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Mass Determination of Francium and Radium Isotopes by a Penning Trap Mass Spectrometer

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Abstract

A tandem Penning trap mass spectrometer is used for mass measurement of radioactive isotopes produced at the on-line isotope separator ISOLDE/ CERN. The mass is determined directly and with high accuracy via a measurement of the cyclotron frequency of the stored ions. Measurements were performed on ^{209,210,211,212,221,222}Fr and ^{226,230}Ra. A resolving power of 5×10^5 was used and an accuracy of 1.8×10^{-7} has been achieved.

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Nuclear masses are key input parameters for nuclear models. The masses of stable isotopes are now generally known with an accuracy of a few keV [1]. However, the errors grow rapidly for unstable isotopes away from the valley of β-stability. It is the binding energies of these short-lived nuclei which provide the most stringent tests of nuclear models and allow the discovery of new phenomena.

The masses of most radioactive nuclei have been obtained from the determination of Q-values in nuclear decays or in nuclear reactions. The need for increased accuracy for unstable isotopes has led to the development of more direct techniques such as the use of magnetic [2] and time-of-flight [3] spectrometers on-line at accelerators. We report here on an application of a Penning trap which allows the direct mass determination of heavy unstable isotopes by the measurement of the cyclotron frequency of radioactive ions.

An ion of mass m and charge q will orbit in a magnetic field B at a frequency $\omega_c = \frac{qB}{m}$, hence it is confined radially. In a Penning trap, an electrostatic quadrupole potential is superimposed on the magnetic field to provide axial confinement [4]. The quadrupole potential leads to an axial harmonic oscillation decoupled from the radial motion. However, it also affects the normal cyclotron motion ω_c by slightly decreasing its frequency to a reduced value ω_+. Finally, it gives rise to another radial motion, the magnetron motion, at a frequency ω_-. This is a slow precession around the trap axis in the crossed electric and magnetic fields.

For a pure electrostatic quadrupole potential, the sum of the modified cyclotron and magnetron frequencies is equal to the unperturbed cyclotron frequency. The measurement of this sum frequency therefore allows, from the known magnetic field, a direct determination of the mass of the stored ions. The ion's radial motion can be excited directly by an azimuthal quadrupole r.f. field (i.e. a field that results from an electric potential $V = V(r,z) \sin 4\varphi \sin \omega_{r.f.} t$) which couples the magnetron and cyclotron motions [5] (where r, z, and φ are cylindrical coordinates). If a collection of ions is prepared in a state of pure magnetron motion, applying such a field at a well choosen amplitude for a fixed amount of time will lead to a conversion of the initial magnetron motion into a pure cyclotron motion. This cyclotron motion, which will be of the same radius as the initial magnetron radius, is of much larger energy due to its higher frequency compared to the magnetron motion. If the trap is opened after excitation and the ions are allowed to drift slowly out of the magnetic field, the kinetic energy of the cyclotron motion is transferred adiabatically into longitudinal kinetic energy. This results in a shorter time of flight (TOF) from the trap to a detector. A resonance with the r.f. field at the sum frequency $\omega_c = \omega_+ + \omega_-$ is therefore observed as a decrease in this TOF.

The experimental set-up is shown in figure 1. It consists of two Penning traps, a transfer section between them and a drift section leading to a channelplate detector. The first trap is in the 0.7 Tesla field of an electro-magnet and is used as an accumulating-cooling-bunching device. The second trap is a compensated high-precision trap. It is placed in the homogeneous and highly stable 5.7 Tesla field of a superconducting solenoid. The transfer section is fitted

with adjustable diaphragms which allow precise control of the injection conditions into the second trap. In addition, differential pumping is applied between the traps to maintain the high vacuum (10^{-9} mbar) required in the high-precision trap.

The continuous 60 keV radioactive ion beam from ISOLDE is first accumulated on a rhenium foil situated in the lower endcap of the lower trap. After collection, the foil is turned to face the inside of the trap and the ions are released and surface-ionized by heating the foil with an electric current pulse [7]. The ions are captured and centered in the trap by a new mass selective cooling technique. It is a combination of buffer gas and sideband cooling described in more details in Ref. 6. The ions are then ejected as a bunch and transferred to the high-precision trap where they are captured in flight [8]. After r.f. excitation the ions are ejected and allowed to drift to the channelplate detector where the TOF signal is detected. The cyclotron resonance frequency is obtained by a fit to the mean time of flight as a function of the applied frequency. The theoretical lineshape is known to be symmetric but an analytical expression is not available. A gaussian was found to be a good approximation and is therefore used as fitting function.

