]).

With this preparation of the RFQ cooler-buncher, the accuracy of MR-ToF mass measurements was tested with the PI loop off and on. The results of many consecutive measurements performed without the PI regulation are presented in Fig. 13 as difference between the

Fig. 12. Time-of-flight of ^{85}Rb with 1000 revolutions inside the MR-ToF MS with the phase of the RF field of the RFQ cooler-buncher at the moment of ion ejection scanned over three RF periods as in Fig. 10, but using the timing pattern of Fig. 11 with optimized RF delay.

Fig. 13. Series of measured mass of ^{87}Rb with ^{85}Rb and ^{133}Cs as references, the PI loop off and 1000 revolutions inside the MR-ToF MS. During a single time-of-flight measurement with a length of about minute, 250–350 ions were recorded. The measurement index denotes the number of the ^{85}Rb - ^{87}Rb - ^{133}Cs cycle. The error bars do only account for the uncertainty of the fit of the mean time-of-flight of the single species. Additionally, the weighted mean and the standard deviation are stated. The histogram on the right is the projection on the y-axis weighted with the statistical weight and normalized.

Fig. 14. Measurement of the mass of ^{87}Rb as in Fig. 13, but with the PI loop switched on.

determined mass of ^{87}Rb and the AME2016 value. The ToF measurement for each ion species takes about a minute before changing ion species (which with a moderate ion rate corresponds to about 250–350 recorded ions per peak) in an automated sequence which ensures reproducibility.

One notices first of all that the experimental results scatter around their mean with a standard deviation of about 70 keV, which is significantly larger than the individual statistical uncertainties, as illustrated by the Birge ratio [43] of 2.70. Second, the mean of all results is only about 7 keV away from the AME value. This means that in an on-line experiment a result of very good accuracy can be achieved if enough measurements are performed in order to average all voltage fluctuations. If, however, only a single, or a limited number of measurements can be performed, the systematic uncertainty is given by the standard deviation of the distribution in Fig. 13, so about 70 keV in the case of no PI regulation.

Fig. 14 depicts a series of mass measurements performed for the exact same conditions as for Fig. 13, but with the PI regulation on. As the ion ToF is more stable with the activated PI loop, the standard deviation of the scattering around the AME mass value is reduced from 70.3 keV to 40.3 keV. One notices that the average of all results differs from the AME value by about the same amount of ≈ 7 keV, which suggests that this error originates from other sources than the voltage drifts of the MR-ToF MS. The systematic uncertainty of a single measurement, however, is reduced by almost a factor 2. The reduced Birge ratio of only 1.6 shows that, combining the PI regulation with very short measurements, which reduce the available time for voltage fluctuations (significantly below a minute) and increase the statistical uncertainty, would lead to experimental results scattering with the normal, statistical standard deviation.

4. Summary and outlook

We have shown that we can stabilize a voltage supply to a level of 0.5 ppm by use of a low-pass filter and a software-based PI controller. This method was applied to the voltage of the most crucial element in an MR-ToF device, the mirror electrode which defines the turnaround point of the ion bunch. Significant improvements in terms of short- and long-term stability were observed which leads to a more stable total flight time of the ions in the MR-ToF device and thus to a mass resolving power which is stable even on time scales of hours. This is particularly useful when a subsequent purification of the ion bunch (using the in-trap lift [1] or a Bradbury Nielsen beam gate [44,36]) is intended for extended time durations. Also the precision of mass measurements using reference species that are non isobaric with respect to the ion of interest is enhanced. It is planned to add a PI-regulation of the voltage which is responsible for the next to leading order fluctuations, in our case the one of the neighbouring electrode.

