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Octupolar excitation of ions stored in a Penning trap mass spectrometer—A study performed at SHIPTRAP

S. Eliseev[a,b,*], M. Block[a], A. Chaudhuri[a,c], F. Herfurth[a], H.-J. Kluge[a,d], A. Martin[a], C. Rauth[a], G. Vorobjev[a,b]

[a] *Gesellschaft für Schwerionenforschung, Planckstrasse 1, 64291 Darmstadt, Germany*
[b] *St. Petersburg Nuclear Physics Institute, Orlova Roscha, 188300 Gatchina, Russia*
[c] *Institut für Physik, Ernst-Moritz-Arndt-Universität, Domstrasse 10a, 17489 Greifswald, Germany*
[d] *Physikalisches Institut, Universität Heidelberg, Philosophenweg 12, 69120 Heidelberg, Germany*

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Abstract

The response of ions stored in a Penning trap to excitation by an octupolar radio frequency field was investigated using the time-of-flight ion-cyclotron resonance method at the SHIPTRAP Penning trap mass spectrometer at GSI. The mass resolving power can be improved at least by an order of magnitude and the accuracy more than doubled as compared to the conventional quadrupolar excitation. It has been shown that the octupolar excitation can be used to determine the mean magnetron radius of the stored ions as well as the radius of their initial spatial distribution in the Penning trap. Along with the experimental investigation, the octupolar excitation in a cylindrical Penning trap was simulated. Excellent agreement between experiment and simulation is observed.

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Keywords: Penning trap; Ion motion; Cyclotron frequency; Octupolar excitation; Resolving power

1. Introduction

Nowadays, the most accurate mass values for stable [1–5] and radioactive isotopes [6–12] are obtained with Penning trap mass spectrometers [6,13,14]. The time-of-flight ion-cyclotron resonance (TOF-ICR) method [15] is employed until now for accurate mass measurements of radionuclides. In the conventional TOF-ICR method the excitation of the ion's motion is accomplished by applying a radio frequency (RF) quadrupolar field at the true cyclotron frequency $\nu_c = (1/2\pi)(q/m)B$ for ions with a charge-to-mass ratio q/m in a magnetic field of strength B [16,17]. The achievable resolving power is proportional to the RF excitation time and the cyclotron frequency. For mass measurements of stable nuclides a higher resolving power and, thus, a higher accuracy are achieved by a longer excitation time [1–5], by use of superconducting magnets with higher magnetic fields [9], or by increasing the charge state [4,18,19]. In the case of short-lived nuclides, the half-life of the nuclide under investigation imposes a boundary condition on the excitation time and, therefore, limits the achievable resolving power. A possible way to overcome this constraint is to increase the resolving power for a given excitation time by the implementation of an azimuthal RF excitation at higher harmonics, i.e., at multiples of ν_c, in particular, the octupolar excitation $\nu_{\mathrm{octu}} = 2\nu_c = (1/\pi)(q/m)B$ instead of the quadrupolar one. First systematic investigation of the potential of octupolar excitation was performed at the LEBIT at NSCL [20].

This paper presents the results of an experimental investigation of the octupolar excitation of Cs ions by use of the TOF-ICR method at the SHIPTRAP Penning trap mass spectrometer [10,21] installed at the SHIP velocity filter at GSI [22]. A similar investigation by Ringle et al. at the LEBIT Penning trap mass spectrometer is published in the same issue of this journal [23].

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[*] Corresponding author at: Gesellschaft für Schwerionenforschung, Planckstrasse 1, 64291 Darmstadt, Germany. Tel.: +49 6159 71 2114; fax: +49 6159 71 2901.
E-mail address: s.eliseev@gsi.de (S. Eliseev).

