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Resolution of nuclear ground and isomeric states by a Penning trap mass spectrometer

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Ground and isomeric states of a nucleus have been resolved for the first time by mass spectrometry. Measurements on ^{78}Rb^{m,g} and ^{84}Rb^{m,g} were performed using a tandem Penning trap mass spectrometer on-line with the isotope separator ISOLDE/CERN. The effects of ion-ion interaction were investigated for two ion species differing in mass and stored simultaneously in the trap.

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The use of mass spectrometers for the investigation of the masses of unstable nuclei far from stability started in the 1970s at the CERN proton synchrotron and at the on-line isotope separator ISOLDE/CERN which delivers a large number of unstable isotopes of a broad variety of elements. These first direct mass measurements were performed [1,2] with a conventional spectrometer of Mattauch-Herzog type with a resolving power of typically 10^4. Such a resolving power is not sufficient to resolve the mass differences of ions in the nuclear ground or isomeric state or even most isobars. Hence corrections had to be estimated in those cases where both species were delivered simultaneously by the isotope separator. For accurate mass measurements on unstable nuclei it is therefore highly desirable to be able to resolve ground and isomeric states.

The very successful use of the Penning trap as a mass spectrometer has been demonstrated in a number of experiments [3]. The mass of a stored charged particle is determined via its cyclotron frequency in a known magnetic field. The good control of the static electric and magnetic fields in which the particle is trapped allows high accuracies to be achieved. The long confinement time leads to high resolving powers. Mass measurements performed on Rb, Sr, Cs, Ba, Fr, and Ra isotopes [4] have demonstrated that such a spectrometer can be used for the investigation of short-lived isotopes with accuracies of typically 10^{-7}. These measurements were carried out with ISOLTRAP, a tandem Penning trap mass spectrometer at ISOLDE/CERN. The present Rapid Communication reports mass measurements on ^{78}Rb and ^{84}Rb ions, for which ground and isomeric states were resolved for the first time in mass spectrometry.

ISOLTRAP has been described in earlier publications [4–6] and will be discussed here only briefly. The apparatus consists of two main parts: one Penning trap acting as an ion cooler and buncher connected to an ISOLDE beam line and a second high-precision Penning trap for mass determination. The radioactive ions delivered by ISOLDE are collected on a rhenium foil mounted in the lower end cap of the first trap. Then the foil is rotated to face the inside of the trap, where the implanted atoms are released and surface ionized by heating the foil. The ions are captured in the first trap and cooled by collisions with a buffer gas [7]. They are then ejected from the trap and transferred to the high-precision trap where the ion bunch is captured in flight [8]. This highly compensated trap is situated in the homogeneous field of a 6 T superconducting magnet. The ring electrode of the trap is split into four segments allowing the creation of an azimuthal quadrupole rf field. Such a field is used to drive the motion of the stored ions at the sum of the two eigenfrequencies ω_+ and ω_-, which equals the cyclotron frequency $\omega_+ + \omega_- = \omega_c = (q/m)B$ of an ion with charge q and mass m in a magnetic field B. For appropriate initial conditions a significant change in radial energy can be achieved [6]. Determining the radial energy as a function of the applied frequency yields a resonance curve centered at $\nu_c = \omega_c/(2\pi)$ from which the mass of the ions can be determined. The linewidth of the resonance $\Delta\nu_c(\text{FWHM}) \approx 0.9/T_{\text{rf}}$ is to first order given by the Fourier limit of the applied rf field of duration T_{rf}. A time-of-flight method is used to detect the gained radial energy. After the excitation of their motion the ions are gently ejected out of the trap and drift through the inhomogeneous part of the magnetic field to an ion detector. In this field gradient the radial energy of the ions is converted into axial energy. At resonance this results in a reduction of the time of flight from the trap to the detector.

Figure 1(a) shows cyclotron resonances of ^{84}Rb obtained for typically 15 detected ions per cycle. The mean time of flight of the ions from the trap to the detector is plotted as a function of the applied frequency. This measurement was obtained immediately after the collection of the ions. The resonances of ground and isomeric states are clearly separated. The isotope ^{84}Rb has a ground state with a half-life of $T_{1/2}=32.8$ d and an isomeric state with $T_{1/2}=20.3$ min. The cyclotron frequency of ^{84}Rb ions is $\nu_c=1085$ kHz. The solid curve shows the result of a fit of the theoretical resonance curve to the experimental data. The half-width of the resonances is $\Delta\nu_c(\text{FWHM})\approx 1$ Hz as expected from the chosen excitation time of $T_{\text{rf}}=900$ ms. This corresponds to a mass resolution of $\Delta m(\text{FWHM})\approx 75$ keV. For comparison Fig. 1(b) shows a cyclotron resonance which was obtained after several half-lives of the isomer. Hence only the resonance for the ground state is observed.

