Phase-Sensitive Cyclotron Frequency Measurements at Ultralow Energies

Sven Sturm, Anke Wagner, Birgit Schabinger, and Klaus Blaum
Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany
(Received 26 January 2011; published 28 September 2011)

A novel technique for a direct and coherent measurement of the modified cyclotron frequency of an ion in a Penning trap at energies close to the thermal cooling limit is presented. This allows a rapid and both precise and accurate determination of the free-space cyclotron frequency in real Penning traps despite the existence of electric and magnetic field imperfections and relativistic shifts. The demonstrated performance paves the way for considerably improved bound-state g-factor measurements on the 10 ppt level and mass measurements in the 1 ppt range and possibly below.

DOI: 10.1103/PhysRevLett.107.143003 PACS numbers: 37.10.Ty, 06.20.Jr, 07.75.+h, 82.80.Nj

A single ion confined in a Penning trap at low energy allows determining a multitude of properties of atomic systems and fundamental constants as well as stringent tests of fundamental theories and symmetries [1]. All relevant experiments include the measurement of the free-space cyclotron frequency of the trapped ion that can be determined by excitation of the different ion oscillation modes by an external radio frequency field. The limiting factors in the current state-of-the-art experiments are the temporal stability of the magnetic and electrostatic trapping fields and systematic shifts arising from residual electric and magnetic field imperfections and ultimately from a relativistic mass shift [2]. Measurement schemes like the coupling of motional modes [3] determine the modified cyclotron frequency as an offset to the axial frequency and allow measurements at thermal temperatures but suffer from long integration times and high sensitivity to electric field drifts [4]. The development of phase-sensitive detection techniques for the modified cyclotron frequency like the pulse and phase technique [5] allows rapid measurements of the eigenfrequencies in a time that is determined by the Cramér-Rao bounds [6] for the signal-to-noise ratio (SNR) of the detector. These measurements, however, require a large cyclotron energy, causing systematic shifts in the eigenfrequencies that have to be corrected for and that tend to limit the precision of the measurement due to the limited reproducibility of the excited mode amplitudes to about 10^{-10} for highly charged ions. A reduction of the uncertainty by 1 order of magnitude or more would have a significant impact in different fields of physics. The mass difference between ^{3}H and ^{3}He, e.g., as an input parameter for neutrino mass determination from ^{3}H beta decay, requires a precision of the individual cyclotron frequencies below 10^{-10} to be significant for ongoing experiments [7]. For g-factor measurements of the electron bound in hydrogenlike ions as a test of bound-state quantum electrodynamics calculations (see, e.g., [8]), the mass of the ion under investigation has to be known to a few parts in 10^{11} in order to match the present precision of the corresponding calculations [9]. This would also lead to a more precise value of the electron mass in atomic units from a comparison of experimental and theoretical g-factor results [10]. A more precise electron mass, in turn, would improve the value of the fine-structure constant as determined from photon recoil measurements [11]. This Letter reports on the development of a novel technique that allows a phase-sensitive cyclotron frequency measurement at considerably lower temperatures, making energy-dependent systematic frequency shifts in most cases negligible and allowing for cyclotron frequency measurements at the uncertainty level of 1 ppt.

The experimental setup used for the studies reported here consists of a cylindrical Penning trap as part of a triple trap system [12]. A single ion is suspended in a 3.76 T magnetic field in a trap of 7 mm inner diameter. The complete setup is cooled down to 4.2 K by liquid helium. A superconducting resonator with high impedance together with an amplifier with extremely low input related noise of less than 600 $\mathrm{pV}/\sqrt{\mathrm{Hz}}$ is connected to a correction electrode and the adjacent end cap and allows the detection of the current induced by the ion's oscillation as shown in Fig. 1. By applying proper voltage ratios to the electrodes, a quadrupole potential is created near the center of the trap. Measurement of all three eigenfrequencies in the trap and application of the invariance theorem [13]

