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August 5, 1991

PROPOSAL TO THE ISOLDE COMMITTEE

HIGH-ACCURACY MASS DETERMINATION OF UNSTABLE NUCLEI  
WITH A PENNING TRAP MASS SPECTROMETER

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SUMMARY

The mass of a nucleus is its most fundamental property. A systematic study of nuclear masses as a function of neutron and proton number allows the observation of collective and single-particle effects in nuclear structure. Accurate mass data are the most basic test of nuclear models and are essential for their improvement. This is especially important for the astrophysical study of nuclear synthesis. In order to achieve the required high accuracy, the mass of ions captured in a Penning trap is determined via their cyclotron frequency  $\nu_c$ . Mass measurements already performed with this technique at ISOLDE-2 demonstrated that resolving powers exceeding  $\nu_c/\Delta\nu_c$  (FWHM) of 1 million can easily be achieved and accuracies of typically  $\delta m/m = 10^{-7}$  are obtained. The technique can in principle be applied to isotopes of all elements available at ISOLDE. Hence a highly accurate, systematic investigation of nearly the complete nuclear mass surface will become possible.

Geneva 1991

216467

# 1 Scientific Motivation

The mass of a nucleus is its most fundamental property and reflects all interactions between the nucleons building up the nucleus. A systematic study of nuclear masses as a function of neutron and proton number allows the observation of effects of nuclear deformation, shell and subshell closure as well as pairing. Accurate mass data are the most basic test of nuclear models and are essential for their improvement as for example for the prediction of masses very far from stability. This is especially important for the astrophysical study of nuclear synthesis. Accurate mass values are in addition needed for the clarification of new phenomena like the proposed neutron halo in  $^{11}\text{Li}$ .

The majority of masses of radioactive isotopes have been measured by determination of Q-values in nuclear reactions or in nuclear decays. In the latter case, masses of isotopes are often linked via long decay chains to a stable nucleus with a very well known mass. It is evident that this procedure can lead to large uncertainties for mass values of isotopes far from stability.

For a long time direct mass determinations have been limited to stable isotopes or isotopes close to stability. This changed in the 70's with magnetic spectrometers put on-line to isotope separators. The Orsay group (Audi et al., 1986) succeeded in measuring the masses in long isotope chains of alkali elements. They demonstrated the possibilities embedded in direct mass determination of isotopes far from stability. The persisting demand for more precise masses of short-lived isotopes (or exotic particles) has prompted during recent years the development of new techniques. One powerful approach is the determination of the cyclotron frequency in a Penning trap, first demonstrated for light stable masses and recently also applied to unstable heavy isotopes in the Penning trap mass experiment at ISOLDE-2. These measurements allowed to determine the masses of about 60 unstable isotopes of alkali and alkali earth elements with an accuracy of  $\delta m/m \simeq 10^{-7}$ . For isotopes far from stability these mass values are now the most accurate obtained so far.

We plan to complete our mass measurements on isotopes of alkali and alkali earth elements with the already existing tandem Penning trap mass spectrometer. After implementation of new bunching techniques for the continuous ISOLDE ion beam (see below) and/or with the ISOLDE laser ion source we will extend our investigations to other elements. In the long term we will be able to investigate isotopes of practically all elements produced at ISOLDE and of some decay products. Of particular interest is the rare earth region where most of the masses far from stability are not known or only linked by floating  $\alpha$ -chains. We aim for mass measurements on neutron-deficient mercury isotopes in order to look for correlations between their masses and the large odd-even staggering in nuclear deformation and the shape coexistence which were observed by laser spectroscopy. In a later stage we plan to extend these investigations to the neighbouring gold and platinum isotopes. Beside the systematic exploration of masses we plan to investigate particular cases, like the nuclei around  $^{146}\text{Gd}$  or the very neutron-rich nucleus  $^{11}\text{Li}$ , for which we hope to achieve an accuracy of about 10 keV.

