Phase-Imaging Ion-Cyclotron-Resonance Measurements for Short-Lived Nuclides

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A novel approach based on the projection of the Penning-trap ion motion onto a position-sensitive detector opens the door to very accurate mass measurements on the ppb level even for short-lived nuclides with half-lives well below a second. In addition to the accuracy boost, the new method provides a superior resolving power by which low-lying isomeric states with excitation energy on the 10-keV level can be easily separated from the ground state. A measurement of the mass difference of ^{130}Xe and ^{129}Xe has demonstrated the great potential of the new approach.

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Investigating atomic nuclei and exploring the borders of their existence, nuclear physics has pushed from the valley of stability to the still largely unexplored regions of rare and very short-lived nuclides close to the drip lines and in the region of superheavy elements [1]. One of the most basic properties and a sensitive indicator of nuclear structure is the atomic mass comprising all atomic and nuclear forces. The studies of more and more rare and shorter-lived nuclides require ever faster mass measurements.

At present, three complementary techniques are applied in precision mass measurements of short-lived nuclides: Schottky and isochronous mass spectrometry in storage rings [2], and Penning-trap mass spectrometry (PT-MS) [3,4]. Schottky mass spectrometry allows mass measurements of nuclides with half-lives down to several seconds with an uncertainty of a few tens keV. In contrast, isochronous mass spectrometry provides access to very short-lived nuclides with half-lives in the microsecond range [2]. However, its typical uncertainty of about 100 keV is not sufficient for detailed nuclear structure investigations. As soon as masses with uncertainties of only a few keV are demanded for nuclides with half-lives below a second, Penning-trap mass spectrometry becomes the method of choice.

The era of high-precision Penning-trap mass measurements on short-lived nuclides began with the time-of-flight ion-cyclotron-resonance (TOF-ICR) technique [5,6] introduced at the first Penning-trap mass spectrometer for short-lived nuclides ISOLTRAP [7]. For masses of nuclides with half-lives down to about a second TOF-ICR MS reaches a relative uncertainty of a few parts in 10^8 and provides a resolving power of up to 10^6.

However, it is difficult to achieve such a low uncertainty for nuclides with very short half-lives often faced in the "terra incognita" in the chart of nuclides. Thus, faster mass measurement techniques have been developed. The Ramsey TOF-ICR technique, suggested 20 years ago [8], has recently shown to provide a threefold gain in precision [9–11].

Nevertheless, for singly charged ions of medium-heavy nuclides with half-lives of about 100 ms the mass resolution does not exceed 1 MeV/c^2. This is often not even sufficient to separate nuclear isobars. Other approaches include the production of highly charged ions (see, e.g., Ref. [12]), which requires an additional step of ion-charge breeding, and the replacement of the quadrupolar by an octupolar excitation field [13,14], which increases the resolving power by up to a factor of ten, but does not seem to improve the precision as compared to the Ramsey TOF-ICR technique [15].

In this Letter we present a novel approach to mass measurements on very short-lived nuclides which is 25 times faster and provides a 40-fold increased resolving power compared to the Ramsey TOF-ICR technique. The new phase-imaging ion-cyclotron-resonance (PI-ICR) technique is based on the determination of the cyclotron frequency $\nu_c = qB/(2\pi m)$ (of ion of charge q and mass m in a magnetic field B) via the projection of the ion motion in the trap onto a high-resolution position-sensitive microchannel plate (MCP) detector. The use of such a detector to monitor the ion motion is the most novel aspect of this work. It was implemented at the SHIPTRAP Penning-trap mass spectrometer [16] and compared with the Ramsey TOF-ICR method by determining the mass difference between ^{130}Xe and ^{129}Xe. The mass difference of these stable xenon isotopes is known with a very low uncertainty (about 10 eV [17]) and thus provides a benchmark for the present measurement.