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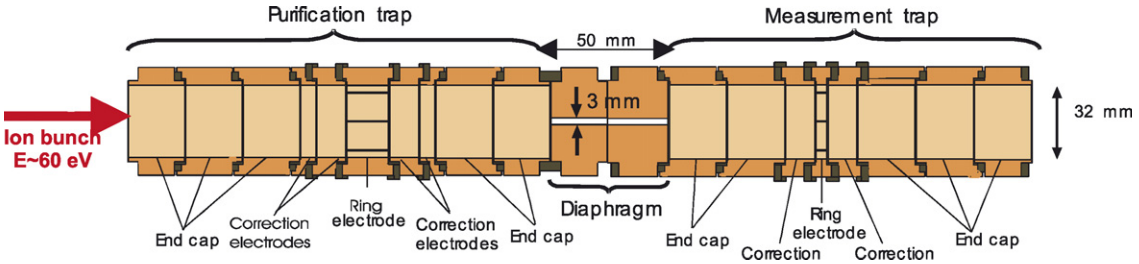

Fig. 1. The SHIPTRAP Penning trap system. The purification trap, a seven-electrode cylindrical trap, and the measurement trap, an orthogonalized five-electrode trap, are separated by a diffusion barrier that is 50 mm long and has an inner diameter of 3 mm. The ring electrodes of the purification trap and the measurement trap are segmented into eight equal segments as indicated in the figure.

2. Experimental

In order to implement an octupolar excitation scheme, a Penning trap is required with a ring electrode that is split into eight equal segments in the radial plane. Although there is quite a number of Penning trap mass spectrometers worldwide, only a few of them possess such a ring electrode. In addition, as shown in this article, a small transversal emittance and a small energy spread of the ion bunch injected into the trap are prerequisites for a successful application of octupolar excitation. All these ingredients are available at the SHIPTRAP mass spectrometer [10,21], which make it an excellent tool for investigation of octupolar excitation.

The SHIPTRAP Penning trap mass spectrometer consists of two cylindrical traps: the purification trap and the measurement trap. In the purification trap mass-selective buffer gas cooling [24] is implemented in order to separate isobars. In the measurement trap the mass to charge ratio m/q of stored ions is determined by measuring their true cyclotron frequency using the TOF-ICR method. The schematic layout of the traps is shown in Fig. 1.

The investigation of the octupolar excitation was performed with ions of the stable isotope ^{133}Cs, which were produced with a surface ion source. Before injection into the measurement trap, the ^{133}Cs ions were cooled and centered in the purification trap applying a mass-selective buffer gas cooling technique [24] to bring them into thermal equilibrium with the buffer gas. In the measurement trap the magnetron motion was excited for 10 ms at a frequency of 1300 Hz. The amplitude of the magnetron excitation was varied from 0 to 140 mV (which represents the typical range used at SHIPTRAP). This brings the ions to a mean magnetron radius R_m. Then, a conversion of the magnetron motion into the reduced cyclotron motion was observed for various amplitudes of the octupolar excitation at twice the cyclotron frequency (octupolar conversion curve). After that, the frequency of the octupolar excitation was varied for a fixed amplitude of the octupolar excitation (octupolar resonance curve).

Along with the experimental investigation of the octupolar excitation a detailed simulation of the total cycle including the magnetron as well as the octupolar excitation was performed using SIMION 7.0 [25]. For this the following initial conditions for the ions in the measurement trap were assumed:

(1) The ions with mass number 133 are singly charged.
(2) The initial position of the ions is random within a sphere of radius ΔR with its origin at the center of the trap ($R_m = 0$).
    ΔR is called the radius of the initial spatial distribution of the ions and was varied in the simulation.
(3) The initial kinetic energy of the ions is random within a range of 0–0.1 eV, which is close to thermal distribution at room temperature.
(4) The initial direction of the ion's motion is random.
(5) The initial phase of the magnetron excitation is fixed. The initial phase of the octupolar excitation is random.