## 2 The Penning Trap Mass Spectrometer

### 2.1 The Principle

The principle used in the Penning trap mass spectrometer is to determine the resonance of the cyclotron frequency

$$\omega_c = \frac{q}{m} \cdot B$$

of an ion with charge to mass ratio  $q/m$  in a homogeneous magnetic field  $B$ . In a Penning trap the confinement of a charged particle is achieved by three electrodes, a ring electrode and two endcaps, which create an axially symmetric electrostatic quadrupole field. Due to the additional electric force the ion motion is not a pure cyclotron motion with frequency  $\omega_c$  but a superposition of three harmonic eigenmotions: A slow drift around the trap axis called magnetron motion with frequency  $\omega_-$ , a modified cyclotron motion with frequency  $\omega_+$  and an axial oscillation with frequency  $\omega_z$ . The Penning trap provides the relation

$$\omega_c = \omega_+ + \omega_-$$

which is the key for a precise mass determination. The direct determination of this sum frequency and the knowledge of the magnetic field is sufficient for the determination of the mass of a stored ion. The ion motion can be excited directly at this sum frequency by means of an azimuthal quadrupole field (Bollen et al., 1990). After an excitation time  $T_{RF}$ , an initially pure magnetron motion is transformed into a pure cyclotron motion. This transformation leads to a large change in radial energy (since  $\omega_+ \gg \omega_-$ ) which is detected by a time-of-flight technique described below. The width of the cyclotron resonance curve  $\Delta\nu(FWHM) \simeq 0.9/T_{RF}$  is determined by the Fourier-limit of the driving RF-field switched on for a time  $T_{RF}$ .

### 2.2 The Experimental Set-up and its Present Performance

Figure 1 shows the experimental set-up of the tandem Penning trap mass spectrometer as it was used at ISOLDE-2. It consists of two main parts, the lower Penning trap (trap #1) acting as ion beam buncher and cooler and the upper high-precision trap (trap #2) in which the mass determination takes place. The first trap is placed in a vacuum chamber mounted in a 0.7 T electromagnet. The continuous 60 keV ISOLDE beam is collected on a rhenium foil mounted in the lower endcap of this trap. The foil is rotated by 180° and current-heated for 500 ms so as to release and surface-ionize the collected material. By buffer gas collisions the ions are trapped and cooled. An additional excitation of the ion motion with  $\omega_c$  prevents an increase of the magnetron motion and a loss of ions. Since the cyclotron frequency is involved, this new cooling technique is mass selective (G. Savard et al., 1991). Use of He gas at a pressure of  $p = (10^{-4} - 10^{-3})$  mbar yields a capture efficiency of  $\epsilon_{capture} > 10^{-4}$ . A mass resolution of up to  $m/\Delta m(FWHM) = 500$  is obtained in this trap, sufficient to remove isotopic contaminations present in the ISOLDE beam. After a cooling period of 200 ms the ions are ejected. The ion bunch is accelerated to 1 keV, transferred to the upper Penning trap with an electrostatic lens system, again retarded and captured in-flight in trap #2 (H. Schnatz et al., 1986).

With regard to the high accuracy aimed for in this experiment it is important to use a trap with minimum electric and magnetic field imperfections which can cause frequency shifts. A careful investigation of possible sources of distortion allowed the construction of a Penning trap with very low magnetic and electric field imperfections (Becker et al., 1990). The azimuthal quadrupole field for the excitation is created by splitting the ring electrode into four quadrants. After excitation of their motion by the RF-field the ions are gently kicked out of the trap and drift along the magnetic field lines to an ion detector placed in the weak fringe field of the magnet. The radial energy gained by the excitation is converted into axial energy in the inhomogeneous part of the magnetic field. Therefore the measurement of the drift time as a function of the applied

