

Separated Oscillatory Fields for High-Precision Penning Trap Mass Spectrometry

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Ramsey's method of separated oscillatory fields is applied to the excitation of the cyclotron motion of short-lived ions in a Penning trap to improve the precision of their measured mass. The theoretical description of the extracted ion-cyclotron-resonance line shape is derived out and its correctness demonstrated experimentally by measuring the mass of the short-lived ^{38}Ca nuclide with an uncertainty of $1.6 \cdot 10^{-8}$ using the ISOLTRAP Penning trap mass spectrometer at CERN. The mass value of the superallowed beta-emitter ^{38}Ca is an important contribution for testing the conserved-vector-current hypothesis of the electroweak interaction. It is shown that the Ramsey method applied to mass measurements yields a statistical uncertainty similar to that obtained by the conventional technique ten times faster.

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In 1989 the Nobel prize in physics was awarded to N.F. Ramsey [1] in recognition of his molecular beam resonance method with spatially separated oscillatory fields proposed 40 years earlier [2, 3]. In 1992 G. Bollen *et al.* [4] demonstrated the use of time-separated oscillatory fields for the excitation of the cyclotron motion of an ion confined in the Penning trap mass spectrometer ISOLTRAP. This and experiments performed later at SMILETRAP [5] showed that the method may improve the precision of mass measurements with Penning traps - on the condition that the shape of the detected ion-cyclotron-resonance could be put on a sound theoretical basis. In the case of exotic nuclides, the extremely low production rates make precision measurements challenging because of the time required to accumulate the necessary statistics. The Ramsey method would therefore allow accumulation of sufficient statistics in a much shorter time, which is of prime importance for on-line measurements.

In this Letter, we introduce the correct theoretical description of the application of the Ramsey method to ions stored in a Penning trap. We also demonstrate its validity for the first time with an on-line mass measurement of ^{38}Ca ($T_{1/2} = 440(8)$ ms). This short-lived nuclide is used for testing the conserved-vector-current hypothesis (CVC) of the Standard Model of fundamental interac-

tions, which postulates a vector-current part of the weak interaction unaffected by the strong interaction. Thus, the decay strength of all superallowed $0^+ \rightarrow 0^+$ β -decays is independent of the nuclei except for theoretical corrections [6, 7]. Here, a relative mass precision of 10^{-8} is required.

Comparisons with the conventional excitation scheme [8, 9] show that the line width of the resonance is reduced by almost a factor of two and the statistical uncertainty of the extracted resonance frequency is more than a factor of three smaller. It is shown that the Ramsey method allows a measurement with the same statistical uncertainty, but ten times faster. This is an enormous improvement in the very active field of on-line high-precision mass measurements with Penning traps at radioactive beam facilities [10].

The prerequisite for the successful implementation of the Ramsey method to stored ions is the detailed understanding of the observed time-of-flight cyclotron resonance curves using time separated oscillatory fields. While here only the most important parts of the theory and its experimental confirmation is given, a detailed presentation will be published elsewhere.

In a Penning trap an ion with a charge-to-mass ratio q/m is stored in a strong homogeneous magnetic field B_0 combined with a weak electrostatic quadrupole field. The mass measurement is performed via the determination of the cyclotron frequency $\nu_c = qB_0/(2\pi m)$. For the basic theory of the ion motion in Penning traps we refer to the review article of Brown and Gabrielse [11]. Our

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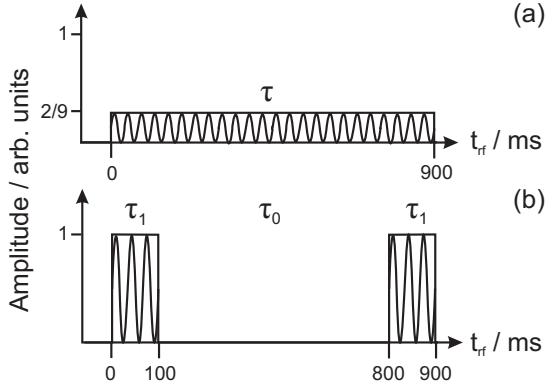