Starting with these assumptions the following sequence was repeated for different parameters as, for instance, different initial spatial distributions or RF amplitudes A_m, but with a RF excitation time fixed to $T_m = 1$ ms:

(1) A dipolar radio frequency at the magnetron frequency was applied with various amplitudes of the radio frequency. By this, the ions were brought to a magnetron orbit of radius $R_m \pm \Delta R$.
(2) The RF amplitude of an octupolar excitation A_{octu} was scanned with the frequency set to twice the cyclotron frequency.
(3) The frequency of an octupolar excitation was scanned for different RF amplitudes A_{octu}.

The magnetron (T_m) and octupolar excitation (T_{octu}) times were chosen in the simulation to be much shorter than in the experiment in order to save CPU time. In view of that, the simulated data were adjusted to the experimental excitation times. The total kinetic energy K of the ions after excitation was the observed parameter in the simulations. Since the experimental observable is the time-of-flight T_{TOF} of the ions from the trap to the detector, the kinetic energies obtained in the simulations were converted to time-of-flights according to the formula $T_{TOF} = a - b\sqrt{K}$, where a and b are parameters that have been found from the experimental data.

2.1. Determination of the mean magnetron radius and the radius of the initial spatial distribution of ions

The response of ions to the octupolar excitation is strongly correlated to their initial state in the trap. In particular,

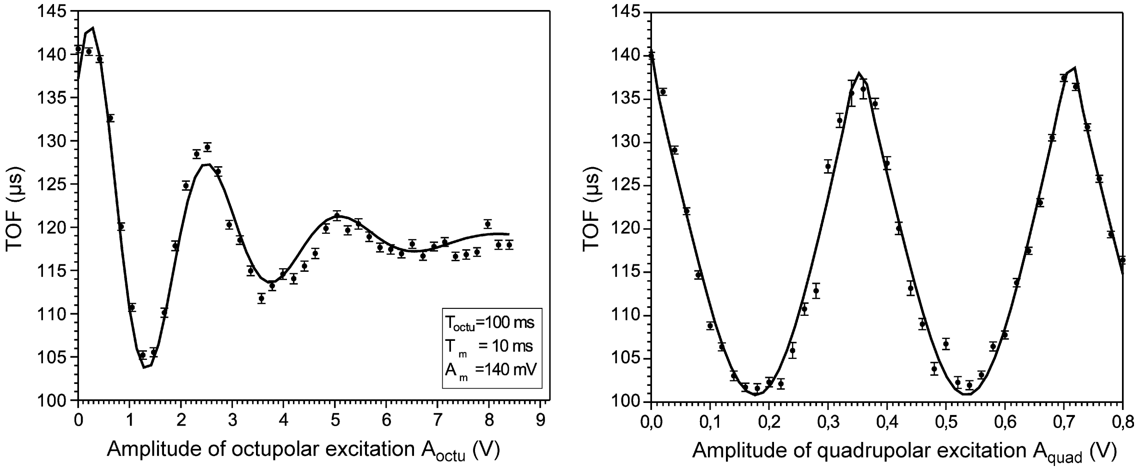

Fig. 2. Measured mean time-of-flight of the Cs ions ejected from the measurement trap as a function of the amplitude A_{octu} of the octupolar radio frequency field at frequency $\nu_{\mathrm{RF}} = \nu_{\mathrm{octu}} = 2\nu_{\mathrm{c}}$ (octupolar conversion curve) (left picture) and as a function of the amplitude A_{quad} of the quadrupolar RF field at frequency $\nu_{\mathrm{RF}} = \nu_{\mathrm{c}}$ (quadrupolar conversion curve) (right picture).

information on the mean radius R_{m} of the ion's magnetron motion and the radius of the initial spatial distribution of ions ΔR can be obtained from the conversion curve of the ion's magnetron motion into their cyclotron motion performed at twice the true cyclotron frequency (octupolar conversion curve). Octupolar conversion curves were measured for an octupolar excitation time of $T_{\mathrm{octu}} = 100$ ms and various magnetron excitation amplitudes A_{m} between 0 and 140 mV. The octupolar conversion curve for an amplitude of the magnetron excitation of $A_{\mathrm{m}} = 140$ mV is shown in Fig. 2. For comparison, a conventional quadrupolar conversion curve for the same magnetron excitation is presented.

