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Precision Measurements of Masses of Radioactive Atoms using ISOLDE and Ion Traps

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Abstract

A mass spectrometer has been set up at the on-line isotope separator ISOLDE at CERN/Geneva. Mass-separated radioactive ions are stored in a Penning trap. Their mass is determined by a measurement of the cyclotron frequency in the magnetic field of a superconducting magnet. A resolving power of up to 300 000 and a precision of some 10 keV were determined in case of mass measurements of neutron-deficient *Rb* and *Cs* isotopes.

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1 Introduction

The nuclear mass represents as a fundamental nuclear property, which embodies all the effects of nuclear forces, a key input parameter for nuclear matter calculations: For example, its knowledge is important for a determination of the limits of nuclear stability against spontaneous emission of nuclear matter, the binding energy is sensitive to shell and deformation effects as recently shown in case of a possible stable octupole deformation of heavy *Ra* isotopes, and it serves as basis for astrophysical calculations. Until now, precise direct mass determinations are restricted to the valley of stability or its direct neighbourhood [1]. Far away from this valley nuclear binding energies are measured indirectly by means of mass differences obtained as Q values from nuclear decays or reactions. The only exceptions until now are a series of direct mass measurements on alkali isotopes performed by the Orsay group at ISOLDE by means of a Mattauch-Herzog spectrometer [2,3] and very recently performed time-of-flight experiments at Los Alamos [4] and GANIL [5,6] on light isotopes.

Mass measurements of heavier isotopes far away from stability by a determination of Q values suffer from errors introduced by summing up the errors of the many mass differences which link the mass of the isotope under investigation to that of a well known mass and, in addition, from uncertainties due to an insufficient knowledge of the nuclear level schemes of nuclei far away from stability. Hence, a method is very desirable where the masses are directly determined, i.e. independently of the knowledge of level schemes and masses of neighbouring isotopes. This can be achieved by measuring the cyclotron frequency of ions stored in a Penning trap. Such a system which can be loaded with ions delivered from an on-line isotope separator has been developed at Mainz and recently installed at the ISOLDE facility at CERN [7]. G. Bollen, P. Dabkiewicz, P. Egelhof, H. Kalinowsky, F. Kern, H.-J. Kluge, H. Schnatz, L. Schweikhard, H. Stolzenberg and H. Stürmer/Mainz were involved in the conception, construction and setting-up of the apparatus at CERN. First test experiments were performed in collaboration with K. Kunz and Th. Hilberath/Mainz, G. Audi/Orsay, R. B. Moore/Montreal, G.M. Temmer/Rutgers University and G. Ulm/CERN.

2 Principles of the Technique

2.1 Penning Trap

Particles with charge e and mass m perform a cyclotron motion in a homogeneous magnetic field B given by

$$\omega_c = (e/m)B. \quad (1)$$

In order to achieve a high resolving power $R = \nu_c/\Delta\nu_c$, the cyclotron frequency ν_c should be as large and the line width $\Delta\nu_c$ as small as possible. The ultimate line width depends by the uncertainty principle on the coherent interaction time T_{RF} with the applied radio frequency field ($\Delta\nu_c \simeq 1/T_{RF}$). Hence, it is straightforward to use a Penning trap in the magnetic field of a superconducting magnet because it combines long confinement with maximum ν_c .

A combination of static, magnetic and electric fields is used to establish a three-dimensional trapping potential [8,9]. Due to the special (hyperbolic) shape of the electrodes which are used for a restoring force in the direction of the magnetic field (z axis) the z motion of an ion stored in an ideal trap is decoupled from those in the $x - y$ direction and furthermore all motions are harmonic. They can be separated into an oscillation ω_z along and the modified cyclotron frequencies ω_+ and ω_- perpendicular to the z direction. The frequencies deviate from the pure cyclotron frequency ω_c due to the presence of the electrostatic field. The harmonic oscillations are described by the frequencies

