Offline commissioning and performance of a multi-reflection time-of-flight mass analyzer with a new configuration Postprint

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Abstract

To precisely measure atomic masses and select isotopes of interest, a multi-reflection time-of-flight mass spectrometer is being built at SHANS (Spectrometer for Heavy Atom and Nuclear Structure). Its mass analyzer features a new configuration that differs from existing designs and has been successfully commissioned offline using a chopped 133Cs beam with a Bradbury-Nielsen gate. The potentials on all electrodes were optimized using a two-dimensional potential scan method. With the optimized parameter set, an efficiency of 50% was achieved. The mass resolving power was measured to be 74,000 at a time-of-flight of 12.9 ms, reaching 90,000 with a longer time-of-flight of approximately 20 ms.

Full Text

Preamble

Offline commissioning and performance of a multi-reflection time-of-flight mass analyzer with a new configuration

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Abstract

To precisely measure atomic masses and select isotopes of interest, a multi-reflection time-of-flight mass spectrometer is being built at SHANS (Spectrometer for Heavy Atom and Nuclear Structure). Its mass analyzer features a new configuration that differs from existing designs and has been successfully commissioned offline using a chopped 133Cs beam with a Bradbury-Nielsen gate. The potentials on all electrodes were optimized using a two-dimensional potential scan method. With the optimized parameter set, an efficiency of 50% was achieved. The mass resolving power was measured to be 74,000 at a time-of-flight of 12.9 ms, reaching 90,000 with a longer time-of-flight of approximately 20 ms.

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1. Introduction

In recent years, more than a dozen multi-reflection time-of-flight mass spectrometers (MRTOF-MS) have been constructed or are under development worldwide [1–11]. These instruments perform accurate mass measurements and isobaric separation, yielding fruitful experimental results [12–19]. Plaß et al. [20] reviewed MRTOF-MS developments for research with short-lived nuclei and various instrumental implementations. At the Institute of Modern Physics, Chinese Academy of Sciences (IMP/CAS), we are developing an MRTOF-MS to be attached to the end of SHANS (Spectrometer for Heavy Atom and Nuclear Structure [21]) for precise atomic mass measurements and isotope selection.

The conceptual design of the low-energy mass spectrometer at SHANS was described in Ref. [22]. It comprises a degrader, cryogenic gas catcher, quadrupole ion beam guide, radiofrequency quadrupole cooler and buncher, MRTOF mass analyzer, and time-of-flight detector. The MRTOF mass analyzer plays a crucial role, as ions fly through it and are separated in time according to their mass-to-charge ratios. The electrode design and applied potentials must be carefully chosen to ensure the analyzer is time-focusing—that is, to minimize time-of-flight differences arising from the initial energies, positions, and angles of injected ions.

Since the number of electrodes affects the potential smoothness along the optical axis and determines the number of required power supplies, we designed a new type of MRTOF mass analyzer with a different configuration, where each mirror consists of five electrodes rather than four [23] or eight [3, 24, 25]. The design and computer simulations were reported in previous papers [8, 26]. This paper focuses on the offline commissioning of the MRTOF mass analyzer and reports the achieved performance in detail.

2. Experimental setup

Fig. 1 [Figure 1: see original paper] shows a schematic of the experimental beam line used for offline commissioning. A continuous 133Cs1+ beam is produced by a heated alkali ion source emitter (a) from HeatWave Labs [27], followed by a skimmer (b) and an extraction electrode (c). The emitter connects to a supporting electrode floating at approximately 190 V above ground, thereby defining the initial kinetic energy of the ions. The potentials on the skimmer and extraction electrode are 155 V and 100 V, respectively. A Bradbury-Nielsen gate (BNG, e) [28], made of stainless steel wires with a diameter of 50 µm and wire spacing of 250 µm, is employed to bunch the beam by switching $±$28 V potentials on two sets of parallel wires, designated BNG1 and BNG2.