The octupolar conversion curve can be approximated by a damped sinusoid (a purely phenomenological conclusion drawn from the view of the experimental conversion curve):

$$T_{\mathrm{TOF}} = a\,\mathrm{e}^{-DA_{\mathrm{octu}}} \sin(bA_{\mathrm{octu}} + \varphi) + c \qquad (1)$$

with the parameters a, b, c, the damping constant D and the initial phase φ. All these parameters are constants.

The first minimum corresponds to the RF amplitude $A_{\mathrm{RF}} = A_{\mathrm{octu}}^{\mathrm{full}}$ required for one full conversion (equal to 1.4 V in Fig. 2) of the magnetron motion of the ions into reduced cyclotron motion. The magnitude of the RF amplitude for a full conversion depends on the mean radius R_{m} of the ion's magnetron motion. R_{m} is proportional to the RF amplitude A_{m} of the magnetron excitation (at a fixed period of time for magnetron excitation T_{m}). Thus, from the amplitude for a full conversion the mean radius of the magnetron motion R_{m} can be determined. In Fig. 3 the measured relation is plotted between the RF amplitude required for a full conversion $A_{\mathrm{octu}}^{\mathrm{full}}$ and the RF amplitude of a magnetron excitation A_{m} applied prior to octupolar excitation.

It can be seen, firstly, that a higher RF amplitude of the magnetron excitation A_{m} results in a lower RF amplitude for a full conversion $A_{\mathrm{octu}}^{\mathrm{full}}$ and, secondly, that the inverse of $A_{\mathrm{octu}}^{\mathrm{full}}$ is proportional to T_{m}^2.

Ion optical simulations established the relation between the RF amplitude of the magnetron excitation A_{m} (measured in V) and the mean radius of the magnetron motion R_{m} (measured in mm) as

$$R_{\mathrm{m}}\,(\mathrm{mm}) \approx 15.7 A_{\mathrm{m}}\,(\mathrm{V}) \qquad (2)$$

Thus, the mean radius of the magnetron motion of the ions that corresponds to $A_{\mathrm{m}} = 140$ mV (Fig. 2) is approximately $R_{\mathrm{m}} = 2.2$ mm.

From the value of the damping constant D in (1) the radius of the initial spatial distribution of the ions can be estimated. This is achieved by simulating octupolar conversion curves for different radii ΔR of the initial spatial distribution of the ions and comparing those with experimental ones (Fig. 4). For this, a series of octupolar conversion curves was simulated for different ΔR (Fig. 4) and R_{m} (Fig. 5). Fig. 4 shows octupolar conversion curves for $R_{\mathrm{m}} = 2.2$ mm and different ΔR between 0 and 0.8 mm. With the choice of $\Delta R = 0.3$ mm the simulated curve reproduces the experimental one very well, fixing the radius of the initial spatial distribution of ions in the SHIPTRAP measurement trap

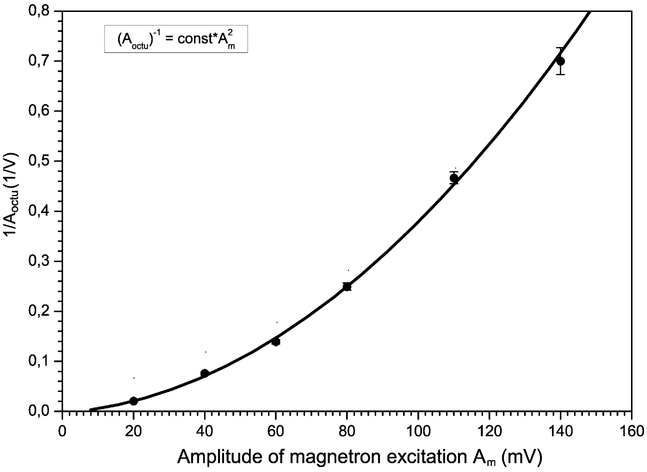

Fig. 3. Inverse of the experimentally determined radio frequency amplitude $(A_{\mathrm{octu}}^{\mathrm{full}})^{-1}$ required for a full conversion by octupolar excitation as a function of the amplitude of the magnetron excitation that is proportional to the magnetron radius. The solid line is a parabolic fit to the experimental points.