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References

- [1] F. Wienholtz, S. Kreim, M. Rosenbusch, L. Schweikhard, R. Wolf, Mass-selective ion ejection from multi-reflection time-of-flight devices via a pulsed in-trap lift, *Int. J. Mass Spectrom.* 421 (2017) 285–293.
- [2] R. Wolf, F. Wienholtz, D. Atanasov, D. Beck, K. Blaum, C. Borgmann, F. Herfurth, M. Kowalska, S. Kreim, Y.A. Litvinov, D. Lunney, V. Manea, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, K. Zuber, Isoltrap's multi-reflection time-of-flight mass separator/spectrometer, *Int. J. Mass Spectrom.* 349–350 (2013) 123–133 100 years of Mass Spectrometry.
- [3] P. Schury, M. Wada, Y. Ito, D. Kaji, F. Arai, M. MacCormick, I. Murray, H. Haba, S. Jeong, S. Kimura, H. Koura, H. Miyatake, K. Morimoto, K. Morita, A. Ozawa, M. Rosenbusch, M. Reponen, P.-A. Söderström, A. Takamine, T. Tanaka, H. Wollnik, First online multireflection time-of-flight mass measurements of isobar chains produced by fusion-evaporation reactions: Toward identification of superheavy elements via mass spectroscopy, *Phys. Rev. C* 95 (2017) 011305.
- [4] T. Dickel, W. Pla, A. Becker, U. Czok, H. Geissel, E. Haettner, C. Jesch, W. Kinsel, M. Petrick, C. Scheidenberger, A. Simon, M. Yavor, A high-performance multiple-reflection time-of-flight mass spectrometer and isobar separator for the research with exotic nuclei, *Nucl. Instrum. Methods Phys. Res., Sect. A* 777 (2015) 172–188.
- [5] T.Y. Hirsh, N. Paul, M. Burkey, A. Arahmanian, F. Buchinger, S. Caldwell, J.A. Clark, A.F. Levand, L.L. Ying, S.T. Marley, G.E. Morgan, A. Nystrom, R. Orford, A.P. Galvn, J. Rohrer, G. Savard, K.S. Sharma, K. Siegl, First operation and mass separation with the caribu mr-tof, *Nucl. Instrum. Methods Phys. Res., Sect. B* 376 (2016) 229–232 Proceedings of the XVIIth International Conference on Electromagnetic Isotope Separators and Related Topics (EMIS2015), Grand Rapids, MI, U.S.A., 11–15 May 2015.
- [6] O. Heber, P.D. Witte, A. Diner, K.G. Bhushan, D. Strasser, Y. Tokar, M.L. Rappaport, I. Ben-Itzhak, N. Altstein, D. Schwalm, A. Wolf, D. Zajfman, Electrostatic ion beam trap for electron collision studies, *Rev. Sci. Instrum.* 76 (2004) 013104.
- [7] O. Heber, P.D. Witte, A. Diner, K.G. Bhushan, D. Strasser, Y. Tokar, M.L. Rappaport, I. Ben-Itzhak, N. Altstein, D. Schwalm, A. Wolf, D. Zajfman, Electrostatic ion beam trap for electron collision studies, *Rev. Sci. Instrum.* 76 (2005) 013104.
- [8] W. Paul, Electromagnetic traps for charged and neutral particles, *Rev. Mod. Phys.* 62 (1990) 531–540.
- [9] A. Naaman, K.G. Bhushan, H.B. Pedersen, N. Altstein, O. Heber, M.L. Rappaport, R. Moalem, D. Zajfman, Metastable states of negative carbon clusters: C_n^- , $n = 2-6$, *J. Chem. Phys.* 113 (2000) 4662–4667.
- [10] Y. Tokar, O. Aviv, M. Eritt, M.L. Rappaport, O. Heber, D. Schwalm, D. Zajfman, Radiative cooling of Al_4^- clusters, *Phys. Rev. A* 76 (2007) 053201.
- [11] M. Lange, M. Froese, S. Menk, J. Varju, R. Bastert, K. Blaum, J.R.C. López-Urrutia, F. Fellenberger, M. Grieser, R. von Hahn, O. Heber, K.-U. Kühnel, F. Laux, D.A. Orlov, M.L. Rappaport, R. Repnow, C.D. Schröter, D. Schwalm, A. Shornikov, T. Sieber, Y. Tokar, J. Ullrich, A. Wolf, D. Zajfman, A cryogenic electrostatic trap for long-time storage of kev ion beams, *Rev. Sci. Instrum.* 81 (2010) 055105.