To reduce emittance by limiting the radial beam spread and thus improve beam quality, three apertures are used in combination. The first aperture is in the skimmer with a diameter of 3 mm, the second is in the flange (d) before the BNG with a diameter of 6 mm, and the third (f) is behind the BNG with a diameter of 2 mm. Subsequently, an einzel lens (g) with potentials of 0, 35 V, 0 is used to focus the pulsed beam to the middle of the acceleration pulsed drift tube (Acc-PDT, h), where ion bunches are accelerated to a kinetic energy of 1500 eV by switching its potential from 0 to approximately 1310 V to match the MRTOF mass analyzer requirements. A cylindrical deflector (i) cut equally into four parts is placed between the Acc-PDT and mass analyzer to adjust the ion beam radially, with applied potentials of 0, 28 V, 38 V, and 0, respectively.

The MRTOF mass analyzer consists of four pairs of mirror electrodes M1–M4 (j, k, l, m), a pair of lens electrodes L (n), and a drift tube DT (o). Details of the MRTOF structure design can be found in [8, 26]. The mirror electrodes, from the outermost ends to the middle, have lengths of 20, 16, 26, and 26 mm, respectively; the lens electrode has a length of 46 mm, and the intermediate drift tube has a length of 400 mm. Adjacent electrodes are insulated by 4 mm thick ceramic spacers, and the total analyzer length is 708 mm. The electrode diameter is 60 mm, and four reduced apertures with a diameter of 30 mm located on the outer ends of M1 and DT are added for electric shielding.

Ions are captured using the in-trap potential lift method [29], where the kinetic energy of ions is reduced below the mirror electrode barrier by switching off the potential on the drift tube. After flying a certain number of laps, bunches are released by switching the drift tube potential back on and hit a MagneTOF detector from ETP Ion Detect™ [30] placed at the end of the beam line. The time of flight (TOF) from the BNG to this detector is recorded by an SR430 multichannel scaler (MCS) from Stanford Research Systems [31].

Two sets of vacuum pumps from Edward Vacuum [32] achieve a residual gas pressure better than 3 × 10−8 mbar. High voltage (HV) modules with high precision and stability from ISEG Spezialelektronik GmbH [33] drive the electrodes. Fast solid-state switches from BEHLKE Power Electronics [34] and CGC Instruments [35] switch the time-varying voltages.

The entire system is controlled remotely by a specialized distributed real-time control system based on EPICS [36]. Fig. 2 Figure 2: see original paper shows the fast control diagram. A PCI-7813R field programmable gate array (FPGA) module from National Instruments [37] followed by a TTL line driver triggers the BNG, Acc-PDT, DT switches, and SR430 MCS. The measurement cycle is depicted in Fig. 2(b), consisting of five channels synchronized with 25 ns precision. A cycle begins when the BNG opens. The opening time was optimized to 0.15 µs. Twenty-three µs later, the Acc-PDT potential switches on when the bunched ions reach the middle of the Acc-PDT, lasting 1 ms—sufficient to ensure all ions leave the Acc-PDT and are accelerated. Ions are captured 40 µs after bunching when they reach the middle of DT and released after flying a certain number of laps, which determines the duration. Since the total ion TOF exceeds the maximum recordable by the SR430 MCS, the MCS trigger must be delayed to capture the entire TOF spectrum. This delay varies according to the number of laps flown, as well as the MCS bin width and bins per record. Typically, bin widths of 40 ns or 5 ns (the narrowest available) were used during offline commissioning.

3. Optimization of the MRTOF mass analyzer

The optimization goal is to find electrode potentials that maximize resolving power while maintaining good TOF peak symmetry. Mass resolving power is defined as MRP = TOF / (2 × ΔTOF), where TOF is the total time-of-flight and ΔTOF is the full width at half maximum (FWHM) of the peak.