$$\omega_z = \sqrt{(2eU/mr_0^2)} \quad (2)$$

$$\omega_+ = \omega_c/2 + \sqrt{(\omega_c^2/4 - \omega_z^2/2)} \quad (3)$$

$$\omega_- = \omega_c/2 - \sqrt{(\omega_c^2/4 - \omega_z^2/2)} \quad (4)$$

U is the potential difference between the ring electrode and the end caps, r_0 is the minimum radius of the ring electrode. In case of an ion with mass $m = 40 \text{ amu}$, $B = 5.87 \text{ T}$, $U = 8 \text{ V}$ and $r_0 = 0.8 \text{ cm}$ the frequencies are $\nu_c = 2253 \text{ kHz}$, $\nu_z = 124 \text{ kHz}$, $\nu_+ = 2250 \text{ kHz}$ and the magneton frequency $\nu_- = 3.4 \text{ kHz}$ almost independent of the mass of the stored particle.

Note that ω_z as well as the modified cyclotron frequencies depend on the voltage U applied to the electrodes. These frequencies correspond to one-quantum transi-

tions in the harmonic oscillators. However, the sum frequency $\omega_+ + \omega_-$ equals to ω_c . This frequency depends only on B (see equ. (1)). Although the ω_c resonance has a small strength because it is a two-quantum transition, it has the advantage of being independent of the trapping voltage. Even more important, this resonance is in contrast to ω_+ not very sensitive to space charge effects caused by a larger number of stored ions, and is not very much influenced by imperfections of the hyperbolic shape of the electrode caused, for example, by the necessary holes for capture and ejection of the ions [10].

2.2 Detection of the Cyclotron Resonance

In resonance the trapped ions gain energy out of the applied radio frequency (RF) field. This is detected by a time-of-flight (TOF) method [11]. The charged particles are ejected out of the trap by applying an electrical pulse to the ring electrode. For a short time the potential between the endcaps and the ring electrode is lowered so that the ions with highest energy can just escape the trap. These ions drift to a channel plate detector and their TOF is determined (Fig. 1). This procedure is repeated by continuously lowering the trapping potential until the trap is empty. The mean TOF as a function of the RF shows a resonance which gives the mass of the stored ions (Fig. 2).

The basic principle of this detection technique is easily understood. In resonance the cyclotron orbit becomes larger. As a consequence the orbital magnetic moment μ_L increases, leading to a higher energy of the charged particle in the magnetic field. If the ions are ejected out of the trap into a region of lower magnetic field, the ions experience a force $\mu_L \partial B / \partial z$ in the direction of magnetic field gradient. Conservation of energy demands that the radial energy $\mu_L B$ is transformed into longitudinal kinetic energy. This is just the reverse of the magnetic-bottle effect. Hence in resonance the ions reach the detector faster than off resonance.

Fig. 1 shows the TOF effect for the case of ^{124}Cs . If the frequency $\nu_c(^{124}\text{Cs})$ is applied, the mean TOF is shifted to smaller values. This is seen in the bottom part of Fig. 1, which shows the difference between the TOF pattern in and off resonance. The change in the mean TOF has to be kept small if high resolution of the mass determination is required. Hence the change in TOF seen in Fig. 1 amounts only

to a few percent. If the strength of the RF field is increased, the ν_c resonance is power broadened. Finally at very high RF power, the ions gain such a high radial energy that they can no longer escape through the hole of the ion trap. In this case, they strike the electrodes and are lost. This effect can be used for purification of the sample, i.e. to get rid of unwanted contaminating species in the trap.

2.3 Loading a Penning Trap

The ion storage technique has not yet been used in nuclear physics for the study of short-lived nuclei. The reason is an experimental difficulty: The ions investigated so far in an ion trap were generally created inside the trap. However, in case of the study of short-lived isotopes these rare species have to be produced outside the trap and then guided with high efficiency into the trap. We have developed and tested a technique by which a bunch of ions can be captured in flight in a Penning trap [10].

3 Experimental Setup

The experimental setup for the mass measurements is shown in Fig. 3. The apparatus consists of a Penning trap (trap 1) placed in the pole gap of an electro magnet, a transfer line with a number of electrostatic lenses and deflectors, a Penning trap (trap 2) placed in the stable and homogeneous field of a superconducting magnet, a drift tube and finally a channel plate detector.