$$\nu_c = \sqrt{\nu_+^2 + \nu_-^2 + \nu_z^2} \tag{1}$$

enables precise determinations of the free cyclotron frequency. Because of the typical hierarchy of the modified cyclotron, axial, and magnetron frequencies $\nu_+ \gg \nu_z \gg \nu_-$, the accuracy attained in ν_c depends mostly on ν_+. Since magnetic and electric field drifts generally tend to increase with time, it is desirable to perform the measurement as fast as possible. This can be provided by the measurement of the integrated phase of a coherently excited ion. The initially resistively cooled ion is dipolar excited by a radio frequency field with a fixed phase, resulting in a well-defined cyclotron amplitude and phase. After allowing the cyclotron phase to evolve freely for a

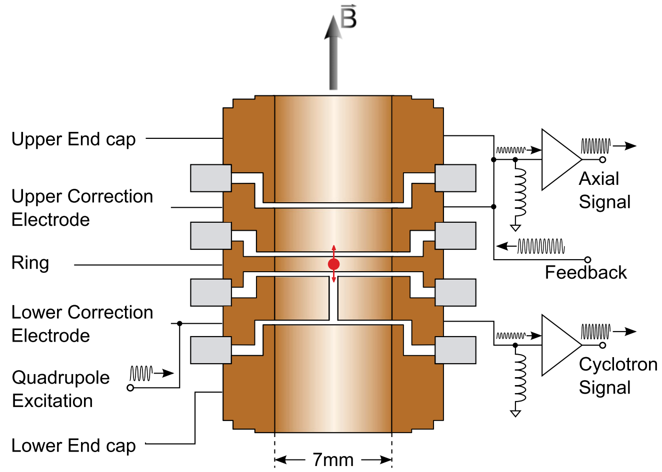

FIG. 1 (color online). Orthogonal and compensated cylindrical Penning trap with detection electronics and excitation ports. Two superconducting tank circuits allow for cooling of all eigenmodes. For details, see the text.

given period, the axial and cyclotron modes are coupled by a radio frequency field and the integrated phase is detected (Fig. 2). After adding the appropriate multiples of 2π, the slope of the unwrapped phase evolution represents the sought-after modified cyclotron frequency. The precision attained will now depend on the SNR of the detection system. The SNR can be increased by applying negative electronic feedback to the ion during detection in order to increase the possible observation time or even use a self-excited oscillator [14] that is seeded with the excited ion axial frequency and thus maintains the axial phase and amplitude during the measurement. All these techniques depend on an axial amplitude well above the thermal limit at the beginning of the observation time. The technique used to date suffers from the premise that the axial amplitude reached after coupling is connected to the cyclotron amplitude [15], leading to the need for a macroscopic amplitude, in our case 30 μm, of the cyclotron motion during the measurement. This amplitude causes both systematic shifts through the residual electric and magnetic field errors and ultimately by virtue of the relativistic mass increase. These lowest order shifts can be quantized as a function of the mode radii:

$$\frac{\delta\nu_+}{\nu_+} = \frac{3}{4}\frac{c_4}{d^2}\frac{\nu_-}{\nu_c}\left(-\frac{r_+^2}{2} + r_z^2 - r_-^2\right) \quad \text{(electrostatic)}$$
$$+ \frac{B_2}{2B_0}(-r_+^2 + r_z^2 - r_-^2) \quad \text{(magnetic)}$$
$$- \frac{2\pi^2\nu_c^2}{c^2} r_+^2 \quad \text{(relativistic).} \tag{2}$$

Here, B_0 and B_2 denote the magnetic field coefficients of lowest and second order, respectively, d is the trap-size parameter, and c_4 is the electrostatic field coefficient of fourth order [2]. For highly charged ions in a well-compensated trap, the relativistic mass increase accounts for the largest energy-dependent systematic error, leading to unacceptable shifts when applying the pulse and phase technique. This limitation results from the finite temperature of the detection circuit that needs a macroscopic mode amplitude in order to overcome the Johnson noise of the tank circuit. The method proposed within this Letter makes the mode amplitude during the phase evolution time essentially independent of the detection amplitude, allowing a very small cyclotron amplitude during the measurement period that becomes amplified just for detection, and thus eliminates to a large extent the shifts of the observed frequencies. For this purpose, a method is required to increase the mode energy while conserving the phase information in a deterministic way without explicit detection of the motion, which would not be possible for the small amplitudes used. The mode coupling at the upper sideband at $\nu_+ + \nu_z$ can provide this, as we will show. The ion oscillation in the presence of a radio frequency coupling field of quadrupolar shape in the z, ρ plane satisfies the equations of motion [5]