FIG. 1: Excitation schemes: (a) conventional excitation with a continuous rf pulse (here $\tau=900$ ms), (b) excitation with two 100-ms Ramsey pulses τ_1 interrupted by a 700 ms waiting period τ_0 .

considerations here focus on the process of conversion of the magnetron mode into the cyclotron mode under the influence of the quadrupolar rf-field in the Penning trap, extending previous studies by one of us [12] of this process in a quantum mechanical framework. In this work the effective interaction of a quadrupolar field with driving frequency $\nu_d \approx \nu_c$ with a trapped ion is described in terms of the annihilation (a) and creation (a^\dagger) operators of the magnetron (-) and cyclotron (+) oscillators, the phase of the quadrupole field $\phi_d(t) = \omega_d t + \chi_d$ with $\omega_d = 2\pi\nu_d$, and a coupling constant g that is proportional to the amplitude of the quadrupolar field:

$$H_1(t) = \hbar g \left(e^{-i\phi_d(t)} a_+^\dagger(t) a_-(t) + \text{h.c.} \right), \quad (1)$$

“h.c.” denotes the Hermitian conjugate of the first term. Addition of this interaction to the Hamiltonian of an ion in an ideal Penning trap yields a model system for which Heisenberg’s equations of motion can be solved rigorously. The interaction (1) has the important property that the total number of quanta in the magnetron and cyclotron oscillators is conserved, $N_{\text{tot}} = N_+(t) + N_-(t) = a_+^\dagger(t)a_+(t) + a_-^\dagger(t)a_-(t) = 2T_0$. The conversion process can be studied in terms of the “Bloch vector operator” $\mathbf{T} = T_1\mathbf{e}_1 + T_2\mathbf{e}_2 + T_3\mathbf{e}_3$, introduced in [12], with

$$T_1(t) = \frac{1}{2} \left(a_+^\dagger(t) a_-(t) + a_-^\dagger(t) a_+(t) \right), \quad (2)$$

$$T_2(t) = \frac{1}{2i} \left(a_+^\dagger(t) a_-(t) - a_-^\dagger(t) a_+(t) \right), \quad (3)$$

$$T_3(t) = \frac{1}{2} \left(a_+^\dagger(t) a_+(t) - a_-^\dagger(t) a_-(t) \right). \quad (4)$$

The components obey the same commutation rules as an angular momentum, $[T_j, T_k] = i\epsilon_{jkl}T_l$ ($j, k, l = 1, 2, 3$) and $T_1^2 + T_2^2 + T_3^2 = T_0(T_0 + 1)$. The expectation value $\langle \mathbf{T} \rangle$ is a real 3-dimensional vector of constant length that describes a precessional motion on a “Bloch sphere” during the quadrupolar excitation. The model Hamiltonian

expressed in terms of \mathbf{T}

$$H(t) = \hbar\omega_1(T_0 + \frac{1}{2}) + \hbar\omega_c T_3(t) + \hbar \cdot 2g (\cos \phi_d(t) \cdot T_1(t) + \sin \phi_d(t) \cdot T_2(t)) \quad (5)$$

governs the time development of the Bloch vector operator. Compare this result to the Hamiltonian H_{mag} that describes the precession of the nuclear spin \mathbf{I} in magnetic resonance experiments, $\mathbf{I} = I_1\mathbf{e}_1 + I_2\mathbf{e}_2 + I_3\mathbf{e}_3$,

$$H_{\text{mag}} = -\vec{\mu} \cdot \mathbf{B} = -\hbar\omega_L I_3 - \hbar\gamma B_1 (\cos \omega t \cdot I_1 - \sin \omega t \cdot I_2), \quad (6)$$

where $\vec{\mu} = \gamma\mathbf{I}$ is the nuclear magnetic moment, γ the gyromagnetic ratio, and ω_L the Larmor frequency. Both Hamiltonians govern the time development of a vector operator, thus exhibiting a dynamical similarity between nuclear magnetic resonance on the one hand and ion motion in a Penning trap with quadrupole excitation on the other hand. This structural analogy provides deeper insight why Ramsey’s idea of using separated oscillating fields can be successfully applied also to Penning trap physics.