The first trap is essentially a bunching device which collects the continuous ISOLDE beam. The second trap is used for high-precision measurements of the cyclotron frequency. Hence ultra-high vacuum, excellent homogeneity of the magnetic field and perfect geometry of the second trap are essential. Since the operating conditions of a bunching trap are incompatible with those of a precision trap, the layout of Fig. 3 was chosen with two completely separated traps connected only by a transfer tube. This configuration allows efficient differential pumping between traps 1 and 2 and the installation of electro-optical devices for steering the ion beam.

Bunching Trap: Alkali ions delivered by ISOLDE are implanted into a turnable rhenium foil mounted in front of a hole in the lower endcap of the Penning trap, then surface ionized by heating the foil, trapped [12,13] and finally ejected by a sudden change of the trapping potential.

Transfer Tube: After leaving the trap the ion bunch is accelerated to 1 keV by some electrodes. A system of electrostatic lenses guides the bunch into the fringe field of the superconducting magnet in such a way that the radial energy is not increased. Extensive electro-optical calculations have been performed to find an optimum design [14]. This is most important for mass measurements, which require a very small energy spread of the trapped ions. The transmission of the transfer tube has been measured to be 80 % (including the transmission through the holes in the endcaps of trap 2). At the entrance of trap 2 the length of the bunch is shorter than 30 μ s.

Precision Trap: Here the bunched ion beam is captured in flight by retarding it to a few eV just before entering the trap. The potential of the lower endcap of trap 2 is switched to the potential of the ring electrode just at the moment the ion bunch arrives. When the ions pass the center of trap 2 the potential of the encap is raised again and the ions can no longer escape. More details of the apparatus built for mass measurements are given in [10].

4 Mass Measurements

The first test experiments with radioactive isotopes have only been performed recently and concerned neutron-deficient *Rb* and *Cs* isotopes, which are produced with high yields up to 10^{11} mass-separated ions per second and mass number at ISOLDE. The isotope with the shortest half life investigated was ^{122}Cs with $T_{1/2} = 4.2 \text{ min}$ and 21.0 s. The aim of these experiments was to check the performance of the apparatus under the realistic conditions of a beamtime at an on-line isotope separator. The results given below were obtained during a 2-days run in December 1986 ($^{77,78,85,86,88}\text{Rb}$ and ^{88}Sr) and a 8-hour run in June 1987 ($^{122,124,125,127,135,136}\text{Cs}$). A further test run is scheduled for September 1987.

5 Performance

Trapping Efficiency: Up to 70 % of an ion bunch ejected out of trap 1 can be retarded and captured in flight in trap 2. More details are given in [10].

Sensitivity: Typically 50 ions were stored in trap 2 at a time for the measurements of the cyclotron frequency. Since the detection scheme for the resonance by TOF is destructive, the trap has to be refilled after each cycle. This involves preparing ions in trap 1, ejecting them as an ion bunch and injecting into trap 2, inducing the RF, ejecting the ions and measuring the TOF. The cycle time is about 1 s. Minimum 10^4 to 10^5 stored and detected ions are required for a resonance curve with sufficient statistics. Hence one mass measurement takes typically 30 min to 1 hour. The overall efficiency was determined to be $\varepsilon = 5 \cdot 10^{-6}$ defined as the ratio between the number of ions implanted into the foil of trap 1 to those detected by the channel plate detector. Since the trapping efficiency for a bunched ion beam in trap 2 is up to 70 %, the main loss is clearly due to evaporation, ionization, trapping and ejection in the bunching trap 1. Much better efficiencies will be obtained if a laser ion source is installed at the on-line mass separator which delivers directly a pulsed ion beam [15] or the continuous beam of the on-line separator is bunched in a Paul trap as discussed during this conference by R. B. Moore [16].

Storage Time: The storage time of trap 2 has been determined to be $\tau \simeq 15 \text{ min}$ for Cs ions at a vacuum pressure of $p \leq 10^{-9} \text{ mbar}$.