Figure 2 shows cyclotron resonances for ^{78}Rb. This measurement was performed with 25 detected ions per cycle. ^{78}Rb has a ground state with a half-life of $T_{1/2}=17.7$ min and an isomeric state with $T_{1/2}=5.7$ min. The cyclotron frequency of ^{78}Rb ions is $\nu_c=1168$ kHz. The excitation time was again $T_{\text{rf}}=900$ ms leading to a mass resolution of $\Delta m(\text{FWHM})\approx 70$ keV. Here the mass difference between the two states is comparable to the resolution but both resonances can still be distinguished.

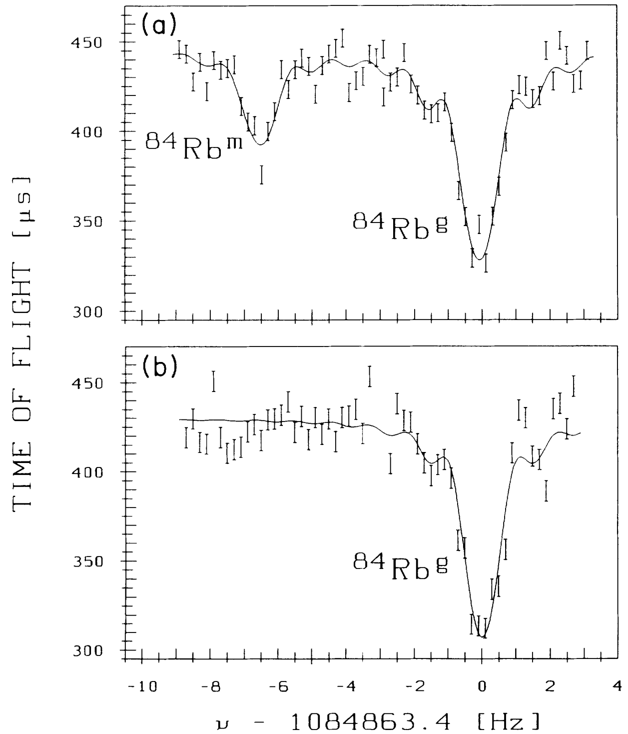

FIG. 1. Cyclotron resonances for ^{84}Rb ions. The measurements were performed (a) shortly after the collection of the ions and (b) with a delay of several half-lives of the isomeric state.

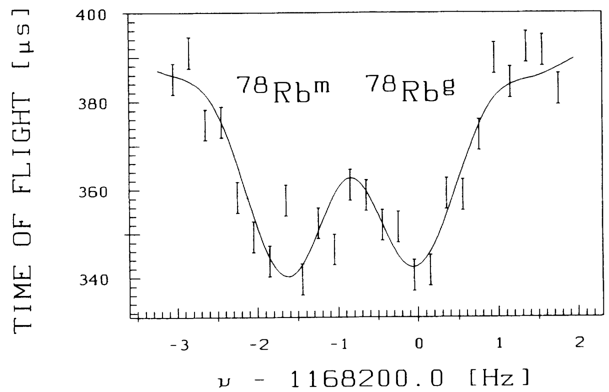

FIG. 2. Cyclotron resonances of the ground and isomeric states of ^{78}Rb.

Typically the measurements are performed with ten to thirty ions loaded in the precision trap of the ISOLTRAP spectrometer. Hence effects of the Coulomb interaction on the ion motion have to be considered. If the simultaneously stored ions have equal mass a driving field will act on the mass center of the cloud and no frequency shifts will be observed. This is different when two ion species differing in mass ($m_1\neq m_2$) are confined at the same time. It was observed experimentally that such contaminations cause frequency shifts. A systematic study of these shifts was performed with stable and unstable isotopes. The results are briefly summarized here.

It was found that the size of the shifts increases with the total number of stored ions. It was also found that the sign of the frequency shifts depends on the magnitude of the difference in the cyclotron frequencies compared to the half-width of the resonances. When the unperturbed resonances cannot be resolved, one single resonance is observed which is narrower than expected from a simple superposition. The position of this resonance corresponds to the center of mass of all of the ions in the trap. However, when the mass differences are large enough for the unperturbed resonances to be separated by more than their widths, the measured resonances are both shifted to lower frequencies. The size of the resonance shift for the ions with mass m_1 was found to be proportional to the number of ions with mass m_2 and vice versa.

A similar behavior was observed for the axial frequencies of two trapped ion species in experiments performed with a Paul trap [9] and qualitatively explained by a model based on two coupled oscillators. A quantitative description of the observed frequency shifts must take into account the coupling of all eigenmodes by the Coulomb interaction.

