

# A time-of-flight mass spectrometer to resolve isobars

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## Abstract

In order to determine nuclear binding energies of short-lived nuclei we have developed a multi-pass time-of-flight mass spectrometer in which ions are repeatedly reflected between two coaxially arranged electrostatic ion mirrors. These ions are passed up to several hundred times between the ion mirrors. Within this system the overall ion flight time can be several milliseconds and the overall flight distance more than 100 m. There are no intensity losses in the grid-free ion mirrors and the transmission is limited only by angle scattering of the ions by residual gas atoms. The achieved mass resolving power exceeded  $m/\Delta m \approx 40000$ .

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## 1. Introduction

To determine nuclear binding energies by direct mass measurements one must use mass spectrometers of very high mass resolving powers  $m/\Delta m$  and accuracies of approximately 1 ppm. To perform such measurements for short-lived nuclei, which are produced with small cross sections, one requires the used mass spectrometer to feature also a high transmission. The transmission is proportional to the initially accepted transversal phase-space areas  $\varepsilon_x = \Delta x \cdot dx/dz$  and  $\varepsilon_y = \Delta y \cdot dy/dz$  with  $z$  denoting the coordinate along the

ion beam axis and  $\Delta x$  as well as  $\Delta y$  the initial transversal extension of the ion beam [1] perpendicular to  $z$ . In case of transversally dispersive systems this demand of high transmission, i.e. large  $\varepsilon_x \cdot \varepsilon_y$ , is in conflict with the demand on high mass resolving power  $m/\Delta m$  which requires narrow slits  $\Delta x$  and reasonably small angles of divergence  $dx/dz$  and  $dy/dz$ . In longitudinally dispersive systems, i.e. in time-of-flight mass analyzers, there is no such conflict [1–3] since in such systems the mass resolving power requires only short ion pulses  $\Delta t$  and reasonably small energy spreads  $\Delta K$ .

## 2. The design of a time-of-flight mass spectrometer of high resolving power

Time-of-flight mass spectrometers must differentiate between ions of mass-to-charge ratios

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$(m_0/q_0) + \Delta(m/q) = (m_0/q_0)(1 + \delta_m)$  with  $|\delta_m| \ll 1$  even if they have different energy-to-charge ratios  $(K_0/q_0) + \Delta(K/q) = (K_0/q_0)(1 + \delta_K)$  with  $|\delta_K| \ll 1$ . This separation relies on the different flight times  $t = t_0 + \Delta(t) = t_0(1 + \delta_t) = l/v$  along a trajectory of length  $l$ . If this system must operate as a mass analyzer and the ions have velocities  $v = v_0\sqrt{(1 + \delta_K)/(1 + \delta_m)}$  with  $v_0 = \sqrt{2K_0/m_0}$ , it is necessary to design the flight path such that  $l = l_0\sqrt{1 + \delta_K}$  with causes  $t = l/v = l_0/[v_0/\sqrt{1 + \delta_m}] \approx (l_0/v_0)(1 + \delta_m/2 + \dots)$ . In other words the system must sent ions of increased energy along properly dimensioned detours. Such a system we call an isochronous time-of-flight mass spectrometer [1,2].

Since even mono-mass ions start at slightly different times  $\pm\Delta t$  the mass resolving power  $1/\delta_m \approx 1/2\delta_t$  is limited to  $t_0/2\Delta t$ . In order to achieve high mass resolving powers one must either increase  $t_0$  or reduce  $\Delta t$ . Since it is not practical to reduce  $\Delta t$  below certain limits, it is advisable to increase  $t_0$  as much as possible. One way to do this is to increase the length of the ion flight path  $l_0$ . This will, however, cause the time-of-flight mass spectrometer become unwieldy unless one can design it such that its physically given flight path is used  $N$  times as a “race track” through which the ions pass again and again. To allow this with stable beam envelopes the transversal phase-space advance  $\mu$  per lap must stay [1] below but close to  $\pi/2$ . Such systems we refer to [4] as isochronous multi-pass time-of-flight mass spectrometers (MTOF-MS). There are two types of such systems:

- (1) those that use magnetic or electrostatic sector fields forming a ring [1,2,5,6] or
- (2) those that reflect ions repeatedly between ion mirrors [7–9].

Both systems can achieve energy isochronicity. In sector fields [1] more energetic ions move along trajectories of radii  $\rho$  that grow with  $\delta_K$ . In electrostatic mirrors [3] more energetic ions penetrate deeper into the repeller fields of the mirrors with this penetration depth being proportional to  $\delta_K$ .