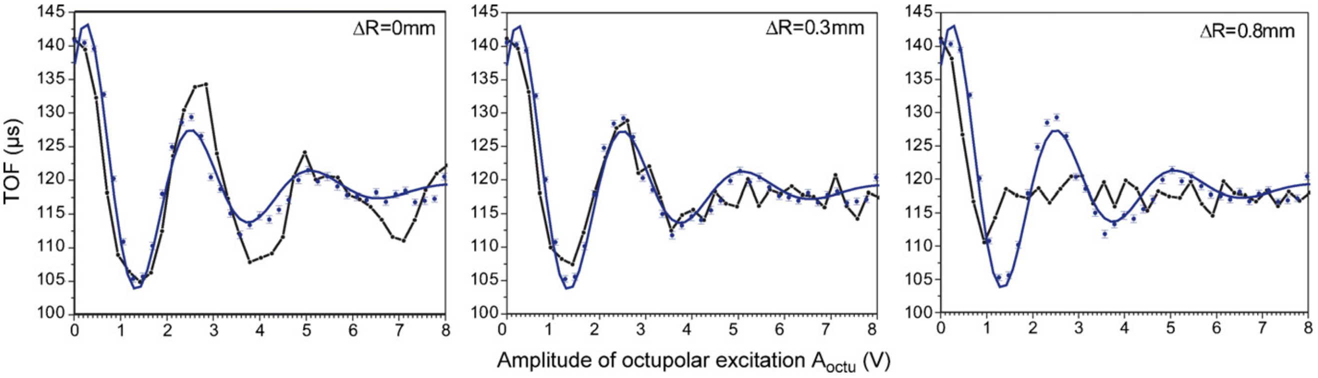

Fig. 4. Mean time-of-flight of ions ejected from the measurement trap as a function of the amplitude A_{octu} of the octupolar RF field at frequency $\nu_{\mathrm{RF}} = \nu_{\mathrm{octu}} = 2\nu_{\mathrm{c}}$ (octupolar conversion curve) for an excitation time $T_{\mathrm{octu}} = 100$ ms. The black line presents the simulation and the blue line shows the measurement. The mean radius of the magnetron motion obtained from the simulations is $R_{\mathrm{m}} = 2.2$ mm that corresponds to an excitation time $T_{\mathrm{m}} = 10$ ms and an amplitude of $A_{\mathrm{m}} = 140$ mV. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of the article.)

to $\Delta R \approx 0.3$ mm. Fig. 5 presents a series of octupolar conversion curves for different R_{m} and $\Delta R = 0.3$ mm. As can be seen, simulation and experiment are in good agreement for $R_{\mathrm{m}} = 2.2$ mm.

It can be seen from Figs. 4 and 5 that the damping of the octupolar conversion curve increases with the ratio $\Delta R/R_{\mathrm{m}}$, resulting in a reduced TOF contrast. Thus, in the case of the octupolar excitation one should keep the ratio $\Delta R/R_{\mathrm{m}}$ as small as possible by increasing R_{m} to a still acceptable value and/or by decreasing ΔR. However, for large R_{m} the systematic uncertainties due to imperfections of the electrical trapping potential and/or due to inhomogeneities of the magnetic field reduce the precision of a mass measurement. Thus, a compromise between a large R_{m} and the desired accuracy has to be found. In the SHIPTRAP case ΔR is mostly defined by the thermal energy distribution of the buffer gas in the purification trap at room temperature. By implementing a cryogenic cooler Penning trap, the energy spread of the ions could be reduced, which would result in a smaller ΔR.

