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A multi-reflection time-of-flight mass separator for isobaric purification of radioactive ion beams

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Abstract A multi-reflection time-of-flight mass separator (MR-ToF-MS) for the enhancement of the performance of the Penning-trap mass spectrometer ISOLTRAP at the on-line isotope separator ISOLDE/CERN has been built and investigated at an off-line test facility. The MR-ToF-MS consists of two ion-optical mirrors between which oscillating ions are separated according to their different mass-over-charge ratios m/q. Flight paths of several hundreds of meters are folded to an apparatus length of less than one meter. Preliminary tests resulted in a mass-resolving power of up to $m/\Delta m \approx 80{,}000$, and the separation was demonstrated for the isobaric ions CO^+ and N_2^+. The MR-ToF-MS will support the existing purification methods of ISOLTRAP and will extend the access to nuclides produced with high isobaric contamination yields at the ISOLDE facility.

Keywords Isobaric contamination · Isobar separator · Multi-reflection time-of-flight mass separator · ISOLTRAP · Precision mass measurement · Exotic nuclei

1 Introduction

As early as in the late 1950's the so called "Farvitron" trapped ions electrostatically between two ion mirrors [1, 2]. Only several decades later, the use of multiple-reflection ion trajectories was studied by Wollnik and coworkers [3]. They suggested that by folding the trajectory of the ions in ion-mirror arrangements one could

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increase the mass resolving power of time-of-flight mass spectrometers without increasing their physical dimensions. This was demonstrated by Piyadasa et al. [4] in combination with a MALDI ion source and by Casares et al. [5] with a three-mirror arrangement where a mass resolving power of $m/\Delta m \approx 50{,}000$ was achieved. Further devices were constructed mainly consisting of two rotationally symmetric ion mirrors [6–8] or of planar mirrors [9, 10]. These setups are referred to as multi-reflection time-of-flight mass spectrometers (MR-ToF-MS). Due to their high mass-resolving powers, potential short operation cycles and compact dimensions, it has been suggested to use MR-ToF-devices as mass separators for the purification of isobaric ion ensembles [7, 11–13], as spectrometers in precision mass measurements of rare isotopes [14, 15] and for nuclear-decay studies [16]. The capability of MR-ToF-MS to achieve mass spectra with a resolving power in the order of $m/\Delta m \approx 10^5$ as necessary for these purposes has been demonstrated [10, 12, 13].

Devices with similar electrode configurations are also referred to as electrostatic ion beam traps. After their simultaneous introduction by Benner et al. [17] and Zajfman et al. [18] several such devices have been built for various applications in atomic, molecular and cluster research [19–31]. Their operational principle is in many cases related to beam bunching caused by space-charge effects [32].

Accurate atomic mass information provides a key towards the understanding of nuclear structure. More specifically, the nuclear binding energies derived from experimental mass values are important input parameters for, e.g., the modeling of stellar nucleosynthesis and serve as test for the standard model (for examples see [33–35]). At the Penning-trap mass spectrometer ISOLTRAP [36] at ISOLDE/CERN, for which the present setup has been developed, as well as at several other facilities [37, 38] various measurements of short-lived nuclides have been performed with a mass accuracy of down to $\delta m/m = 10^{-8}$ [39] by determination of the cyclotron frequency of stored ions in a Penning trap.

One important condition for high-accuracy mass determinations is a pure ensemble of the ions of interest. The presence of other ions in the trap weakens the strength of the resonance and thus increases the statistical uncertainty. In addition, the presents of ions with similar masses, such as of isobars, lead to systematic shifts of the resonance frequencies. To remove contaminating ions a preparation Penning trap in front of the high-precision trap acts as purifier by use of resonant buffer-gas cooling [40]. However, contaminant ions of similar mass and of amounts significantly exceeding that of the ions of interest lead to problems [41–45]. With the mass separator described below we plan to overcome these limits.

2 Experimental setup

The multi-reflection time-of-flight mass separator for isobar separation at the ISOLTRAP experiment at ISOLDE/CERN (Geneva) has been built up and tested at the University of Greifswald. The test setup as described in the following is shown in Fig. 1.