The geometry of our MTOF-MS is shown in Fig. 1. It consists of a field-free region and two

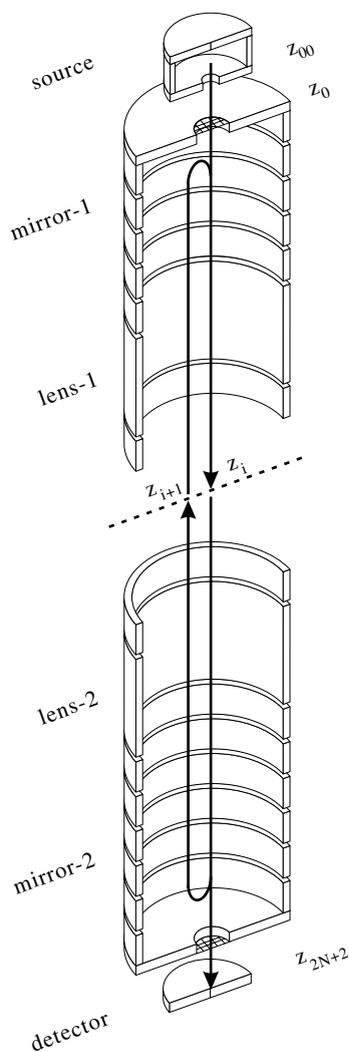


Fig. 1. Sketch of a coaxial multi-pass time-of-flight mass spectrometer (MTOF-MS) that consists of a pulsed ion source, a fast ion detector as well as two coaxially arranged electrostatic ion mirrors and lenses all built from ring electrodes. To avoid ion losses the ion mirrors should not contain grids in the repeatedly used path of the ions. However, grids may be placed in the back of both mirrors since the ions need to pass through those only once when they enter or leave the “race track”.

coaxially arranged electrostatic ion mirrors that are built from ring-electrodes only. In this system the ions must pass  $N$  times through the “race track”, i.e. for each lap they must pass twice through the field-free region between the ion mirrors and once through each mirror. In order to

reach an overall time accuracy of one ppm it is necessary that the overall system geometry is mechanically stable to the same precision which corresponds to 0.4  $\mu\text{m}$  in our  $\approx 400$  mm long system. This stability can only be achieved if either the system is build such that the overall temperature coefficient is very small or that the temperature is very constant. For simplicity we have build the system such that it is mounted in a vacuum chamber with only weak connections to it. This design ensures that the system temperature follows changes in the laboratory temperature only with a long time constant.

Assuming that positively charged ions have been produced in a storing ion source [9] at potential 0 they must move in a system as shown in Fig. 1:

- (1) From the ion source to the midpoint between the ion mirrors. In this case the ions must pass through mirror-1 in order to enter the “race track”, which requires that mirror-1 is deactivated for a short time. For this purpose the potential of its back-electrode must be lowered to a potential below the ion source potential.
- (2)  $N$  times through the “race track” during which time mirror-1 as well as mirror-2 must be activated, i.e. their back electrodes must be at potentials that are higher than the ion source potential.
- (3) From the midpoint between the ion mirrors to the ion detector. In this case the ions must pass through mirror-2 in order to leave the “race track”, which requires that mirror-2 is deactivated for a short time. For this purpose the potential of its back-electrode must be lowered to a potential below the ion source potential.

To achieve a high time resolving power it is necessary that the MTOF-MS is energy isochronous. For such a system the difference in ion flight times for ions of mass  $m$  and energy  $K_0$  and for ions of the same mass  $m$  but different energy  $K = K_0(1 + \delta_K)$  should be negligibly small. This time difference can be described as

$$\Delta t_K = \{(t|K)_0 + N[(t|K)_1 + (t|K)_2] + (t|K)_{2N+1}\} \delta_K, \quad (1)$$

where  $N$  is the number of laps in the “race track”. Since  $N$  is a large number for most settings of an MTOF-MS, the middle expression of Eq. (1) is usually much larger than the others and the postulate  $(t|K)_1 + (t|K)_2 = 0$  already reduces  $\Delta t_K$  to a very small value.

### 3. Experimental tests of the MTOF-MS

For experimental tests of the MTOF-MS the ions were produced in an electron impact source, extracted by an electric field of  $\approx 100$  V/mm and then accelerated to 1500 eV. This system provided ion pulses of  $\approx 3$  ns duration with repetition rates of 1 kHz. As detector we used a double channel plate of 25 mm diameter. The ion mirrors are simple stacks of ring electrodes. Each ring has an inner diameter of 40 mm and is 8 mm high except for the second to last electrode that has a height of 26 mm. The apertures in the back of the last mirror electrodes were 6 mm.

So far we have recorded ions of narrow mass multiplets only. One of those is the CO–N<sub>2</sub>–C<sub>2</sub>H<sub>4</sub> mass triplet that contains ions of the masses  $m_{\text{CO}} = 27.99491$  u,  $m_{\text{N}_2} = 28.00615$  u,  $m_{\text{C}_2\text{H}_4} = 28.03130$  u and thus features mass differences of  $m_{\text{CO}} - m_{\text{N}_2} \approx 0.01124$  u  $\approx 10.4638$  MeV/c<sup>2</sup> and  $m_{\text{C}_2\text{H}_4} - m_{\text{N}_2} \approx 0.02515$  u  $\approx 23.4290$  MeV/c<sup>2</sup>. The obtained spectrum in Fig. 2 shows a mass