Initially ($t = 0$) the ions are prepared in a pure magnetron mode ($N_{\text{tot}} = N_-(0) = -2\langle T_3(0) \rangle$), with the objective to convert the magnetron motion as completely as possible into cyclotron motion and thus to bring the radial energy to its maximum. For the conventional excitation scheme with a single pulse (see Fig. 1a) of duration τ and constant amplitude the expectation value for the percentage of converted quanta is obtained as

$$F_1(\omega_R, \tau, g) = \frac{N_+(\tau)}{N_{\text{tot}}} = \frac{4g^2}{\omega_R^2} \cdot \sin^2(\omega_R\tau/2), \quad (7)$$

where $\omega_R = \sqrt{(2g)^2 + \delta^2}$ is the analog of the Rabi frequency and $\delta = \omega_d - \omega_c$ the detuning of the quadrupolar field. The “conversion time”, *i.e.* the time required for complete conversion exactly on resonance, is seen to be $\tau_c = \pi/(2g)$.

If two pulses of quadrupolar radiation, each of duration τ_1 , and separated by a waiting period τ_0 (see Fig. 1b), are used for the excitation, the expectation value for the percentage of converted quanta becomes

$$F_2(\delta, \tau_0, \tau_1, g) = \frac{16g^2}{\omega_R^2} \cdot \sin^2\left(\frac{\omega_R\tau_1}{2}\right) \cdot \left[\cos\frac{\delta\tau_0}{2} \cdot \cos\frac{\omega_R\tau_1}{2} - \frac{\delta}{\omega_R} \cdot \sin\frac{\delta\tau_0}{2} \cdot \sin\frac{\omega_R\tau_1}{2} \right]^2. \quad (8)$$

In order to compare experimentally the different excitation methods the total duration of the excitation cycle was chosen equal for both, namely 900 ms. The calculated energy conversion is shown in Fig. 2 as a function of the frequency detuning $\delta' = \delta/(2\pi)$ and the waiting time τ_0 . The conventional single pulse excitation appears here as the limiting case with waiting period $\tau_0 = 0$. The experimental resonance spectra for a conventional scheme

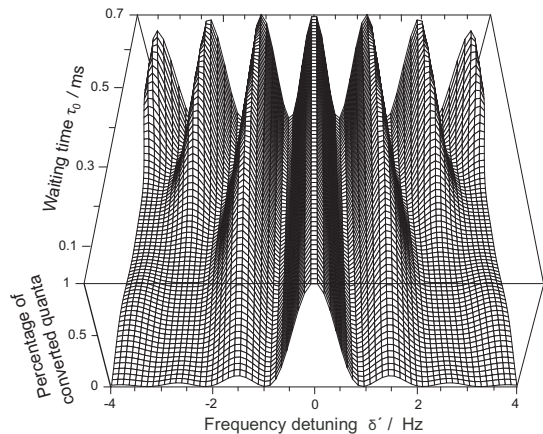


FIG. 2: Percentage of converted quanta versus the frequency detuning $\delta' = \delta/(2\pi)$ and versus the waiting time τ_0 of the two-pulse Ramsey scheme of Fig. 1b. Each pulse has a duration of $\tau_1 = \tau_c/2$ to obtain complete conversion at resonance, the total duration of the excitation cycle is fixed to 900 ms.

as well as for a scheme with two excitation periods of $\tau_1 = 100$ ms and $\tau_0 = 700$ ms are shown in Fig. 3. In the latter case the sidebands are very pronounced and the full-width-at-half-maximum (FWHM) is considerably reduced. The solid line represents the fit of the theoretical line shape to the data points.