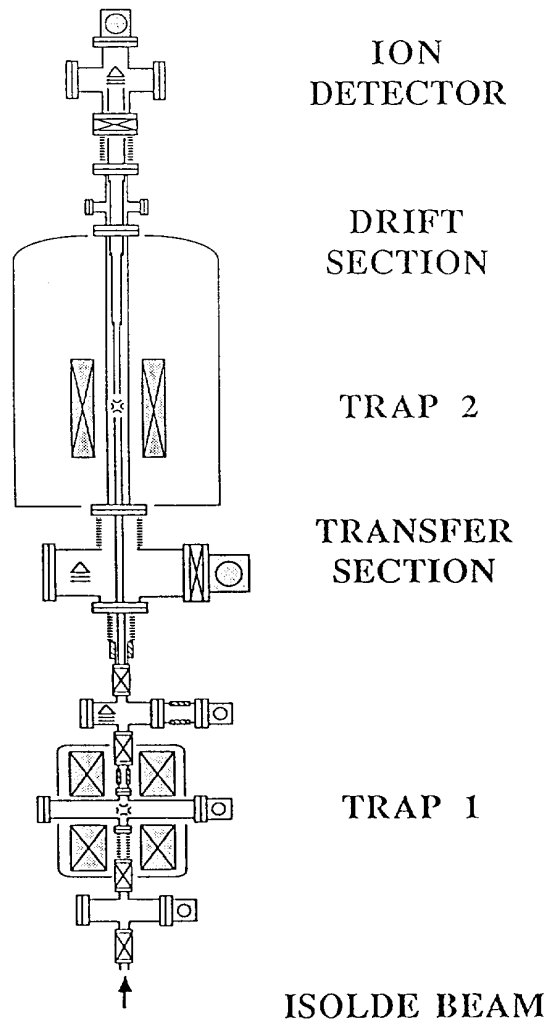


Figure 1: Experimental set-up of the tandem Penning trap mass spectrometer

radio-frequency yields a resonance curve with a minimum at  $\omega_c = \omega_+ + \omega_-$ , corresponding to a maximum of gained radial energy. As an example, figure 2 shows a resonance curve obtained for  $^{120}\text{Cs}$  ( $T_{1/2} = 64 \text{ s}$ ). As expected from the excitation time of 1.8 s a Fourier-limited linewidth of 0.5 Hz is achieved corresponding to a resolving power of 1.5 million. This measurement took 7 minutes with 2000 ions detected. The overall efficiency (ratio of the number of detected ions to the the number of ions implanted in the foil) was determined to be  $\epsilon_{total} = 5 \cdot 10^{-5}$ . An accuracy of  $\delta m/m = 2.1 \cdot 10^{-7}$  could be achieved for this isotope, corresponding to  $\delta m = 24 \text{ keV}$ .

During runs several cyclotron resonances are measured for each isotope under investigation. The calibration of the magnetic field is performed via the cyclotron resonance of a stable reference isotope with well known mass. Such measurements of the reference isotopes are repeated frequently during the run in order to detect drifts and changes of the magnetic field.

