# Optimization of multi-reflection time-of-flight mass analyzer operating in in-trap-lift mode postprint

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## Abstract

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## Full Text

## Preamble

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### Optimization of Multi-Reflection Time-of-Flight Mass Analyzer Operating in In-Trap-Lift Mode

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### Abstract

**Purpose:** We are building an MRTOF-MS (Multi-Reflection Time-of-Flight Mass Spectrometer) for isobaric separation for the Lanzhou Penning Trap. The potentials applied to the electrodes of our MRTOF mass analyzer operating in in-trap-lift mode must be optimized to achieve very high mass resolving power.

**Methods:** Our method for designing and optimizing an MRTOF mass analyzer has been updated to introduce constraints on the potentials. This method can now be used to optimize the parameters of MRTOF-MS operating in both mirror-switching mode and in-trap-lift mode. Using this method, the optimal potential parameters of the electrodes have been obtained for our MRTOF mass analyzer operating in in-trap-lift mode.

**Results and Conclusion:** With a beam size of 2.8 mm diameter and an initial average ion kinetic energy of 1500 eV, the maximal mass resolving power has been achieved to be $3.2 \times 10^4$ with a total TOF of 7.0 ms for an ion species of $^{40}\mathrm{Ar}^{1+}$. It can reach up to $5.6 \times 10^4$ for a beam size of 0.3 mm diameter. The simulation shows that the inaccuracy of the potentials applied to the outermost mirror electrodes M1-M2 must be less than 50 ppm, or preferably 20 ppm.

**Keywords** Time-of-flight mass spectrometer · Multiple-reflection · Mass measurement · Isobaric separation · Exotic nuclei

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## 1 Introduction

The Multi-Reflection Time-of-Flight Mass Spectrometer (MRTOF-MS) has been developed as a new device in recent years. By extending the flight path using multi-reflection between electrostatic ion mirrors, an MRTOF-MS can achieve a very high mass resolving power of >100,000 in a compact structure. Moreover, it also has other unique advantages, such as extremely short measurement time, a large mass range, very high sensitivity, and non-scanning operation. Plaß et al. [?] reviewed MRTOF-MS developments for research with short-lived nuclei and different instrumental implementations. Up to now, many MRTOF-MS systems for mass measurements and isobaric separations have been commissioned \cite{2–6} or are under construction

\cite{7–11}. Many excellent results have been achieved; for example, the masses of $^{82}$Zn [?], $^{53}$,$^{54}$Ca [?], and $^{52}$,$^{53}$K [?] have been measured using MRTOF-MS at ISOLDE/CERN, and the masses of heavy nuclides produced by fusion-evaporation reactions at GARIS-II/RIKEN [?].

Injection and ejection of ions into/from MRTOF-MS have been achieved by switching the electric potentials of the electrodes to appropriate values while the ions are passing. Thus, according to the switching mode, all MRTOF-MS systems can be categorized into two types: mirror-switching and in-trap-lift modes. In the former mode, the electric potentials of the entrance and exit mirrors are switched to lower values while the potential of the intermediate drift-tube remains at 0. In the latter mode, all the potentials of the mirrors remain at their optimal values and the intermediate drift-tube is switched to its appropriate value; in other words, the drift-tube acts as a lift to the ions and the energies of the ions vary, thus the ions can be injected, trapped, and ejected by only switching the potential of the drift-tube. Wolf et al. [?] provided a detailed theoretical study, and many MRTOF-MS systems [?, ?, ?] have adopted the in-trap-lift mode.

The Lanzhou Penning Trap (LPT) [?], whose main task is to perform direct mass measurements on atoms and nuclei with high precision, is presently under construction at the Institute of Modern Physics, Chinese Academy of Sciences. We are building an MRTOF-MS for isobaric separation for LPT. The design and optimization procedures have been reported in detail in Ref. [?, ?], where the mirror-switching mode was assumed and a maximal resolving power of 1.3 $\times$ $10^5$ was achieved with a total time-of-flight of 6.5 ms for an ion species of $^{40}$Ar$^{1+}$. In this paper, we report the optimization of our MRTOF mass analyzer operating in in-trap-lift mode and the corresponding results.