Resolution: A resolving power of $3 \cdot 10^5$ for K and Cs has been achieved. This corresponds to a line width of 4 Hz (2 Hz) and allows to resolve the cyclotron frequencies of isobars and in many cases also of isomers. Fig. 4 shows the signal obtained for the isobars $^{88}\text{Rb}/^{88}\text{Sr}$ which have a mass difference of 5.3 MeV. Both were produced at ISOLDE with comparable yields and cannot be separated by the ISOLDE mass separator. Although the resonances are broadened to $FWHM = 20 \text{ Hz}$ by RF power broadening they are clearly resolved owing to the still high resolving power of the Penning trap technique.

The Orsay group has investigated a number of isomeric states in neutron-deficient Cs isotopes by laser spectroscopy [17]. The same group could not resolve in their mass experiments [2,3] the ground and isomeric states. We have observed

in a measurement at $A = 122$ a broadened structure (Fig. 5), which can be fitted by two Gaussians with center frequencies differing by 3 Hz (500 keV), line widths of 2.7 Hz and heights of 3 and 6 %. Two half-lives are known for ^{122}Cs ($T_{1/2} = 4.2\text{ min}$ and $T_{1/2} = 21.0\text{ s}$). The signals of Fig. 5 were obtained by starting immediately the experiment after collection of the $A = 122$ ions on the foil. When the start of the experiment was delayed by some minutes, the resonance at lower frequency disappeared and we observed only one resonance with a line width of about 2 Hz and a height of the typical magnitude of 7 %. Hence, there is strong evidence that we have resolved for the first time in a mass experiment the ground and isomeric states. Due to lack of beam time, the measurement could not be repeated in June 1987.

Accuracy: The statistical uncertainty of the centroid of the resonances is about 5 % of the line width corresponding to $\Delta\nu_c/\nu_c \leq 2 \cdot 10^{-7}$ or 20 keV for ions with mass number $A = 100$. The magnetic field at the place of trap 2 is measured repeatedly by the determination of the cyclotron frequency of a stable alkali isotope. It was found in test experiments with those reference isotopes that the ratios of the cyclotron frequencies coincide with the tabulated mass ratios within the statistical uncertainties. Similar results were obtained in case of the test experiments with short-lived isotopes. In almost all cases the masses determined by the Penning trap technique coincide with the Orsay results within one standard deviation.

The dependence of ν_c of the space charge in the trap was checked experimentally. Varying the number of ions detected by the MCP detector from 4 to 400 corresponding to about 10 to 10^3 stored ^{133}Cs ions, we observed a space charge dependence of ν_c of 3 mHz or 0.6 keV per detected event in each cycle.

6 Conclusion

We were able to demonstrate that the Penning-trap technique is well suited to mass measurements far away from the valley of stability. After calibration of the magnetic field the masses are measured in an absolute way without the necessity of knowing the masses of neighbouring isotopes. The high resolving power of the technique enables to avoid problems and uncertainties by the isobaric impurities of on-line beams and even allows to separate resonances of isomers and groundstates. The accuracy of the technique is better than 100 keV for $A \simeq 100$ and sufficient for tests of nuclear models by masses of nuclei far from stability.

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Figure Captions

Fig.1: Time-of-flight spectra obtained for ^{124}Cs ($T_{1/2} = 30.8\text{ s}$) ions in and off resonance with the applied RF field. The spectra correspond to the indicated points (a,b) in Fig. 2.

Fig.2: Cyclotron resonance of ^{124}Cs ($T_{1/2} = 30.8\text{ s}$) as obtained by the mean time of flight as a function of the frequency of the applied RF field. The points are indicated which are used for the TOF spectra shown in Fig. 1.

Fig.3: Experimental set-up used for mass measurements of short-lived isotopes at the on-line separator ISOLDE at CERN.

Fig.4: Simultaneously recorded cyclotron resonance of the isobars ^{88}Rb and ^{88}Sr which have a mass difference of 5.3 MeV . The power broadened line width is due to higher RF power used during this measurement.

Fig.5: Mean time of flight of ^{122}Cs ions as a function of applied RF frequency.

Fig. 1

Fig. 2

Fig. 3

Fig. 5