2.2. *Time-of-flight octupolar resonances*

In the conventional TOF-ICR method a conversion of the magnetron motion into reduced cyclotron motion is observed when the excitation frequency of an azimuthal quadrupolar RF field matches the ion's true cyclotron frequency. The line shape that is observed by scanning the frequency is described analytically by the theoretical line shape as given in Ref. [17]. In contrast, the response of the ions and the line shape of the resonance when an octupolar RF field is applied cannot be analytically derived and has to be calculated numerically. The line shape defines the resolving power and, hence, the achievable accuracy of the cyclotron frequency determination.

Time-of-flight octupolar resonances were measured for different excitation amplitudes A_{octu}. The excitation time was set to 100 ms and the amplitude of the magnetron excitation was equal to 140 mV ($R_{\mathrm{m}} \approx 2.2$ mm). Fig. 6 shows a series of measured time-of-flight octupolar resonances together with simulated ones. The red solid curves represent the simulated time-of-flight octupolar resonances. The octupolar conversion curve that corresponds to this measurement is presented in Fig. 2.

The experimental time-of-flight octupolar resonances agree very well with the simulations. At octupolar excitation amplitudes that are much below the amplitude for a full conversion ($A_{\mathrm{octu}}^{\mathrm{full}} = 1.4$ V), the resonance curves are slightly asymmetric and resemble a Gaussian line shape. The resolving power is comparable with that of the quadrupolar excitation. With increasing amplitude A_{octu}, the TOF octupolar resonance changes in two ways: firstly, the base of the resonance broadens and, sec-

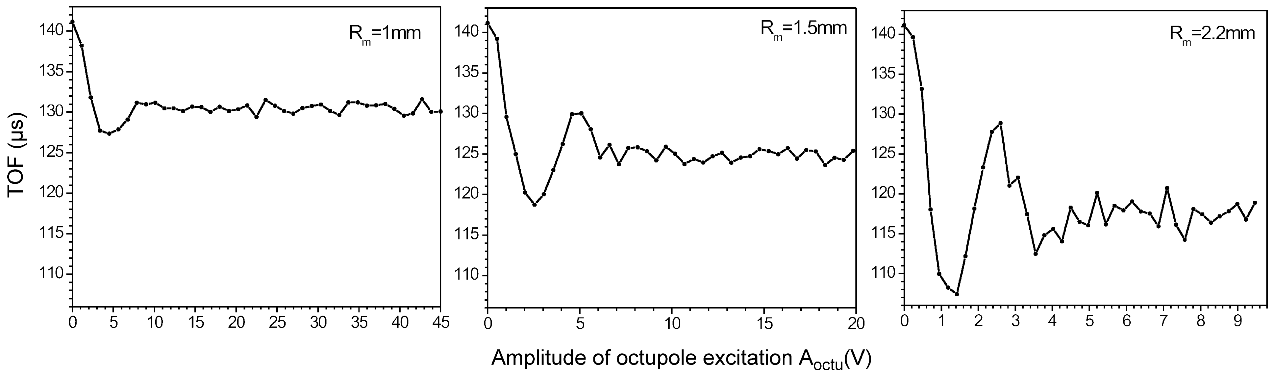

Fig. 5. Mean time-of-flight of ions ejected from the measurement trap as a function of the amplitude A_{octu} of the octupolar RF field at frequency $\nu_{\mathrm{RF}} = \nu_{\mathrm{octu}} = 2\nu_{\mathrm{c}}$ (octupolar conversion curve) for an excitation time $T_{\mathrm{octu}} = 100$ ms for different magnetron radii R_{m}. The radius of the initial spatial distribution of the ions is $\Delta R = 0.3$ mm.