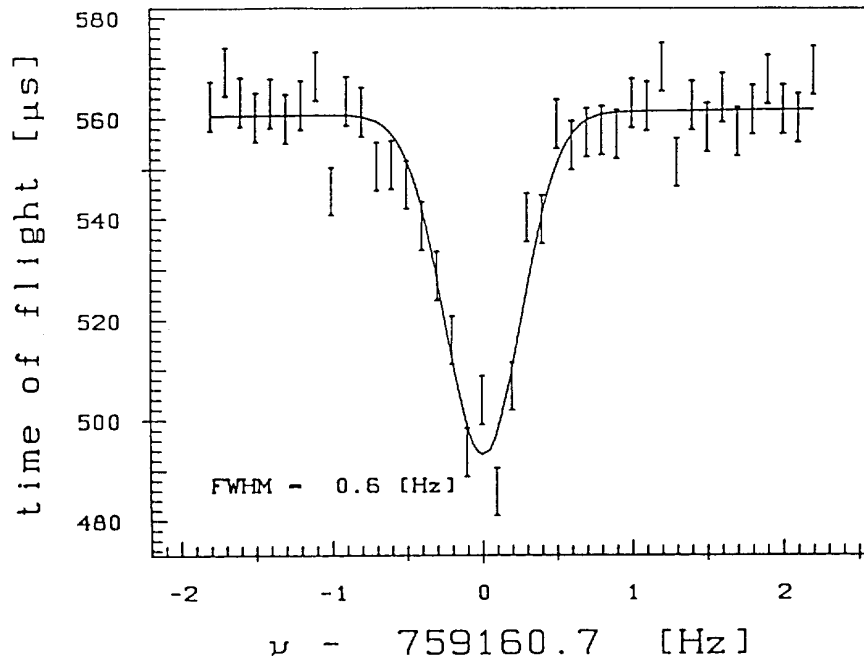


Figure 2: Cyclotron resonance of  $^{120}\text{Cs}$  ( $T_{1/2} = 64\text{ s}$ ) with a resolving power of 1.5 million.

### 3 Summary of Results as obtained at ISOLDE-2

Table 1 summarizes the isotopes investigated so far with the Penning trap mass spectrometer. The stable isotopes  $^{39}\text{K}$ ,  $^{85,87}\text{Rb}$ ,  $^{133}\text{Cs}$  listed in the table were used for the magnetic field calibration. Experience with the treatment of more than one ion species in the trap and the high resolving power of the Penning trap mass spectrometer allowed for the first time in the history of mass spectrometry to resolve isomeric and ground state of a nucleus in the cases of  $^{78}\text{Rb}$  and  $^{84}\text{Rb}$  (fig. 3). Masses could be determined for the first time in the cases of  $^{78,79}\text{Sr}$ ,  $^{122m}\text{Cs}$ ,  $^{124,126}\text{Ba}$ , and  $^{230}\text{Ra}$ . The high performance of the Penning trap mass spectrometer allowed the determination of the mass values of all isotopes with accuracies of  $\delta m/m \approx 10^{-7}$ . The measurements performed on neutron-deficient Cs isotopes increased the accuracy of the masses far from stability by one order of magnitude (Stolzenberg et al., 1990). This has resolved the conflict between the mass values from  $Q_\beta$ -studies and values obtained by a Mattauch-Herzog-spectrometer (Audi et al., 1986). In addition wrong mass values were found for  $^{130}\text{Cs}$  and  $^{135}\text{Cs}$ , both close to stability, due to an underestimation of the errors of  $Q_\beta$ -values. Presently, calculations with an extended macroscopic-microscopic model are performed for the Cs-region with the parameters adjusted to give the best fit to the accurate experimental masses. The model then allows the evaluation of spins, magnetic dipole and electric quadrupole moments which have to be compared with experimental data. The data evaluation for the other elements is nearly finished. In the case of the neutron-deficient Rb and Sr isotopes an analysis in the frame of macroscopic-microscopic models reveals a pronounced drop in the neutron pairing energy for the lightest isotopes. We are presently studying this behaviour, taking into account information from laser and gamma spectroscopy. The performed measurements did not only improve the accuracy of the mass values of the investigated nuclei but also improved those of neighbouring isotopes of other elements due to the manifold links between them.

### 4 Future Developments

The main aim is to extend in the future the mass measurements to elements which cannot be surface-ionized and to increase the efficiency of the apparatus. In order to achieve this, we plan