2.1 Ionization and pulsing

A commercial electron-impact ion source (PRISMA QMS 200 from Pfeiffer Vacuum) delivered the isobaric doublet N_2^+/CO^+ for the off-line tests of the

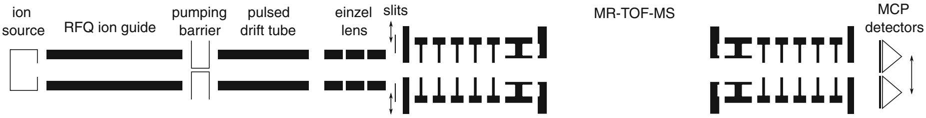

Fig. 1 Test setup of the MR-ToF-MS (not to scale)

MR-ToF-MS. An RFQ ion guide ($f = 2.57$ MHz, $r_0 = 7$ mm) was used to transfer the continuous ion beam from the high pressure region of the source to the UHV region of the injection ion optics and the mass separator. The injection optics consisted of a pulsed drift tube (length 120 mm, inner diameter 20 mm) and an einzel lense. By switching the pulsed drift tube rapidly with a solid-state switch (Behlke Power Electronics, HTS 06-01-GSM), a $\Delta t \approx 100$ ns section of the continuous beam of $E_{\mathrm{kin}} = 150$ eV was cut out and accelerated to $E_{\mathrm{kin}} = 2.2$ keV. The increase in energy distribution due to ions flying between pulsed drift tube and einzel lens while the acceleration potential was switched on was reduced by the energy-depended focusing of the negative einzel lens voltage and the injection diaphragm of the mass separator. According to simulations, the energy spread was $\Delta E \approx 45$ eV (FWHM).

2.2 Multi-reflection time-of-flight mass separator

The mass separator was based on the design of the electrostatic ion beam trap at Rehovot/Israel [25, 32]. As shown in Fig. 2, it consisted of two electrostatic mirrors (each a set of five electrodes) in combination with einzel lenses and shielded by grounded inner and outer end-electrodes. The mirror electrodes had an inner aperture of 16 mm, a thickness of 3 mm at the aperture and were equally spaced by 20 mm. The lens electrode had an inner diameter of 26 mm, length of 30 mm and was positioned 11.5 mm from the neighboring mirror electrode. The mirrors were mounted on three support rods at a distance of 480 mm, separated by a nearly field-free drift region. This resulted in a total length of around 800 mm, as required by the limited space of the ISOLTRAP-beamline where the device will be used. The spacing and insulation between adjacent electrodes was accomplished by ceramic balls (0.3125 inch diameter), placed in three conical holes on both sides of each electrode.

The mass separator was mounted in a CF 200-CF 100 4-way reducing-cross vacuum chamber that was evacuated by a CF 200 cryopump (Oerlikon Leybold, CV 1500 BV-L). The residual gas pressure without gas flow from the ion source was $p \approx 1 \times 10^{-10}$ mbar measured with an extractor gauge (Oerlikon Leybold, IE 514) in the middle of the field-free section.

Corresponding electrodes on both mirrors were connected to the same high-voltage pulser to reduce the technical effort and conserve the symmetry of the system. Each pair of electrodes was activated/deactivated by a fast solid-state switch (Behlke Power Electronics, HTS-151-01-GSM). The supply voltage was produced by high-precision high-voltage modules (Applied Kilovolts), model HP010P/N (0 to $+/-10$ kV) for the mirror electrodes and HP015N (0 to -15 kV) for the lens electrodes. These modules were controlled by a PC with a multi-channel 16 bit digital-analog-converter (National Instruments, NI-6703), allowing voltage steps of 150 mV for model HP010P/N and 225 mV for model HP015N. Each of the six high-

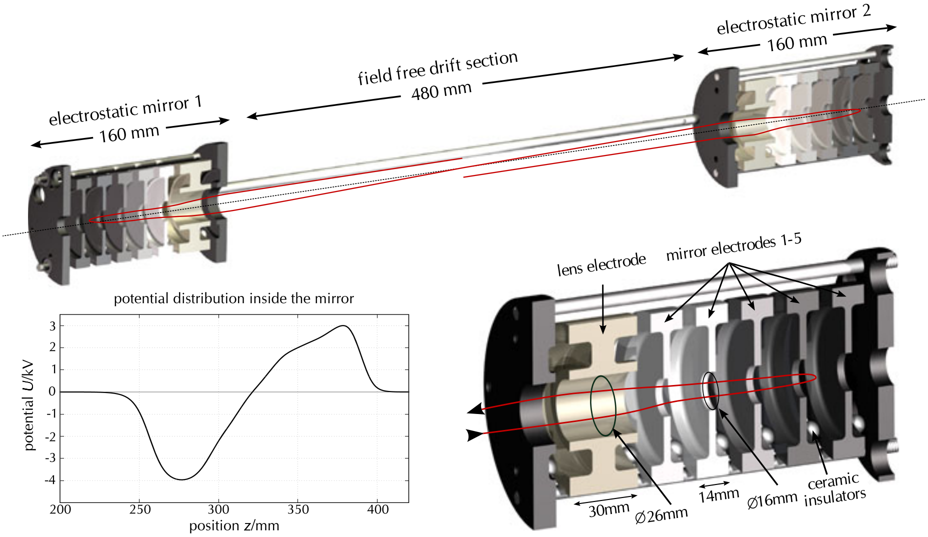

Fig. 2 Overview of the electrodes of the multi-reflection time-of-flight mass separator (*top*), detailed drawing (*bottom right*) and potential as function of position on the axis (*bottom left*, $z = 0$ is the center of the apparatus) of the electrode set on the right