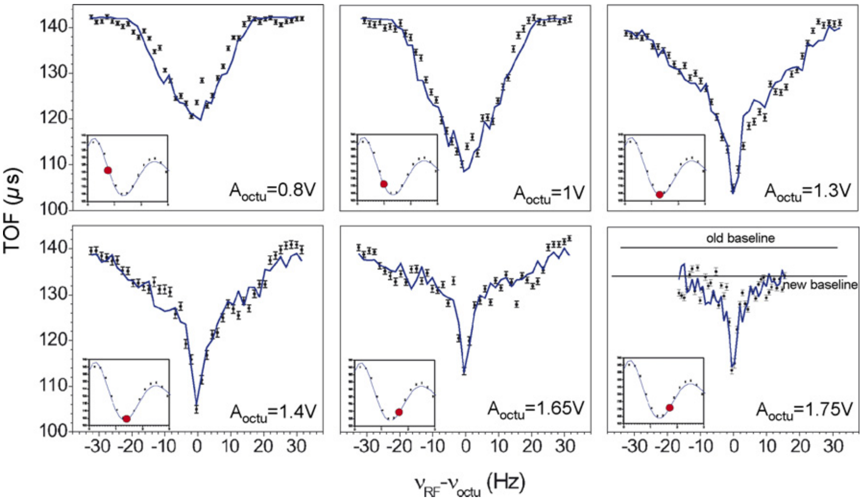

Fig. 6. Measured cyclotron resonances for Cs ions obtained from the mean time-of-flight plotted as a function of the frequency ν_{RF} of the octupolar RF field for an excitation time $T_{\mathrm{octu}} = 100$ ms and different amplitude A_{octu}. An excitation time $T_{\mathrm{m}} = 10$ ms and an amplitude $A_{\mathrm{m}} = 140$ mV were chosen for the magnetron excitation. The blue solid curves represent the simulated time-of-flight octupolar resonances. A full conversion from magnetron motion into cyclotron motion is obtained for $A_{\mathrm{octu}} = 1.4$ V (see Fig. 2). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of the article.)

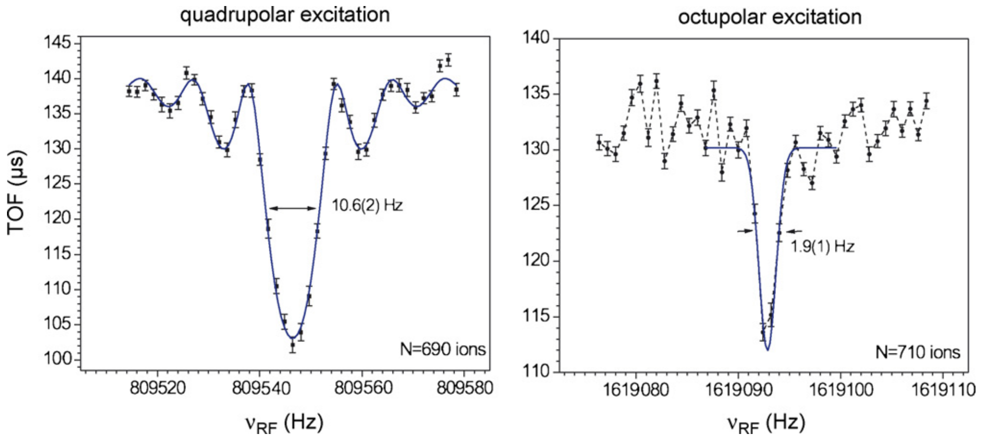

Fig. 7. Comparison of a measured time-of-flight quadrupolar resonance at the amplitude for a full conversion (left) and a time-of-flight resonance (right) for an octupolar excitation amplitude of $A_{\mathrm{octu}} = 1.75$ V, see Fig. 6. Quadrupolar and octupolar excitation times were the same and equal to 100 ms. An excitation time of $T_{\mathrm{m}} = 10$ ms and an amplitude of $A_{\mathrm{m}} = 140$ mV were used. The data points with error bars are fitted by the theoretical line shape [17] in the case of the quadrupolar resonance and by a Gaussian in the case of the octupolar one. The almost identical number of detected ions is indicated.

ondly, a narrow dip develops in the central part of the resonance structure.