- [12] M.W. Froese, M. Lange, S. Menk, M. Grieser, O. Heber, F. Laux, R. Repnow, T. Sieber, Y. Tokar, R. von Hahn, A. Wolf, K. Blaum, The decay of ion bunches in the self-bunching mode, *New J. Phys.* 14 (2012) 073010.
- [13] F. Wienholtz, D. Beck, K. Blaum, C. Borgmann, M. Breitenfeldt, R.B. Cakirli, S. George, F. Herfurth, J.D. Holt, M. Kowalska, S. Kreim, D. Lunney, V. Manea, J. Menéndez, D. Neidherr, M. Rosenbusch, L. Schweikhard, A. Schwenk, J. Simonis, J. Stanja, R.N. Wolf, K. Zuber, Masses of exotic calcium isotopes pin down nuclear forces, *Nature* 498 (2013) 346.
- [14] E. Leistenschneider, M.P. Reiter, S. Ayet San Andrés, B. Kootte, J.D. Holt, P. Navrátil, C. Babcock, C. Barbieri, B.R. Barquest, J. Bergmann, J. Bollig, T. Brunner, E. Dunling, A. Finlay, H. Geissel, L. Graham, F. Greiner, H. Hergert, C. Hornung, C. Jesch, R. Klawitter, Y. Lan, D. Lascar, K.G. Leach, W. Lippert, J.E. McKay, S.F. Paul, A. Schwenk, D. Short, J. Simonis, V. Somà, R. Steinbrügge, S.R. Stroberg, R. Thompson, M.E. Wieser, C. Will, M. Yavor, C. Andreoiu, T. Dickel, I. Dillmann, G. Gwinner, W.R. Plaß, C. Scheidenberger, A.A. Kwiakowski, J. Dilling, Dawning of the $N = 32$ shell closure seen through precision mass measurements of neutron-rich titanium isotopes, *Phys. Rev. Lett.* 120 (2018) 062503.
- [15] Y. Ito, P. Schury, M. Wada, F. Arai, H. Haba, Y. Hirayama, S. Ishizawa, D. Kaji, S. Kimura, H. Koura, M. MacCormick, H. Miyatake, J.Y. Moon, K. Morimoto, K. Morita, M. Mukai, I. Murray, T. Niwase, K. Okada, A. Ozawa, M. Rosenbusch, A. Takamine, T. Tanaka, Y.X. Watanabe, H. Wollnik, S. Yamaki, First direct mass measurements of nuclides around $Z = 100$ with a multireflection time-of-flight mass spectrograph, *Phys. Rev. Lett.* 120 (2018) 152501.
- [16] S. Kreim, D. Atanasov, D. Beck, K. Blaum, C. Böhm, C. Borgmann, M. Breitenfeldt, T. Cocolios, D. Fink, S. George, A. Herlert, A. Kellerbauer, U. Köster, M. Kowalska, D. Lunney, V. Manea, E.M. Ramirez, S. Naimi, D. Neidherr, T. Nicol, R. Rossel, M. Rosenbusch, L. Schweikhard, J. Stanja, F. Wienholtz, R. Wolf, K. Zuber, Recent exploits of the isoltrap mass spectrometer, *Nucl. Instrum. Methods Phys. Res., Sect. B* 317 (2013) 492–500 XVth International Conference on ElectroMagnetic Isotope Separators and Techniques Related to their Applications, December 2–7, 2012 at Matsue, Japan.
- [17] M. Mukherjee, D. Beck, K. Blaum, G. Bollen, J. Dilling, S. George, F. Herfurth, A. Herlert, A. Kellerbauer, H.J. Kluge, S. Schwarz, L. Schweikhard, C. Yazdijian, Isoltrap: an on-line penning trap for mass spectrometry on short-lived nuclides, *Eur. Phys. J. A* 35 (2008) 1–29.
- [18] E. Kugler, D. Fiander, B. Johnson, H. Haas, A. Przewłoka, H. Ravn, D. Simon, K. Zimmer, The new CERN-ISOLDE on-line mass-separator facility at the PS-Booster, *Nucl. Instrum. Methods Phys. Res., Sect. B* 70 (1992) 41–49.
- [19] A. Welker, N.A.S. Althubiti, D. Atanasov, K. Blaum, T.E. Cocolios, F. Herfurth, S. Kreim, D. Lunney, V. Manea, M. Mougeot, D. Neidherr, F. Nowacki, A. Poves, M. Rosenbusch, L. Schweikhard, F. Wienholtz, R.N. Wolf, K. Zuber, Binding energy of ^{79}Cu : Probing the structure of the doubly magic ^{78}Ni from only one proton away, *Phys. Rev. Lett.* 119 (2017) 192502.