$$\ddot{z} = -\omega_z^2 z + \mathrm{Re}\left(\frac{qU_{\mathrm{rf}}}{d_q^2 m} e^{\imath\omega_{\mathrm{rf}} t}\right)x, \tag{3a}$$

$$\ddot{u} = \frac{\omega_z^2}{2} u + \imath\omega_c \dot{u} + \mathrm{Re}\left(\frac{qU_{\mathrm{rf}}}{d_q^2 m} e^{\imath\omega_{\mathrm{rf}} t}\right)z, \tag{3b}$$

where $u \equiv x + \imath y$, d_q is the typical electrode distance for the quadrupole excitation field, and $U_{\mathrm{rf}} = |U_{\mathrm{rf}}|e^{\imath\varphi_{\mathrm{rf}}}$ denotes the complex radio frequency excitation signal. The coupling field adds a driving force proportional to the radial displacement which depends on the phase of the radial motion. It is readily possible to guess the solution of the equations of motion:

$$z(t) = \mathrm{Re}[\zeta(t)e^{\imath\omega_z t}], \tag{4a}$$

$$u(t) = \kappa_+(t)e^{\imath\omega_+ t} + \kappa_-(t)e^{\imath\omega_- t}. \tag{4b}$$

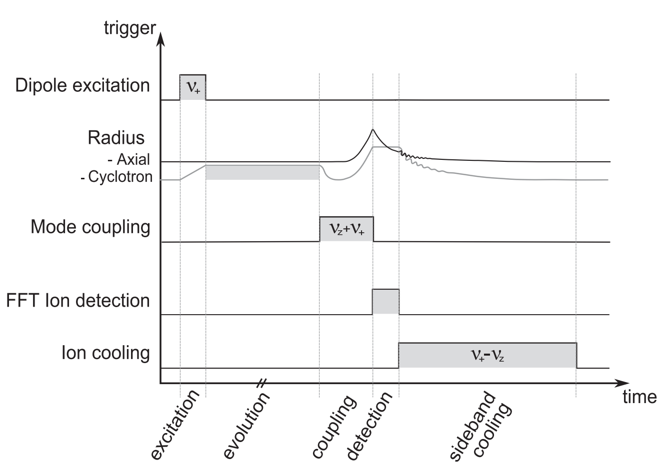

FIG. 2. Excitation pulses scheme for the advanced integrated phase measurement method. For details, see the text.

A solution for almost resonant excitation frequencies $\omega_{\rm rf} \simeq \omega_+ + \omega_z$ is sufficient here. Inserting this into Eq. (3a) and neglecting nonsecular terms at the sum frequency (rotating wave approximation) yields by comparison of coefficients

$$\dot{\zeta} = \frac{qU_{\rm rf}}{4md_q^2\imath\omega_z}\kappa_+^* = \frac{\Gamma}{\omega_z}\kappa_+^*, \quad (5a)$$

$$\dot{\kappa}_+ = \frac{qU_{\rm rf}}{4md_q^2\imath(\omega_+ - \omega_-)}\zeta^* = \frac{\Gamma}{\omega_+ - \omega_-}\zeta^*, \quad (5b)$$

where * denotes complex conjugate and $\Gamma \equiv \frac{qU_{\rm rf}}{4md_q^2\imath}$. The magnetron radius is assumed to be constant $[\kappa_-(t) = \kappa_{-,0}]$, since it is not resonantly driven at any sideband. The general solutions to these equations are