Already at the voltage of a full conversion $A_{\mathrm{octu}}^{\mathrm{full}}$ this composite structure of the resonance is well pronounced. The TOF quadrupolar resonance (at the amplitude for full conversion) is compared in Fig. 7 with the TOF octupolar resonance ($A = 1.75$ V, see Fig. 6). In both cases, a similar number of ions were detected by the channel plate detector behind the measurement trap. The experimental TOF quadrupolar resonance was fitted by the theoretical line shape [17] while a Gaussian was used for the TOF octupolar resonance. The full widths at half maximum $\Delta\nu_{\mathrm{FWHM}}$, the central frequencies ν_{c} and ν_{octu}, and the resulting uncertainties $\delta\nu$ were determined for both resonances. The quadrupolar resonance yielded a width of $\Delta\nu_{\mathrm{FWHM}} = 10.6(2)$ Hz at the fundamental cyclotron frequency which allowed one to determine the center of the resonance with an uncertainty of $\delta\nu_{\mathrm{c}}/\nu_{\mathrm{c}} = 6 \times 10^{-8}$. The fit of the octupolar resonance yielded a five-fold narrower line width $\Delta\nu_{\mathrm{FWHM}} = 1.9(1)$ Hz at the second harmonic of the cyclotron resonance. The center of the resonance can be determined with an uncertainty of $\delta\nu_{\mathrm{octu}}/\nu_{\mathrm{octu}} = 2.5 \times 10^{-8}$, which is a factor of 2.5 more accurate than obtained by the quadrupolar excitation. Hence, a resolving power of $R = \nu_{\mathrm{octu}}/\delta\nu_{\mathrm{octu}} \approx 800{,}000$ could be achieved for an excitation time as short as $T_{\mathrm{octu}} = 100$ ms for $^{133}\mathrm{Cs}^{+}$, a medium-heavy singly charged ion.

The octupolar resonance presented in the right part of Fig. 7 contains only five data points. Therefore, a much higher increase in accuracy will be achieved by reducing the range of the RF scan in proportion to the reduced line width.

3. Summary

An experimental investigation has been performed at the SHIPTRAP Penning trap mass spectrometer in which the response of the ion's motion to the excitation by an azimuthal octupolar RF field in geometry was studied. It has been experimentally shown that the mass resolving power for an octupolar excitation with an excitation time of only $T_{\mathrm{octu}} = 100\,\mathrm{ms}$ for Cs ions is as high as $R = \nu_{\mathrm{octu}}/\delta\nu_{\mathrm{octu}} \approx 800{,}000$. This exceeds the resolving power of the TOF-ICR method based on a quadrupolar excitation by a factor of 10. For an excitation time of 900 ms (the standard excitation time at SHIPTRAP), a resolving power of $R \approx 7 \times 10^6$ can be expected which corresponds to a resolution in mass by 18 keV for Cs^+. Thus, the resolution of isomeric states with excitation energies of only a few 10 keV is now possible even for moderate observation times and hence short half-lives of the isotope under investigation.

In addition, the improved resolving power of the octupolar excitation results in a gain in accuracy by at least a factor of 2.5 with which the cyclotron frequency can be determined. An even higher gain is expected by reducing the range of the scan around the sharp resonance. Alternatively, octupolar excitation allows one to study still shorter-lived isotopes with the same accuracy as achieved by quadrupolar excitation.

It has been shown that octupolar excitation can be used to determine the mean radius of the magnetron motion as well as the initial spatial distribution of the ions confined in a Penning trap. The typical mean radius of the magnetron motion ranges from 1 to 2 mm. It has been shown experimentally that the ions occupy in the SHIPTRAP measurement trap initially a volume with a radius of $\Delta R \approx 0.3\,\mathrm{mm}$.

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