First operation and mass separation with the CARIBU MR-TOF

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Abstract

The recent installation of a Multi-Reflection Time-Of-Flight (MR-TOF) isobar separator at the CARIBU facility has the promising potential to significantly improve the mass separation and selection of short-lived neutron-rich beams. Ions cycled in the km-long isochronous trajectories between two electrostatic mirrors can be separated to high levels of mass-resolving power within a short time (tens of ms). The installation process is described and results from the first operation are discussed. Following an optimization of the mirror voltages a mass-resolving power of $6.8 \cdot 10^4$ was achieved and a separation of isobars was demonstrated. The higher purity beams provided by the MR-TOF and delivered to the Canadian Penning Trap (CPT) will provide access to further measurements of neutron-rich nuclei along the astrophysical r -process path.

Keywords: MR-TOF Time-of-flight Mass-separator

1. Introduction

The low-energy beamline at the Californium Rare Isotope Breeder Upgrade (CARIBU) [1] at the ATLAS facility of Argonne National Laboratory was recently upgraded with the installation of a Multi-Reflection Time-Of-Flight (MR-TOF) mass separator [2, 3]. The MR-TOF provides fast, high-resolution mass separation by multiple reflections of ions between two electrostatic mirrors, composed of six pairs of voltage-adjustable electrodes. By achieving isochronous time-focusing, different masses can be separated by time of flight in the kilometers-long folded trajectory. Due to its fast and simple operation, several MR-TOF systems are in operation or under construction at various labs around the world [4, 5, 6, 7]. In particular, the ISOLTRAP MR-ToF-MS system at CERN, has been fully operational since 2012 and performs precision mass measurements of rare isotopes or on-line mass selection for the Penning trap system [8, 9, 10]. The CARIBU MR-TOF is a scaled-up version of the ISOLTRAP MR-ToF-MS, realizing the same operation principle of a single in-trap lift electrode [11], however providing longer isochronous phase-space volume and smaller field aberrations.

CARIBU provides low energy and reaccelerated beams of neutron-rich isotopes obtained from ^{252}Cf fission products that are stopped in and extracted from a high-intensity gas catcher. The ion beam is then accelerated to 36 kV and sent for primary mass separation in a compact isobar separator providing a mass-resolving power of ~ 14000 (FWHM,

Figure 1: CARIBU platform with the newly installed MR-TOF and other main components.

same units are used below). Downstream, a helium-filled RFQ bunches and lowers the energy of the beam to provide short pulses at ~ 3 kV to the MR-TOF. The higher mass-separated beams provided by the MR-TOF are then delivered to various experiments in the low-energy experimental area, including the Canadian Penning Trap mass spectrometer (CPT). A picture of the CARIBU platform with annotations of its main components is shown in Figure 1.

High-precision mass measurements at the CPT are generally limited by the long (few 100 ms) in-trap preparation process that is deployed to remove mass contamination. The introduction of the MR-TOF to the beamline assists in the selection of the desired mass to be measured

Figure 2: Drawing of the MR-TOF showing a center cut of the geometry of the two electrostatic mirrors separated by a pulsed drift tube, the *lift*. The mirror electrodes are numbered ①-⑥ and the shielding electrodes are labeled ⑤. The simulated flight path of the ions inside the device is indicated in red and the potential distribution inside the trap is shown in blue.

and the fast removal of the contaminating isobars with a mass-resolving power of more than 10^5 , allowing for measurements of more exotic neutron-rich nuclei along the astrophysical r -process path [12]. The design considerations and simulations of the MR-TOF and associated beamline components as well as first operation results, are described below.

2. Design and simulations

The mechanical and electrical design of the MR-TOF is heavily based on the ISOLDE MR-ToF-MS [13] which itself is based on the design of the electrostatic ion beam trap of the Weizmann Institute [14]. The MR-TOF is 135 cm long and consists of two electrostatic mirrors, 75 cm apart, each composed of six coaxial electrodes, surrounded and separated by shielding electrodes (see Figure 2). The voltage on each of the six electrodes is controlled independently while the shielding electrodes are resistively-chained to their adjacent electrodes and held at "midway" voltages to provide a smooth distribution of the potential throughout the mirrors and to reduce aberrations. Electrodes 2-6 (see Figure 2), are identically shaped as thick coaxial rings made of stainless steel with an inner diameter of 26 mm and outer diameter of 94 mm. The first electrode controls the focusing of ions inside the device and has a longer shape, similar to an einzel lens design, with a larger borehole of 40 mm and 50 mm in length.