$$\zeta(t) = \zeta_1 e^{(t/\tau)} + \zeta_2 e^{-(t/\tau)}, \quad (6a)$$

$$\kappa_+(t) = \kappa_1 e^{(t/\tau)} + \kappa_2 e^{-(t/\tau)} \quad (6b)$$

with $\tau \equiv \sqrt{\omega_z(\omega_+ - \omega_-)/\Gamma\Gamma^*}$. The free coefficients are determined by the initial conditions $\zeta_0 \equiv \zeta(0) = \zeta_1 + \zeta_2$ and $\kappa_0 \equiv \kappa(0) = \kappa_1 + \kappa_2$. After a transient with typical length τ, the final phase of the axial motion will be given by the factor ζ_1 that can be expressed as

$$\zeta_1 = \frac{\zeta_0}{2} + \kappa_0^*\frac{\omega_+ - \omega_-}{2\tau\Gamma^*} = \frac{\zeta_0}{2} + \frac{\kappa_0^*}{2}\gamma e^{\imath\varphi_{\rm rf}}, \quad (7)$$

with $\gamma \equiv \sqrt{(\omega_+ - \omega_-)/\omega_z} \simeq \sqrt{\omega_+/\omega_z}$. The final axial phase will be determined entirely by the initial cyclotron phase and the phase of the coupling pulse if $|\kappa_0|\gamma \gg |\zeta_0|$. For small to intermediate cyclotron amplitudes, the readout phase is a mixture of the axial phase during the coupling pulse and the sought-after cyclotron phase. However, since the axial phase is statistical, this causes a technical jitter on the measured phase that depends on the ratio $\gamma|\frac{\kappa_0}{\zeta_0}|$ as illustrated in Fig. 3. We note that a large axial mode amplitude during detection, allowing an excellent SNR, does not imply any minimum cyclotron radius $|\kappa_0|$ during the phase evolution. Figure 3 demonstrates the almost complete elimination of all cyclotron energy-dependent errors. Allowing a maximum phase jitter of 30° that is sufficient for efficient phase unwrapping yields a cyclotron frequency measurement precision that corresponds to a final uncertainty of 1×10^{-12} within less than 100 s, while the systematic shift amounts to 6×10^{-12}, a value that can be easily corrected for. The lower limit for the necessary cyclotron energy does not originate from the finite detector temperature but from the ion temperature before excitation and could thus be decreased further by advanced cooling methods. Already the direct cooling via a cyclotron resonator that was not available for the measurements in this Letter would decrease the systematic shifts by a factor of about 40. By coupling the axial mode to the cyclotron mode via a radio frequency field and bringing the cyclotron mode into thermal equilibrium with a resonator attached to a split electrode, axial mode amplitudes of 50 mK and below have been demonstrated in our setup. Furthermore, the parametric squeezing techniques outlined in Ref. [16] can be used to further decrease the necessary cyclotron excitation amplitude by applying a squeezing pulse in phase with the cyclotron excitation pulse, decreasing the phase uncertainty at the cost of the amplitude uncertainty. Applying this scheme additionally allows for a further reduction of the systematic shifts by an order of magnitude for a highly charged ion in our trap. Ultimately, sympathetic laser cooling could be used to generate ion temperatures low enough to decrease the measurement time by another order of magnitude.

The necessary calibration is achieved by phase measurements with increasing waiting time. The slope of the unwrapped phase does not depend on fixed phase offsets arising from the unknown transfer function of the excitation lines and filters or delays during the acquisition. In Fig. 4, the reconstructed phase evolution is plotted. The residuals are well below 2° and demonstrate the successful unwrapping procedure and the negligible nonlinearities. The slope of the unwrapped phase represents the difference

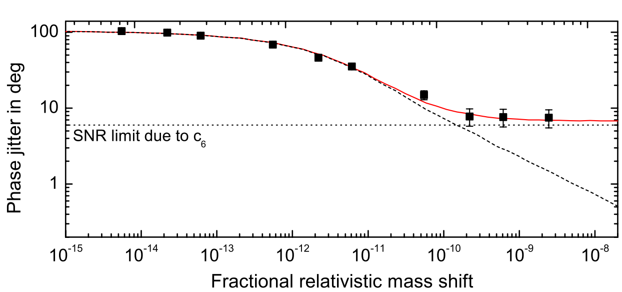

FIG. 3 (color online). Phase jitter due to finite thermal amplitudes at the beginning of the coupling process for a $^{28}Si^{13+}$ ion cooled to thermal equilibrium with the axial detector. Coupling to the cyclotron detector would further reduce the systematic shifts by a factor of about 40. The red curve includes the limitation in the phase resolution arising from the SNR achievable in our setup due to electrostatic anharmonicities.