## 2 Parameters of the MRTOF Mass Analyzer

[Figure 1: see original paper] shows the geometry of our MRTOF mass analyzer. It consists of two identical electrostatic mirrors (each containing a set of four electrodes) in combination with two einzel lenses and a drift-tube. All electrodes of this analyzer have a cylindrical shape. The total length is 708 mm with an inner diameter of 60 mm. The mirror electrodes M1-M4 (numbered from the outermost) have lengths of 20, 16, 26, and 26 mm, respectively, and the lens electrode L has a length of 46 mm. The intermediate drift-tube D has a length of 400 mm. The distance between two adjacent electrodes is 4 mm. The time focus plane, where a micro-channel-plate detector or a Bradbury-Nielsen gate [?] is placed, is located 255 mm after the analyzer. Different from our previous optimization, we set the mass analyzer operating in the in-trap-lift mode.

## 3 Update of the Optimization Procedure

In our previous paper [?], we reported an optimization strategy in detail to find the optimal parameters for our mass analyzer operating in mirror-switching mode. It includes two sub-procedures: a global search and a local refinement. The ion trajectories are calculated in SIMION [?] according to our specified user program as usual, and the variations of the parameters are performed in a separate optimization program that we coded ourselves using the Nelder-Mead simplex algorithm [?]. Because we search all possibilities in the whole parameter space, the best parameters can be found for certain.

We discovered a drawback in our optimization program when we attempted to optimize our MRTOF mass analyzer operating in in-trap-lift mode. Practically, the kinetic energy of the ions from the RFQ cooler and buncher upstream must be fixed at a certain value, say 1500 eV. This constrains all the potentials applied to the electrodes of the mass analyzer to <1500 V. Thus, the optimization must be performed according to boundaries on the possible potentials on the electrodes. Our previous program cannot handle this, so we had to update it.

The Nelder-Mead simplex algorithm originally does not support constraints (upper and lower boundaries). A penalty function or domain transformation has been proposed for use. Box [?] proposed a way to handle some explicit and some implicit constraints, and Guin [?] later refined the idea. Le Floc'h [?] compared all these methods. To solve our problem, we implement the following strategy. When a trial point is outside the boundaries and the centroid's coordinate $P_i$ respects the constraints, the point is reset at $l+w(P_i-l)$ if the lower boundary $l$ is broken, or at $h-w(P_i-l)$ if the upper boundary $h$ is broken, where $w$ is a uniform random number between 0.00001 and 0.5. If the centroid breaks the constraints, the point is reset at $l+0.00001$ if the lower boundary is broken, or at $h-0.00001$ if the upper boundary is broken.

## 4 Results and Discussion

To calculate the mass resolving power with respect to the number of revolutions, we considered the following initial beam conditions. The beam consists of 100 ions of $^{40}\mathrm{Ar}^{1+}$, with an average kinetic energy of 1500 eV, and the bunch width is 20 ns (FWHM) at the middle of the analyzer. (Since the calculation is very time-consuming, we cannot increase the number of ions due to our existing computational capacity.) In the middle between the ion mirrors, the ion beam coordinates $x$ and $y$ orthogonal to the optical axis $z$, the angles $\alpha = dx/dz$ and $\beta = dy/dz$ with respect to the axis $z$, and the energy $K$ are approximately represented by Gaussian distributions with standard deviations $\sigma_x = \sigma_y = 1.0$ mm, $\sigma_\alpha = \sigma_\beta = 1.5$ mrad, and $\sigma_K = 8.5$ eV. All these parameters are the same as those in Ref. [?]. For comparison, we also reduce the beam spot from $\sigma_x = \sigma_y = 1.0$ mm to 0.5 and 0.1 mm, corresponding to beam sizes with diameters of 2.8, 1.4, and 0.3 mm, respectively. If not specified, the results are obtained for the beam size with 2.8 mm diameter.