The system must fulfill two important requirements. The first is to transfer the short-lived activity, available only in minute quantities, into the trap with an acceptable efficiency. The second is to achieve the necessary resolution and accuracy. For the first requirement, the 60 keV continuous beam of ISOLDE must be transformed into a pulsed low-energy ion beam. This is achieved by the implantation-reionization-cooling-ejection sequence in the first trap. The efficiency of this sequence is typically of the order of some parts in 10^{-4}. Once the ions are bunched, they can be transferred and captured in flight in the second trap with nearly 100% efficiency. The overall efficiency of the system in on-line experiments, defined as the ratio of the number of ions detected by the channelplate detector to the number of ions implanted into the foil, is about 10^{-4}.

For the second requirement, the accuracy needed was achieved by careful design, machining and alignment of the system [9]. The trap is made of oxygen-free copper to minimize distortion of the magnetic field homogeneity. It is gold plated to eliminate static charge build-up and has two pairs of correction electrodes to tune out higher order multipoles in the electrostatic potential. This is essential for achieving both high resolution and high accuracy. Resolving powers of over 2 000 000 have been achieved with this system for isotopes as heavy as ^{133}Cs. The resolution used in this work was Fourier-limited to about $\Delta\nu_c(FWHM) = 1$ Hz by the excitation time of $T_{r.f.} = 0.9$ seconds. This corresponds to a resolving power of 500 000 for the francium and radium isotopes investigated.

The accuracy of the system has been checked by measuring the mass of well known stable isotopes to high statistical precision. In particular, we have performed numerous measurements on ^{85}Rb, ^{87}Rb and ^{133}Cs whose masses are known to an accuracy of a few keV [1]. A single determination of the cyclotron resonance frequency of these stable isotopes has a typical fit error of some parts in 10^{-8}. Repeating the measurements several times one obtains an instrumental

error below 10^{-8}. In these measurements, as in on-line runs, the chosen reference isotope is measured at regular time intervals to monitor the decay of the magnetic field (observed to be about $\delta B/B = -2.5 \times 10^{-9}$ per hour) which is then taken into account in the mass determinations. If ^{85}Rb is taken as reference isotope, the masses of ^{87}Rb and ^{133}Cs are determined with a precision of about 3×10^{-8}. This error is mainly due to the uncertainty of the mass of the reference isotope (2.8 keV [1]). We call this error primary error in the following. Comparing the determined masses with the tabulated ones [1] we observe agreement within the primary error for ^{87}Rb and better than 1×10^{-7} for ^{133}Cs. Hence we conclude that calibration errors are less than 10^{-7} within a mass range of up to $\approx 60\%$. More tests on possible calibration errors will be carried out in the near future using cluster ions which provide a very accurate mass scale over a large mass range. The use of carbon clusters in particular will provide an absolute calibration for the system.

Measurements have also been carried out with different numbers of ions loaded into the trap. No space charge dependence has been observed for up to 100 ions of equal mass within the instrumental uncertainty. A more serious problem is the presence of contaminants in the ion cloud which has been observed to lead to shifts in the cyclotron frequency. These contaminant ions, which are often present at on-line mass separators, must be eliminated. The scheme used for buffer gas cooling in the lower trap is mass selective and removes isotopic contaminants from the collection. In cases where isobaric contamination is also present, it can be removed in the second trap by applying a dipole excitation at the reduced cyclotron frequency. This will excite the contaminants to larger radii until they hit the ring electrode and are lost from the trap. This method has been used to remove contaminants with relative mass differences as small as 10^{-5}.

In on-line runs, where less time can be devoted to each isotope, the accuracy is determined by the limited statistics available for each isotope and the accuracy with which the various parameters can be controlled. The most important parameter is the magnetic field which is affected by the magnetic field variations in the experimental hall due to the synchrocyclotron, mass separator magnets, elevator, etc. These magnetic field perturbations can only be monitored with a limited accuracy in run conditions. In addition, the measurements on francium and radium reported here are the first extension of this mass measuring technique to such heavy masses where the possible systematic shifts have not been checked thoroughly. We therefore assign a systematic error of 10^{-7} to each cyclotron resonance frequency determination which includes possible calibration errors. This is a conservative estimate for all possible errors other than statistical uncertainties. The future tests with heavy clusters, together with a better control of external magnetic field perturbations will allow us to determine this systematic error more accurately.

The system has been used previously to measure the masses of radioactive isotopes of Rb, Sr, Cs and Ba [10]. In this paper we report mass measurements performed on francium and radium isotopes. As an example, figure 2 shows a resonance curve obtained for ^{230}Ra. This resonance was obtained in 19.5 minutes with 15 000 detected ions.