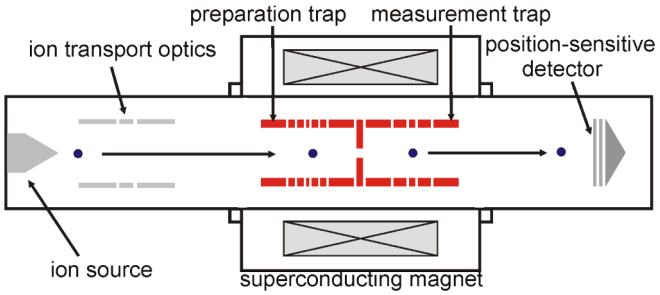

FIG. 1 (color online). Schematic of the setup used for the measurement of the mass difference between ^{130}Xe and ^{129}Xe.

A detailed description of the part of SHIPTRAP used for this measurement can be found, e.g., in Ref. [18]. The setup is schematically presented in Fig. 1. Singly charged ions of ^{129}Xe and ^{130}Xe produced with an electron-impact ion source were injected into the preparation trap, where the radial motions of the ions were cooled via mass-selective buffer-gas cooling [19]. After cooling, the ions were transferred to the measurement trap for the determination of ν_c. Similar to the TOF-ICR technique, the cyclotron frequency of the ion is determined as the sum of the magnetron ν_- and modified cyclotron ν_+ frequencies, i.e., $\nu_c = \nu_- + \nu_+$ [20,21].

ν_- and ν_+ are determined via independent measurements. Since the corresponding measurement schemes are similar, the subscripts $(-)$ and $(+)$ which relate various parameters to the magnetron and cyclotron motion, respectively, are used only when these motions must be distinguished. Otherwise, the subscripts are in the following omitted and both motions are called "radial motion." All quantitative examples are given for singly charged ions of mass 130u. The principle of the radial-motion-frequency measurement is presented in Fig. 2. The FWHM width of the spatial distribution $2\Delta R$ of ions after transfer to the measurement trap (position 1 in Fig. 2) is defined by the cooling of the radial motions in the preparation trap and is on the order of a few tens of micrometers. By applying a dipolar rf field with a certain initial phase at the radial-motion frequency the ion can be prepared on an average radius R (position 2 in Fig. 2). After free evolution for a time t, the ion radial motion accumulates a total phase of $\phi + 2\pi n = 2\pi\nu t$ (position 3 in Fig. 2). ϕ is the polar angle between position 2 and 3 of the ion, n is the number of the full revolutions the ion performs in the time t and ν is the frequency of the radial motion. Positions 1, 2, and 3 are called in this manuscript the center, the reference phase and the final phase of the radial motion, respectively.

The frequency of the radial motion and the uncertainty of its determination can be calculated according to (see Fig. 2)

$$\nu = \frac{\phi + 2\pi n}{2\pi t}; \qquad \delta\nu = \frac{\delta k}{2\pi t\sqrt{1-k^2}}, \tag{1}$$

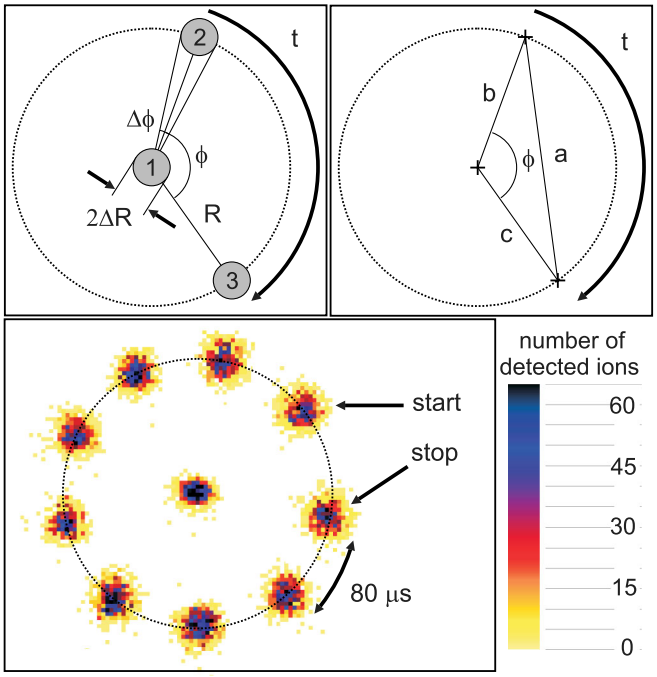

FIG. 2 (color online). Top: Principle of the measurement of the radial motions in a Penning trap (see text for details). Bottom: Experimentally recorded projection of the position of $^{130}Xe^+$ ions for several phases of the magnetron motion with radius of approximately 0.5 mm in the measurement trap of SHIPTRAP.