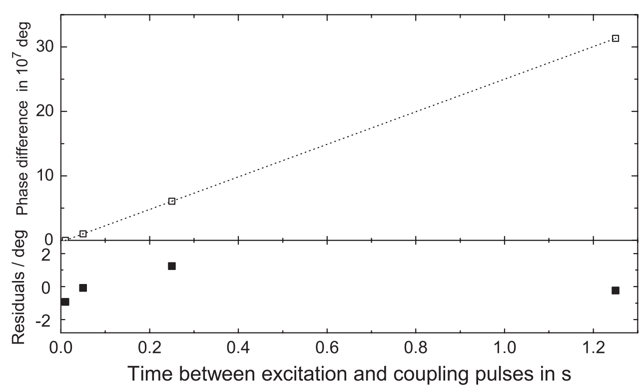

FIG. 4. Unwrapping of the integrated phase allows a frequency calibration. The slope of the unwrapped phase represents the difference of the modified cyclotron frequency and the coupling frequency $\nu_{\rm rf} \simeq \nu_+ + \nu_z$.

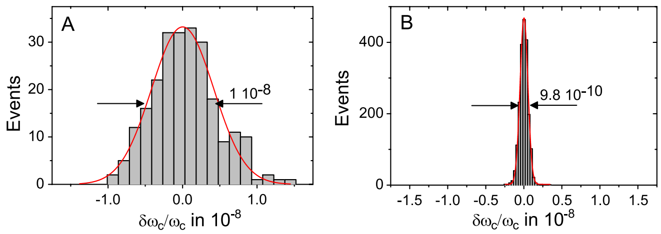

FIG. 5 (color online). Fractional difference of consecutive measurements of the modified cyclotron frequency measured with the conventional mode coupling technique (a) and with the new phase-sensitive method (b). The fluctuation is reduced by more than an order of magnitude. The measurement time for (a) was 60 s, while (b) needs only 5 s. Further shielding of external fluctuations [19] and simultaneous coherent measurements will allow longer integration times and even better stability.

between the coupling frequency ν_{rf} and the modified cyclotron frequency ν_+. This difference is very close to the axial frequency ν_z. The precision attained for the modified cyclotron frequency is thus given by the precision of the slope times a factor ν_z/ν_+. For precision measurements, it is not necessary to perform the phase unwrapping for each single measurement since the offset phases are constant to better than $1°$ per day. However, it can be advantageous to perform at least one additional measurement with a medium integration time in order to rule out phase jumps larger than 2π if the final integration time is chosen large compared to the magnetic field fluctuations. Since the stability of the offset phases is better than the typical root-mean-square phase measurement jitter, the precision of the measurement is not limited by the calibration procedure. Moreover, the precision of the mean cyclotron frequency determination can be increased virtually arbitrarily by increasing the phase evolution time. Measurements of the cyclotron frequency in our experimental setup [12] (Fig. 5) show significantly improved stability compared to conventional measurement techniques. This is partly due to the lower sensitivity to voltage fluctuations and to the better frequency determination performance. Furthermore, the necessary measurement time is reduced by more than 2 orders of magnitude for comparable precision.