The mass measurements on the short-lived nuclide ^{38}Ca using the Ramsey method were performed using the Penning trap mass spectrometer ISOLTRAP [13] installed at the on-line mass separator facility ISOLDE/CERN. The calcium isotope was produced by bombarding a heated titanium metal foil target with 1.4-GeV protons from the CERN proton-synchrotron-booster accelerator. A hot tungsten surface was used to ionize the released atoms. In order to suppress isobaric contaminations by $^{38}\text{K}^+$ ions, a CF_4 leak was added and the ions of interest were delivered to ISOLTRAP in form of the molecular sideband $^{38}\text{Ca}^{19}\text{F}^+$. Ions extracted from the source were accelerated to 30 keV, mass separated in ISOLDE's high-resolution mass separator, and injected into the first part of the ISOLTRAP apparatus, a gas-filled linear radiofrequency quadrupole ion trap for accumulation, cooling and bunching of the ion beam [14]. From here the ions were transferred at lower energy as short bunches to a cylindrical Penning trap for further buffer-gas cooling and isobaric purification [15]. The actual mass measurement was performed in a second, hyperboloidal Penning trap [16] via the cyclotron frequency determination.

The time-of-flight ion cyclotron resonance detection technique [8] relies on the coupling of the ion's orbital magnetic moment to the magnetic field gradient after excitation of the ion motion with rf-fields and axial ejection from the trap into a time-of-flight section. The sum frequency of the modified cyclotron mode and the magnetron mode $\nu_c = \nu_+ + \nu_-$ is probed by the measurement of the radial energy [17, 18]. A typical time-of-flight

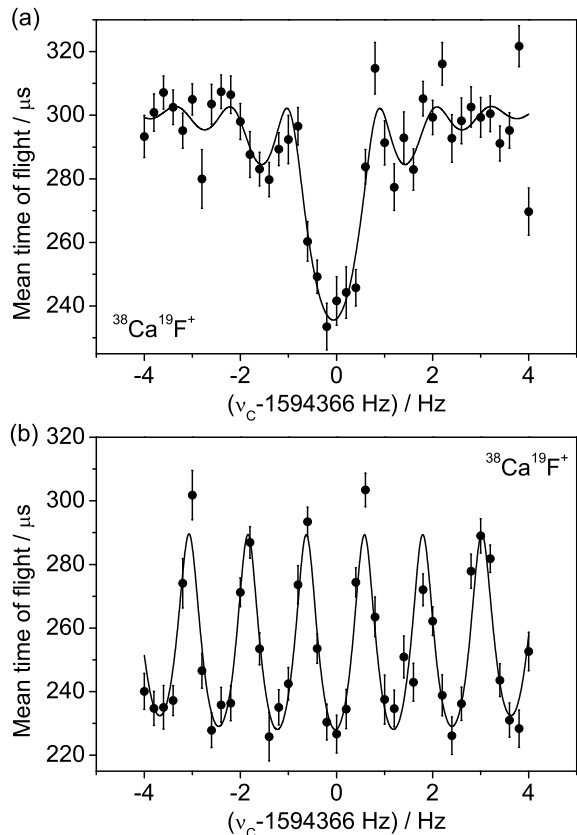


FIG. 3: Time-of-flight ion-cyclotron-resonance spectra of $^{38}\text{Ca}^{19}\text{F}^+$ with the conventional quadrupolar excitation (a) and with a two-pulse Ramsey scheme with two 100 ms duration excitation periods interrupted by a 700 ms waiting period (b). The solid curves are fits of the theoretical line shapes to the data. The center frequency, where the detuning is 0, is at 1594365.92 Hz.

cyclotron resonance of $^{38}\text{Ca}^{19}\text{F}^+$ ions using the conventional excitation technique with one continuous rf pulse (Fig. 1a) is shown in Fig. 3a. The mass of the ion of interest is obtained from a comparison of its cyclotron frequency with the one of a reference ion with well-known mass, here $^{39}\text{K}^+$.

The uncertainty of the center frequency depends mainly on three parameters. First, the relative uncertainty is inversely proportional to the square root of the number of recorded ions. Second, it is proportional to the linewidth of the resonance and third it depends on the shape of the resonance and its sidebands. The stronger the sidebands are, the more precisely the center frequency can be determined. For the latter two aspects Ramsey's excitation method with separated oscillatory fields is superior to the conventional method.