where $\phi = \arccos(k)$ for $0 \le \phi \le \pi$ and $\phi = 2\pi - \arccos(k)$ for $\pi < \phi < 2\pi$, and $k = (b^2 + c^2 - a^2)/(2bc)$. a, b, and c are the distances as defined in Fig. 2. Thus, the determination of the radial-motion frequency consists of the determination of the angle ϕ for a chosen phase accumulation time t. Although, k yields two values for the angle ϕ, in practice the correct value is easily inferred from the projection figure. A quick measurement of the radial-motion frequency with a moderate precision by the conventional TOF-ICR technique performed every few days is sufficient due to a high temporal stability of the magnetic field and trap electric potentials to determine the number of full revolutions n which the ion performs in the time t.

In order to measure the accumulated phase, the ion's radial motion is projected onto a position-sensitive detector (a delayline MCP detector) located on the axis of the measurement trap outside of the strong magnetic field, as described earlier [22]. The ideal projection preserves the value of ϕ.

In order to determine the angle ϕ, the coordinates of the center, reference phase, and final phase on the detector for each radial motion have to be measured. Since the coordinates of the center and the reference phase are constant in contrast to those of the final phase, it is sufficient to measure them just once a day. Thus, the determination of the cyclotron frequency ν_c reduces to the measurements of the final phases for the magnetron and cyclotron motions.

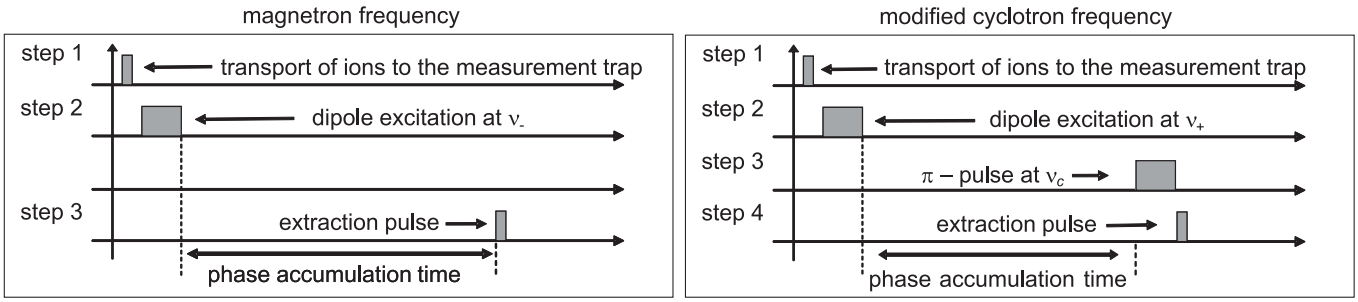

FIG. 3. Excitation pulse scheme for the measurement of the magnetron (left) and modified cyclotron (right) frequencies. For details see text.

Excitation-pulse schemes for the measurement of the magnetron and the modified cyclotron frequencies are presented in Fig. 3 (left) and Fig. 3 (right), respectively: First, an ion of interest is transferred from the preparation trap to the measurement trap (step 1) with a subsequent dipolar excitation of the radial motion (step 2). Then, the ion spends the time t in the measurement trap accumulating the phase $\phi + 2\pi n$. After that, the ion is ejected from the measurement trap and its radial position is projected onto the position-sensitive detector. An intermediate step is performed for the measurement of the modified cyclotron frequency ν_+. Before extraction the fast cyclotron motion is converted into the slow magnetron motion via a quadrupolar π pulse at the cyclotron frequency ν_c. A direct projection of the cyclotron motion would result in its substantial smearing on the detector due to the time-of-flight distribution of the ions between the measurement trap and detector. The measurement cycles for the magnetron and modified cyclotron frequencies are performed alternately many times until sufficient ion events are acquired in order to determine the ion's cyclotron frequency ν_c with the precision aimed at.