- [20] V. Manea, D. Atanasov, D. Beck, K. Blaum, C. Borgmann, R.B. Cakirli, T. Eronen, S. George, F. Herfurth, A. Herlert, M. Kowalska, S. Kreim, Y.A. Litvinov, D. Lunney, D. Neidherr, M. Rosenbusch, L. Schweikhard, F. Wienholtz, R.N. Wolf, K. Zuber, Collective degrees of freedom of neutron-rich $A \approx 100$ nuclei and the first mass measurement of the short-lived nuclide ^{100}Rb , *Phys. Rev. C* 88 (2013) 054322.
- [21] M. Rosenbusch, P. Ascher, D. Atanasov, C. Barbieri, D. Beck, K. Blaum, C. Borgmann, M. Breitenfeldt, R.B. Cakirli, A. Cipollone, S. George, F. Herfurth, M. Kowalska, S. Kreim, D. Lunney, V. Manea, P. Navrátil, D. Neidherr, L. Schweikhard, V. Somà, J. Stanja, F. Wienholtz, R.N. Wolf, K. Zuber, Probing the $N = 32$ shell closure below the magic proton number $Z = 20$: Mass measurements of the exotic isotopes $^{52,53}\text{K}$, *Phys. Rev. Lett.* 114 (2015) 202501.
- [22] M. Mougeot, D. Atanasov, K. Blaum, K. Chrysalidis, T.D. Goodacre, D. Fedorov, V. Fedosseev, S. George, F. Herfurth, J.D. Holt, D. Lunney, V. Manea, B. Marsh, D. Neidherr, M. Rosenbusch, S. Rothe, L. Schweikhard, A. Schwenk, C. Seiffert, J. Simonis, S.R. Stroberg, A. Welker, F. Wienholtz, R.N. Wolf, K. Zuber, Precision mass measurements of $^{58-63}\text{Cr}$: Nuclear collectivity towards the $N = 40$ island of inversion, *Phys. Rev. Lett.* 120 (2018) 232501.
- [23] R.N. Wolf, D. Beck, K. Blaum, C. Böhm, C. Borgmann, M. Breitenfeldt, N. Chamel, S. Goriely, F. Herfurth, M. Kowalska, S. Kreim, D. Lunney, V. Manea, E. Minaya Ramirez, S. Naimi, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, F. Wienholtz, K. Zuber, Plumbing neutron stars to new depths with the binding energy of the exotic nuclide ^{82}Zn , *Phys. Rev. Lett.* 110 (2013) 041101.
- [24] D. Atanasov, P. Ascher, K. Blaum, R.B. Cakirli, T.E. Cocolios, S. George, S. Goriely, F. Herfurth, H.-T. Janka, O. Just, M. Kowalska, S. Kreim, D. Kislser, Y.A. Litvinov, D. Lunney, V. Manea, D. Neidherr, M. Rosenbusch, L. Schweikhard, A. Welker, F. Wienholtz, R.N. Wolf, K. Zuber, Precision mass measurements of $^{129-131}\text{Cd}$ and their impact on stellar nucleosynthesis via the rapid neutron capture process, *Phys. Rev. Lett.* 115 (2015) 232501.
- [25] A. Welker, P. Filianin, N.A.S. Althubiti, D. Atanasov, K. Blaum, T.E. Cocolios, S. Eliseev, F. Herfurth, S. Kreim, D. Lunney, V. Manea, D. Neidherr, Y. Novikov, M. Rosenbusch, L. Schweikhard, F. Wienholtz, R.N. Wolf, K. Zuber, Precision electron-capture energy in ^{202}Pb and its relevance for neutrino mass determination, *Eur. Phys. J. A* 53 (2017) 153.
- [26] N.A. Althubiti, D. Atanasov, K. Blaum, T.E. Cocolios, T. Day Goodacre, G.J. Farooq-Smith, D.V. Fedorov, V.N. Fedosseev, S. George, F. Herfurth, K. Heyde, S. Kreim, D. Lunney, K.M. Lynch, V. Manea, B.A. Marsh, D. Neidherr, M. Rosenbusch, R.E. Rossel, S. Rothe, L. Schweikhard, M.D. Seliverstov, A. Welker, F. Wienholtz, R.N. Wolf, K. Zuber, Spectroscopy of the long-lived excited state in the neutron-deficient nuclides $^{195,197,199}\text{Po}$ by precision mass measurements, *Phys. Rev. C* 96 (2017) 044325.