The mirror and shielding electrodes are mechanically constrained by silicon-nitride balls that provide precise alignment and electrical insulation. The mirrors are separated by the *lift*, a 75-cm long drift tube made out of stainless steel mesh.

The operation concept of the MR-TOF is similar to the ISOLDE approach, where ions are trapped by pulsing the central lift electrode instead of gating the entrance and exit mirrors. In addition to its simplicity of operation, this method has the advantage of controllability of time-focusing inside the MR-TOF that allows matching an optimal separation for a given number of cycles. Assuming the voltage settings of the mirror electrodes are isochronous and optimized, the operation of the MR-TOF requires the tuning of only two parameters, the lift voltage and timing, for any given mass.

The MR-TOF and associated low-energy beamline were simulated using SIMION [15]. The geometry of the MR-

Figure 3: Simulation results showing the mass-resolving power for a given number of cycles. The plot shows different evolutions of mass separation with the cycle number that correspond to changes in the mirrors voltage set (① and ②) and the lift voltage (② and ③). See discussion in the text.

TOF electrodes and shielding electrodes as well as beamline components downstream of the buncher were simulated. By optimizing the voltages of the six mirror-electrodes, using a simplex minimum-search algorithm, an isochronous ion-motion and mass-resolving power of over 10^5 has been achieved. Figure 3 shows the mass-resolving power evolution as a function of the number of cycles simulated using SIMION for three given sets of mirror and lift voltages and an initial beam energy of 3 keV. While optimizing the mirror voltages may result in a higher mass-resolving power, as demonstrated by curve ②, tuning the lift voltage has the effect of changing the time-focus within the trap, reflected in a higher shifted peak of the mass-resolving curve ③. A list of potential voltage sets generated by the optimization procedure has been used as the basis for the voltage set used in lab. The potential distribution associated with the manually optimized voltage set ③ is visualized as a blue curve over the MR-TOF drawing in Figure 2. The curve is linearly scaled to the actual voltages with minimum (maximum) voltage of -3850 V (2884 V) that is applied to electrode 1 (6). The transverse acceptance of the device was also simulated, requiring an emittance of less than 21.5π mm-mrad for the converging injection beam. Additionally, the extraction from the MR-TOF results in a significant beam divergence and emittance of up to 45π mm-mrad due to beam expansion and strong focusing as the beam exit through the high voltage region of

the mirror.

3. The experimental setup

In accordance with the simulation results, the MR-TOF beamline was designed to include an einzel lens upstream and downstream of the device as well as steerers to maneuver the beam into the MR-TOF and through the differential pumping stages. In preparation for the MR-TOF installation, the low-energy beamline was modified by repositioning the buncher upstream. The MR-TOF was installed downstream of the RFQ buncher, separated by two differential pumping sections. In between the devices, a pulsed drift tube, the *elevator*, is used to reduce the beam energy from 36 keV to 3 keV, an appropriate energy for the MR-TOF operation. The voltages for all electrodes are supplied by precision high voltage (HV) modules (ISEG, DPS/DPR series) that can be set and read back independently by a 16-bit DAC system (HYTEC DAC8402 module). The HV supplies are mounted inside a thermal-insulated box, temperature controlled by a Peltier heat-exchange circuit to prevent voltage drift. To further suppress noise and eliminate ground loops, the ground of the entire system is wired in a star-ground configuration and low-pass filters are applied to both the control and high voltages. At the MR-TOF exit, an interchangeable Micro-Channel Plate (MCP) detector and a Bradbury-Nielsen Gate (BNG) [16] are used to monitor and select the desired masses. The BNG is based on the one being used by the ISOLDE MR-ToF-MS [8] and is composed of pairs of 20 micron thin gold plated tungsten wires, evenly spaced 0.5 mm apart and mounted to a PEEK frame. A pulsed voltage of ± 250 V applied to each of the odd/even wires is used to deflect ions on demand and select the desired masses. The MCP signals are digitized and recorded using an oscilloscope.

The trapping region of electrodes in the RFQ buncher were shortened from 20 to 10 mm in length to reduce the time spread of the bunched beam and allow a higher mass-resolving power. As a result of this modification and by operating the buncher with extraction pulse of increased amplitude, the bunch time-spread has been reduced from 600 to 70 ns FWHM. During the installation, the glass cover of the buncher was replaced with a stainless steel tube providing improved durability and easier installation.