The mass resolving power $R$, which is one of the most important quantities in mass spectrometry, is calculated as $R = \mathrm{TOF}/2\Delta\mathrm{TOF}$, where TOF and the overall spread $\Delta$TOF are the centroid and the full-width-at-half-maximum

(FWHM) of the time-of-flight peak.

### 4.1 Optimized Results for the Beam Size with 2.8 mm Diameter

Considering the symmetric shape of our MRTOF mass analyzer and the practical potential controls through the power supplies during experiments, we set the potentials applied to the four mirror electrodes, one lens electrode, and the intermediate drift-tube as the optimization parameters—six in total. Because the incoming ions have an average energy of 1500 eV, all potentials applied to the electrodes have been set with an upper limit of 1400 V.

In the global search step, we considered a total of $4 \times 4 \times 4 \times 7 \times 9 \times 3 = 12,096$ potential combinations as initial parameter sets and finally chose only the best five new sets obtained from the search for further local refinement. Using the Nelder-Mead simplex algorithm, all parameters varied and a large number of local minima were obtained. We chose the parameter set that gives the largest value of mass resolving power as the optimal one, rather than the smallest peak width of the TOF when the mass analyzer operates in mirror-switching mode, because in the in-trap-lift mode the average kinetic energy of the ions traveling in the trap varies according to the formula $K_{\text{intrap}} = e[1500 - U_{\text{drift}}]$, where $U_{\text{drift}}$ is the potential of the drift tube, and the total TOF varies with different parameter sets.

We optimized the potentials for different numbers of revolutions and obtained the maximal resolving power that can be achieved, as shown in [Figure 2: see original paper]. For the beam size with 2.8 mm diameter, when increasing the number of revolutions $N$, the maximal resolving power increases steadily and then reaches its maximum around $N \approx 150$. This means we can achieve very good separation of ions in only 3 ms for $^{40}\text{Ar}^{1+}$.

For every optimization for a specific number of revolutions, we obtained one potential set with the highest mass resolving power. lists the potentials optimized for the number of revolutions of $N = 350$. [Figure 3: see original paper] shows the potential distribution along the optical axis in one of the mirrors of the analyzer when the ions are injected into/ejected from the analyzer and when they are trapped inside. It has two regions with approximately constant electric fields [**?**, **?**], $E_1 < E_2$. The ions turn around in the weaker constant electric field $E_1$.

Applying the potentials in to the electrodes, we calculated the temporal width at the detector plane with respect to the number of revolutions. [Figure 4: see original paper] shows the calculation results of the total TOF, the temporal width $\Delta$TOF, and the mass resolving power $R$ as functions of the number of revolutions. As the number of revolutions increases, the total time-of-flight increases almost linearly, and the initially high temporal spread decreases until a minimum—only 61 ns at the number of revolutions of $N \approx 100$—and then increases. The maximal resolving power $R = 3.2 \times 10^4$ is achieved at 350 laps, corresponding to a TOF of 7.0 ms.

### 4.2 Effects of Potential Inaccuracy

[Figure 5: see original paper] shows the relative variation of TOF and mass resolving power as functions of relative variation of bias potentials as determined by our optimization code. For the total TOF, the behavior from the mirror electrode M3 acts differently from other electrodes, and the largest variation comes from the mirror electrode M1. The maximal achievable mass resolving power decreases as the potentials applied to the electrodes vary from the ideal values, as expected. Except for M1 and M2, potential variations from all other electrodes (M3, M4, L, and D) almost have no effect on the final resolving power. Those from the mirror electrode M1 have the biggest effect: the resolving power decreases by 22% from $3.2 \times 10^4$ to $2.5 \times 10^4$ with a relative potential variation of 500 ppm. Thus, we must ensure the inaccuracy of the potentials applied to the mirror electrodes M1 and M2 is less than 50 ppm, or preferably 20 ppm. It is easy to understand that the potentials on electrodes M1 and M2 form the mirrors and the ions turn around at the position between M1 and M2.