voltage pulsing units was buffered by a stack of high-voltage capacitors ranging from 1 nF to 1 μF to deliver the necessary current for a fast switching, charging the electrodes with a precision of $< 1 \times 10^{-3}$, and to prevent large load changes on the power supplies. The pulsing circuits were able to drive the system with a maximum repetition rate of about 220 Hz while still keeping the specifications of the high voltage devices. The stability of the power supplies was crucial with respect to the reproducibility and thus to the achievable mass resolving power. For a high throughput of separated ions, the ability to repeat the separation cycle with high rates was as well significant.

2.3 Ion detection and data acquisition

After ejection from the MR-ToF-MS, the ion signal was detected either with a conventional 25 mm-diameter multichannelplate detector in chevron arrangement with metal anode (Photonis USA, APD3025) or with a 50 mm-diameter MCP detector with phosphor screen (Tectra DE, MCP-50-D-R-P43) for beam-profile analysis. The data was recorded with a multi-channel analyzer (Becker & Hickl, MSA300 or Stanford Research Systems, SR430) for the metal anode MCP detector and with a low-speed CCD-camera for the phosphor-screen MCP detector.

3 Principle of operation and optimization procedure

When the mirror-electrode potentials are switched down ions can enter the MR-ToF-MS. Afterwards the mirror potentials are activated again and the ions perform oscillations inside the device if the electric potential in the mirrors is at least at one point of the optical axis higher than the kinetic energy of the ions divided by their charge. As in a time-of-flight mass analyzer of several hundred meters length that is folded to less than one meter, ion species separate due to their different mass-over-charge ratios m/q while oscillating in axial direction between the two mirrors.

The mass resolving power R, defined as mass m divided by the difference of neighboring masses that can still be distinguished, Δm, is related to the time of flight t and the peak width Δt by $R = \frac{m}{\Delta m} = \frac{t}{2\Delta t}$. To achieve a high mass resolving power, either the flight time t or the signal width Δt has to be maximized or minimized, respectively. Reducing the width of the signal is limited by several parameters like the type of ion source and the ions' temperature. The second approach to increase the mass resolving power is to enlarge the flight time of the ions. That can be achieved by either decreasing their energy or increasing the flight path. The former also increases the relative energy spread of the ion-bunch, making it more difficult to compensate the resulting time-of-flight aberrations. The latter is in principal not limited for a MR-ToF-MS. If the mean oscillation period of an ensemble of ions of one species inside the MR-ToF-MS is denoted as T, their initial bunchwidth as Δt_i and their time-of-flight aberration per revolution as ΔT, the mass resolving power after n revolutions is given by

$$R = \frac{t}{2\Delta t} = \frac{nT}{2\sqrt{\Delta t_\mathrm{i}^2 + n^2 \Delta T^2}} = \frac{T}{2\sqrt{(\Delta t_\mathrm{i}/n)^2 + \Delta T^2}} \xrightarrow[n\to\infty]{} \frac{T}{2\Delta T}. \tag{1}$$

Equation 1 shows that after n revolutions the contribution of the initial bunchwidth is reduced by a factor of n. The maximal achievable mass resolving power is in first approximation defined by the aberrations per revolution ΔT. The time required to approach this plateau depends on the initial bunchlength Δt_i [46].

To cope with the very large parameter space of the five mirror electrode potentials, the lens potential and the entrance einzel-lens potential (see Fig. 1), a numerical optimization algorithm was used in SIMION [47] simulations as well as in the experimental control system. The system was first tuned manually to reasonably good parameters, were the ion trajectory meets the point-to-parallel/parallel-to-point focusing condition [9, 46] for highest trajectory stability. Furthermore, the time-of-flight aberrations related to the energy distribution were reduced by tuning the three outer mirror electrodes. Within the numerical optimization, all parameters were varied for a specified number of revolutions inside the MR-ToF-MS by a Nelder-Mead simplex algorithm [48] and the peak width of one species (N_2^+) in combination with the aberration from the point-parallel/parallel-point focusing was measured and treated as the goal function to be minimized. After approaching a local minimum of the time-of-flight (hyper-)surface within a certain radius, these optimized parameters where multiplied by random numbers close to 1 and the optimization was restarted. With this procedure, a large number of local minima could be obtained.