Altogether eight resonances of $^{38}\text{Ca}^{19}\text{F}^+$, each with a scan width of ± 4 Hz around the expected cyclotron fre-

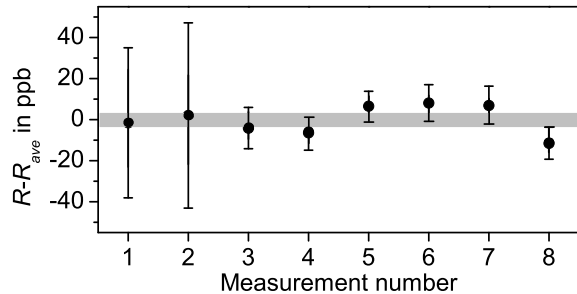


FIG. 4: Difference of the measured cyclotron frequency ratios R and their average value R_{ave} between $^{38}\text{Ca}^{19}\text{F}^+$ and $^{39}\text{K}^+$. The error bars are the statistical uncertainties. Each data point contains the same number of recorded ions in the resonance.

quency were measured with the Penning trap mass spectrometer ISOLTRAP. The first two employed the conventional excitation scheme with 900 ms, the others used the Ramsey-type scheme of two 100-ms excitation pulses and one 700-ms waiting period (Fig 1). Thus, the total excitation period of both schemes was 900 ms and they can be easily compared. Taking only into account the statistical error, the mass excess can be determined to within a 1.03 keV uncertainty for the two resonances with conventional excitation. The uncertainty of the mass excess obtained from two Ramsey type measurements under identical experimental conditions is here drastically reduced to 0.25 keV. All six measurements lead to an overall statistical uncertainty of the mass excess of only 0.12 keV. After consideration of systematic uncertainties of ISOLTRAP [19] we obtain finally a frequency ratio R between the ion of interest and the reference ion $\nu_{ref}/\nu_{ion} = 1.462\,257\,6087(50)(229)$, where the first error derives from the statistics and the second one denotes the systematic error of the measurement. Together with the mass excess of ^{19}F $ME(^{19}\text{F}) = -1487.39(07)$ keV [20] we obtain a mass excess $ME(^{38}\text{Ca}) = -22058.01(65)$ keV, which is in agreement with the previously accepted value [20] as well as with the recent value $ME(^{38}\text{Ca}) = -22058.53(28)$ keV [21], measured by the new Penning trap

facility at MSU and being a factor of two more precise.

The 12 best known superallowed Fermi-type beta decays ranging from ^{10}C to ^{74}Rb yield an average Ft value of $Ft = 3072.7(0.8)\text{s}$ [7], which confirms the CVC hypothesis at a level of $3 \cdot 10^{-4}$. Further mass measurements on superallowed β -emitters for an even more stringent CVC test have been performed recently [21, 22, 23], demonstrating the continuous interest in this weak interaction and Standard Model test. ^{38}Ca is a new candidate for testing the CVC hypothesis. Its large isospin symmetry-breaking correction of 0.73(5)% makes it a proper candidate for testing the validity of the calculations. Until recently the Ft value had a relative uncertainty of $4 \cdot 10^{-3}$, limited by the uncertainty of the ground-state mass, the uncertainty of the $0^+ \rightarrow 0^+$ branching ratio, and the accuracy of the half-life ($T_{1/2} = 440(8)\text{ms}$), which was measured and improved in parallel to our experiment at ISOLDE. Here, the data analysis is presently under way.

Obviously, our accuracy is not limited by the statistical uncertainty, but by the systematic error, which comes from magnetic field fluctuations and mass dependent error of the experimental setup. In the future these systematic errors can be reduced by a stabilization of the magnetic field and a local determination of mass dependent deviations with carbon clusters [19].

In conclusion, the method of separated oscillatory fields has been for the first time theoretically derived and applied to the mass measurement of radioactive ions confined in a Penning trap. It allows a considerable reduction in data-taking time by its improvement of precision with much less statistics. Thus, it is ideally suited for radionuclides, especially of half-lives below 100 ms and production rates of only a few 100 ions/s where the uncertainty is often limited by the statistical error, as in the case of ^{32}Ar ($T_{1/2} = 98\text{ms}$) [24] and ^{74}Rb ($T_{1/2} = 65\text{ms}$) [25].

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