- [27] R. Wolf, D. Atanasov, K. Blaum, S. Kreim, D. Lunney, V. Manea, M. Rosenbusch, L. Schweikhard, A. Welker, F. Wienholtz, K. Zuber, Background-free beta-decay half-life measurements by in-trap decay and high-resolution mr-tof mass analysis, *Nucl. Instrum. Methods Phys. Res., Sect. B* 376 (2016) 275–280 Proceedings of the XVIIth International Conference on Electromagnetic Isotope Separators and Related Topics (EMIS2015), Grand Rapids, MI, U.S.A., 11–15 May 2015.
- [28] V. Fedosseev, K. Chrysalidis, T.D. Goodacre, B. Marsh, S. Rothe, C. Seiffert, K. Wendt, Ion beam production and study of radioactive isotopes with the laser ion source at isolde, *J. Phys. G: Nucl. Part. Phys.* 44 (2017) 084006.
- [29] B. Marsh, B. Andel, A. Andreyev, S. Antalic, D. Atanasov, A. Barzakh, B. Bastin, C. Borgmann, L. Capponi, T. Cocolios, T.D. Goodacre, M. Dehairs, X. Derckx, H.D. Witte, D. Fedorov, V. Fedosseev, G. Focke, D. Fink, K. Flanagan, S. Franchoo, L. Ghys, M. Huyse, N. Imai, Z. Kalaninova, U. Köster, S. Kreim, N. Kesteloot, Y. Kudryavtsev, J. Lane, N. Lemesne, V. Liberati, D. Lunney, K. Lynch, V. Manea, P. Nicol, D. Pauwels, L. Popescu, D. Radulov, E. Rapisarda, M. Rosenbusch, R. Rossel, S. Rothe, L. Schweikhard, M. Seliverstov, S. Sels, A. Sjödin, V. Truesdale, C.V. Beveren, P.V. Duppen, K. Wendt, F. Wienholtz, R. Wolf, S. Zemlyanov, New developments in the in-source spectroscopy method at RILIS/ISOLDE, *Nucl. Instrum. Methods Phys. Res., Sect. B* 317 (2013) 550–556 XVth International Conference on ElectroMagnetic Isotope Separators and Techniques Related to their Applications, December 2–7, 2012 at Matsue, Japan.
- [30] J.G. Cubiss, A.E. Barzakh, M.D. Seliverstov, A.N. Andreyev, B. Andel, S. Antalic, P. Ascher, D. Atanasov, D. Beck, J. Biero, K. Blaum, C. Borgmann, M. Breitenfeldt, L. Capponi, T.E. Cocolios, T. Day Goodacre, X. Derckx, H. De Witte, J. Elseviers, D.V. Fedorov, V.N. Fedosseev, S. Fritzsche, L.P. Gaffney, S. George, L. Ghys, F.P. Heßberger, M. Huyse, N. Imai, Z. Kalaninova, D. Kislser, U. Köster, M. Kowalska, S. Kreim, J.F.W. Lane, V. Liberati, D. Lunney, K.M. Lynch, V. Manea, B.A. Marsh, S. Mitsuka, P.L. Molkanov, Y. Nagame, D. Neidherr, K. Nishio, S. Ota, D. Pauwels, L. Popescu, D. Radulov, E. Rapisarda, J.P. Revill, M. Rosenbusch, R.E. Rossel, S. Rothe, K. Sandhu, L. Schweikhard, S. Sels, V.L. Truesdale, C. Van Beveren, P. Van den Bergh, Y. Wakabayashi, P. Van Duppen, K.D.A. Wendt, F. Wienholtz, B.W. Whitmore, G.L. Wilson, R.N. Wolf, K. Zuber, Charge radii and electromagnetic moments of $^{195-211}\text{At}$, *Phys. Rev. C* 97 (2018) 054327.
- [31] N.A. Marsh, T. Day Goodacre, S. Sels, Y. Tsunoda, B. Andel, A.N. Andreyev, N.A. Althubiti, D. Atanasov, A.E. Barzakh, J. Billowes, K. Blaum, T.E. Cocolios, J.G. Cubiss, J. Dobaczewski, G.J. Farooq-Smith, D.V. Fedorov, V.N. Fedosseev, K.T. Flanagan, L.P. Gaffney, L. Ghys, M. Huyse, S. Kreim, D. Lunney, K.M. Lynch, V. Manea, Y. Martinez Palenzuela, P.L. Molkanov, T. Otsuka, A. Pastore, M. Rosenbusch, R.E. Rossel, S. Rothe, L. Schweikhard, M.D. Seliverstov, P. Spagnoletti, C. Van Beveren, P. Van Duppen, M. Veinhard, E. Verstraelen, A. Welker, K. Wendt, F. Wienholtz, R.N. Wolf, A. Zadovnaya, K. Zuber, Characterization of the shape-staggering effect in mercury nuclei, *Nat. Phys.* 14