These numerically optimized parameter sets served as starting values for the optimization of the device in the laboratory. We used the same algorithm to search for local minima in the nearby region around the numerical values. The peak width of the ion signal at the MCP detector served as the goal function.

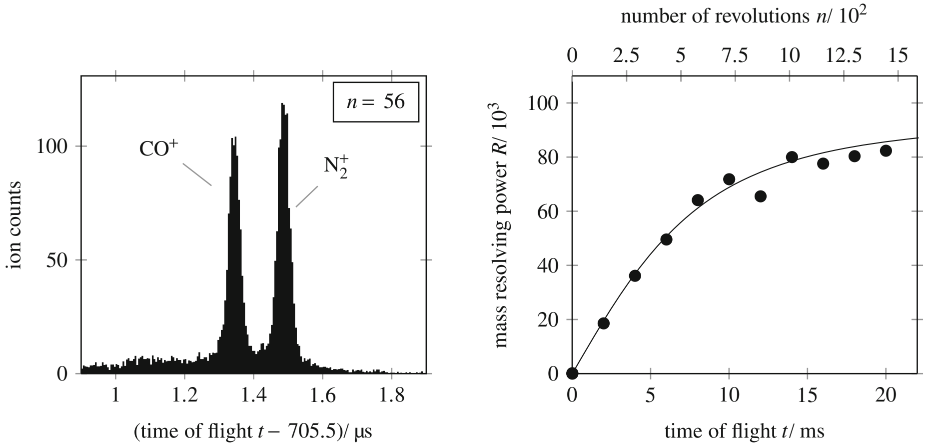

Fig. 3 Ion signal as a function of time of flight demonstrating mass separation of CO^+ and N_2^+ (*left*). Experimental mass resolving power achieved as a function of flight time and number of revolutions (N_2^+, 2.2 keV), curve to guide the eye (*right*)

4 Preliminary experimental results

The ability to separate isobaric ions has been confirmed experimentally. Figure 3 shows a time-of-flight spectrum (ion counts versus time of flight) for the isobaric doublet N_2^+/CO^+ ($m_{N_2^+} = 28.0056$ u, $m_{CO^+} = 27.9944$ u [49]). The mass resolving power required to separate these ions is $R_{\mathrm{FWHM}} \approx 2{,}500$ (for equal signal intensities). This is reached after a flight time of $t \approx 400$ μs and corresponds to 32 revolutions in the MR-ToF-MS. After $n = 56$ revolutions (Fig. 3 left), the signals are separated to the baseline, which corresponds to a mass resolving power of $R_{\mathrm{FWHM}} = 6{,}000$. In a typical case, the ratio of contamination ions to ions of interest is higher than one. Assuming the time-of-flight distribution to be Gaussian, the required mass resolving power for a separation to the half height will increase, e.g. by a factor of 2 for a ratio of $10^2 : 1$ or by a factor of 2.6 for a ratio of $10^4 : 1$. Reproducible mass resolving powers of up to $R_{\mathrm{FWHM}} = 80{,}000$ have been achieved for flight times of the order of 14 ms (Fig. 3 right) what corresponds to more than 1000 revolutions for N_2^+ at 2.2 keV.

The main limiting factor for transfer efficiency is the background gas pressure since collisions lead to momentum transfer and thus instable ion trajectories. In addition, losses due to a mismatch of pulse emittance and MR-ToF-MS acceptance are observed in the first revolutions, and further losses caused by electronic noise from the pulsing units appear at fixed times after injection. The mean lifetime at background pressures of about 2×10^{-8} mbar was measured to be $\tau \approx 15$ ms.

5 Conclusion and outlook

A multi-reflection time-of-flight mass separator was built and tested successfully with the isobaric doublet CO^+ and N_2^+. The mass resolving powers achieved reproducibly are in the order of $R_{\mathrm{FWHM}} = 80{,}000$. For the selection of the separated isobars, a

Bradbury-Nielsen ion gate [50] will be constructed to deflect the contaminant species and only transmit the ions of interest [12]. To further increase the mass resolving power, the stability and signal-to-noise ratio of the high-voltage powersupplies will be improved by additional filter stages and a temperature-regulated environment. The system will be installed at the ISOLTRAP mass spectrometer to support the purification of rare ion beams.

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