### 4.3 Effects of Different Beam Size

[Figure 2: see original paper] also shows the maximal mass resolving power achieved at different beam sizes. For the same number of resolutions, we keep all parameters of the initial beam conditions except the beam size. For all three beam sizes, the maximal resolving powers increase steadily and then reach their maxima as the number of revolutions increases, but the reaching point delays from $N \approx 150$ to $\approx 200$ and $\approx 300$ for beam sizes reducing from $\Phi 2.8$ mm to $\Phi 1.4$ mm and $\Phi 0.3$ mm, respectively.

For different beam sizes, the maximal resolving power increases as the beam size reduces. It reaches $3.2 \times 10^4$ for a beam size of $\Phi 2.8$ mm, increases to $4.4 \times 10^4$ for $\Phi 1.4$ mm, and to $5.6 \times 10^4$ for $\Phi 0.3$ mm. The smaller beam size benefits the resolving power; thus, we should inject ion beams with smaller beam size into the mass analyzer to achieve higher mass resolving power.

### 4.4 Comparison Between the Mirror-Switching and In-Trap-Lift Modes

Since we have obtained the optimal parameters for the same mass analyzer operating in both mirror-switching and in-trap-lift modes, we can compare them directly. In our optimization, we used the same initial beam conditions described above: the initial beam size is 2.8 mm diameter and the initial average kinetic energy of the ions is 1500 eV. The outcome differences only come from the different operating modes.

In the mirror-switching mode, the maximal resolving power has been achieved to be $1.3 \times 10^5$ with a total time-of-flight of 6.5 ms for the ion species $^{40}\mathrm{Ar}^{1+}$; but in the in-trap-lift mode, it is only $3.2 \times 10^4$ with a total time-of-flight of 7.0 ms. It appears that the mirror-switching mode is much better than the in-trap-lift mode if we ignore the financial costs of the power supplies and switches needed to switch the potentials applied to the different electrodes. The difference in mass resolving power may come from the difference in kinetic energy of the ions traveling in the trap, which is 1500 eV in the mirror-switching mode and 1040 eV in the in-trap-lift mode in our case. We may improve the mass resolving power in the in-trap-lift mode by increasing the kinetic energy of the ions in the trap. A paper about the relationship between maximal resolving power, kinetic energy in the trap, mass of ion species, beam size, and other parameters will be prepared after more extensive calculations.

## 5 Summary

Our previous method for designing a multiple-reflection time-of-flight mass analyzer has been updated. Now constraints can be applied to the potentials. The method can be used to search for optimal potentials applied to the electrodes in MRTOF mass analyzers operating not only in mirror-switching mode but also in in-trap-lift mode.

Using this updated method, the potentials applied to the electrodes of our MRTOF mass analyzer operating in in-trap-lift mode have been searched and optimized. With a beam size of 2.8 mm diameter and an initial average ion kinetic energy of 1500 eV, the maximal mass resolving power has been achieved to be $3.2 \times 10^4$ with a total time-of-flight of 7.0 ms for an ion species of $^{40}\mathrm{Ar}^{1+}$. The maximal resolving power can reach up to $5.6 \times 10^4$ for a beam size of 0.3 mm diameter. The simulation also shows that variations of the potentials applied to the mirror electrodes M3-M4, the lens electrode L, and the drift-tube D almost have no effect on the final resolving power. Crucially, the inaccuracy of the potentials on the mirror electrodes M1-M2 needs to be less than 50 ppm, or preferably 20 ppm.

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