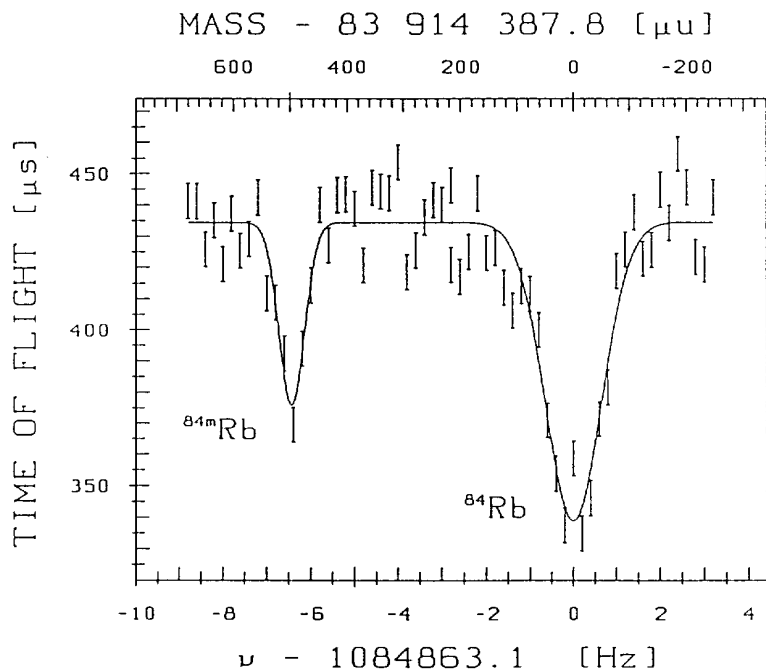


Figure 3: Cyclotron resonances of  $^{84}\text{Rb}$  and  $^{84m}\text{Rb}$ . The mass difference determined to be  $463(10) \text{ keV}$  is in full agreement with the  $\gamma$ -spectroscopically known energy of the isomer of  $463.62(10) \text{ keV}$ .

to divide the apparatus into three functional parts, each one optimized for its particular task: A bunching device, a Penning trap ion cooler and the Penning trap mass spectrometer, which will be the already existing trap #2. The other two parts are presently under construction.

#### Ion Beam Buncher(s)

*Paul Trap Ion Beam Buncher.* This system can retard and collect the continuous ISOLDE beam and deliver it to the ion cooler as an ion bunch (R.B. Moore et al., 1991). A prototype system was successfully tested at ISOLDE-3. The achieved bunching efficiency of already 0.2% was limited by the RF-supplies used. With a new design of the trap, more powerful supplies and a pre-buncher, bunching efficiencies exceeding 10% are expected. This buncher which converts the existing tandem Penning trap mass spectrometer into a triple trap mass spectrometer will allow us to investigate all isotopes of those elements which are not surface-ionizable and are produced relatively free of isobaric contamination. In the case of the rare earth elements this can be achieved by means of the ISOLDE laser ion source.

*Laser-Induced-Surface-Ionization (LISI) Source.* The ISOLDE beam will be collected on an appropriate target and released and surface-ionized by heating the surface with a short intense laser pulse. The capture efficiency for the produced ion bunches in the ion cooler will be much higher than the present efficiency of trap #1 since the laser induced-ion bunch has a very short pulse length (about 100ns). The system will be able to produce positive as well as negative ions (halogens) and to extend the mass determination of isotopes of surface-ionizable elements to regions far away from stability due to its high efficiency. In addition, the source will enable the creation of carbon clusters over a large mass scale. As already tested in Mainz the clusters can be used for the calibration of the magnetic field. Hence not only direct but also absolute mass determinations will be performed. The system will be open for a future use of laser-induced desorption and resonance ionization for the investigation of isotopes of refractory elements only available as daughter products. Such a scheme has already been successfully applied at ISOLDE-2 for the investigation of spin, moments and charge radii of gold and platinum isotopes.

Table 1: Isotopes for which cyclotron resonances have been determined with the Penning trap mass spectrometer. The stable isotopes  $^{39}\text{K}$ ,  $^{85,87}\text{Rb}$ ,  $^{133}\text{Cs}$  have been used for the calibration of the magnetic field.