4. First operation and results

Following the installation and the modification of the buncher, the MR-TOF was first operated using radioactive beams from CARIBU. A pulsed beam of ions with $A=144$ extracted from the buncher at 3 keV with a repetition rate of 10 Hz was time-focused on a close-range MCP detector by adjusting the RFQ trapping and extraction voltages. The 70-ns (FWHM) time structure of the beam expands as it is transferred to the MR-TOF due to

the energy spread of the emerging ions. During the cycling process of ions in the MR-TOF, by choosing the lift voltage properly, the time-focusing is gradually restored with each revolution until a time spread of around 80 ns and an associated high mass-resolving power is achieved. After a certain number of cycles the ions are ejected and measured at an MCP detector that is located on a manipulator at the BNG position. By manually tuning the mirror voltages a mass-resolving power higher than $5 \cdot 10^4$ was achieved.

Figure 4: Energy-time heatmap for $A=144$ ions (green). Recorded at the close-range MCP detector following 40 cycles by keeping the mirror voltage fixed and changing the lift voltage. A quadratic fit to the signal peak indicated by a dashed red line visualizes the clear isochronous condition within the trap.

Figure 4 shows the measured time-energy curve using the manually optimized voltage set of the mirrors. This is an MCP measurement of mass 144 following 40 cycles (~ 2 ms), recorded for different lift voltages which provides an effective measure of the trapped ions' energy. The parabola shaped curve indicates an almost isochronous condition inside the trap, showing a change of only 400 ns over a span of 80 V. Fixing the lift near the minimum of this curve, at around 790 V, yields the narrowest time distribution and best mass-resolving power for the specific number of cycles. The transmission through the MR-TOF was estimated to be $\sim 30\%$ for 20 ms time-of-flight, as measured by comparing the counts of silicon detectors located upstream and downstream to the device.

The mass separation is demonstrated in Figure 5 showing the evolution of resolution with the increasing number of cycles for separation of mass $A=92$ ions. All three measurements shown in Figure 5 were performed using the same manually optimized mirror voltage set and a fixed lift voltage thereby keeping a gradual time-focusing restoration over the cycles, as discussed above. The top plot shows the time structure of the beam, averaged over 100 bunches following 40 cycles that correspond to a time of flight of ~ 2 ms. The beam width is 167 ns and the corresponding mass-resolving power is $m/\Delta m=6100$. The different masses that constitute the beam are still indistinguishable. The middle plot shows the beam after 200 cycles, ~ 10 ms. A structure of five distinctive peaks comes

Figure 5: Evolution of mass separation with the number of cycles for mass $A=92$ ions. The different peaks correspond to different isobars. The mass structure is gradually revealed as the mass-resolving power increases with the number of cycles. Some of the isobars are identified according to the time differences and penning trap measurements, the others may correspond to contaminant molecular ions.

into view, the center peak is 80 ns wide resulting in an estimated mass-resolving power of $m/\Delta m = t/2\Delta t = 62000$. The identification of two of the peaks, as ^{92}Rb and ^{92}Kr was performed by matching the time to mass differences and by a subsequent measurement at the CPT. As the number of laps is increased to 600, a prominent separation of the peaks can be seen in the bottom plot, along with a concurrent widening of the peaks that is clearly visible. The width of the central peak is now 220 ns with a corresponding $m/\Delta m = 68000$. This is an expected behavior that indicates that a full restoration of time-focusing inside the MR-TOF has occurred sometime between the 200 and 600 cycles which corresponds to the narrowest peak signals and highest mass-resolving power that can be achieved with this specific mirror and lift voltage set. By only changing the lift voltage, the time-focusing restoration can be tuned and optimized for a specific number of cycles. By alternating the MCP with the BNG that shares the same manipulator, the selection of any desired mass is straightforward.

5. Summary and outlook

The MR-TOF was successfully installed at the CARIBU facility low-energy beamline. This installation provides a significant upgrade to the mass-resolving power that can be achieved at CARIBU by cycling the ions between two

isochronous electrostatic mirrors, gaining separation by the time-of-flight difference of the different masses. The MR-TOF and associated beamline is based on the successful design and method of operation of the ISOLTRAP MR-ToF-MS. It was optimized for our setup with the aid of SIMION simulations, allowing a mass-resolving power higher than 10^5 (FWHM). During first operation of the MR-TOF, a mass-resolving power of $6.8 \cdot 10^4$ was achieved and mass separation of several isobars was demonstrated. The optimization of the MR-TOF voltages, performed manually so far, will be upgraded to include an automatic process that could provide even higher mass-resolving power. Using the BNG to deliver specific purified beams to the CPT located downstream will reduce the preparation and measurement time at the CPT and allow the precise mass measurements of more neutron-rich isotopes. The fast separation, ease of operation, and high resolving power of the MR-TOF provide an excellent opportunity to make mass measurements of short-lived isotopes.

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