The results extracted from our data are shown in table 1 together with the previous accepted values [1] and the results of a new adjustment. Both the primary error and the total error, which is the quadratic sum of the primary and the systematic errors, are given for the measured mass excesses. Our results show a good general agreement with the previous values which were obtained by the francium measurements of the Orsay group [11] and indirectly via Q_β or Q_α determinations. The excellent agreement with the accurately known mass values for ^{226}Ra and ^{221}Fr points towards negligible calibration errors when extending measurements from mass A=133 to the mass region around A=226 and gives confidence that our estimation of the systematic error is adequate. The mass of ^{230}Ra was measured for the first time, having previously only been estimated from systematics. The measurements reported here provide not only more accurate masses for most of the measured isotopes but also a firm backbone for the α-chains of this mass region. For example, the slightly shifted and more accurate value for the mass of ^{212}Fr is related directly to the masses of ^{216}Ac, ^{208}At and ^{204}Bi, all members of the same α-chain which is now more accurately linked to the rest of the mass table by the better mass of ^{212}Fr.

In conclusion, a Penning trap mass spectrometer for radioactive ions has been set up on-line at the isotope separator ISOLDE/CERN. It has and will be used to provide direct and accurate mass measurements of radioactive isotopes of surface-ionizable elements. Its use to measure the mass of francium and radium isotopes has improved considerably the mass values in this mass region. ^{230}Ra is the heaviest isotope for which its mass has been measured in a trap. The mass measuring technique used in this system is universal. We are currently developing a new collection system [12] to replace the collection-reionization sequence (which is only applicable to surface-ionizable elements) performed in the first trap. This will allow mass measurements to be performed on isotopes of all elements produced at ISOLDE.

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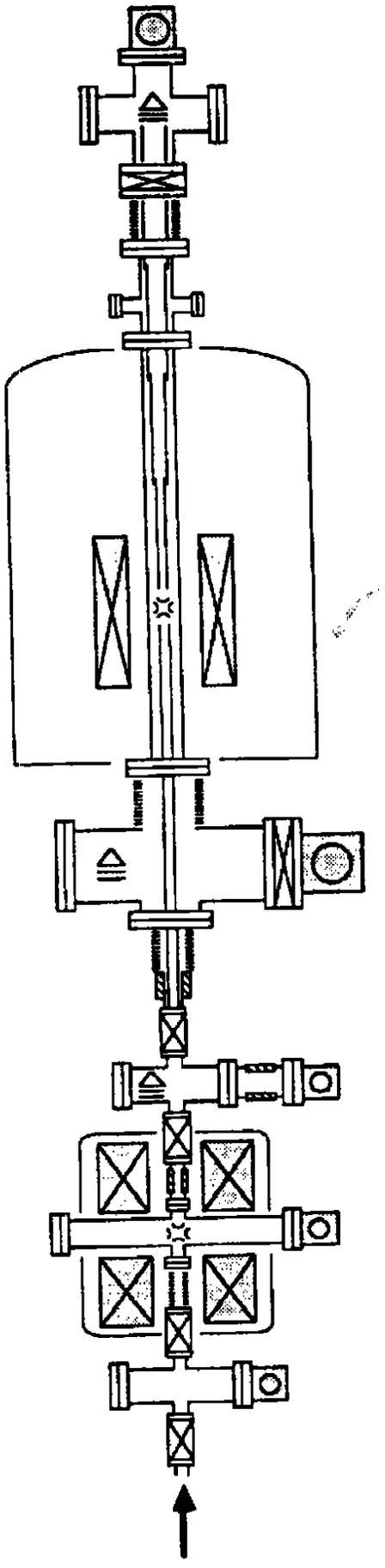
FIGURE CAPTIONS

Fig. 1 Experimental set-up used for direct mass measurements of short-lived isotopes.

Fig. 2 Cyclotron resonance of ^{230}Fr ($T_{1/2}$ = 93 min) obtained in 20 minutes of observation.

Table 1 Half-lives, measured cyclotron frequencies with statistical errors and mass excesses for the studied Fr and Ra isotopes. For the measured mass excess, both the primary (round brackets) and the total error (square brackets) are given. The last two columns present the previous accepted ground-state mass excesses [1] and the result of a new least-squares adjustment of all mass data available. The mass excess of the reference isotope ^{133}Cs results from an adjustment including the Penning trap mass measurements in the Cs isotopic chain [9].