The advantage of the lower cyclotron energy is of particular benefit for experiments on highly charged ions due to the large relativistic mass shift at high cyclotron frequencies. If other error sources such as magnetic field wander and cavity shifts can be reduced, all mode energy-dependent frequency shifts known to date will be reduced compared to the present state-of-the-art measurement precision. If, furthermore, the electrostatic field stability can be improved as it is planned for future experiments [17], all limitations known to date can be either excluded or well controlled. For the measurement of the g factor of the electron bound in highly charged ions [12], this technique will allow 10 ppt accuracy due to the negligible energy-dependent corrections. The limitation originating from the cavity interaction that creates systematic shifts can be overcome by increasing the trap radius due to its strong trap-size dependence. If, furthermore, magnetic field fluctuations can be suppressed by a simultaneous phase measurement in two adjacent traps as it is envisaged in the high-precision Penning trap mass spectrometer PENTATRAP experiment [18], mass comparisons at an unrivaled precision of $\frac{\delta m}{m} \leq 10^{-12}$ become feasible, setting the stage for intriguing measurements in the field of fundamental metrology.

This work is supported by the Max-Planck Society and the DFG under Contract No. BL981/2-1.

[1] K. Blaum, Phys. Rep. **425**, 1 (2006).
[2] L. S. Brown and G. Gabrielse, Rev. Mod. Phys. **58**, 233 (1986).
[3] J. Verdú, S. Djekić, S. Stahl, T. Valenzuela, M. Vogel, G. Werth, H.-J. Kluge, and W. Quint, Phys. Scr. **T112**, 68 (2004).
[4] J. Verdú, S. Djekić, H. Haffner, S. Stahl, T. Valenzuela, M. Vogel, G. Werth, T. Beier, H.-J. Kluge, and W. Quint, Phys. Rev. Lett. **92**, 093002 (2004).
[5] E. A. Cornell, R. M. Weisskoff, K. R. Boyce, and D. E. Pritchard, Phys. Rev. A **41**, 312 (1990).
[6] H. Cramér, *Mathematical Methods of Statistics* (Princeton University, Princeton, NJ, 1946).
[7] J. Wolf, Nucl. Instrum. Methods Phys. Res., Sect. A **623**, 442 (2010).
[8] S. Sturm, A. Wagner, B. Schabinger, J. Zatorski, Z. Harman, W. Quint, G. Werth, C. H. Keitel, and K. Blaum, Phys. Rev. Lett. **107**, 023002 (2011).
[9] K. Pachucki, A. Czarnecki, U. D. Jentschura, and V. A. Yerokhin, Phys. Rev. A **72**, 022108 (2005).
[10] T. Beier, H. Häffner, N. Hermanspahn, S. G. Karshenboim, H.-J. Kluge, W. Quint, S. Stahl, J. Verdú, and G. Werth, Phys. Rev. Lett. **88**, 011603 (2001).
[11] R. Bouchendira, P. Cladé, S. Guellati-Khélifa, F. Nez, and F. Biraben, Phys. Rev. Lett. **106**, 080801 (2011).
[12] S. Sturm, K. Blaum, B. Schabinger, A. Wagner, W. Quint, and G. Werth, J. Phys. B **43**, 074016 (2010).
[13] G. Gabrielse, Int. J. Mass Spectrom. **279**, 107 (2009).
[14] B. D'Urso, R. Van Handel, B. Odom, D. Hanneke, and G. Gabrielse, Phys. Rev. Lett. **94**, 113002 (2005).
[15] E. A. Cornell, R. M. Weisskoff, K. R. Boyce, R. W. Flanagan, Jr., G. P. Lafyatis, and D. E. Pritchard, Phys. Rev. Lett. **63**, 1674 (1989).
[16] V. Natarajan, F. DiFilippo, and D. E. Pritchard, Phys. Rev. Lett. **74**, 2855 (1995).
[17] D. B. Pinegar, K. Blaum, T. P. Biesiadzinski, S. L. Zafonte, and R. S. V. Dyck, Rev. Sci. Instrum. **80**, 064701 (2009).
[18] K. Blaum, Y. N. Novikov, and G. Werth, Contemp. Phys. **51**, 149 (2010).
[19] R. Van Dyck, Jr., D. Farnham, S. Zafonte, and P. Schwinberg, Rev. Sci. Instrum. **70**, 1665 (1999).