Up to now no general analytical solution has been found for the equations of motion, neither for the Paul nor the Penning trap. To be sure that the observed shifts of the cyclotron resonance in the Penning trap can be ascribed to Coulomb interaction, a three-dimensional simulation was performed for two confined ions. As in the experiment the ions were excited by an azimuthal quadrupole field and the radial energy that was gained was determined as a function of the applied frequency. The shift of the center of these "resonances" as a function of the mass difference is shown in Fig. 3. A good qualitative agreement between the experimental observations and the results from the simulation is achieved. The frequency shifts are always negative for frequency differences large compared to the half-width whereas for small differences the resonances attract each other. A quantitative description was out of the scope of the simulations due to the available CPU time. The rf excitation time and Coulomb interaction had to be scaled by several orders of magnitude to perform the calculations in a finite time.

The measurements shown in Figs. 1 and 2 were performed with ions both in the ground and isomeric states. In order to examine the effect of Coulomb interaction cyclotron resonances were measured for different time intervals T_d between collection of the ions on the foil and start of the measurement. In this way the ratio of the number of ions in the isomeric state to that in the ground state was varied from an initial value of $N_m/N_g \approx 1$ for ^{84}Rb and $N_m/N_g \approx 2$ for ^{78}Rb down to practically zero.

Such measurements were made on ^{84}Rb for three different numbers of stored ions. Figures 4(a) and 4(b) show the cyclotron frequencies that were obtained for about 15 detected ions as functions of the delay T_d. No frequency shifts are observable within the statistical uncertainty. Hence effects of ion-ion interaction can be neglected and the mean values $\nu_{c,m}$ and $\nu_{c,g}$ of the centers of both resonances are the unperturbed cyclotron frequencies with $\Delta\nu = \nu_{c,m} - \nu_{c,g} = 6.4(1)$ Hz. This corresponds to a mass difference of $\Delta m = 464(7)$ keV which is in perfect agreement with the more precise value of $\Delta m = 463.62(10)$ keV known from nuclear spectroscopy [10].

The situation changes drastically if the number of ions in the trap is increased. Figures 4(c) and 4(d) show the frequency shifts for 25 and 70 detected ions as functions of the delay T_d (for 70 ions the cyclotron resonance of the isomeric states was outside the scanned frequency re-

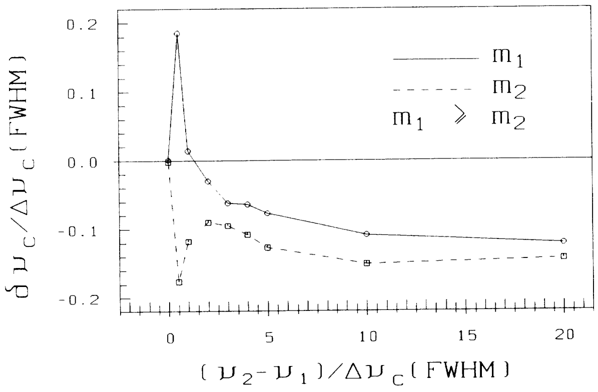

FIG. 3. Frequency shifts $\delta\nu_c$ of the cyclotron resonances for two simultaneously trapped ions with different masses $(m_1 \geq m_2)$ as obtained from a numerical simulation of the ion motion. The shifts are plotted versus the difference of the cyclotron frequencies $\nu_2 - \nu_1$ of both ions. Both quantities are normalized to the theoretical half-width $\Delta\nu_c$(FWHM) of the cyclotron resonances.