The new PI-ICR technique exhibits a substantial gain in precision and resolving power for the determination of an ion's cyclotron frequency as compared to the ToF-ICR method. In order to estimate the maximal gain in precision Eq. (1) can be simplified by assuming that $b = c$, $\Delta R_- = \Delta R_+ = \Delta R$, $R_- = R_+ = R$ and $a \ll R$. Then,

$$\delta\nu_c \approx \frac{1}{\pi t\sqrt{N}} \times \frac{\Delta R}{R}, \tag{2}$$

where N is the number of ion events acquired during the measurement of the magnetron and modified cyclotron frequencies. A comparison with a typical precision obtained at SHIPTRAP with the Ramsey TOF-ICR method under the same experimental conditions yields that the expected gain in precision with the PI-ICR technique is approximately a factor of five.

Since the difference of the magnetron frequencies of two ionic species is much smaller than that of the modified cyclotron frequencies, the resolving power of the technique is defined by the ability to resolve the phase difference accumulated for time t of the cyclotron motions of two species

$$\frac{\nu_c}{\Delta\nu_c} \approx \frac{\nu_+}{\Delta\nu_+} = \frac{\phi + 2\pi n}{\Delta\phi} = \frac{\pi\nu_+ t R_+}{\Delta R_+}. \tag{3}$$

For a phase-accumulation time t of 100 ms, a typical radius $R_+ = 1$ mm, a frequency $\nu_+ = 800$ kHz, and an estimated FWHM width of the spatial distribution of the cooled ions $2\Delta R_+ = 100$ μm, the resolving power is approximately 5×10^6, which exceeds that of the TOF-ICR technique $\nu_c/\Delta\nu_c = 1.6\nu_c t = 1.3 \times 10^5$ by a factor of 40 under similar conditions. Note that in contrast to the TOF-ICR method the resolving power of the new technique is not only given by t, but is also a function of the ratio $R_+/\Delta R_+$ and thus can be further increased either by using a larger radius or by reducing the radial spread. ΔR_+ can be decreased by cooling the buffer gas in the preparation trap, e.g., from room temperature to liquid nitrogen temperature as already demonstrated at TRIGATRAP [23].

The PI-ICR technique with the phase-accumulation time of 200 ms and the Ramsey TOF-ICR method with two-pulse pattern 10 ms-180 ms-10 ms were employed to determine the mass difference between Xe isotopes ^{130}Xe and ^{129}Xe by the measurement of the cyclotron-frequency ratio $r = \nu_c(^{129}\mathrm{Xe}^+)/\nu_c(^{130}\mathrm{Xe}^+)$ of singly charged ions of $^{129}\mathrm{Xe}^+$ and $^{130}\mathrm{Xe}^+$. ν_c of $^{130}\mathrm{Xe}^+$ and $^{129}\mathrm{Xe}^+$ were measured alternately with both methods. Each single measurement lasted about 5 min accumulating about 800 ions. During the measurement with the PI-ICR technique the center and the reference position of the ions on the detector were measured once before the experiment. Each single measurement of ν_c consisted of an alternating series of ν_- and ν_+ measurements (see description of the method above). The acquired number of ions were equally distributed between the magnetron and modified cyclotron frequency measurements. A set of projection figures belonging to the same measurement of ν_c are shown in Fig. 4 (left). The average x and y coordinates of the phase spots on the detector were determined by fitting a Gaussian to the $N(x)$ and $N(y)$ projections, respectively. On the right panel of Fig. 4 such projections are shown for the final phase of the modified-cyclotron-motion projection figure.