Due to image aberration, field imperfections, and mechanical misalignment, TOF spectrum peaks are asymmetrical [38]. To quantify peak symmetry and easily identify potential parameters with good symmetry during optimization, we calculated the coefficient of determination (COD) for Gaussian fits. The COD is expressed as $R^2 = 1 - \Sigma(yi - f(xi))^2 / \Sigma(yi - \bar{y})^2$, where yi is experimental data and f(xi) is the fitted value from the statistical model. The minimum R^2 value is 0 and the maximum is 1. The COD measures how well a statistical model predicts an outcome.

Since the Gaussian distribution is a primary component of TOF peaks and is distinctly symmetrical, we can employ Gaussian fitting COD to describe peak symmetry. The closer the Gaussian fitting COD is to 1, the more the TOF peak approximates a Gaussian distribution and the more symmetrical it is. While one cannot predetermine a COD range for reasonable symmetry, analysis of spectra revealed that long tails appear when R^2 falls below 0.980. Note that Gaussian fitting is used throughout this paper unless otherwise specified.

A Python code was developed to set timing and potential parameters and analyze experimental results online. TOF, ΔTOF, and R^2 values were calculated in real time after each measurement and stored for future analysis along with the potential parameters.

Switching times for ion acceleration by Acc-PDT, and ion capture and release by DT, were scanned to ensure these operations begin when ions are near the middle of Acc-PDT and DT. One-dimensional potential scans on the skimmer, extraction electrode, BNG, and einzel lens, and two-dimensional scans on the deflector, were performed to achieve narrow beam width and high transport efficiency. Optimized values are described in Section 2.

For MRTOF mass analyzer optimization, potentials on adjacent electrodes were scanned two-dimensionally over given ranges, following a linear progression through electrode pairs: M1 vs. M2, M2 vs. M3, M3 vs. M4, M4 vs. L, and L vs. DT. Searching a large space with small step sizes would be extremely time-consuming, so we employed a global search followed by local refinement. During global search, potential ranges spanned tens of volts with step sizes of 0.5 V or larger. This identified sub-regions with relatively narrow distributions and good symmetry, which were then selected for local refinement with step sizes of 0.02 V or smaller.

SIMION [39] simulated potentials served as optimization starting points. All MRTOF analyzer electrodes were optimized based on 300 laps. To ensure sufficient statistics for TOF spectrum fitting, 10,000 bunches were recorded per measurement.

As an example, Fig. 3 Figure 3: see original paper-(c) shows two-dimensional contour plots of TOF, MRP, and R^2 as functions of potentials applied to M1 and M2. Fig. 3(d) shows the MRP contour plot with $R^2 > 0.980$. Here, the maximum MRP with good R^2 occurs at M1 = 1073.5 V and M2 = 886.0 V, representing the optimal choice. We adopted 1073.5 V as the optimal potential for M1 and continued with a two-dimensional scan of potentials on M2 and M3. This process repeated until all electrode potentials were optimized.

4. Results and discussion

Table 1 lists the optimal potentials applied to mirror electrodes, lens electrode, and drift tube after complete optimization. With this parameter set, we investigated TOF spectra at different lap numbers and studied power supply stability effects.

4.1. Performances at different laps

Fig. 4 [Figure 4: see original paper] shows measured TOF, ΔTOF, MRP, and R^2 as functions of lap number. All values depend on lap number. TOF from BNG to detector increases linearly with lap number at 42.758(1) µs per lap, and ions can be stored in the analyzer for up to 65 ms or longer. MRP increases with lap number until approximately 400 laps, then plateaus around 90,000. Fig. 5 [Figure 5: see original paper] shows TOF spectra obtained at 300 laps (where optimization was performed) and 500 laps (where maximum MRP was obtained). MRP reaches 74,000 at 300 laps (TOF = 12.9 ms) and 90,000 at 500 laps (TOF = 21.4 ms). As shown in Fig. 4(d), R^2 exceeds 0.980 in most cases but decreases—and peak symmetry worsens—when lap number deviates significantly from 300, where optimization was performed.