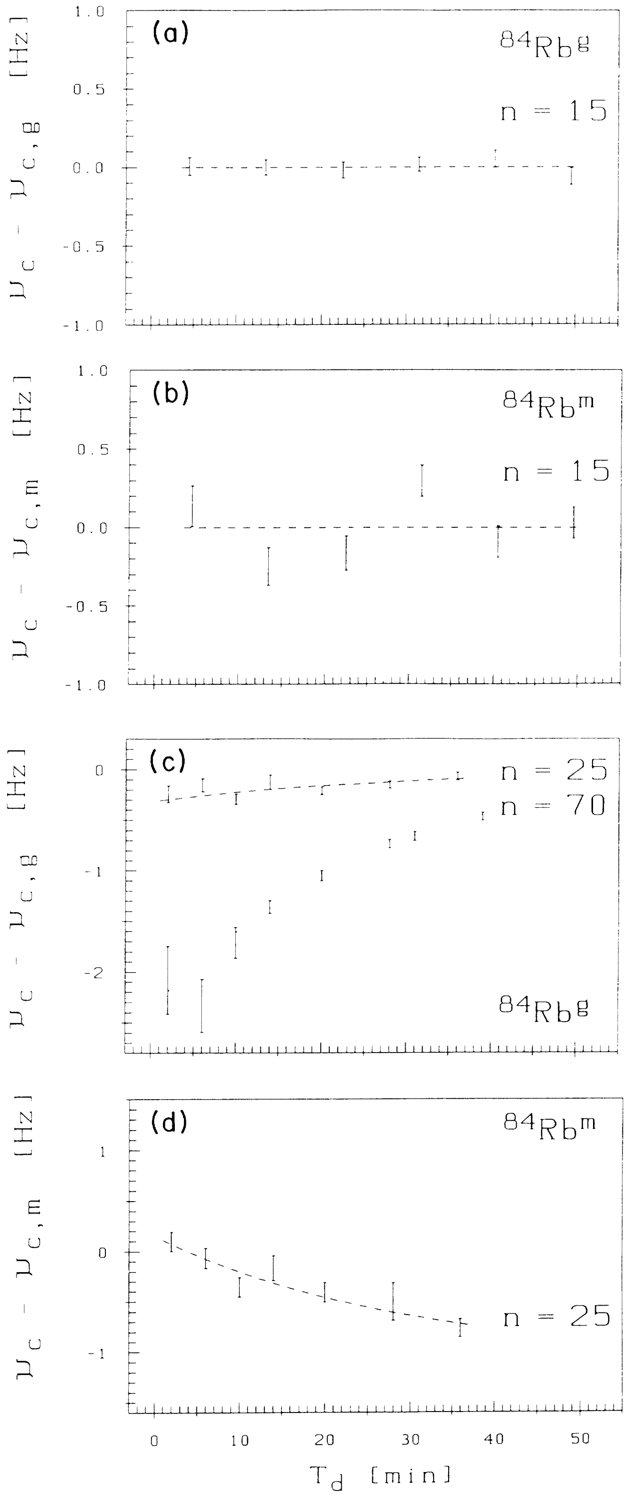

FIG. 4. Cyclotron frequencies of ground and isomeric states of ^{84}Rb as a function of the delay T_d between collection of the ions and measurement. Series of measurements were performed with about 15, 20, and 70 detected ions as indicated. The dashed horizontal lines in (a) and (b) correspond to the unperturbed cyclotron frequencies $\nu_{c,g}$ and $\nu_{c,m}$ obtained from the mean value of the measurements. The dashed and dotted curves in (c) and (d) represent a fit (see text) describing the effect of ion-ion interaction.

gion). The size of the shift increases with the total number of confined ions. The dashed and dotted curves show the result of a fit with the ansatz that the frequency shift for the ground state is proportional to the number of ions in the isomeric state and vice versa. The shifts are always negative within the statistical accuracy and decrease with T_d for the ground state and increase for the isomeric state.

In the case of ^{78}Rb all measurements were performed with 25 detected ions. Again the cyclotron frequencies for both states were examined as a function of the delay between collection and measurement. Due to the limited beam time available only a few measurements could be performed. It was not possible to follow accurately the frequency shifts as a function of T_d. Therefore only limits on the mass difference of ground and isomeric states can be given with $71\ \mathrm{keV} < \Delta m < 132\ \mathrm{keV}$. The result agrees with the value of 111.2 keV recently determined by γ spectroscopy [11].

A natural way to eliminate frequency shifts due to Coulomb interaction is to confine only one species in the trap. In principle this can be achieved by removing unwanted ions by selectively driving them at their eigenfrequency ω_+. This procedure was found to work well in cases where both masses are resolved. It was routinely applied to isotopes and isobars with mass differences down to about 1 MeV. It should therefore also be possible in cases like ^{84}Rb to remove selectively ions in the ground or the isomeric state.

The mass measurements with the ISOLTRAP spectrometer presented in this Rapid Communication showed for the first time the resolution of isomeric and ground states of a nucleus. It was shown for the cases of the radiative isotopes ^{78}Rb and ^{84}Rb that it is possible to determine directly the energy of nuclear levels. Beside the fascinating point that nuclear spectroscopy can be performed by "weighing," the resolution of isomeric and ground states has some important advantages. For the mass measurements on unstable nuclei it allows a determination of ground state masses with high accuracy since the resonances can be resolved and no corrections have to be estimated for the isomeric contamination. The effects of Coulomb interaction in the trap can be treated if as few ions as possible are stored in the trap and the resonances are recorded as a function of the delay time between collection and measurement. In this way the unperturbed cyclotron frequencies of ground and isomeric states can be obtained with high accuracy as demonstrated in the case of ^{84}Rb. If this is not feasible because of short half-lives or limited statistics limits for the excitation energy can still be obtained as shown for ^{78}Rb.

A future application could be to operate the trap as an isomer separator. After loading the trap with a number of ions those in the ground state can be removed by driving their cyclotron motion. Then the ions in the isomeric state can be ejected and delivered to other experiments.

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