Isotope	Mass Number
K	39
Rb	75, 76, 77, 78, 78m, 79, 80, 81, 82m, 83, 84, 84m, 85, 86, 87
Sr	<span style="border: 1px solid black; padding: 0 2px;">78</span> <span style="border: 1px solid black; padding: 0 2px;">79</span> 80, 81, 82, 83, 87
Cs	118, 119, 120, 121, 122+122m, <span style="border: 1px solid black; padding: 0 2px;">122m</span> 123, 124, 125, 126, 127 128, 129, 130, 131, 132, 133, 134, 135, 136, 137, 138, 139, 140
Ba	<span style="border: 1px solid black; padding: 0 2px;">124</span> <span style="border: 1px solid black; padding: 0 2px;">126</span> 138, 139, 140, 141, 142, 143, 144
Fr	209, 210, 211, 212, 221, 222
Ra	226, <span style="border: 1px solid black; padding: 0 2px;">230</span>

#### The Penning Trap Ion Cooler

For the application of the buffer gas cooling scheme (see section 2.2) a new Penning trap is presently designed which will be optimized for accepting a large phase space volume of the incoming beam, for short cooling times and a cooling limit of 77 K, as well as higher mass selectivity for the removal of contaminating ions. The trap will be placed in a 5 T superconducting magnet presently being tested at Mainz.

## 5 The Mass experiment at the PS-Booster ISOLDE

The move to the PS-Booster-ISOLDE implies a number of changes to the Penning trap mass spectrometer. Since the spectrometer was designed for vertical operation at ISOLDE-2, a two-stage platform is required. A discussion about the size and location of this platforms already took place and will not be repeated here. Furthermore the supporting structure for the apparatus and parts of the ion optics of the apparatus have to be modified or renewed in order to fit the changed requirements. Due to the more 'dirty' environment (temperature variations, changes of the magnetic field) as compared to ISOLDE-2, additional field and temperature stabilization have to be installed.

In the beginning it is planned to bend the ISOLDE beam by 90° and to deliver it to the existing trap # 1 which will be placed on the first platform. After completion of the new ion beam bunchers and the ion cooler, the LISI-device will be placed at the intercept of the ISOLDE beam axis and the axis of the apparatus. Later trap # 1 will be replaced by the superconducting ion cooler. Finally the Paul trap will be installed in the horizontal beam line in front of the mass set-up.

## 6 ISOLDE Requirements

Most of these requirements have already been discussed in the answers to the questionnaire given to all ISOLDE users and in later discussions with the Technical Committee. Therefore only the most important points will be repeated here.

Space: For the installation of the apparatus a two-stage platform is required. This platform should surround the self supporting structure of the mass experiment. The first floor should be able to carry locally the load of the electromagnet (1500 kg).

Two barracks are needed: One for lasers and one for preparation, storage and on-line data evaluation.

Beam Quality: The mass experiment needs beams with as little contamination by neighbouring isotopes and isobars as possible. Therefore the operation of the HRS will be required in cases where the resolving power is high enough to get rid of isobaric contaminations. The operation of the laser ion source will allow the investigation of rare earth elements.

Environment: Changes of the magnetic field in the surrounding of the mass set-up result in shifts of the cyclotron frequency. In order to minimize these effects no crane operation should take place during our beam times and for certain test periods. Magnets which are close by should not be energized during these periods. Especially the magnetic field of the NICOLE experiment should be shut off during our experiments..

## 7 Beamtime

For the two years after the start of the PS-Booster ISOLDE we ask for 50 shifts of radioactive beamtime. Based on our experience from ISOLDE-2 we estimate to be able to measure the mass of about 100 isotopes during these 50 shifts. We intend to use both the GPS and the HRS separator and are strongly interested in an operation of the laser ion source.

In addition to unstable beams we apply for 20 shifts of stable beam for testing of the apparatus and the new ion bunchers. For the test of the beam lines and the bending of the beam into our apparatus additional beam time will be needed, the amount depending on the status of the beam observation systems and the ion optics properties.



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