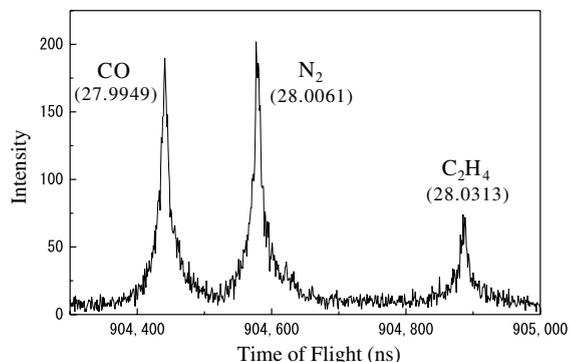


Fig. 2. The mass spectrum of a mixture of CO, N<sub>2</sub> and C<sub>2</sub>H<sub>4</sub> as obtained in the MTOF-MS of Fig. 1. The achieved FWHM mass resolving power in this spectrum is  $m/\Delta m \approx 25,000$  after  $N = 121$  laps. Assuming two of these masses to be known, one can determine the third one with an accuracy of  $< 1$  ppm.

resolving power of  $m/\Delta m \approx 25,000$  for ions that had performed 121 laps in  $\approx 905 \mu\text{s}$ . Using the masses of CO and  $\text{N}_2$  as known masses one finds the mass of  $\text{C}_2\text{H}_4$  as 28.031276(42) u which differs

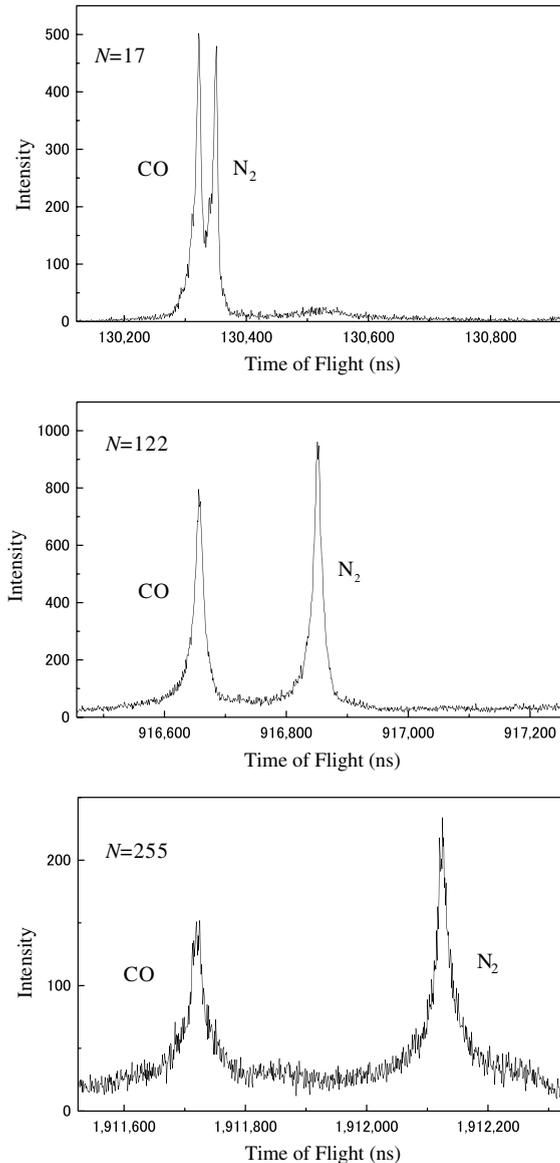


Fig. 3. Mass spectra of slightly different mixtures of CO and  $\text{N}_2$  as recorded in these experiments in which the corresponding ions had performed  $N = 17, 122$ , or 255 laps in the MTOF-MS. These spectra demonstrate that the mass resolving power increases drastically with  $N$  though the intensity is reduced by 0.1% per lap in our vacuum of  $\approx 10^{-6}$  mbar.

from the correct value by  $22 \text{ keV}/c^2$  or 0.85 ppm. Although this accuracy is not better than that obtained from storage rings [6] or Penning traps [10] an MTOF-MS is a considerably smaller and simpler system, and can be used as a mass separator to purify a beam or to study nuclear decay of a nucleus of fixed neutron and proton number [11].

As one would expect the mass resolving power increases with the number of performed laps in the “race track”. This is shown in Fig. 3 for the CO– $\text{N}_2$  mass doublet after 17, 122 and 255 laps. As one can see, the separation of the two mass peaks increases with  $N$  the number of laps, while their widths stay approximately constant. The maximally achieved mass resolving power was higher than  $m/\Delta m = 40,000$ . The signal-to-noise ratios in the mass spectra, however, decreased with  $N$ . The reason for this decrease is that for large  $N$  the ion flight paths become so long that noticeable percentages of the ions experience large-angle scattering events with residual gas atoms and thus are lost. Since the vacuum in the “race track” of our system is only  $\approx 10^{-6}$  mbar we lose about 0.1% of the intensity per lap.

To determine the masses of short-lived nuclei we plan to collect them in a gas cell before they are passed to the MTOF-MS through an RF ion-guide system [12]. In this setup the presently used storage ion source [9] will be replaced by a small Paul trap [13].

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