Isotope	$T_{1/2}$	Cyclotron Frequency [Hz]	Measured Mass Excess [keV]	Literature Mass Excess [keV]	Adjusted Mass Excess [keV]
^{133}Cs	stable	684961.640 (2)	reference	-88086 (5)	-88080 (4)
^{209}Fr	50.0 s	435582.671 (60)	-3817 (28) [34]	-3830 (50)	-3811 (27)
^{210}Fr	3.18 min	433507.415 (26)	-3363 (13) [24]	-3400 (50)	-3365 (21)
^{211}Fr	3.10 min	431454.625 (27)	-4177 (14) [24]	-4200 (40)	-4180 (20)
^{212}Fr	20.0 min	429418.055 (36)	-3553 (18) [26]	-3600 (40)	-3565 (21)
^{221}Fr	4.9 min	411896.443 (48)	13267 (25) [32]	13266 (8)	13266 (8)
^{222}Fr	14.2 min	410035.108 (22)	16323 (13) [24]	16380 (40)	16336 (21)
^{226}Ra	1600 y	402764.375 (24)	23652 (14) [25]	23662.7 (2.7)	23662.7 (2.7)
^{230}Ra	93 min	395740.436 (44)	34533 (25) [33]	34660 (360)	34533 (33)



ION DETECTOR

DRIFT SECTION

TRAP 2

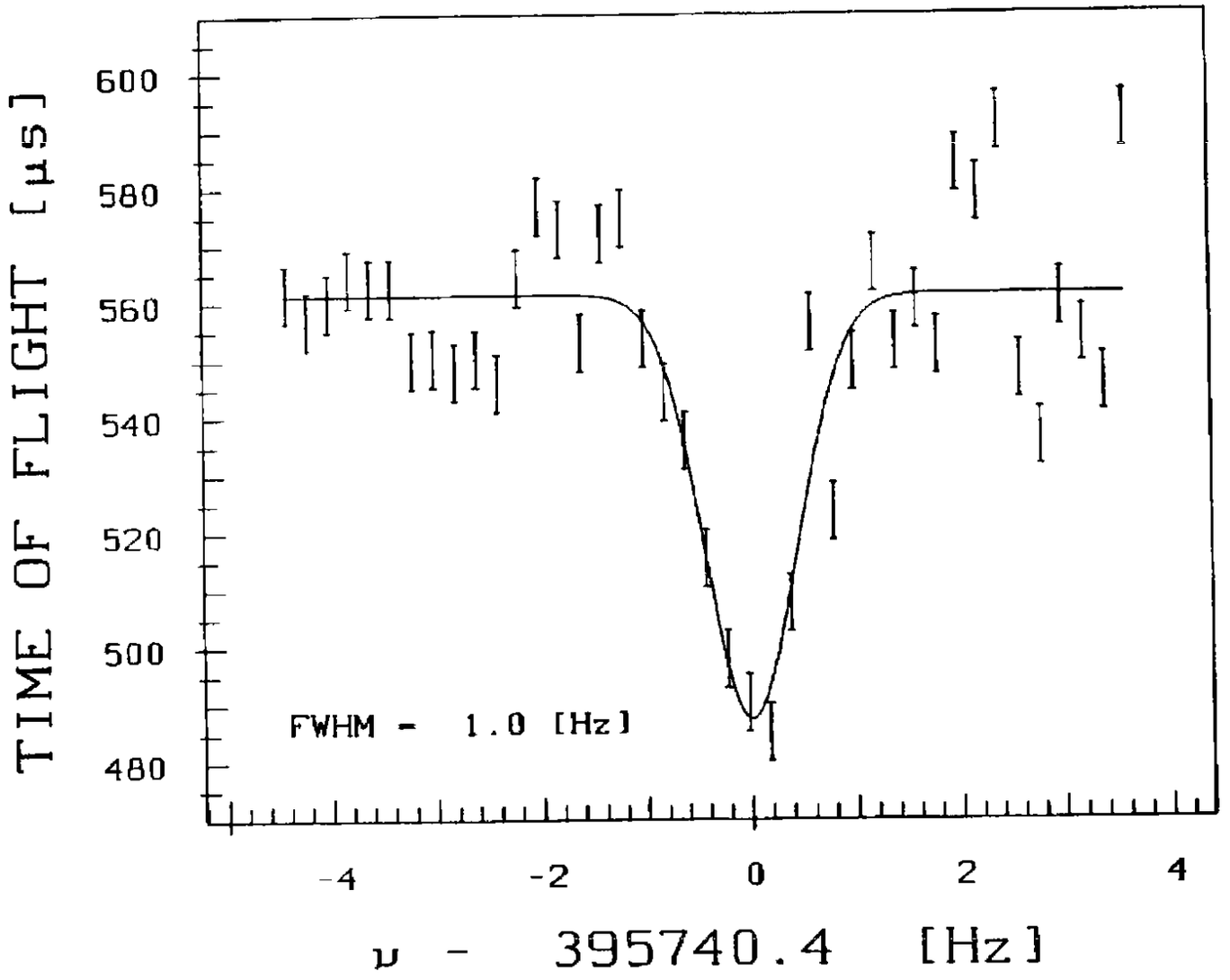
TRANSFER SECTION

TRAP 1

ISOLDE BEAM

Figure 1

Figure 2


TIME OF FLIGHT [μs]
600
580
560
540
520
500
480
FWHM = 1.0 [Hz]
-4
-2
0
2
4
μ - 395740.4 [Hz]