- (2018) 1163–1167.
- [32] J. Cubiss, A. Barzakh, A. Andreyev, M.A. Monthery, N. Althubiti, B. Andel, S. Antalic, D. Atanasov, K. Blaum, T. Cocolios, T.D. Goodacre, R. de Groote, A. de Roubin, G. Farooq-Smith, D. Fedorov, V. Fedosseev, R. Ferrer, D. Fink, L. Gaffney, L. Ghys, A. Gredley, R. Harding, F. Herfurth, M. Huyse, N. Imai, D. Joss, U. Köster, S. Kreim, V. Liberati, D. Lunney, K. Lynch, V. Manea, B. Marsh, Y.M. Palenzuela, P. Molkanov, P. Mosat, D. Neidherr, G. O'Neill, R. Page, T. Procter, E. Rapisarda, M. Rosenbusch, S. Rothe, K. Sandhu, L. Schweikhard, M. Seliverstov, S. Sels, P. Spagnoletti, V. Truesdale, C.V. Beveren, P.V. Duppen, M. Veinhard, M. Venhart, M. Veselský, F. Wearing, A. Welker, F. Wienholtz, R. Wolf, S. Zemlyanoy, K. Zuber, Change in structure between the $I=1/2$ states in ^{181}Tl and $^{177,179}\text{Au}$, *Phys. Lett. B* 786 (2018) 355–363.
- [33] A. Gottberg, T. Mendonca, R. Luis, J. Ramos, C. Seiffert, S. Cimmino, S. Marzari, B. Crepieux, V. Manea, R. Wolf, F. Wienholtz, S. Kreim, V. Fedosseev, B. Marsh, S. Rothe, P. Vaz, J. Marques, T. Stora, Experimental tests of an advanced proton-to-neutron converter at ISOLDE-CERN, *Nucl. Instrum. Methods Phys. Res., Sect. B* 336 (2014) 143–148.
- [34] F. Herfurth, Segmented linear RFQ traps for nuclear physics, *Nucl. Instrum. Methods Phys. Res. Section B* 204 (2003) 587–591 14th International Conference on Electromagnetic Isotope Separators and Techniques Related to their Applications.
- [35] R.N. Wolf, G. Marx, M. Rosenbusch, L. Schweikhard, Static-mirror ion capture and time focusing for electrostatic ion-beam traps and multi-reflection time-of-flight mass analyzers by use of an in-trap potential lift, *Int. J. Mass Spectrom.* 313 (2012) 8–14.
- [36] R. Wolf, D. Beck, K. Blaum, C. Böhm, C. Borgmann, M. Breitenfeldt, F. Herfurth, A. Herlert, M. Kowalska, S. Kreim, D. Lunney, S. Naimi, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, F. Wienholtz, K. Zuber, On-line separation of short-lived nuclei by a multi-reflection time-of-flight device, *Nucl. Instrum. Methods Phys. Res., Sect. A* 686 (2012) 82–90.
- [37] D.W. Allan, Statistics of atomic frequency standards, *Proc. IEEE* 54 (1966) 221–230.
- [38] D.W. Allan, Should the classical variance be used as a basic measure in standards metrology? *IEEE Trans. Instrum. Measure.* IM-36 (1987) 646–654.
- [39] J. Levine, Introduction to time and frequency metrology, *Rev. Sci. Instrum.* 70 (1999) 2567–2596.
- [40] A. de Roubin, D. Atanasov, K. Blaum, S. George, F. Herfurth, D. Kisler, M. Kowalska, S. Kreim, D. Lunney, V. Manea, E. Minaya Ramirez, M. Mougeot, D. Neidherr, M. Rosenbusch, L. Schweikhard, A. Welker, F. Wienholtz, R.N. Wolf, K. Zuber, Nuclear deformation in the $A \approx 100$ region: comparison between new masses and mean-field predictions, *Phys. Rev. C* 96 (2017) 014310.
- [41] W.J. Huang, G. Audi, M. Wang, F.G. Kondev, S. Naimi, X. Xu, The ame2016 atomic mass evaluation (i). evaluation of input data; and adjustment procedures, *Chinese Phys. C* 41 (2017) 030002.
- [42] T. Steinsberger, Microwave injection for the ALPHATRAPP experiment and developments of the multi-reflection time-of-flight technique of the ISOLTRAPP experiment, Ruprecht-Karls-Universität, Heidelberg, 2018 Master's thesis.
- [43] R.T. Birge, The calculation of errors by the method of least squares, *Phys. Rev.* 40 (1932) 207–227.
- [44] N.E. Bradbury, R.A. Nielsen, Absolute values of the electron mobility in hydrogen, *PR* 49 (1936) 388–393.