Transport efficiency is defined as the ratio of total counts after a given number of laps to total counts when shooting through the analyzer. Fig. 6 [Figure 6: see original paper] shows measured efficiency at different lap numbers. Efficiency decreases with increasing lap number. Approximately 40% of ions are lost in the first 10 laps, while remaining ions tend to stay for subsequent laps. Efficiency remains at approximately 50% for 100-500 laps and decreases slightly thereafter. At 1500 laps, efficiency still exceeds 40%. Losses comprise two components: rapid loss likely due to analyzer acceptance, beam emittance, or mechanical misalignment; and slow loss due to collisions with residual gas. The relatively higher efficiency compared to Ref. [10] likely results from our analyzer's larger inner diameter and thus larger acceptance.

4.2. Effect of potential inaccuracy

Fig. 7 [Figure 7: see original paper] shows how TOF, ΔTOF, and MRP change when varying potentials on each electrode pair. Potential deviation is defined as $(V - Vopt)/Vopt$, where V is the applied potential and Vopt is the optimized potential from Table 1. For all analyzer electrodes, TOF changes linearly with potential, with sensitivity quantified by linear fit slopes listed in Table 1. TOF is most sensitive to M1 and M2 potentials: a 0.1 V deviation on M1 and M2 causes 0.46 µs and 0.25 µs TOF changes, respectively. For MRP, M1 and DT potentials are most sensitive. The M1 potential is most critical to analyzer performance, as ions turn around at M1 and small deviations from ideal potential significantly alter ion trajectories. MRP changes from DT potential variations result from changes in trapped ion energy. MRP is insensitive to other electrode potentials except M1 and DT.

4.3. Long time stability test

To observe TOF spectrum behavior over time, we performed a long-duration measurement with optimized potentials from Table 1 at 300 laps. Fig. 8 [Figure 8: see original paper] shows measured TOF and MRP during continuous 52-hour measurement. TOF drifts with elapsed time with a period of approximately 24 hours, likely due to electrical power drift between day and night and possibly environmental temperature changes. Maximum TOF change reached 0.2 µs. MRP also fluctuates but without regular period.

TOF distribution symmetry can be studied using 1167 spectra from this 52-hour measurement. Fig. 9 [Figure 9: see original paper] shows the sum peak after re-centering TOF peaks to correct temporal drift and summing them. The peak is symmetric except for a small long tail on the right side, possibly resulting from non-ideal trapping potential and asymmetric beam distribution at the analyzer entrance. The sum peak fits well using a Hyper-EMG function [38], yielding ΔTOF = 67.9(1) ns and $R^2 = 0.999$. Gaussian fitting yields 88.9(1) ns and 0.989.

5. Summary

A multi-reflection time-of-flight mass analyzer with a new configuration has been constructed and successfully commissioned. Potentials on all electrodes were optimized using a two-dimensional potential scan method. The coefficient of determination from Gaussian fitting was calculated and used to quickly and accurately identify potential parameters with good symmetry. With optimized electrode potentials, ions can be stored for up to 65 ms or longer. An efficiency of 50% was achieved. Mass resolving power reached 74,000 at 300 laps (TOF = 12.9 ms) and can reach 90,000 at longer TOF of approximately 20 ms. Mass resolving power is currently limited by the relatively broad beam chopped by the Bradbury-Nielsen gate and will improve with higher-quality pulsed beams having narrow bunch width and small emittance. The analyzer will be installed at SHANS for direct mass measurement and isobar separation of fusion evaporation reaction products.

CRediT authorship contribution statement

W.X. Huang: Supervision, Conceptualization, Investigation, Methodology, Software, Formal analysis, Visualization, Writing - original draft & review & editing, Funding acquisition. Y.S. Wang: Investigation, Methodology, Writing - review & editing. Y.L. Tian: Investigation, Conceptualization, Methodology, Formal analysis, Visualization, Writing - original draft, Funding acquisition. J.Y. Wang: Investigation, Methodology, Writing - review & editing. Y. Wang: Investigation. Z.G. Gan: Supervision, Conceptualization, Investigation, Funding acquisition. H.S. Xu: Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Note: Figure translations are in progress. See original paper for figures.