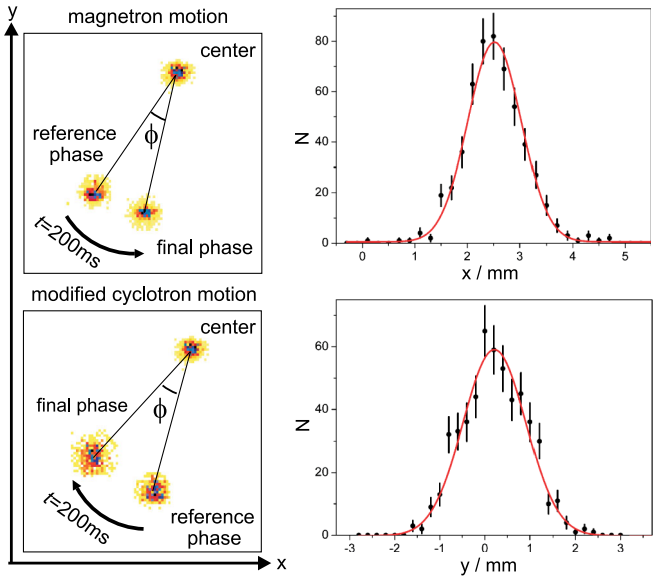

FIG. 4 (color online). (left-top) Magnetron and (left-bottom) modified-cyclotron projection figures of the ν_c measurement of $^{130}Xe^+$. (right) $N(x)$, and $N(y)$ projections of the final phase of the cyclotron motion. The phases were fitted with a Gaussian.

From three consecutively measured cyclotron frequencies at times t_1, t_2, and t_3, a ratio r obtained at time t_2 can be linearly interpolated from the frequencies measured at t_1 and t_3 to time t_2. The final ratio is the weighted mean of 16 such single frequency ratios (Fig. 5). The total measurement time with each method did not exceed three hours. The nonlinear drift of the magnetic field between two successive frequency measurements was negligible

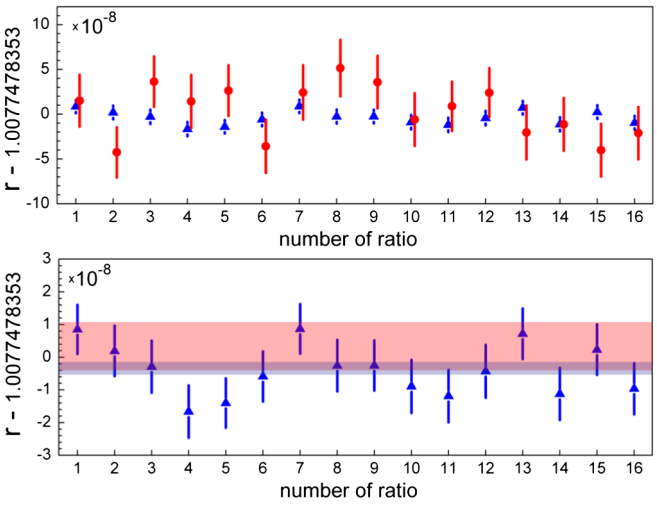

FIG. 5 (color online). Cyclotron-frequency ratios r of $^{129}Xe^+$ to $^{130}Xe^+$ measured in this work with the PI-ICR (blue triangles) and Ramsey TOF-ICR (red circles) techniques, respectively. The blue and red shaded bands are the uncertainties of the final ratios determined with the PI-ICR and Ramsey TOF-ICR method, respectively. The lower panel is a zoom of the upper panel.

[24] and thus was not taken into account. Systematic errors associated with the projection of the ion's motion onto the detector were negligible on the level of the obtained uncertainty.

The final frequency ratios obtained with the PI-ICR and Ramsey ToF-ICR techniques are 1.007 747 831 6(20) and 1.007 747 838 9(73), respectively.

Using the frequency ratios, the mass difference $\Delta M_{\mathrm{SHIPTRAP}} = M(^{130}Xe^+) - M(^{129}Xe^+)$ was calculated and compared with the mass difference ΔM_{FSU} determined by the FSU group in a cryogenic Penning trap with highly charged ions resulting in $\Delta M_{\mathrm{FSU}} - \Delta M_{\mathrm{SHIPTRAP}}$ 180(240) eV for the PI-ICR and −690(880 eV) for the Ramsey TOF-ICR technique. Compared to the measurement with the Ramsey ToF-ICR technique, the new method provided a factor of almost four gain in precision thus reducing the measurement time needed to obtain the same uncertainty as in the Ramsey method by more than an order of magnitude.

In conclusion, a novel technique based on the image detection with a position-sensitive detector is presented for the first time for measurements of the cyclotron frequency in a Penning trap. The technique can be employed in Penning traps for mass measurements of very short lived nuclides or for high-precision measurements on stable nuclides. Compared to the presently used TOF-ICR methods, the new technique offers a 40-fold increase in the resolving power and fivefold gain for the precision of the cyclotron-frequency determination.

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